Study of Irradiation Effects on Physical Properties of Some Magnetic Semiconductor Materials

A Thesis
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Abstract

The effect of γ-irradiation on the structure, magnetic and electrical properties of ferrite samples with chemical formula Mg$_x$Cu$_{0.5-x}$Zn$_{0.5}$Fe$_2$O$_4$ (where $x = 0.0$, 0.2 and 0.4) were studied. The samples were prepared by conventional ceramic method. X-ray diffraction parameters, magnetization, initial permeability, electrical resistivity and dielectric constant were measured for the investigated samples before and after irradiation. Both magnetization and initial permeability were measured on toroidal samples used as transformer cores. The initial permeability ($\mu_i$) was measured as a function of temperature at constant frequency of 10 kHz and then Curie temperatures ($T_C$) were determined. The DC resistivity ($\rho_{DC}$) and the dielectric constant are measured on tablet form samples using the two probes technique. The real and imaginary parts of dielectric constant $\varepsilon$ and $\varepsilon''$ were measured as a function of frequency within the range 45 Hz - 5 MHz before and after irradiation.

X-ray diffraction patterns (XRDPs) indicated the presence of a single spinel phase before and after irradiation for all investigated samples. The lattice parameter and porosity increased while the average crystallite size decreased after irradiation. Both of magnetization and initial permeability decreased as a result of irradiation. The frequency dependence of $\mu_i$ showed a resonance peak within the range 1-3 MHz for all samples. The irradiated samples showed a peak shift to the lower frequency for samples with $x = 0.0$ and 0.2 and to the higher frequency for that of $x = 0.4$. In general, it was noticed that irradiation caused decrease in Curie temperature with significant one from 195 °C to 165 °C.
Abstract

for the sample of $x = 0.0$. Moreover, the value of $\mu_i$ decreased due to irradiation for all investigated samples.

The value of resistivity at room temperature decreased after irradiation for both DC and ac applied electric fields. Temperature dependence of the DC resistivity curves, before and after irradiation, showed three regions with different activation energies. The conduction mechanism in the first region was attributed to impurity conduction one. It is also observed that the activation energy decreased after irradiation at every region. Both $\varepsilon$ and $\varepsilon''$ decreased with increasing frequency. After the irradiation process, it was found that the values of $\varepsilon$ and $\varepsilon''$ increased for all samples. These results were discussed in the view of $\gamma$-rays interaction with ferrite lattice.
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Chapter 1
Introduction

Magnetic semiconductor materials are referring to mixed metal oxides combined to ferric oxides. The electrical and magnetic properties of these materials cover a wide range of electronics industry and applications. The most famous type of these materials is ferrites, which are widely used in satellite control system, microwave communications, digital systems and power transformers. Ferrites are the main materials to produce multilayer chip inductors (MLCI) that are the most important in printed circuits board in mobiles and computing systems. Spinel ferrites have been investigated in recent years for their useful electrical and magnetic properties and applications in information storage systems, magnetic bulk cores, magnetic fluids, microwave absorbers and medical diagnostics. The literature investigated the preparation and characterization of spinel ferrites, M\(_{\text{Fe}_2\text{O}_4}\) (divalent metal M = Co, Mg, Mn, Zn, etc.) by using conventional ceramic techniques or by wet chemistry to form nanoparticles. Several parameters affect the structure and properties of ferrites. The metal ions substitution play an important role in lattice structure and affect the magnetic moments interactions which leads to wide variety in the electrical and magnetic characteristics.

1.1 Historical Overview

There are basically two types of ferrite: soft and hard. Naturally occurring magnetite is a weak 'hard' ferrite. 'Hard' ferrites possess magnetism, which is essentially permanent while 'Soft ferrite' does not retain significant magnetization. During the 1930's research on 'soft' ferrites continued, primarily in Japan and the Netherlands. However, it was not until 1945 that [Snoeck J. L. 1945] of the Phillips Research Laboratories in the Netherlands succeeded in producing a 'soft' ferrite for
commercial applications. Originally manufactured in a few select shapes and sizes, primarily for inductor and antenna applications, 'soft' ferrite has proliferated into countless sizes and shapes for a multitude of uses.

Ferrites are used predominately in three areas of electronics: low-level applications, power applications, and *Electro Magnetic Interference* (EMI) suppression. The breadth of application of ferrites in electronic circuitry continues to grow. The wide range of possible geometries, the continuing improvements in material characteristics and their relative cost-effectiveness make ferrite components the choice for both conventional and innovative applications. In 1947, studies were done on spinel ferrites by Snoeck and others at Phillips laboratories and led ferrites of strong magnetic properties, high electrical resistivity and low relaxation losses [Snoeck 1947]. At about the same time, in 1948, Neel introduced his famous theory on ferrimagnetism, which described the basics of "spin-spin interaction" phenomenon taking place in the magnetic sublattices in ferrites [Neel 1948].

In 1952, Hogan from Bell Labs developed the first non-reciprocal microwave devise at 9 GHz that was based on the Faraday rotation effect. Improvement of spinel ferrites were investigated by various cation substitutions to get suitable magnetic properties for different frequency ranges, power requirements, and phase shift applications [Hogan 1952]. Developments have been made on the magnetic characteristics of ferrite materials since the 1950s that have improved microwave device performances. These involve both compositional and processing modifications. New application of ferrite materials continue to be realized, such as in the cellular phone, medical, and automotive markets.

In 1956, Neel, Bertaut, Forrat, and Pauthenet discovered the garnet ferrites of three sublattices, and referred to as rare-earth iron
garnets. In addition, hexagonal ferrites were also developed during this time. They have three basic sub lattices combined in different numbers in a hexagonal structure.

The ferrite cores used for power transformers are working in the range of low frequencies (1 to 50 kHz usually) and are quite big in size, can be toroid, shell or C shape and are useful in all kind of switching electronic devices (especially power supplies from 1 watt to 100 watts maximum because usually powerful applications are out of range of ferritic single core and required grain oriented laminations cores). The ferrite cores used for signal have a range of applications from 1 kHz to many MHz perhaps also 300 and found the main application in electronics.

With the rapid development of mobile communication and information technology, small, inexpensive, high performance electronic devices are in high demand [Qi 2002]. Usually, Ni-Cu-Zn and Mg-Cu-Zn ferrites are employed as a magnetic material for (MLCI) because of its magnetic properties at high frequencies and its low sintering temperature [Atassi 2006], [Hak et al.1996]. In addition, Mg-Cu-Zn ferrites are considered promising materials are useful in the fabrication of cores of intermediate frequency transformers (IFT) for amplitude modulation [Parak 1997].

Recently, the synthesis of magnetic materials on the nanoscale has been a field of intense study, due to the novel mesoscopic properties shown by particles of quantum dimensions located in the transition region between atoms and bulk solids. Quantum size effects and the large surface area of magnetic nanoparticles dramatically change some of the magnetic properties and exhibit superparamagnetic phenomena and quantum tunneling of magnetization, because each particle can be
considered as single magnetic domain. [Mathew et al. 2006] reviewed the synthesis and magnetic structure characterization of spinel metastable nanoferrites by using conventional ceramic techniques or by wet chemistry to form nanoparticles.

1.2 Magnetic Materials

The magnetic properties of materials depend on the atomic spin ordering in the lattice structure. In general there are two types of materials; magnetic and nonmagnetic. The theory of magnetism is classified according to magnetic behavior of the materials to five categories [Chikazumi 1964]:

(i) Diamagnetism.
(ii) Paramagnetism.
(iii) Ferromagnetism.
(iv) Antiferromagnetism.
(v) Ferrimagnetism.

1.2.1 Diamagnetism

They are characterized by a small and negative magnetic susceptibility of the order $10^{-5}$ to $10^{-6}$ H/m. The origin of these magnetic materials is an orbital rotation of the electrons about the nuclei induced electromagnetically by the application of an external field. According to "Lenz law", the induced current produces a magnetic flux, which opposes any change in external field. The negative susceptibility means that the magnetization occurs in the opposite direction of the applied magnetic field. The diamagnetic atoms or ions have no resultant magnetic moment, as their orbits form completely filled shells. Since the electronic structure of materials is independent of temperature, thus the diamagnetic
susceptibility is also independent of temperature. Diamagnetic substances include inert gases (helium, argon... etc.), metals (bismuth, copper, zinc, gold, mercury, silver... etc.) and many other organic compounds (water, glass, marble... etc).

### 1.2.2 Paramagnetism

These materials contain atoms or ions whose spins are randomly oriented and isolated from their surrounding as shown in Figure (1.1-a). Upon application of an external magnetic field, the spins are oriented in the direction of the field producing a weak magnetization as shown in Figure (1.1-b). The susceptibility $\chi$ in this case is positive and small susceptibility ($10^{-2}$ - $10^{-5}$) H/m. At finite temperature, the spins take different orientations and the susceptibility obeys the Curie law. Paramagnetic substances include aluminum, potassium, manganese, rare-earth elements, alkaline earth materials etc.

![Diagram](image)

(a) The atomic spins of paramagnetic material at a finite temperature. (b) Magnetization change as a function of magnetic field.
1.2.3 Ferromagnetism

The atomic magnetic moments of a ferromagnetic material are aligned parallel to each other as shown in Figure (1.2). The parallel alignment of spins is attributed to the strong positive interaction between them. Susceptibility of ferromagnetic materials is within the range $10^3 - 10^6$ H/m. Ferromagnetic substance is not usually magnetized but exists in a demagnetization state. This is because the interior of the block is divided into many magnetic domains each of them is spontaneously magnetized ($M_{sp}$) and the directions of domains magnetizations are random.

By applying a magnetic field, the magnetization increases until it reaches its saturation value ($M_s$) as shown in Figure (1.3-a). Ferromagnetic substances normally include iron, cobalt, nickel and a number of alloys. Curie temperature $T_c$ is the temperature at which there is a change from ferromagnetic to paramagnetic state as shown in Figure (1.3-b).
1.2.4 Antiferromagnetism

They have weak magnetization and small positive susceptibility $\chi$. In antiferromagnetic materials, the atomic magnetic moments are distributed between two sublattices. The moments are equal in magnitude but aligned antiparallelled to each other. The net magnetic moment is zero as shown in Figure (1.4). The temperature dependence of antiferromagnetic materials is characterized by a kink in the susceptibility curve. This point is called Neel temperature as shown in Figure (1.5). The spin magnetic moments are antiparallel below Neel temperature while above this point, they are randomly oriented and the material becomes paramagnetic.
1.2.5 Ferrimagnetisms

In this case, the magnetic ions occupy two different kinds of lattice sites (A and B sites). The net magnetic moments of the two sites are oriented in opposite directions and are not equal in magnitude as shown in Figure (1.6). This may arise from the fact that the ions forming the ferrimagnetic materials have different moments, or the number of atoms having one spin direction is different from that having the opposite spin direction. Since such magnetization is produced without applying any external magnetic field, it is called spontaneous magnetization \( M_s \). By increasing the temperature, the spins are disturbed by thermal agitation, which is accompanied by a decrease in spontaneous magnetization and vanishes at Neel temperature as shown in Figure (1.7).
Fig. 1.6 Spin arrangement of ferrimagnetic crystal, A and B are the tetrahedral and octahedral sites respectively.

Fig. 1.7 M and 1/χ versus T curves in ferrimagnetic materials.

1.3 Chemical Composition and Crystal Structure of Ferrites

Mixed metal oxides with ferric oxides as the main components are known as ferrites. According to the ion concentration and the chemical formula, different types of ferrites are established. In these types of ferrites, the crystal structure, the distribution of the metal ions and the different interactions between these ions in ferrites determine the properties of this material.
1.3.1 Spinel ferrites

There are two general chemical formulae for ferrite compounds as follows:

(i) \( \text{M}^{2+}\text{Fe}^{3+}_2\text{O}_4 \) spinel ferrite

(ii) \( \text{Fe}^{2+}\text{Fe}^{3+}_2\text{O}_4 \) natural ferrite (magnetite)

where M is a divalent metal, such as Mn, Co, Ni, Cu, Mg, Zn or Cd. M may be one or mixture of ions having average valence = 2 producing various ferrites with different intrinsic magnetization and properties. Thus, a great variety in this chemical composition of oxides with spinel structure is possible. The lattice of ferrites has cubic shape of size 8.3 - 8.5 Å. The unit cell contains 8 molecules of \( \text{MFe}_2\text{O}_4 \) or 56 ions per unit cell (8 divalent metal, 32 oxygen and 16 trivalent iron ions) [Chikazumi (1) 1964]. The relatively large oxygen ions are packed quite close together in a face centered cubic arrangement, and the much smaller metal ions occupy the space between them as shown in Figure (1.8). The radius of the oxygen ion is about 1.32 Å, which is much larger than that of the metal ions (0.6-0.8 Å), so the oxygen ions in this lattice touch each other, and form a closed packed face centered cubic shape.

Fig. 1.8. Arrangement of metal and oxygen ions in spinel ferrite unit cell.
In the oxygen lattice the metal ions take interstitial positions and can be classified into two sites: Tetrahedral site (A-site): in this site each metal ion is surrounded by 4 oxygen ions as shown in Figure (1.9-a); and octahedral site (B-site) where each metal ion is surrounded by six oxygen ions as shown in Figure (1.9-b). The complete shape of the unit cell is shown in Figure (1.9-c) and the structure of two octants sharing an edge is illustrated in Figure (1.9-d).

Fig. 1.9. (a) Cation on the tetrahedral lattice site (b) Cation on the octahedral lattice site. (c) Eight octants of a unit cell of spinel ferrites. The locations of ions in four shaded and unshaded octants are identical. (d) Locations of the oxygen and metal ions in two octants of single unite cell of spinel ferrite.
On the basis of the cation distribution ferrite may be classified into three classes: The possible distribution of the metal ions can be represented by the general formula

\[ (M^{2+}_δ Fe^{3+}_{1-δ})[M^{2+}_{1-δ}Fe^{3+}_{1-δ}]O_4 \]  

(1.1)

where δ is degree of inversion. The ions inside the brackets ( ) are located in tetrahedral sites while those inside the brackets [ ] occupy the octahedral sites. According to this distribution, there are three categories of spinel ferrites:

(i) Normal spinel (δ = 1): the formula becomes \( (M^{2+})[Fe^{3+}_2]O_4 \) and the divalent metal ions are in tetrahedral sites. \( ZnFe_2O_4 \) and \( CdFe_2O_4 \) are examples for normal spinel ferrites.

(ii) Inverse spinel ferrite (δ = 0): the formula becomes \( (Fe^{3+})[M^{2+}Fe^{3+}_2]O_4 \). In this case, the divalent metal ions occupy completely the octahedral sites while the iron is equally divided between the tetrahedral and octahedral sites. \( NiFe_2O_4 \) and \( CoFe_2O_4 \) are examples of inverse spinel ferrites.

(iii) Intermediate ferrite in which the \( M \) and \( Fe^{3+} \) ions are distributed uniformly over the tetrahedral and octahedral sites. \( MnFe_2O_4 \) is an example for the intermediate ferrites.

The cation distribution in the lattice is affected by the ionic radius, the shell electromagnetic distribution, and electrostatic interaction.

### 1.3.2 Garnet ferrites

The chemical formula of this type of ferrites is \( 3M_2O_3.Fe_2O_3 \), where \( M \) is a rare earth element such as Sm, Eu, Gd, Dy, Ho, Er, Lu or Y. The advantage of garnet ferrites are their high electrical resistivity and
very low losses in the microwave region. Therefore, they are used in microwave applications.

### 1.3.3 Hexagonal ferrites

In this type of ferrites the most important groups, according to their chemical formula, are:

**M-type**: The general formula of this type is \((\text{MO})_6(\text{Fe}_2\text{O}_3)\), where \(\text{M}\) represents a large divalent ion such as \(\text{Ba}^{2+}\), \(\text{Sr}^{2+}\) or \(\text{Pb}^{2+}\) ions. This type has hexagonal crystal structure, which is composed of accumulation of several spinel layers, separated by single ionic layers consisting of \(\text{M}^{2+}\), \(\text{O}^2-\) and \(\text{Fe}^{3+}\). Such a type has large magnetocrysaline anisotropy and thus it is used as permanent magnet, for example Ba-ferrites.

**W-type**: This type has the general chemical formula \((\text{BaO})_2(\text{MO})_8(\text{Fe}_2\text{O}_3)\), where \(\text{M}\) denotes a divalent ion such as \(\text{Mn}^{2+}\), \(\text{Fe}^{2+}\), \(\text{Co}^{2+}\), \(\text{Ni}^{2+}\), \(\text{Zn}^{2+}\) or \(\text{Mg}^{2+}\).

**Y-type**: The general chemical formula of this type is \(2(\text{BaO})_2(\text{MO})_6(\text{Fe}_2\text{O}_3)\), where \(\text{M}\) denotes also a divalent ion.

**Z-type**: The general chemical formula of this type is \(2(\text{BaO})_2(\text{MO})_12(\text{Fe}_2\text{O}_3)\).

### 1.4 Theory of Magnetic Moments Interactions

#### 1.4.1 Super-exchange interaction

The superexchange or an indirect exchange interaction was suggested by Kramers. The main point of superexchange is the spins moments of the metal ions on the opposite sides at the p-orbit of the oxygen ion \([\text{Kramers 1934}]\).
By considering, a system composed of two metal ions \( M_1 \) and \( M_2 \) separated by an oxygen ion \( O \). The ground state of the oxygen should be doubly charged as \( O^{2-} \) has the electronic configuration \((2s^2 \ 2p^6)\). In this state there is no spin coupling with the metal ions. There is however a possibility of having one of the two electrons of the \( O^{2-} \) ion excited and transferred to the neighboring metal ion (say \( M_1 \)), in which the strong exchange interaction tends to direct the spin of the transferred electron in such direction that the ion has a maximum spin magnetic moment (Hund's rule). In other words, if the \( M_1 \) ion with transferred electron is less than five 3d electron, all the 3d electron spins tend to align themselves parallel to each other, while if it is more than five electrons, the transferred electron must have its spin pointing antiparallel to the resultant magnetic moment of the ion. At the same time, the unpaired electron left in the p orbit of the \( O^{2-} \) ion will be coupled with the other metal ion \( M_2 \), in which the transferred electron should interact with \( M_2 \) electron in the manner described above.

Since, according to Pauli principle, the two electrons, which were in the p orbit of the \( O^{2-} \), must have opposite spins, both metals should have antiparallel magnetic moments in order to fulfill Hund's rule for both ions. Such a superexchange interaction is expected to be stronger when \( M_2-O-M_2 \) lie along a straight line, because the p orbit of \( O^{2-} \) ions is stretched as shown in figure (1.10) so it can overlap the orbits of both metal ion in this configuration [Chikazumi\(^{(4)}\) 1997].

If, however, \( M_1-O-M_2 \) makes an angle, say 90° the superexchange is expected to be small. In spinels, an A-O-B angel is about 125°, while that of A-O-A is about 80° and that of B-O-B is 90°. Thus, superexchange interaction (negative interaction) should be greater
between A and B sites. The weakest interaction will be the A-A interaction while B-B interaction will be intermediate [Gorter 1954].

![Fig. 1.10 The p-orbit of the O\(^{2-}\) ion through which exchange interaction acts between the spins on magnetic ions M\(_1\) and M\(_2\)](image)

### 1.4.2 Double exchange interaction

The other type of interaction is called double-exchange reaction, which can be described by Figure (1.11). The transfer of an electron from Fe\(^{2+}\) ion to the nearby oxygen ions accompanied by the simultaneous transfer of an electron with parallel spin to the Fe\(^{3+}\), where the jumping electrons are shown in a circle. This process was proposed by Zener in 1951 for explaining the adjacent ions of parallel spins via neighboring oxygen ion [Zener 1951].
In the molecular field theory, the magnetic interactions between ions are considered. The most significant theories are the molecular field theory by Culity in 1972, or molecular field theory (Neel theory) [Culity 1972].

1.4.3 Temperature dependence of the susceptibility

According to the molecular field theory, one considers an atom and replaces its interaction with the reminder of the crystal by internal field, called molecular field. In ferrites there are several interactions between magnetic ions to be considered and they may be classified to A-A, B-B, A-B and B-A interactions, where A-A refer to the interaction between A ions with its neighbors also on A site. Similar definitions stand for the other terms. In Neel theory, it is assumed that the A-B and B-A interactions are identical and predominate over A-A and B-B interactions.
Let there be $N$ identical ions per unit volume with a fraction $\lambda$ located on A sites and fraction $\nu = (1 - \lambda)$ on B sites. Let $\mu_A$ be the average moment of A ion in the direction of the field.

The magnetization of the A substance is:

$$M_A = \lambda N \mu_A$$

Let

$$N \mu_A = M_a$$

Thus

$$M_A = \lambda M_a$$

Similarly, the magnetization of B substance is:

$$M_B = \nu M_b$$

The total magnetization is

$$M = M_A + M_B = \lambda M_a + \nu M_b$$

(1.2)

Weiss also assumed that the molecular field is proportional to the magnetization in the material and the constant of proportionality is $\lambda$ which is called molecular field coefficient.

The molecular field acting on sublattice A is:

$$H_{mA} = H_{AA} + H_{AB}$$

where $H_{AB}$ is the molecular field on A site cased by its neighbors in B site.

$$H_{mA} = \gamma_{AA} M_A + \gamma_{AB} M_B$$

where the molecular field coefficients $\gamma$ are to be regarded as positive quantities and the signs correspond to the assumption of a negative (antiparallel) interaction between A and B ions and a positive (parallel) interaction between A ions.

Similarly,

$$H_{mB} = \gamma_{BB} M_B + \gamma_{AB} M_A$$
The coefficients $\gamma_{AA}$ and $\gamma_{BB}$ are now unequal and could be expressed as function of $\gamma_{AB}$ as follows.

$$\alpha = \frac{\gamma_{AA}}{\gamma_{AB}} \text{ and } \beta = \frac{\gamma_{BB}}{\gamma_{AB}}$$

The molecular fields are then

$$H_{mA} = w (\lambda \alpha M_a - \nu M_b) \quad H_{mB} = w (\nu \beta M_b - \lambda M_a)$$

(1.3)

where ($w = \gamma_{AB}$) is positive coefficient and $\alpha$ and $\beta$ are factors represent the intensity of intersite of interactions relative to the intersite interactions. Assuming that ionic magnetic moment is $\mu_m$, the thermal equilibrium value of the magnetization of each sublattice is given by

$$M_a = 2N\mu_mL(\alpha)\left(\frac{\mu_mw(\alpha\lambda M_a - \nu M_b) + \mu_mH}{kT}\right)$$

$$M_b = 2N\mu_mL(\alpha)\left(\frac{\mu_mw(\beta\nu M_b - \lambda M_a) + \mu_mH}{kT}\right)$$

(1.4)

where $H$ is the intensity of the external field and $L (\alpha)$ is called the Langevin function. For paramagnetic state, one can use the approximation $L (\alpha) = 1/3$.

$$\frac{\partial M_a}{\partial H} = \frac{2N\mu_m^2}{3kT}\left[1 + w\left(\alpha\lambda \frac{\partial M_a}{\partial H} - \nu \frac{\partial M_b}{\partial H}\right)\right]$$

$$\frac{\partial M_b}{\partial H} = \frac{2N\mu_m^2}{3kT}\left[1 + w\left(\beta\nu \frac{\partial M_b}{\partial H} - \lambda \frac{\partial M_a}{\partial H}\right)\right]$$

(1.5)

Solving for $\partial M_a/\partial H$ and $\partial M_b/\partial H$ in the above two equations and substituting in the formula

$$\frac{\partial M}{\partial H} = \lambda \frac{\partial M_a}{\partial H} + \nu \frac{\partial M_b}{\partial H}$$

we have

$$\frac{\partial M}{\partial H} = \frac{T - Cw\lambda \nu (\alpha + \beta + 2)}{T^2 - Cw(\alpha\lambda + \beta \nu)T + C^2w^2\lambda \nu (\alpha \beta - 1)}$$

(1.6)

where

$$C = 2N\mu_m^2/3k$$

The latest equation may be written in simplified form
where $\chi$ is the magnetic susceptibility; $\chi = \frac{\partial M}{\partial H}$,

$$\frac{1}{\chi} = \frac{T}{C} + \frac{1}{\chi_0} - \frac{\sigma}{T - \Theta}$$ (1.7)

The relationship between $1/\chi$ and $T$ given in (1.7) is graphically shown in Fig. 1.12. At high temperatures, the third term on the right-hand side of (1.7) becomes negligibly small, so that the remaining terms are approximated by a straight line, which has slope $1/C$ and intersects the ordinate at $1/\chi$. When the temperature is lowered from well above the Curie point, the magnitude of the third term in (1.7) increases as the temperature approaches 0, so that the $1/\chi$~$T$ curve departs from the asymptotic line and finally drops to zero at the Curie point $\Theta_f$. The asymptotic Curie point in this case is given by $\Theta_a = -C/\chi_0$.

Fig. 1.12. The $1/\chi$ vs. $T$ curves of ferrimagnetic material.

1.5 Net Magnetic Moments of Ferrites
The maximum spin magnetic moment of each transition metal ion can be calculated by applying Hund's rule on the alignment of its 3d-electrons spins. Since Mn$^{2+}$, Fe$^{2+}$, Co$^{2+}$, Ni$^{2+}$, Cu$^{2+}$ and Zn$^{2+}$ have 5, 6, 7, 8, 9 and 10 electrons in the 3d-orbit, respectively, their corresponding net spin magnetic moment are 5, 4, 3, 2, 1 and zero Bohr magneton ($\mu_B$). where $\mu_B$ is given by

$$\mu_B = \frac{\mu_0 e \hbar}{2m} = 1.165 \times 10^{-29} \text{ (Wb.m)}$$

where $\mu_0$ is magnetic permeability of vacuum, $\hbar$ is Planck's constant divided by $2\pi$ and $m$, $e$ are the mass and charge of electron respectively. Figure (1.13) shows the net spin magnetic moments of various ferrites as function of the number of 3d-electrons of their respective metal ions. The solid lines show the theoretical magnetic moment, which expected, where the net magnetization is assumed due to the spin magnetic moment of the metal ions. The difference between the theoretical and experimental pointes is due to the following:

1. Slight deviation of the distribution of the metal ions from that of an ideal inverse spinel.
2. The contribution of the orbital moment (hatched area).
3. Angle formation by the ionic moment on the sites.
Fig. 1.13 Molecular absolute saturation moment of various ions.

The molecular magnetic moment of ferrite cannot exceed $5 \, \mu_B$ as long as it takes the inverse structure. It is expected that the molecular magnetic moments of a ferrite can be increased by adding some moment of normal spinel ferrite; for instance $\text{ZnFe}_2\text{O}_4$. As the $\text{Zn}^{2+}$ ions should occupy the A-sites, the same number of $\text{Fe}^{3+}$ ions will migrate from A-sites to B-sites. Thus, if a fraction $x$ of $\text{ZnO.Fe}_2\text{O}_3$ is added to an inverse ferrite, the resultant structure is expressed as

$$\left(\text{Fe}_{1-x}^{3+}\text{Zn}_x^{2+}\right)O_2 \cdot \left[\text{Fe}_{1+x}^{3+}\text{Me}_{1-x}^{2+}\right]O_3$$  \hspace{1cm} (1.9)
The net magnetization moment of this molecule is called as

\[ M = [5(1 + x) + n(1 - x) - 5(1 - x)]\mu_B = [n + (10 - n)x]\mu_B \]  \hspace{1cm} (1.10)

where \( n \) is the number of Bohr magnetons (\( \mu_B \)) of \( M^{2+} \) moment. Thus, \( M \) should increase from \( n\mu_B \) toward 10 \( \mu_B \) with addition of \( Zn^{2+} \) ions.

[Guillaud 1950] have actually found that this addition makes increase of the total magnetic moment. Figure (1.14) shows the dependence of the molecular magnetic moment of the various mixed \( Zn \) ferrites on their composition. There is tendency of over curve toward the moment 10 \( \mu_B \). The deviation of the actual magnetic from theoretical lines at higher \( Zn \) concentration is discussed by assuming a combination of parallel and antiparallel moments. Since pure \( Zn \) ferrite is known to be antiferromagnetic, it is expected that the B-B interaction will be antiferromagnetic even in mixed ferrimagnetic \( Zn \) ferrite. The effect of this interaction is usually masked by the strong A-B interaction, which cause the spins in B-site to aligned parallel to each other. However, if the A-B interaction is weakened by say, the interaction of non-magnetic \( Zn \) ions into A-sites. Then the tendency toward antiferromagnetic arrangement in the B-sits will be increased.
1.6 Structure Analysis-using XRD

Characterization of ferrite crystals by x-ray diffraction (XRD) gives much information about the crystal structure and the crystallite size. As mentioned above, each kind of ferrite has chemical formula that determines its crystal structure. To ensure that the composition is formed, x-ray diffraction is used. The interplaner spacing is donated by $d_{hkl}$, where $(hkl)$ is Miller index of the scattering plane. According to Bragg’s equation of diffraction

$$2d_{hkl} \sin \theta = n \lambda$$  \hspace{1cm} (1.11)
where $d_{hkl}$ is the interplaner space, $\theta$ is the diffraction angle and $\lambda$ is the x-ray wavelength, $n$ is the diffraction order. The diffraction of the scattered x-rays leads to formation of characteristics spectrum of every ferrite structure. Figure (1.15) shows an example of x-ray for Mg-Cu-Zn ferrite [Haque et al. 2008]. From XRD spectrum, lattice parameter can be determined from the following relation

$$d_{exp} = d_{hkl}(h^2 + k^2 + l^2)^{\frac{1}{2}}$$  

(1.12)

![XRD pattern of Mg-Cu-Zn ferrites](image)

Fig. 1.15 XRD pattern of Mg-Cu-Zn ferrites [Haque et al. 2008].

The porosity of the samples is defined by the percentage of the apparent density to the x-ray density and is given by

$$\rho_{app} = \frac{\rho_{app}}{\rho_{xray}}$$

(1.13)
where $d_{\text{app}}$ and $d_x$ are the apparent and x-ray densities, respectively. The apparent density $d_{\text{app}}$ of the samples is determined by applying Archimedes principle. The x-ray density is determined using the following relation:

$$d_x = \frac{8M}{N_A a^3} \quad (1.14)$$

where, $M$ is the molecular weight, $N_A$ is Avogadro’s number and $a$ is the lattice parameter [Cullity B.D. 1959].

### 1.7 Magnetic Parameters of Ferrites

#### 1.7.1 Magnetization

It is defined as the dipole moment per unit volume. The magnetization ($M$) depends on the magnitude of the atomic magnetic moment $\mu_m$, and the number of atoms per unit volume $n$ and is given by the following formula [Chikazumi 1997].

$$M = n \mu_m \quad (1.15)$$

#### 1.7.2 Permeability and susceptibility

In general, the permeability ($\mu$) is a function of the induced magnetic field of the magnetic materials. In order to characterize the properties of a given magnetic material it is necessary to measure $B$ (induced field) as a function of $H$ (applied field) over a continuous range
of H to obtain the Hysteresis curve. From electromagnetic theory, the relation between B and H is given by the formula:

\[ B = M + \mu_0 H \, (\text{Web/m}^2) \]  

(1.16)

where \( \mu_0 \) is the magnetic permeability of vacuum, since

\[ M = \chi H \]  

(1.17)

where \( \chi \) is the magnetic susceptibility (H/m). Thus from Eq. (1.16) and Eq. (1.17) we get

\[ B = (\chi + \mu_0) H = \mu H \]  

(1.18)

where \( \mu \) is the magnetic permeability of the sample.

1.7.3 Magnetic loss

If the magnetic martial is magnetized by the oscillating magnetic field \( H = H_0 e^{i\omega t} \), where \( H_0 \) is amplitude of the applied field, the induced
magnetic flux density $B$ is generally delayed by the phase angle $\delta$ and is thus expressed as $B = B_0 e^{i(\omega t - \delta)}$. Then the permeability is given by

$$\mu = \frac{B}{H} = \frac{B_0 e^{i(\omega t - \delta)}}{H_0 e^{i\omega t}} = \frac{B_0}{H_0} e^{-i\delta} = \mu' - i\mu''$$

(1.19)

where, $\mu' = \frac{B_0}{H_0} \cos \delta$ and $\mu'' = \frac{B_0}{H_0} \sin \delta$ are the complex terms of permeability in AC fields. $\omega = 2\pi f$ is angular frequency, $\mu'$ expresses the real component of $B$ which is in phase with $H$, so it corresponds to the normal permeability. If there are no losses, we should have $\mu = \mu'$. The term $\mu''$ expresses the imaginary component of $B$ which is delayed by the phase angle $\delta$ from $H$. From equation (1.18) the ratio between $\mu''$ and $\mu'$ becomes

$$\frac{\mu''}{\mu} = \frac{H_0 B_0 \sin \delta}{H_0 B_0 \cos \delta} = \tan \delta$$

(1.20)

where $\tan \delta$ is known as magnetic loss factor or power factor. It is a measure of the energy loss inside the magnetic material due to phase delay between applied and induced field [Chikazumi\(^{(1)}\) 1997].

### 1.7.4 Hysteresis parameters (H-B loop)

The typical H-B curve of ferromagnetic material is represented in Figure (1.16) by plotting the magnetic induction $B$ as a function of field strength $H$. At low values of the applied field, the induced magnetization varies linearly with the field and reversible. When the field is gradually increased, the magnetization show non-linear variation with the field and
becomes irreversible. If the field increased gradually further, the magnetization also increased gradually and ultimately attains a saturation value $M_s$. A further increase in the field produces no change in the magnetization, however on the other hand when the field is gradually decreased; the decrease in magnetization is called the residual or remnant of magnetization $M_r$ [Jilies 1991].

Fig. 1.16 Hysteresis loop.

A finite field equal to the coercive field ($H_c$) is equal to bring the magnetization back to zero further increase in the reverse field results in complete reversal of $M_s$. A second reversal of the field completes the hysteresis curve. The saturation of magnetization depends mainly on the magnitude of the atomic magnetic moment $\mu_m$, and the number of atoms per unit volume $n$.

1.8 Electrical Parameters of Ferrites
1.8.1 Conduction in ferrites

The ferrites materials are classified as ferrimagnetic semiconductors, which restrict their electrical conductivity to small values in comparison with the conventional magnetic materials. The electrical conductivity is within the range $10^2 - 10^{11} \, \Omega^{-1} \, \text{cm}^{-1}$. This electrical behavior made the ferrites widely used in electronics applications at microwave frequencies. The conductivity is due to simultaneous presence of Fe$^{2+}$ and Fe$^{3+}$ ions on equivalent lattice sites (usually the octahedral sites). It also, due to the presence of divalent metal ion in two different valance states (e.g. Cu$^{2+}$, Cu$^{3+}$ and Ni$^{2+}$, Ni$^{3+}$). The charge carriers in ferrites are localized at the magnetic ions (cations). These cations are surrounded by close-packed oxygen anions and can be considered isolated ions. Hence, the localized model is more appropriate in ferrite case rather than the band model. According to this model, the conduction is due to the exchange of the 3d electrons, localized at metal ions, from ferrous to ferric ions.

The conduction mechanism in ferrites is quite different from that in semiconductors. In ferrites the temperature dependence of mobility affects the conductivity while the carrier concentration is almost unaffected by temperature. Unlike in semiconductors, wherein the charge carriers occupy states in the wide energy band, the charge carriers in ferrites are localized at the magnetic atoms. In ferrites the cations are surrounded by closed packed oxygen, anion's charge clouds or orbitals.

In addition to the above-mentioned differences, the hopping electron model could differentiae the electrical behavior of ferrites from that of semiconductors [Samokhvalov et al. 1965]. An appreciable conductivity in these systems is found to be due to the presence of iron with different valance states at crystallographically equivalent points [De
Boer et al. 1937]. Conduction is due to the exchange of the 3d electrons localized at the metal ions, from Fe$^{2+}$ to Fe$^{3+}$. Assuming all Fe$^{2+}$ ions in the B-sites to participate in hopping transport, the number of charge carriers (n) works out to be of the order of $10^{20}$/cm$^3$.

1.8.2 DC permittivity

The relative permittivity or the dielectric constant of a medium is defined as the ratio between capacitance of a condenser in vacuum, $C_0$, and that with existence of the medium $C$

$$ \epsilon_r = \frac{C}{C_0} $$  

(1.20)

The permittivity of the medium is defined as

$$ \epsilon = \epsilon_0 \epsilon_r $$  

(1.21)

where $\epsilon_0 = 8.854 \times 10^{-12}$ F.m$^{-1}$ is the permittivity of vacuum and $\epsilon_r$ is the relative permittivity.
1.8.3 AC permittivity

If an AC sinusoidal voltage source is applied across a capacitor filled with dielectric medium, there will be energy dissipated due to phase shift between applied wave and the dipole alignment. The total current in the circuit will consist of two components charging and loss currents. The permittivity will thus be complex number $\varepsilon_r$, consisting of a real part $\varepsilon'$, which represents the storage energy and an imaginary part $\varepsilon''$, which represents the loss.

$$\varepsilon_r = \varepsilon' - i\varepsilon''$$  \hspace{1cm} (1.22)

The relation between $\varepsilon'$ and $\varepsilon''$ is represented in Figure (1.17) using a phase diagram. The loss tangent or tan $\delta$ is defined as the ratio of the imaginary part of the dielectric constant to the real part i.e.

$$\tan \delta = \frac{\varepsilon''}{\varepsilon'}$$  \hspace{1cm} (1.23)

Fig. 1.17 Phase diagram of AC permittivity.
The AC permittivity and tan δ are important for improving ferrite absorption and packaging designs. The permittivity of ferrites is changing with frequency, temperature, orientation, pressure and molecular structure of the material.

1.9 Interaction of Radiation with Matter

The interaction of radiation with substances is an extremely important from the viewpoint of theory and practice. This interaction may be considered from various aspects. When radiation acts on a material, its properties may change. In the recent years, a broad development of nuclear engineering, use of radioactive isotopes, accelerators of elementary particles put forward much attention to study the radiation effect on materials used in electronic devices and control systems.

The interaction processes depend on both the type and energy of the incoming particles (or photons). For example, the electromagnetic spectrum (photons) covers many decades of frequencies. The situation is similar for charged particles. Their energy ranges from fractions of eV to $10^{20}$ eV as in the case of ultra-high energy cosmic rays. For instance, in gas medium the energy dissipation process creating ion pairs (i.e., electrons and positive ions) which are separated and move in the medium. Typically, about 30 eV are required to create an ion pair. In semiconductors, the medium is denser and an electron-hole pair requires about 3.6 eV to be generated. A much stronger effect is exerted on materials by various kinds of ionizing radiation (X- and gamma rays, fluxes of α- and β-particles, neutrons, etc.). These interactions can be
elastic, inelastic scattering or nuclear reactions according to the type and energy of the incident radiation [Tareev 1979], [Turner 2007].

1.9.1 Types of radiation

Radiation can be classified into two general groups:

(i) Charged particles: light ions, heavy ions, electrons, β, α,… etc.

(ii) Neutral particles: photons (γ-rays, x-rays, laser) and neutrons

Therefore, it may be expected that interactions with matter fall into two general types. Charged particles directly ionize the media through which they pass, while uncharged particles and photons can cause ionization only indirectly or by secondary radiation. A moving charged particle has an electrical field surrounding it, which interacts with the atomic structure of the medium through which it is passing. This interaction decelerates the particle and energized electrons in the atoms of the medium. The energized electrons may acquire enough energy to escape from the parent atom. This process, whereby radiation "strips" off one or more of the orbital electrons, is called ionization. Uncharged moving particles have no electrical field, so they can only lose energy and cause ionization by such means as collisions or scattering. A photon can lose energy by the photoelectric effect, Compton effect, or pair production. It should be pointed out that the outer space is filled with radiation that comes from a variety of sources such as burning (for example, our Sun) and exploding (for example, Supernovae) stars. These bodies produce immense amounts of radiation.

α- Gamma ray sources

There are large numbers of radioactive elements that emit γ-rays. These γ-rays are often accompanied by α- and β-particles. Besides
naturally occurring sources, it is possible to produce these isotopes in laboratory. This is normally done by bombarding a source material by neutrons, accelerated charged particles or high energy photons. The nuclei, as a result, go into unstable states and try to get rid of the acquired energy. In the process, they also release energy in the form of γ-rays.

The most commonly used radioactive sources of γ-rays are $^{60}\text{Co}$, $^{137}\text{Cs}$ and $^{192}\text{Ir}$. The easiest way to produce $^{60}\text{Co}$ is by bombarding $^{59}\text{Co}$ with slow neutrons as represented by the following reaction.

$$^{59}\text{Co} + n \rightarrow ^{60}\text{Co} + \gamma$$ \hspace{1cm} (1.24)

The resultant $^{60}\text{Co}$ isotope is radioactive with half-life 5.27 years and gives off two energetic γ-rays (1.17 and 1.33 MeV). The decay scheme is shown in Figure (1.18).

![Decay scheme of cobalt-60](image)

**Fig. 1.18. Decay scheme of cobalt-60.**

**b- X-ray sources**

Production of x-rays is a relatively simple process in which a high Z target is bombarded by high velocity electrons as shown in Fig.1.19.
This result in the production of two types of x-rays: Bremsstrahlung and characteristic x-rays.

Fig. 1.19. Sketch of a typical x-ray tube.

1.9.2 Charged particles sources

Accelerators are the main tools of generating charged particle beam of wide energy range. Cockcroft-Walton, Van de Graaf, tandem, cyclotron accelerators are commonly used to accelerate light and heavy ions to moderate energies (up to several hundred MeV). For example, protons, deuterons and $\alpha$ particles are generated from hydrogen, deuterium and helium gas, respectively.
The basic principle is to ionize the gas to generate plasma and the required charged particles are extracted by a puller to start acceleration in the main chamber. In cyclic accelerators, the accelerated particles take a circular path by the effect of vertical magnetic field lines generated by the electromagnet. The radius of the particle trajectory increases with increasing energy. Fig. 1.20 shows construction of a classical cyclotron.

**1.10 Units of Radioactivity**

Since the most natural way to measure, the activity of a material is to count how many disintegrations occur per unit time. Therefore, the units of activity are defined in terms of disintegrations per second. For example, 1 Becquerel corresponds to 1 disintegration per second and 1 Curie is equivalent to $3.7 \times 10^{10}$ disintegrations per second. For most
practical sources used in laboratories, Curie is too big. Therefore, its subunits of milli-Curie and micro-Curie are more commonly found in literature. The subunits of Curie and interconversion factors of Curie and Becquerel are given below.

\[ 1Ci = 3.7 \times 10^{10} \text{ disintegrations/sec} \]
\[ 1mCi = 10^{-3}Ci = 3.7 \times 10^{7} \text{ disintegrations/sec} \]
\[ 1\mu Ci = 10^{-6}Ci = 3.7 \times 10^{4} \text{ disintegrations/sec} \]
\[ 1Bq = 1 \text{ disintegration/sec} \]
\[ 1Bq = 2.703 \times 10^{-11}Ci \]

For a radioactive source, activity decays with time according to the relation
\[ A = A_0 e^{-\lambda t} \]  
where \( A_0 = \lambda N_0 \) is the initial activity of the sample, \( t \) is the time and \( \lambda \) is the decay constant. Another parameter, which is extensively quoted and used, is the half-life. It is defined as the time required by half of the nuclei in a sample to decay. It is given by the following formula
\[ T_{1/2} = \frac{\ln 2}{\lambda} \]  

1.11. Radiation Exposure and Dose

Radiation is capable of causing damage to both living and nonliving things through different processes. The severity of such
damage would depend on the amount of radiation, its energy and kind of radiation which absorbed by the material.

1.11.1 Roentgen (R)

Roentgen is a measure of exposure due to photons only (x-rays or gamma rays) and is based on the amount of charge they produce in air. 1 R equals the dose needed to ionize and produce $2.58 \times 10^{-4}$ coulombs of positive and negative charges in one kilogram of air.

1.11.2 Absorbed dose

The amount of energy deposited in a medium per unit mass of the medium by ionizing radiation is called absorbed dose. It is measured in units of J/kg. For absorbed dose, the SI unit of this quantity is gray (Gy) with 1 Gy = 1 J/kg. The non SI unite of absorbed dose is called rad. Even though rad has mostly been replaced by Gy, it is still found in some literature. Rad was introduced in 1953 to replace Roentgen, which was the unit of exposure due to x-rays or γ-rays only. Rad is defined as the dose given rise of 0.01 joule of energy per kilogram.

We next derive a simple formula for computing the exposure rate in air from a point gamma source of activity (A) that emits an average photon energy $E$ per disintegration. The rate of energy release in the form of gamma photons escaping from the source is $AE$. Neglecting attenuation in air, we can write for the energy fluence rate, or intensity, through the surface of a sphere of radius $r$ centered about the source $\dot{\psi} = \frac{AE}{4\pi r^2}$. For monoenergetic photons, the absorbed dose rate in air at the distance $r$ from the source is
where, \( \mu_{en}/\rho \) is the mass energy-absorption coefficient of air for the photons. Experimental data showed that this coefficient has roughly the same value for photons with energies between about 60 keV and 2 MeV: \( \mu_{en}/\rho \approx 0.027 \text{ cm}^2 \text{ g}^{-1} = 0.0027 \text{ m}^2 \text{ kg}^{-1} \). Therefore, Eq. (1.27) becomes

\[
\dot{D} = \frac{\dot{\psi} \mu_{en}}{\rho} = \frac{AE \mu_{en}}{4\pi r^2 \rho}.
\]

(1.27)

with \( A \) in Bq (s\(^{-1}\)), \( E \) in Joule, and \( r \) in m, \( \dot{D} \) is in Gy s\(^{-1}\). This relationship can be brought into a more convenient form. Expressing the activity \( A \) in Ci and the energy \( E \) in MeV, we have

\[
\dot{D} = \frac{2.15 \times 10^{-4} \times A \times 3.7 \times 10^{10} \times E \times 1.60 \times 10^{-13}}{r^2} \text{ Gy s}^{-1}
\]

(1.28)

\[
= \frac{1.27 \times 10^{-6} AE}{r^2} \text{ Gy s}^{-1}
\]

(1.29)

**1.12 Aim of the Work**

In this work ferrite samples of chemical formula \( \text{Mg}_x\text{Cu}_{0.5-} \times \text{Zn}_{0.5}\text{Fe}_2\text{O}_4 \) (where \( x = 0.0, 0.2 \) and \( 0.4 \)) were selected to study the structure, magnetic and electrical properties and their influence by gamma ray irradiation. The selection of this type of ferrite was based on their importance as promising material for multilayer chip inductors (MLCI) with high-performance and low cost. In addition, Mg-Cu-Zn ferrites are useful in the fabrication of cores of intermediate frequency transformers (IFT) for amplitude modulation. Usually, Ni-Cu-Zn ferrites are employed as a magnetic material for MLCIs because of its magnetic
properties at high frequencies and its low sintering temperature. Mg-Cu-Zn ferrites have similar magnetic properties to those of Ni-Cu-Zn ferrites with the advantage that they are economical and easy to synthesize.

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 present study investigates this effect on the selected Mg-Cu-Zn ferrites which is commonly used and can be exposed to high gamma ray doses in sever area.

The investigated samples in this study are prepared using conventional sintering method. To study the structure of the samples XRD is performed and from the resulting spectra lattice, parameter and density of samples can be determined. The same measurements are performed on the irradiated samples to study the change in the structure.

To study the effect of radiation on magnetic properties, the induced magnetization as a function of Mg-concentration is measured for irradiated and unirradiated samples. Similarly, the curves of initial permeability as a function of temperature are deduced for all samples. The Curie temperature is determined from these curves.

The electrical properties are studied by measurements of temperature and frequency dependence of AC and DC resistivity and dielectric constant. These parameters are measured using the two-probe method. A comparison of the results before and after irradiation is performed to study the radiation effect.
Chapter 2

Literature Survey

This chapter deals with literature survey of the structure, magnetic and electrical properties of Mg-Cu-Zn substituted ferrites and the effect of different types of radiation on several types of ferrites.

2.1. Properties of Cu-Zn and Mg-Cu--Zn Ferrites

Rezlescu et al. described the effect of CuO content on the intrinsic properties of Cu$_x$Mg$_{(0.5-x)}$Zn ferrites, prepared by the solid-state reaction method. A high density of about 4.5 g/cm$^3$ was obtained for $x = 0.3$ at a low firing temperature of 1050°C. High electrical resistivities were obtained for $x = (0.0-0.3)$. The dispersion permeability was observed above $x = 0.3$ [Rezlescu et al. 1998].

Murthy prepared group of composition Mg$_{0.6-x}$Cu$_x$Zn$_{0.4}$Fe$_2$O$_4$ with $x = 0.1$ to 0.14 by using the sintering method. It was found that the densification of MgCuZn ferrite is Cu concentration dependent. The sintered ferrite with a density of 4.93 g/cm$^3$ and electrical resistivity $>10^{11}$ Ω-cm was obtained for the ferrite with 0.12 mol Cu at relatively low sintering temperature (910°C). The magnetic properties of the ferrites were improved by the Cu substitution. The chip inductors made of the ferrite fired at 910 °C with 0.12 mol Cu was found to exhibit higher dc resistance [Murthy 2001].

[Julia et al. 2007] prepared ferrite powders of composition Mg$_{0.2+x-y}$Cu$_{y}$Zn$_{0.62-x}$Fe$_{1.98}$O$_{3.99}$ by the standard ceramic route. It was found that, the permeability and Curie temperature have to be optimized by proper selection of composition. Ferrites with < 50 mol% Fe$_2$O$_3$ reveal
enhanced densification behavior. Submicrometer powders, prepared by fine milling, showed good sintering activity and density after firing at 900°C. Maximum shrinkage occurred at T< 800°C. The use of Bi$_2$O$_3$ as a sintering additive further improved the densification, but also affected the microstructure and, hence, the permeability. A maximum permeability of $\mu_i = 450–500$ was obtained.

The structural, electrical and magnetic properties of Cu$_{1-x}$Zn$_x$Fe$_2$O$_4$ ferrites, prepared by the standard solid-state technique, were studied by [Muhammad et al. 2007]. It was found that, the substitution of Zn in the Cu$_{1-x}$Zn$_x$Fe$_2$O$_4$ ferrites caused appreciable changes in its structural, electrical and magnetic properties. Unit cell parameters ‘a’ increased linearly with the increase in Zn concentration due to the larger ionic radius of Zn as compared to that of Cu. Sintered density increased with increasing Zn concentration. This was due to the ionic radius difference between Cu and Zn. X-ray density and porosity both of them decreased with the increase in Zn concentration. Temperature dependent dc electrical resistivity decreased with the increase in temperature. Dielectric constant and dielectric loss factor both had decreasing trend with frequency. This decrease had been explained on the basis of space charge polarization resulting from the electron displacement. Saturation magnetization and magnetic moment both increased with the increase in Zn concentration up to $x = 0.2$ and then decreased with the increase of Zn concentration.

[Bhosale et al. 1997] prepared ferrites samples of structure Cu$_x$0.5Mg$_x$Zn by co-precipitation method using oxalate precursors. They studied the structure and the initial permeability ($\mu_i$) and found that, the lattice parameter showed a gradually decreasing trend with increasing
Mg$^{2+}$ content, which was attributed to replacement of Cu$^{2+}$ with Mg$^{2+}$ ions of smaller ionic size. In addition, the density of the samples increased with increasing Mg$^{2+}$ content for the same reason. The Curie temperature did not show appreciable increasing or decreasing trend. The variation in initial permeability in compositions was mainly affected by variations of magnetization (M$_s$) and average particle diameter (D). Also, the samples exhibited thermal hysteresis behavior in the permeability curves. The $\mu_i$ decreased with increasing Mg$^{2+}$ content. This was referred to the lower value of the anisotropy constant (K) for MgFe$_2$O$_4$ than that for CuFe$_2$O$_4$.

The magnetization, initial susceptibility ($\chi_{ac}$) and thermal behavior of this system were studied by [Bhosale et al. 1998], [Bhosale$^{(1)}$ et al.1999], [Bhosale et al., 1999]. It was found that the observed variation in magnetization values was attributed to porosity present in these ferrite compositions. The temperature variation of susceptibility ($\chi_{ac}$) for oxalate complex decomposed at 600 °C did not exhibit maximum with temperature variation. While samples with $x < 0.4$ sintered at 950 °C showed small peak in $\chi_{ac}$ vs. T curve near $T_C$. However, the ferrites sintered at 1000 °C exhibited multi domain (MD) structure. The domain wall width increased with the addition of Mg$^{2+}$ content. Particle size distribution (PSD) indicated absence of fine particles predominantly. To investigate the ferritization temperature of this system, TG, DTG and DTA studies were carried out varying from 599 to 743 K with increasing $x$. The ferritization took place at a very low temperature, ranged from 599 to 743 K varying with Mg$^{2+}$ content. The thermal hysteresis affected around the transition point depends upon the domain wall pinning and competition between intrinsic parameters such
as saturation magnetization ($M_s$) and anisotropy constant ($K_1$). The most sintered sample with $x = 0.2$ has the highest permeability, i.e. $\mu_i = 5034$, which was attributable to continuous grain structure, having clearly defined grain boundaries and without any voids, i.e. having the lowest porosity. The pure Cu-Mg-Zn ferrites were formed by direct thermal decomposition of $\text{Cu}^{2+}$-$\text{Mg}^{2+}$-$\text{Zn}^{2+}$-$\text{Fe}^{3+}$ oxalate complexes (solid solution). The ferritization temperature was far lower than that required for conventional solid-state reaction. The decomposition and ferritization took place simultaneously. XRD studies revealed the formation of single-spinel phase at 337 and 600 °C.

[Zhenxing et al. 2001] prepared Cu-Mg-Zn ferrites by gel auto-combustion method. The effect of Cu ions on the electromagnetic properties has been investigated. It was found that the synthesized powders exhibited high-sintering activity and the ferrite phase could be sintered at temperature less than 950 °C. The prepared samples were found to be of good electromagnetic properties as well as fine-grained microstructures, which is suitable material for multilayer chip inductors with high-performance. Copper content has significant influence on the electromagnetic properties such as initial permeability, quality factor, dc resistivity, dielectric constant and dielectric loss tangent.

[Vital et al., 2007], prepared $\text{ZnFe}_2\text{O}_4$, $\text{Mg}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ and $\text{Mg}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.62}\text{Fe}_{1.98}\text{O}_{3.99}$ powder for the first time by flame spray synthesis (FSS). It was found that the crystalline particles of spinel structure with 6–13 nm primary particle size resulted from the flame process. Particle and crystallite size depended on the flow rate of the atomizing gas, the precursor and its molarity. A sintered density of 5.05 g/cm$^3$ was achieved after firing for 2 h at 900 °C without any sintering.
additives. The permeability of the sintered Mg–Cu–Zn ferrite nanoparticles compacts reached $\mu = 600$ at 1 MHz and the saturation magnetization was 80 emu g$^{-1}$.

[Nath et al. 2004] carried out Mössbauer, x-ray diffraction and ac susceptibility studies on the Mg$_{(1-x)}$Zn$_x$Fe$_2$O$_4$ nanoparticles ferrites prepared by co-precipitation method. It was found that, the crystal size lying in the range 6.4 nm to 21.4 nm. Increase of Zn substitution in Mg-ferrite facilitated the formation of spinel ferrite phase even at lower sintering temperature. Mössbauer spectroscopy showed presence of superparamagnetic (SPM) relaxation in all the samples. At ambient temperature, SPM properties were observed in bigger particle size in samples with higher fraction of Zn substitution. Blocking temperature measured from AC magnetic susceptibility data, showed systematic fall with decrease of particle size in all the three sets of samples with various Zn substitutions. Substitution of 50 % Zn in Mg-ferrite resulted SPM relaxation in 21.4 nm particles at room temperature. The magnetic anisotropy constant increased with decrease of particle size.

Microwave hydrothermal synthesis of nano size PbO added Cu-Mg-Zn ferrites were investigated by [Raju et al. 2006]. It was found that, particle size of the samples synthesized at a low temperature of 160 °C after a treatment time of 1 hour was in the range (10-20 nm) with high surface area. The variations of the sintered density, electrical resistivity, initial permeability, and saturation magnetization with additive concentration had been investigated and the obtained results were compared with one prepared by the conventional ceramic method. It was found that, the addition of PbO improved sintered density, electrical resistivity and permeability.
Similar samples of Mg-Cu-Zn ferrite prepared at low sintering temperature by the citrate precursor method [Atassi et al. 2006]. The fine particle morphology of the powder synthesized by this method was responsible for its higher sintering activity and single-phase ferrite directly from the citrate precursor without any additional heat treatments above 750 °C. The highly active powders could be sintered at relatively low temperatures to obtain highly dense and homogeneous polycrystalline ferrites.

Structure and magnetic properties of the system \( \text{Mg}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4 \) have been studied by [Mazen et al. 2003]. It was found that, the lattice parameter increased with increasing zinc concentration. The composition dependence of the physical properties was divided into two regions. The first one was for \( x \leq 0.3 \) and the second one was for \( x > 0.3 \). From the magnetization measurements, the basic composition (\( \text{MgFe}_2\text{O}_4 \)) showed the lowest magnetization, while the composition of \( x = 0.4 \) showed highest one. The behavior of magnetization \( M \) versus composition showed also two regions for \( x < \) and \( > 0.3 \). The behavior of \( M \) versus \( x \) was discussed on the bases of cation distribution. From the B-H curve, the reminences induction \( B_r \), saturation induction \( B_s \) and the coercive force \( H_c \) were studied with the concentration. It was noticed that the ratio \( B_r/B_s \) and \( H_c \) vs. composition shows that for \( x \leq 0.3 \), the ratio \( B_r/B_s \) increased by 66% while \( H_c \) increased by 14% while for \( x = 0.3 \), \( B_r/B_s \) decrease by 27 % and \( H_c \) decreased by 43%. The Curie temperature \( T_C \) was determined from the measurements of the initial permeability (\( \mu_i \)) versus temperature. It was found that, \( T_C \) decreased with increasing Zn-content. Also, paramagnetic temperature \( T_P \) was determined from the behavior of \( M_S \) vs. \( T \). In general it was found \( T_P > T_C \) by about 7-10 K.
[Haque et al. 2008] studied the magnetic and dielectric behavior of \( \text{Mg}_{0.55-x}\text{Cu}_x\text{Zn}_{0.45}\text{O(Fe}_2\text{O}_3)_{0.97} \) ferrites. It was found that, the presence of Cu ions caused appreciable changes in the physical, magnetic and dielectric properties of the Cu-substituted Mg–Zn ferrite. The lattice parameter ‘\( a \)’ increased with the copper content due to larger ionic radius of \( \text{Cu}^{2+} \) compared to \( \text{Mg}^{2+} \). A remarkable increase in the value of bulk density was found with increasing Cu substitution for Mg in Mg–Zn ferrite. By incorporating CuO, the initial permeability of the samples was increased. This was attributed to the presence of Cu ions activating the sintering processes in ferrites and leading to increased in density. Also, the saturation magnetization increased slightly with increasing copper content for \( x \leq 0.30 \) and then it decreased for \( x = 0.35 \). Dielectric constant (\( \varepsilon' \)) decreased with increasing frequency exhibiting normal dielectric behavior of ferrites. The compositional variation of dielectric constant and resistivity indicated inverse trend with each other.

Magnetic and electrical properties of \( \text{Mg}_{0.80-x}\text{Cu}_{0.20}\text{Zn}_x\text{Fe}_2\text{O}_4 \) ferrite nano-crystalline powders prepared by sol–gel, auto-combustion method were studied by [Barati 2009]. He found that, the nitrate–citrate gels exhibited a self-propagating behavior after ignition in air. The prepared low-temperature sintered MgCuZn ferrites possess good electromagnetic properties, as well as fine-grained microstructures, making them good materials for electronic applications with high-performance and low-cost. The experiments conducted in this study suggested that, the substitution of Zn in the system has a significant influence on there electromagnetic properties. The composition \( \text{Mg}_{0.17}\text{Cu}_{0.20}\text{Zn}_{0.63}\text{Fe}_2\text{O}_4 \) showed highest permeability, magnetization and dielectric constant among all the compositions studied. The dielectric
constant and dielectric loss tangent of the samples were inversely proportional to AC resistivity.

Samples with the composition $\text{Mg}_{0.4569}\text{Cu}_{0.144}\text{Mn}_{x}\text{Zn}_{0.4}\text{Fe}_{2}\text{O}_4$, prepared by microwave sintering method, were investigated by [Bhaskar et al. 2004]. It was found that an increase in permeability is observed as Mn content increase. The increase of permeability due to the incorporation of Mn was attributed to the decrease of magnetostriction constant induced by Mn addition. The change of other electrical properties such as dielectric constant and dissipation factor were caused by the electron exchange between $\text{Fe}^{2+}$ and $\text{Fe}^{3+}$. MgCuZn ferrites with 2 and 4 mol % Mn content and sintered at 910°C/30 min exhibited high density and the electrical properties were found to be optimum among all the present samples.

### 2.2 Irradiation Effect on the Structure, Magnetic and Electrical Properties of Ferrites

#### 2.2.1. Ion beam irradiation

Modifications of dielectric properties of Al$^{3+}$ substituted Mg–Mn ferrite by 50 MeV Li$^{3+}$ were studied by [Dogra et al. 2003]. It was found that compressive strain was generated by the ion irradiation, observed by the x-ray diffraction studies, which had modified the electrical properties. The dispersion of dielectric constant $\varepsilon'$ of unirradiated samples was attributed to Maxwell–Wagner type interfacial polarization and Koops’s theory. The dielectric constant of irradiated samples was decreased in magnitude from that of the unirradiated samples as shown in figure (2.1).
Fig. 2.1 Variation of dielectric constant $\varepsilon$ as a function of frequency for unirradiated and irradiated $\text{Mg}_{0.9}\text{Mn}_{0.1}\text{Al}_x\text{Fe}_{2-x}\text{O}_4$ for $x = 0.0, 0.2$ and $0.4$ \cite{dogra2003}.

This was explained on the basis of space charge polarization and the collective contributions of p and n-type carriers. The electric loss parameter for irradiated samples showed resonance peak, which explained using Maxwell–Wagner interfacial polarization (matching of hopping frequency with frequency of an applied electric field).

In situ monitoring of electrical resistance of $\text{Li}_{0.25}\text{Mg}_{0.5}\text{Mn}_{0.1}\text{Fe}_{2.15}\text{O}_4$ nano ferrite thin film irradiated by 190 MeV $\text{Au}^{14+}$ ions was investigated \cite{ghosh2003}. To probe the swift heavy ion induced modifications in the electrical properties in the film an in situ measurement of electrical resistance using two-probe method was carried out as seen in figure (2.2). It was observed that, the value of resistivity comes down drastically from $1.5 \times 10^8$ to $1 \times 10^5$ $\Omega$ cm after irradiation at the flounce of $1 \times 10^{13}$ ions/cm$^2$. 
Fig. 2.2 The variation in resistivity with fluence when irradiated with 190 MeV Au\(^{14}\) ions [Ghosh et al. 2003].

Influence of 50 MeV Li\(^{3+}\) ion irradiation on structural and magnetic properties of Ti\(^{4+}\)-substituted Li\(_{0.5}\)Al\(_{0.1}\)Fe\(_{2.4}\)O\(_4\) was studied by [Chhantbar et al. 2006]. The consequences of 50 MeV Li\(^{3+}\) ion irradiation on the structural and magnetic behavior of polycrystalline samples of spinel ferrite system were investigated by using x-ray diffraction (XRD), magnetization and \(^{57}\)Fe Mössbauer spectroscopy. The Mössbauer spectra exhibited central enhancement after SHI-irradiation, which was attributed to the paramagnetic centers created due to the irradiation. The reduction observed in the saturation magnetic moment after SHI-irradiation was ascribed to the partial formation of paramagnetic centers and rearrangement of the cations in the lattice corroborated by XRD and Mössbauer Lorentzian intensity analysis. The central enhancement observed in the Mössbauer spectra of the irradiated
samples were not due to amorphization but its origin lies in the formation of localized paramagnetic centers.

The consequences of 85 MeV oxygen ion irradiations on the magnetization behavior of micron-sized and nano-sized powders of hexagonal ferrite (SrFe$_{12}$O$_{19}$) were examined by [Shinde et al. 1998]. The observed changes in the hysteresis loop were attributed to the defect-domain-wall interaction. For the multi-domain particles, the changes were so large that after irradiation loop squareness ($M_3/M_4$) changes from 0.5 to 0.8. The stability of the defect states in the small particles was discussed and the calculation of the energy change due to the introduction of the extended defect showed that the particle size dependent term varies inversely with the particle radius, suggesting instability of the defect state at smaller particle size. The stability of hysteresis loop of nano-sized powders to the irradiation by high-energy particles showed that the nano-sized single domain particles were radiation hard magnetic materials. The electronic stopping power of the oxygen ions in SrFe$_{12}$O$_{19}$ was much smaller than the calculated threshold electronic stopping power, indicating that latent tracks of amorphous material were not formed in this case. The nature of the defects was likely to be in the form of grain refinements along the ion tracks, resulting in the large grain boundary density.

Swift heavy ion induced modifications on the dielectric properties of NiMn$_{0.05}$Ti$_3$(Zn$^{2+}$, Mg$^{2+}$)$_3$Fe$_{1.95-2x}$O$_4$ thin films were studied by [Dogra et al. 2003]. The films were irradiated with 190 MeV Ag$^{1+}$ ions for three different fluencies. The dielectric constant ($\varepsilon'$) and dielectric loss, tan $\delta$, as a function of frequency from 1 KHz to 1 MHz were measured at room temperature for unirradiated and irradiated thin films. The $\varepsilon'$ for
NiMn$_{0.05}$Fe$_{1.95}$O$_4$ and NiMn$_{0.05}$Ti$_{0.2}$Mg$_{0.2}$Fe$_{1.55}$O$_4$ of unirradiated and irradiated film decreased with frequency, which was explained by Koop’s model. The dielectric constant at high frequency decreased with increase in ion irradiation fluence values. The irradiated NiMn$_{0.05}$Ti$_{0.2}$Zn$_{0.2}$Fe$_{1.55}$O$_4$ films showed an anomalous behavior. The dielectric constant below 20 kHz increased with irradiation as compared to pristine film, whereas above 20 kHz the dielectric constant for irradiated samples was less than the pristine samples but it increased with irradiation. Further the dielectric loss showed an increase with the irradiation. All the irradiated films showed a resonance peak, which was not present in the pristine films. Also, with the increase in the dose the resonance peak was found to be shifted to the lower frequency.

The effect of 100 MeV Si$^{7+}$ irradiation on the structural and magnetic properties of Mg$_{0.95}$Mn$_{0.05}$Fe$_2$O$_4$ nanoparticles were studied by [Kumar et al. 2005]. It was found that, after irradiation a slight increase in the particle size. With the room temperature Mössbauer spectroscopy, superparamagnetic relaxation effects were observed in the pristine as well as in the irradiated samples. No appreciable changes were observed in the room temperature Mössbauer spectra after ion irradiation. All the samples showed well-defined magnetic ordering at 5 K, whereas, at room temperature they were in a superparamagnetic state. From the magnetization studies performed on the irradiated samples, it was concluded that the saturation magnetization was enhanced. This was explained on the basis of the coercivity controlled growth of single-domain particles in which the absence of domain walls made the magnetization reversal difficult. By irradiation the surface state pinning
of domains were released and the magnetization of the nanoparticles was increased.

The effect of 50 MeV Li$^{3+}$ ion irradiation on structural, dielectric and permeability studies of In$^{3+}$ substituted Mg–Mn ferrite was studied by [Singh et al. 2002]. It was found that, the 50 MeV Li$^{3+}$ ion irradiation induced modifications on structural, dielectric and magnetic permeability properties of Mg-Mn-In-Fe$_2$O$_4$ ferrites. The x-ray diffraction studies showed that a compressive strain was produced in the lattice after irradiation and the effect of strain was being observed on the dielectric and magnetic permeability properties. The dielectric constant as a function of frequency for unirradiated samples was explained by Koops's model. Whereas, for irradiated samples this was explained by the collective contributions of p and n type carriers. The decrease in dielectric constant of irradiated samples in magnitude was explained on the basis of Maxwell–Wagner interfacial polarization. The electric loss parameter for irradiated sample was increased from that of the unirradiated samples and showed the resonance peak, which was explained by the Maxwell–Wagner interfacial polarization. However, the resonance peaks were not present in unirradiated samples. The dispersion in initial permeability followed the same trend for unirradiated as well as for irradiated samples. A decrease in permeability was observed for irradiated samples as shown in figure (2.3). These results were explained by considering the magneto-mechanical affected generated by stress field induced by the ion irradiation in these samples. The magnetic loss in irradiated samples was drastically reduced due to suppression of imaginary part of permeability.
Fig. 2.3 Initial permeability as a function of frequency for unirradiated and irradiated Mg-Mn-Fe$_2$O$_4$ [Singh et al. 2002].

Magnetic study of nanostructured zinc ferrite irradiated with 100 MeV Oxygen ion beam were done by [Singh et al. 2010]. The irradiated sample possessed less magnetization compared to the pristine as observed by the magnetization versus applied field curve. The thermal magnetization curve exhibited reduction in the blocking temperature of the irradiated sample from 276 to 63 K. Mössbauer spectroscopic measurements at room temperature showed no change of magnetic phase occurs at 300 K. The measurements at 10 K exhibited the presence of magnetic ordering in both the pristine and irradiated samples. Well-resolved spectra, which were obtained by In-field Mössbauer spectroscopic measurements, showed the change in Yafet–Kittel type of canting of spins and also the redistribution of cations in irradiated sample. The reduction in magnetization after the irradiation in the specimen was attributed to the decrease in cation inversion and increase in canting angle after irradiation.
Nanocrystalline Li-Mg ferrite thin films, prepared by R.F. magnetron sputtering technique, irradiated with 190 MeV Au\textsuperscript{14+} ions was investigated by [Ghosh et al. 2004]. It was found that, the existence of stable Au\textsuperscript{14+} ions induced defects at room temperature in the nanocrystals after irradiating with Au\textsuperscript{14+} ions. The formation of defects like columnar tracks, transformed a ferrimagnetic sample to superparamagnetic. The decrease in both magnetic behavior and resistivity with fluence increased due to transformation of Fe\textsuperscript{3+} ions to Fe\textsuperscript{2+} ions. The motion of dislocations in the grain boundaries enhanced the above effect.

### 2.2.2 Laser irradiation

The effect of laser irradiation on the structure and electromechanical properties of Co–Zn ferrite were studied by [Tawfik et al. 2002]. It was found that, the x-ray diffraction patterns of irradiated samples by laser showed a distorted cubic structure. The displacement of some Fe\textsuperscript{3+} ion from its original positions under the effect of laser irradiation changed the Fe\textsuperscript{3+}–O\textsuperscript{2−} bond length and so the frequency of IR absorption band. The resonance frequency increased and reached maximum at 170 °C before irradiation whereas it decreased and reached minimum at the same temperature after irradiation due to the formation of cation vacancies by laser irradiation. These induced vacancies affected domain wall motion led to a decrease of ultrasonic wave velocity. Young’s modulus for unirradiated sample increased with temperature, whereas after irradiation decreased. The electrical conductivity values of the irradiated sample increased due to the increase of Co\textsuperscript{3+}/Co\textsuperscript{2+} ratio after irradiation.
Laser induced structural and transport properties change in Cu–Zn ferrites were investigated by [Ahmed et al. 2007]. It was observed that, the XRDPs of irradiated samples by laser showed a distorted cubic structure, drop in the values of electrical conductivity, dielectric constant and dielectric loss at range of frequencies from 10 to 4000 kHz for irradiated ferrites samples by laser with 18,000 laser shots were detected. Response of these materials to laser irradiation showed different effects on the conductivity depending on the temperature of the samples.

2.2.3 Gamma rays irradiation ($^{60}$Co gamma source with energy $\approx 1.17$ MeV):

[Darwish et al. 1993] as well as [Hemeda et al. 2003] studied the effect of gamma-rays irradiation using a $^{60}$Co source on the structure and the electrical properties of Co-Zn ferrites. It was noticed that XRD peaks were shifted after irradiation. The $\gamma$-radiation changed the ferric ions to ferrous ions of radius 0.69 Å which is larger than that of ferric ions 0.59 Å. This increased the size of the crystal by $\gamma$-irradiation. The values of the lattice parameter and dielectric constant increased after irradiation as seen in figure (2.4). This was attributed to the number of ferrous ions increased at the octahedral sites due to $\gamma$-radiation, leading to increase the lattice parameter and polarization with electric field.
Fig. 2.4 The dependence of lattice parameter (a) with (x) before and after irradiation [Darwish et al. 1993].

It was noticed also that the diffusion coefficient increased after gamma irradiation for all Zn$^{2+}$ concentrations. This was explained on the basis of displacement of metal ions from its original sites under the effect of irradiation leaving behind it lattice vacancies which increase the diffusion coefficient.

[Hamada 2004] and [Dalal 2005] investigated the effect of gamma irradiation on the structure and the diffusion coefficient of Co-Zn-Mn ferrites. A shift was observed in the XRD peak position as shown in figure (2.5). This was indication for the slight distortion of cubic structure. The diffusion coefficient of electrons was found to be increased for the irradiated samples. The IR Spectra showed also the defect which was produced after $\gamma$-irradiation. The activation energy of
hopping electrons decreased after irradiation in the first stage of rising temperature. At high temperature, the hopping holes were the majority carriers in this system.

Fig. 2.5 X-ray diffraction pattern of Co-Zn-Mn-Fe$_2$O$_4$ ferrite: (a) before and (b) after $\gamma$-irradiation [Hamada 2004].

A similar study on Co-Zn-Ce-Fe$_2$O$_4$ ferrites was reported by [Ateia 2006]. It was found that, the crystallite size for unirradiated samples was greater than irradiated one, while the estimated micro strain values for irradiated samples was greater than unirradiated one. Also, the dielectric constant ($\varepsilon'$) increased after irradiation as shown in figure (2.6). This increase in ($\varepsilon'$) was attributed to the production of Fe$^{2+}$ after irradiation and the generation of some vacancies at different depths which acted as trapping centers.
Fig 2.6 Variation of $\epsilon$ with the absolute temperature at different frequencies for irradiated and un-irradiated ferrite samples [Ateia 2006].

[Ahmed et al. 2003, 2007] studied the effect of gamma irradiation on the structural and electrical properties of Mg-Ti-Er$_2$-Fe$_2$O$_4$ system. It was found that, noticeable shift for the XRD peaks occurred after irradiation. This was attributed to the $\gamma$-irradiation may change the ratio of Fe$^{2+}$/Fe$^{3+}$ on the octahedral sites as a consequence of $\gamma + $Fe$^{2+}$ $\leftrightarrow$ Fe$^{3+}$ + e$^-$. This led to decrease in the crystal size. It is also noticed that, there was distinct increase in the dielectric constant. This was attributed to some distortion may occur to the tetrahedral sites due to such displacement and it moved the anions away from the nearest tetrahedral cations. The conductivity increased as the $\gamma$-dose increases. This increase in $\sigma$ was attributed to the increase in the ratio of Fe$^{2+}$/Fe$^{3+}$ on the octahedral sites as a consequence of the hopping process. The values of the activation energy decreased with increasing dose.

[Okasha 2010] achieved an enhancement of magnetization of Mg–Mn nano ferrite by $\gamma$-irradiation. It was noticed an increase of lattice
constant \((a)\) for the irradiated samples was explained due to the ionization of the ferric ions of smaller ionic radius to ferrous ions of larger ionic radius. It was found absence of hysteresis, saturation, remanence, and coercivity at a higher applied magnetic field of 6 kOe indicated the presence of superparamagnetic behavior. Moreover, the values of saturation magnetization \((M_s)\) for irradiated samples were higher than these obtained of the unirradiated ones as shown in figure (2.7). This behavior was explained according to the small particle surface effect and the internal cation disorder.

Fig. 2.7 Hysteresis curve of Mg-Mn-Fe\(_2\)O\(_4\) nanoparticles before and after irradiation [Okasha 2010].

Gamma irradiation induced damage creation on the cation distribution, structural and magnetic properties in Ni-Zn ferrite was
investigated by [Karim et al. 2010]. It was found that, the positions of XRD peaks were shifted to higher 2θ values and the peak intensities were decreased. In addition, the width of the peaks was increased from that of the unirradiated samples. The value of lattice constant decreased after irradiation. It was attributed to the lattice vacancies generated after irradiation caused distortion and deviation from the spinel cubic structure. The cation distributions indicated the redistribution of the cations in the A and B-sites induced. The change in cation distribution data was attributed to the formation of Fe$^{2+}$ ions at octahedral sites after irradiation. An increase in the saturation magnetic moment and Curie temperature after irradiation was observed also after irradiation. It was attributed to the partial formation of ferrimagnetic centers and rearrangement of cations in the lattice and ion induced disorder.
Chapter 3
Experimental Techniques

3.1 Preparation of the Samples

The chemical formula of the ferrite samples under investigation is Mg$_x$Cu$_{0.5-x}$Zn$_{0.5}$Fe$_2$O$_4$, where $x=0.0$, $0.2$ and $0.4$ mol fraction. The standard ceramic method was used to prepare the samples. According to this method, metal oxides (MgO, CuO, ZnO and Fe$_2$O$_3$) in powder form were mixed together relative to their molecular weight ratio in the above-mentioned compound. The chemical purity used for all oxides was 99.9 %. The mixture was milled using mechanical agate for 30 min. The solid reaction can be described by the following equation:

$$x\text{MgO} + (0.5-x)\text{CuO} + 0.5\text{ZnO} + \text{Fe}_2\text{O}_3 \rightarrow \text{Mg}_{x}\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$$  (3.1)

To start the primary reactions between the mixed oxides, the samples were calcined in muffle furnace at 900 °C for 30 hours. The samples were milled again mechanically for 3 hours. Thereafter, the grinded powder was pressed in tablet and toroidal forms under pressure of $4.7 \times 10^8$ Pa. The tablet samples were of diameter 13 mm and the toroid samples were of inner and outer diameters 8.5 and 17 mm, respectively. All the pressed samples were finally sintered at 1050 °C for 10 hours and slowly cooled to room temperature with fixed cooling rate (1 °C/min). The samples were polished to remove any oxide layer formed during the sintering process.
3.2 X-Ray Diffraction Analysis

X-ray diffraction analyses were performed at Central Metallurgical R & D Institute Cairo, Egypt using a diffractometer of type X’Pert Graphics & Identify with CuKα radiation. The radiation source was CuKα (λ = 0.1542 nm). From the obtained spectra of all investigated samples phase compositions were analyzed. The interplaner spacing (d) of each (hkl) plane was calculated using Bragg’s law. The lattice constant (a) was calculated according to the relation (1.12).

The apparent density \( d_{\text{app}} \) (g/cm\(^3\)) was determined by means of hydrostatic weighting in distilled water. The apparent density was calculated according to the relation,

\[
d_{\text{app}} = \frac{m_s}{m_s - m_d} d_w ,
\]

where, \( m_s \) is the mass of the sample in air, \( m_d \) is the mass of the sample in distilled water and \( d_w \) is the density of water (1 g/cm\(^3\)). The porosity percentage P % was calculated using the relation (1.13).

3.3 Measurement of Magnetic Properties

3.3.1 Magnetization

The measurement of magnetization M (A.m\(^{-1}\)) was based on Faraday’s law of electromagnetic induction. A toroidal sample, figure (3.1), of each investigated composition was used as transformer core in primary and secondary induction coils. Figure (3.2) shows the circuit diagram used for magnetization measurement. The
magnetization current $I_p$ (A) in the primary coil was varied from zero to 3.5 A, at constant magnetization frequency $f = 50$ Hz. The circuit contained a power resistance $R_p$ to avoid coil damage. The corresponding magnetizing field strength $H$ (A.m$^{-1}$) is determined by the following equation.

$$H = \frac{N_p I_p}{2\pi r},$$

(3.3)

where, $N_p$ is the number of turns of the primary coil, $I_p$ is the current of the primary coil, $r$ is the mean radius of the toroidal sample, figure (3.1),

$$r = \frac{r_0 + r_i}{2},$$

where $r_0$ and $r_i$ are the outer and inner radii, respectively.

Fig. 3.1 The toroidal sample
Fig. 3.2 Circuit diagram for magnetization measurements.

The induced potential $V_s$ (volt) across the secondary coil is measured using high sensitivity multimeter type BK-5492MH (accuracy at $V_{DC}$ 0.012% and $V_{AC}$ 1% and resolution 10 mv). The corresponding induced magnetic flux $B$ (Tesla) is calculated using the following formula:

$$B = \frac{N_s V_s}{\mu_0 A}$$

(3.4)

Finally, the magnetization $M$ is determined by the relation

$$M = \frac{B}{\mu_0} - H$$

(3.5)

where $\mu_0 = 4\pi \times 10^{-7}$ is the permeability of free space, $N_s$ is the numbers of turns in secondary coils, $A$ is the cross-section area of the toroid [Boll 1977], [Mazen et al. 1996].
3.3.2 Initial permeability

The initial permeability $\mu_i$ was measured as a function of frequency of the applied magnetic field using the above described induction circuit with additional components as shown in Figure (3.3). The specifications of the used components are as follows:

1- Wave generator type BK-4040A (5 Hz-200 MHz) with sensitivity less than 25 mV rms.

2- Standard resistance box $R_s$ type CHAUVIN ARNOX, average accuracy (1 %), by which the magnetization current is measured. The input signal is connected in paralleled with input channel of CRO type PINTEK PS-200, average accuracy (3 %), for measuring $I_p$ by using Ohm’s law $I_p = V_p/R_s$.

3- From the induced voltage $V_s$ measured at the secondary coil, the initial permeability $\mu_i$ could be determined from Poltinnikov formula [Poltinnikov 1966]:

$$V_s = k \cdot \mu_i,$$

where,
ω is the angular frequency = (2πf) and L is the average path of the magnetic flux = (2πr).

Fig. 3.3 Circuit diagram for initial permeability measurement as a function of frequency.

To determine the Curie temperate of the samples, the initial permeability was measured as a function of temperature, the induction circuit including the toroidal sample was inserted in a laboratory made tube furnace as illustrate in figure (3.5). The frequency of the applied current was fixed at f = 10 kHz and the magnetization current was I_p = 4 mA. Figure (3.4) shows the experimental set up of the circuit. The temperature of the sample is measured using thermocouple type K inserted in the toroid center and connected to digital thermometer type BK-710 with (average accuracy ± 0.4).
3.4 Electrical Properties

3.4.1 DC resistivity

The DC electrical resistivity of each composition was measured by inserting the tablet sample between two silver electrodes, using the two-probe method. The investigated sample was polished and coated with silver paste at both surfaces to have good contact. Figure (3.5) shows the setup to measure dc resistivity of the samples from room temperature up to about 570 K. The sample holder consists of:

1. Ceramic tube, which is attached to the system by a spring that exerts a slight pressure on the sample (to get a good contact between the electrodes and the samples).
2. Two silver electrodes to enclose the sample. A mica sheet (3 mm) from a disc of aluminum separates the lower electrode.

Fig. 3.4 Circuit diagram for measuring the initial permeability with temperature.
3. Cr-Al thermocouple type K connected to digital thermometer type BK-710 is used to measure the temperature of the sample. The DC resistivity $\rho$ is measured using the circuit, shown in Figure (3.6), consists of a DC power supply type GP 4303D, with ripple voltage less than $2 \text{ mV}_{\text{p-p}}$, connected to the sample and the standard resistance (type CHAUVIN ARNOX). Both of the sample (S) and standard resistance, ($R_s$) are connected in series.

The resistivity of the sample ($\rho$) is calculated using the relation:

$$\rho = RA/d$$  \hspace{1cm} (3.7)

where $A$ and $d$ are the cross-section area and the thickness of the tablet sample, respectively. The resistance ($R$) of the sample is calculated by Ohm's law. The current ($I$) passing through the sample is obtained by measuring the potential drop across the standard resistance. To avoid the joule heating effect, the current in the sample did not exceed 10 mA.
Fig. 3.5 Experimental set up to measure the dc resistivity with temperature.
Fig. 3.6 Circuit diagram for measuring electrical resistivity.

### 3.4.2 Dielectric constant

Parallel plate capacitor ($C_p$) and dielectric loss tangent (tan $\delta$) were measured for the tablet samples using LCR bridge meter model HIOKI 3532-50-LCR HiTESTER. The real part of the dielectric constant ($\varepsilon'$) is calculated using the formula [Snelling et al. 1988]:

$$
\varepsilon' = \frac{C_p d}{\varepsilon_0 A},
$$

(3.8)

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 $\varepsilon_0$ is the permeability of free space ($\varepsilon_0 = 8.854 \times 10^{-12}$ F/m).

The AC resistivity ($\rho_{AC}$) is obtained from the values of ($\varepsilon'$) and the loss tangent by using the relation [Snelling et al. 1988]:

$$
\rho_{AC} = \frac{1}{\varepsilon_0 \omega \tan \delta}
$$

(3.9)
The sample’s surfaces are rubbed with silver paste as a contact surface for measurements of dielectric parameters. The parameters $\varepsilon'$, $\tan \delta$ and $\rho_{AC}$ were measured as a function of frequency within the range 45 Hz - 5 MHz at room temperature. Figure (3.7) shows the structure of the measuring cell used in dielectric measurements.

Fig. 3.7 Schematic diagram of the sample cell used in dielectric measurements.

### 3.5 Irradiation Process

The samples under investigation were exposed to $^{60}$Co radioactive source in the gamma irradiation cell, (medical sterilizer type CM-20) at Cyclotron facility, Nuclear Research Center, Atomic Energy Authority, Cairo, Egypt. The estimated dose was 1.9 MGy with dose rate 5 kGy/h. Figure (3.8) illustrates live photo of the irradiation chamber.
After irradiation, the samples of all compositions were measured again for XRD characterization, magnetic parameters and electrical parameters and then comparative study were done as shown in the next chapter.
Chapter 4

Results and Discussion

4.1 Physical Properties

4.1.1 XRD spectrum analysis

X-ray diffraction patterns (XRDPs) for ferrite samples with composition \( \text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4 \) (\( x = 0.0, 0.2 \) and 0.4) before and after \( \gamma \)-irradiation are illustrated in figures (4.1), (4.2) and (4.3). XRDPs show single spinel phase for all investigated samples before and after \( \gamma \)-irradiation. Comparison of sample spectra with \( x = 0.0 \), figure (4.1) before and after \( \gamma \)-irradiation shows that the relative intensity of the peaks (400) and (533) decreased after \( \gamma \)-irradiation. Whereas, the relative intensities for the peaks (222), (440) and (511) increased. In addition, it is also noticed that, the intensity of the peak (440) became greater than that of the (311) peak. These results reveal that after \( \gamma \)-irradiation the plane (440) became more dense than (311) plane. In addition, as noticed in figures (4.2) and (4.3), the intensity of peaks (400) and (440) decrease after \( \gamma \)-irradiation, and at \( x = 0.4 \) the peak (200) also decreased. Such results, the modification of the relative intensity for some reflecting planes, were reported by different authors and they attributed that to the distortion occurred in the cubic lattice after \( \gamma \)-irradiation [Dalal 2005], [Ahmed et al. 2003].

The crystallite size (\( d \)), which could be considered as indicator to the grain size, was calculated using Debye-Scherer formula, which is compacted with the program of x-ray diffractometer. It is found
that \( d \) lies in the range (134-160) nm as illustrated in table (4.1). It is noticed that \( d \) decreased as Mg concentration increased. Moreover, it is observed that \( d \) decreased after \( \gamma \)-irradiation. The decrease of grain size with Mg-concentration was reported by different authors such as [Bhosale et al. 1998], [Rezlescu et al. 1998], [Zhenxing et al. 2001]. Such a decrease of grain size by irradiation process was reported for Co-Zn-Ce and Mg-Ti-Er ferrites irradiated by gamma rays [Ahmed et al. 2003], [Ateia 2006].

Table (4.1) Values of crystallite-size with Mg-content for \( \text{Mg}_x\text{Cu}_{0.5} \text{Zn}_{0.5} \text{Fe}_2 \text{O}_4 \) ferrites before and after \( \gamma \)-irradiation.

<table>
<thead>
<tr>
<th>Mg content ( x )</th>
<th>( d ) (nm)</th>
<th>Unirradiated</th>
<th>Irradiated</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>160</td>
<td>144</td>
<td></td>
</tr>
<tr>
<td>0.2</td>
<td>155</td>
<td>140</td>
<td></td>
</tr>
<tr>
<td>0.4</td>
<td>134</td>
<td>92</td>
<td></td>
</tr>
</tbody>
</table>
4.1.2 Lattice parameter

The lattice parameter \((a)\) of the investigated samples, before and after \(\gamma\)-irradiation is calculated using (1.12) relation. The variation of lattice parameter with Mg-concentration before and after \(\gamma\)-irradiation is illustrated in figure (4.4). It is clear that, as Mg-concentration increase the lattice parameter decreases approximately linearly obeying Vegard’s law. This result could be explained on the basis of the ionic radii due to the replacement of ions with larger ionic radius of \(\text{Cu}^{2+}\) ions (0.72 Å) by the smaller one \(\text{Mg}^{2+}\) ions (0.65 Å) [Bhosale et al. 1997]. Furthermore, it is clear that, the lattice parameters for all irradiated samples are larger than that of the unirradiated ones. This could be attributed to the formation of ferrous ion, \(\text{Fe}^{2+}\), with radius (0.78 Å) which is larger than that of ferric ion, \(\text{Fe}^{3+}\), (0.64 Å). The formation of ferrous ions in ferrites, due to irradiation, was reported by [Ahmad et al. 2002], [Hemeda et al. 2003], [Darwish et al. 1994], [Okasha 2010], [Dalal 2005], [Ateia 2006], [Hamada 2004].
4.1.3 Porosity

The variation of porosity (P %) with Mg-concentration, for the investigated samples before and after γ-irradiation, is shown in figure (4.5). It is observed that, as Mg-concentration increase, (P %) decreases. The result indicates that, the Mg-substitutions improve the densification of the ferrite. This behavior could be attributed to the filling of the interstitial positions by the smaller Mg$^{2+}$ ions, then increasing the density [Bhosale et al. 1997]. Also, one can notice that the values of porosity for the irradiated samples are higher than that of unirradiated ones. This increase in the porosity after γ-irradiation could be attributed to generation of some lattice imperfections like voids or vacancies at different depths according to the interaction of
\(\gamma\)-rays with matter [Mazen et al. 2003], [Darwish et al. 1994], [Ateia 2006].

![Graph showing porosity vs. content of Mg ions before and after \(\gamma\)-irradiation.]

**4.2 Magnetic Properties**

**4.2.1 Magnetization**

Figures (4.6), (4.7) and (4.8) show the change of magnetization \(M\) (A.m\(^{-1}\)) with the applied magnetic field \(H\) (A.m\(^{-1}\)) up to \(5 \times 10^4\) A.m\(^{-1}\), before and after \(\gamma\)-irradiation, at room temperature for the investigated samples. It is observed that the magnetization increases with the applied magnetic field, agree with normal behavior. Figure (4.9) shows the change of magnetization with Mg-concentration before and after \(\gamma\)-irradiation at \(H = 2\) (kA.m\(^{-1}\)). It is observed that, the magnetization decreases with increasing Mg-concentration.
Fig. 4.6 Variation of magnetization for sample with \( x = 0.0 \) as a function of magnetic field before and after \( \gamma \)-irradiation.

Fig. 4.7 Variation of magnetization for sample with \( x = 0.2 \) as a function of magnetic field before and after \( \gamma \)-irradiation.
Fig. 4.8 Variation of magnetization for sample with $x = 0.4$ as a function of magnetic field before and after $\gamma$-irradiation.

Fig. 4.9 Variation of magnetization as a function of Mg-concentration before and after $\gamma$-irradiation at $H = 2\text{kA.m}^{-1}$. 
This behavior could be explained on the basis of cation distribution. The cations, as Reynolds supposed, distribute as

\[
\left( \text{Zn}^{2+} \text{Fe}^{3+} \right) \left[ \text{Mg}^{2+} \text{Cu}^{2+} \text{Fe}^{3+} \right]
\]

where the bracts ( ) and [ ] denote to A- and B-sites respectively[Reynolds 1991]. According to the assumed cation distribution, the general formula of the magnetization at A and B sites could be written as

\[
M_B = 5 * 1.5 + (0.5 - x) * 1 + 0 * x = 8 - x
\]

\[
M_A = 0 * 0.5 + 0.5 * 5 = 2.5
\]

Therefore, the net magnetization M is given by

\[
M = M_B - M_A = 8 - x - 2.5 = 5.5 - x
\]

According to eq. (4.2), M decreases linearly with x. The deviation from linearly could be attributed to the presence of Fe\(^{2+}\) ions in B-sites which reduce the magnetization of these sites than expected.

The decrease of magnetization as Mg-concentration increase could be attributed to the replacement of Cu\(^{2+}\) ion with magnetic moment (one \(\mu_B\)) by the non-magnetic Mg\(^{2+}\) ion [Chikazumi 1964], where both of them prefer the occupation of B-site [Bhosale et al. 1998], [Reynolds 1991].

Figure (4.9) shows also that, the value of magnetization decreased after \(\gamma\)-irradiation for all investigated samples. This decrease could be explained on the bases that the formation of Fe\(^{2+}\) ion with magnetic moment (4 \(\mu_B\)) at the expense of Fe\(^{3+}\) with magnetic moment (5 \(\mu_B\)) [Ahmad et al. 2002], [Hemeda et al. 2003], due to irradiation, this leading to decrease the value of magnetization.
The variation of relative permeability ($\mu_r$) with the applied magnetic field before and after $\gamma$-irradiation is shown in figure (4.10) where $\mu_r$ is given by the expression

$$\mu_r = \frac{B}{\mu_0 H}$$

(4.3)

In general, the behavior of $\mu_r$ could be divided into two regions. In the first region, $\mu_r$ increases with magnetizing field until it reaches a maximum value. According to the expression of $\mu_r$, the increase of $\mu_r$ could be related to the ordering effect of the spins as a result of the applied field where a very rapid increase in the flux density ($B$) occurs for small values of $H$. In the second region, $\mu_r$ decreases with increasing $H$. In this region, the alignment effect of the field continues but with a smaller increase of $B$ relative to that of the first region. Accordingly, the relative permeability decreases. The decrease of the relative permeability after $\gamma$-irradiation could be attributed to that the relative permeability is direct proportional to magnetization as mentioned above in magnetization.
Fig. 4.10 The variation of $\mu_r$ with the H before and after $\gamma$-irradiation for $x=0.0$, 0.2 and 0.4.
4.2.2 Frequency dependence of initial permeability

Figures (4.11), (4.12) and (4.13) show the frequency dependence of the initial permeability ($\mu_i$) at room temperature for unirradiated and $\gamma$-irradiated samples with $x = 0.0$, 0.2 and 0.4 Mg content respectively. A resonance peak within frequency range: 1-3 MHz has been noticed. It is observed that, $\mu_i$ decreased after $\gamma$-irradiation for all investigated samples and there are a shift in the peak position to the lower frequency for $x = 0.0$ and $x = 0.2$ and to the higher frequency for $x = 0.4$. The peak deviation after $\gamma$-irradiation near 1 MHz may be returned to the distortions of the spinel structure cased by $\gamma$-rays [Dalal 2005]. The composition dependence of $\mu_i$ is illustrated in figure (4.14). It is obvious that, as Mg-concentration increase the value of $\mu_i$ decreases.

![Fig. 4.11 The initial permeability vs. frequency at room temperature for the sample with $x = 0.0$ Mg content before and after $\gamma$-irradiation.](image_url)
Fig. 4.12 The initial permeability vs. frequency at room temperature for the sample with $x = 0.2$ Mg content before and after $\gamma$-irradiation.

Fig. 4.13 The initial permeability vs. frequency at room temperature for the sample with $x = 0.4$ Mg content before and after $\gamma$-irradiation.
Fig. 4.14 The initial permeability at $f = 40$ kHz vs. Mg content for \(\text{Mg}_{x}\text{Cu}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4\) ($x=0.0, 0.2$ and $0.4$ Mg content) before and after $\gamma$-irradiation.

The effects of both Mg-concentration and $\gamma$-irradiation on the value of $\mu_i$ could be explained in the light of domain theory [Globus et al. 1968]. According to this theory, the initial permeability results from two main sources: domain wall motion ($\mu_w$) and spin rotation ($\mu_{\text{rot}}$) such that,

$$\mu_i = \mu_w + (\mu_{\text{rot}} - 1)$$  \hspace{1cm} (4.4)

However, the value of $\mu_w$ is much greater than $\mu_{\text{rot}}$ ($\mu_w \gg \mu_{\text{rot}}$) according to the Globus's model [Globus 1977], [Kakkar et al. 1996], [Suryawnshi et al. 1999] and [Sankpal et al. 1998] neglecting the spin...
rotation. So, the permeability in ferrites is mainly due to \((\mu_w)\) which given by the formula;

\[
(\mu_i - 1)\mu_w \approx (3\pi M_s^2 D)/4(\Gamma)
\]  

(4.5)

where, \(M_s\) is the saturation magnetization, \(D\) is the grain size and \(\Gamma\) is the magnetic domain wall energy which is proportional to the global anisotropy constant \(k\). Thus, the formula (4.5) could be approximated to,

\[
\mu_i \approx \frac{M_s^2 D}{k}
\]  

(4.6)

According to this equation, \(\mu_i\) is directly proportional to the magnetization and the grain size and inversely proportional to \(k\). The behavior of \(\mu_i\) with Mg-concentration could be explained in terms of equation (4.6). Where, as seen in section (4.2.1) and table (4.1), both magnetization and the crystallite size decrease with Mg concentration. The decrease of both \(M\) and \(d\), which proportional to the grain size \(D\), with Mg-concentration, leading to decrease of \(\mu_i\) as Mg-concentration increase, is in agreement with the behavior that are reported by [Rezlescu et al. 1998] and [Zhenxing et al. 2001]. On the other hand, the value of \(k = -2.5 \times 10^{-4}\) for Mg Fe\(_2\)O\(_4\) [Rado et al. 1956] is lower than that of Cu Fe\(_2\)O\(_4\) \(k = -6 \times 10^{-4}\) [Okamura et al. 1952] this leading to increase \(\mu_i\) as Mg-concentration increase. Thus, the obtained behavior of \(\mu_i\) with \(x\) reveals that the effect of both \(M\) and \(D\) dominates that of \(k\).

The decrease of \(\mu_i\) due to irradiation could be explained also according to eq. (4.6). From table (4.1), it is clear that, the average crystallite size for \(\gamma\)-irradiated samples is smaller than that of
unirradiated ones. Furthermore, the magnetization decreased by irradiation for all the investigated samples, see (4.2.1). Also, it was reported that, the anisotropy field in ferrites results from the presence of Fe\textsuperscript{2+} ions, which is formed mainly during the sintering process \cite{Chikazumi1964, Gorter1954} and its concentration increased by irradiation. Thus, as a result of irradiation process, the values of both M and D are decreased while the value of k is increased leading to decrease \( \mu_i \). In addition to that, the irradiation increases the porosity and the internal strain as reported by \cite{Ateia2006}, which are considered obstacles for the domain-wall motion leading to decrease in the value of initial permeability.

4.2.3 Temperature dependence of initial permeability and Curie temperature

Figure (4.15) shows the variation of \( \mu_i \) with temperature T (\(^\circ\)C) for unirradiated and \( \gamma \)-irradiated samples. It is found that, the curves are typical of multidomain grains showing a sudden drop in \( \mu_i \) at \( T_C \). Curie temperature (\( T_C \)) is determined by drawing a tangent for the curve at the rapid decrease of \( \mu_i \). The intersection of the tangent with the T-axis determines \( T_C \). From figure (4.15), it is noticed that, the permeability increases gradually with increasing temperature up to maximum value and at certain peak there are a sharp drop to zero at Curie temperature (\( T_C \)). This result may be explained as follows: the anisotropy field seems to decrease with temperature much faster than the magnetization \cite{Globus1976, Smit1959} and \cite{Sattar2002}. Hence, the magnetic moment rotate more easily in the direction of external magnetic field leading to increase the
magnetization value. Thus, the highest value of $\mu_i$ corresponding to zero anisotropy field \cite{Bhosale1995, Smit1959}. For temperature higher than Curie temperature, random orientation of the magnetic spins takes place (paramagnetic state) which leads the magnetization to be zero.

Figure (4.15) shows also that, the rate by which $\mu_i$ decreases with temperature at $T_C$ after $\gamma$-irradiation was lower than that before irradiation. This could be explained as follows: It was reported that, the sharp decrease of $\mu_i$ with temperature at $T_C$ reflects the homogeneity of the sample, which can be expressed as $|\Delta\mu_i/\Delta T|$ at $T_C$ \cite{Cedillo1980, Sattar2002}. From table (4.1), one can notice that, the values of $|\Delta\mu_i/\Delta T|$ for irradiated samples are lower that than of unirradiated ones, therefore, the homogeneity of irradiated samples are less than that of unirradiated ones. This could be attributed to two factors:

1. The increase of the porosity and internal strain after $\gamma$-irradiation.
2. The decrease of the crystallite size, after $\gamma$-irradiation, leading to increase the number of grain boundaries.
Fig. 4.15 Temperature dependence of initial permeability of Mg$_x$Cu$_{0.53}$Zn$_{0.5}$Fe$_2$O$_4$ ferrite system ($x=0.0$, 0.2 and 0.4 Mg content) before and after $\gamma$-irradiation.
Table (4.2). Values of $|\Delta \mu_i/\Delta T|$ with Mg content for Mg$_x$Cu$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ ferrites before and after $\gamma$-irradiation.

<table>
<thead>
<tr>
<th>Mg content</th>
<th>Unirradiated</th>
<th>Irradiated</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x$</td>
<td>$97$</td>
<td>$73$</td>
</tr>
<tr>
<td>$0.0$</td>
<td>$114$</td>
<td>$99$</td>
</tr>
<tr>
<td>$0.2$</td>
<td>$134$</td>
<td>$125$</td>
</tr>
</tbody>
</table>

The composition dependence of $T_C$ for unirradiated and irradiated samples is shown in figure (4.16). It is clear that, $T_C$ decreases with increasing Mg-concentration before and after $\gamma$-irradiation. This behavior agrees well with the other studies by different authors [Zhenxing et al. 2001], [Bhosale et al. 1997]. Also, the value of $T_C$ decreased by irradiation for all Mg-concentration with significant decrease for the sample with $x = 0.0$.

The variation of $T_C$ with both compositions and irradiation could be explained using the pair model. According to this model, the exchange energy, which determines the Curie temperature, depends on the following factors [Chikazumi 1964].

- The distance between the spin magnetic moments.
- The exchange energy integral.
- The angle between the spin moments.
- The magnitude of the spin magnetic moments.
Fig. 4.16 Curie temperature vs. Mg-concentration before and after γ-irradiation.

Thus, the behavior of $T_C$ with Mg-concentration could be attributed to the decrease in the magnitude of magnetic moments by replacing the magnetic Cu$^{2+}$ ions by the non-magnetic Mg$^{2+}$ ions. Also, the Curie temperature decreased due to irradiation as a result of the decrease in the value of magnetic moments, as shown in magnetization, and the increase of the separating distance between the moments, as shown in lattice parameter. The significant decreasing in the values of $\mu_i$ and $T_C$ for the sample with $x = 0.0$ due to irradiation may be attributed to the lattice distortion which was observed clearly by the XRDP for that sample.

It is worthy to note that, this part of work is extracted and published in the Journal of Materials Science under the title "Effect of γ-rays irradiation on the structure and magnetic properties of Mg–Cu–Zn ferrites", see appendix.
4.3 Electrical Properties

4.3.1 DC Resistivity at room temperature

Figure (4.17) shows the variation of DC resistivity ($\rho_{DC}$) at room temperature with Mg-concentration ($x$) for Mg$_x$Cu$_{0.5-x}$Zn$_{0.5}$Fe$_2$O$_4$ ($x=0.0, 0.2$ and $0.4$) before and after $\gamma$-irradiation. From figure (4.17), it is obvious that, the resistivity increases with increasing Mg-concentration and their values for irradiated samples are lower than that of unirradiated ones. These results could be explained as follows: It was reported that the main factors, that affected the value of resistivity ($\rho_{DC}$) in ferrites, are the amount of Fe$^{2+}$ ions and the porosity [Miyata 1961], [Sattar et al. 2005]. It was found that the electrical resistivity is inversely proportional to the amount of Fe$^{2+}$ ions. The decrease of Fe$^{2+}$ ions concentration limits the hopping probability of the electrons between Fe$^{2+}$ and Fe$^{3+}$ ions in B-sites, [Mason et al. 1981]. Furthermore, the resistivity is directly proportional to the porosity. The increasing of porosity hinders the motion of the charge carriers. According to [Murthy et al. 2001] and [Zhenxing et al. 2001], in the case of Mg–Zn–Cu ferrite, 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,

$$\text{Fe}^{3+} + \text{Cu}^{+} \leftrightarrow \text{Fe}^{2+} + \text{Cu}^{2+}$$

Under oxidizing conditions, the tendency is towards the right side. Thus, some Fe$^{2+}$ can be formed in ferrite to increase the probability of electron hopping. Therefore, in Mg$_x$Cu$_{0.5-x}$Zn$_{0.5}$Fe$_2$O$_4$ 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 as shown in the figure (4.17).

Fig. 4.17 DC resistivity at room temperature as a function of Mg-concentration.

The decreasing of $\rho_{DC}$ values by $\gamma$-irradiation could be attributed to the increase in the ratio of $\text{Fe}^{2+}/\text{Fe}^{3+}$ on B-sites [Goldenman 1993], [Ateia 2006]. Thus, the decrease of $\rho_{DC}$ after $\gamma$-irradiation indicates that the effect of increasing the $\text{Fe}^{2+}/\text{Fe}^{3+}$ ratio on B-sites dominate that of porosity. Similar behavior was reported by several authors for different ferrites [Hemeda et al. 2003], [Dalal 2005], [Hamada 2004], [Ateia 2006].
4.3.2 Temperature dependence of the DC resistivity

Figures (4.18), (4.19) and (4.20) show the temperature dependence of the DC resistivity, before and after γ-irradiation, expressed as log ρ_{DC} versus 1000/T for Mg_{x}Cu_{0.5-x}Zn_{0.5}Fe_{2}O_{4} ferrite with x = 0.0, 0.2 and 0.4 Mg-content respectively. It is obvious that, the electrical resistivity of all samples decreases with increasing temperature, i.e. the resistivity exhibits a normal semiconductor behavior which could be described by Arrhenius relation \[ Smit (1) 1959 \]

\[ \rho = \rho_o \exp \left( \frac{E_\rho}{K T} \right) \]  \hspace{1cm} (4.7)

where, E_\rho is the activation energy, K is Boltzmann’s constant and \( \rho_o \) is temperature independent constant. Moreover, one can notice that each curve could be divided into three regions. Each region has different activation energy (E_\rho). For all samples, the first region ranged from room temperature up to nearly 335 K. The conduction phenomenon in this region is attributed to the presence of impurities i.e. extrinsic conduction mechanism [Joshi et al. 1987], [Patil et al. 1996], [Ravinder 2000]. The formation of such impurities is due to the oxygen loss during the sintering process. The loss of oxygen leads to the formation of Fe^{2+} ions on the account of Fe^{3+} ions for charge compensation. These Fe^{2+} ions act as donor centers [Bhise et al. 1996], [Patil et al. 1994]. On the other hand, the transition temperature \( T_\rho \) between the second and the third regions are determined for the investigated samples and given in table (4.3). It is observed that, for all samples, the transition temperature, before and
after γ-irradiation, $T_\rho$ have values close to those determined from the magnetic measurements $T_C$ (sec. 4.2.2). Thus, $T_\rho$ could be considered the transition temperature between the ferri and para magnetic regions indicates that $\rho_{DC}$ is sensitive to the magnetic transition.

Table (4.3) Variation of $T_C$ for $\text{Mg}_x \text{Cu}_{0.51-x} \text{Zn}_{0.5} \text{Fe}_2 \text{O}_4$ before and after γ-irradiation with Mg content.

<table>
<thead>
<tr>
<th>Mg content x</th>
<th>Before irradiation</th>
<th>After γ-irradiation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$T_C$ (°C)</td>
<td>$T_C$ (°C)</td>
</tr>
<tr>
<td></td>
<td>mag.</td>
<td>elect.</td>
</tr>
<tr>
<td></td>
<td>$T_C$ (°C)</td>
<td>$T_C$ (°C)</td>
</tr>
<tr>
<td></td>
<td>mag.</td>
<td>elect.</td>
</tr>
<tr>
<td>0.0</td>
<td>193</td>
<td>196</td>
</tr>
<tr>
<td>0.2</td>
<td>140</td>
<td>142</td>
</tr>
<tr>
<td>0.4</td>
<td>115</td>
<td>118</td>
</tr>
</tbody>
</table>

Table 4.3: Variation of $T_C$ for $\text{Mg}_x \text{Cu}_{0.51-x} \text{Zn}_{0.5} \text{Fe}_2 \text{O}_4$ before and after γ-irradiation with Mg content.
The change in the activation energy at $T_{\rho}$ could be attributed to a magnetic transition from the ferrimagnetic to the paramagnetic state. The activation energies in the second region are denoted by $E_{f}$ (activation energy in the ferrimagnetic region), while these in the third region are denoted by $E_{p}$ (activation energy in the paramagnetic region) as indicated in table (4.4). The values of the activation energies are determined by plotting the tangent of the log $\rho_{DC}$ in the three regions that equal $E_{\rho}/K$. From table (4.4) it is clear that, the activation energy in the paramagnetic region $E_{p}$ (region (III)) are greater than that ferrimagnetic one $E_{f}$ (region (II)) for all investigated samples. This increase in activation energy due to magnetic transition is explained as follows: according to [Goodenough 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 expansivity [Zemansky 1952] i.e. an increase in the jumping length between the ions and hence lead to an increase activation energy.

Table (4.4) Variation of activation energy ($E_{f}$ and $E_{p}$) for Mg$_{x}$Cu$_{0.51}$Zn$_{0.5}$Fe$_{2}$O$_{4}$ before and after $\gamma$-irradiation.

<table>
<thead>
<tr>
<th>$E_{p}$ (eV) region (III)</th>
<th>$E_{p}$ (eV) region (III)</th>
<th>$E_{f}$ (eV) region (II)</th>
<th>$E_{f}$ (eV) region (II)</th>
<th>Mg content $x$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>before $\gamma$-irradiation</td>
<td>after $\gamma$-irradiation</td>
<td>before $\gamma$-irradiation</td>
</tr>
<tr>
<td>0.212</td>
<td>0.230</td>
<td>0.163</td>
<td>0.165</td>
<td>0.0</td>
</tr>
<tr>
<td>0.195</td>
<td>0.232</td>
<td>0.122</td>
<td>0.165</td>
<td>0.2</td>
</tr>
<tr>
<td>0.192</td>
<td>0.208</td>
<td>0.168</td>
<td>0.169</td>
<td>0.4</td>
</tr>
</tbody>
</table>

It is also noticed that the values of activation energy decreased after $\gamma$-irradiation in the three regions. This decrease may be attributed to
generation of some vacancies at different depths, which acts as trapping centers cause a depressing of the jumping length leading to decrease the activation energy [Ahmed et al. 2003], [Ateia 2006], [Ahmed et al. 2007].

4.3.3 Frequency dependence of AC resistivity

The variation of the AC resistivity ($\rho_{\text{AC}}$) at room temperature with frequency, before and after $\gamma$-irradiation, is shown in figures (4.21), (4.22) and (4.23). It is observed that, $\rho_{\text{AC}}$ decreases with increasing frequency and its value for irradiated samples are lower than that of unirradiated ones. Similar behavior of AC resistivity with frequency for different ferrites was reported by [Bellad et al. 2000], [Lipare et al. 2004].

Fig. 4.21 Variation of the AC resistivity ($\rho_{\text{AC}}$) with frequency for $\text{Mg}_x \text{Cu}_{0.5-x} \text{Zn}_{0.5} \text{Fe}_2 \text{O}_4$ sample with $x = 0.0$ Mg content before and after $\gamma$-irradiation.
Fig. 4.22 Variation of the AC resistivity ($\rho_{ac}$) with frequency for Mg$_x$Cu$_{0.5-x}$Zn$_{0.5}$Fe$_2$O$_4$ sample with $x = 0.2$ Mg content before and after $\gamma$-irradiation.

Fig. 4.23 Variation of the AC resistivity ($\rho_{ac}$) with frequency for Mg$_x$Cu$_{0.5-x}$Zn$_{0.5}$Fe$_2$O$_4$ sample with $x = 0.4$ Mg content before and after $\gamma$-irradiation.
These results could be explained as mentioned before with DC resistivity; the conduction mechanism in this system is due to the electron hopping between Fe$^{2+}$ ions, Fe$^{3+}$ ions which can change between the 2+/3+ state then the increase in frequency enhances the electron hopping rate and hence increases the conductivity i.e. decreases the resistivity. At high frequencies, the AC resistivity of the samples converges to the value nearly independent of frequency. It is due to the fact that the electron hopping frequency cannot follow the external electric field and thus lags behind it. 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 large and of low resistivity ($\rho_1$). The grain boundaries are thin and of high resistivity ($\rho_2$). Following Koops's model, the total impedance $\rho$ could be written as

$$\rho = \rho_1^0 + \frac{(\rho_1^0 - \rho_2^\infty) / (1 - \omega^2 \tau^2)}{\omega^\infty}$$

where the superscript 0 and $\infty$ refer to low and high frequency value respectively and $\tau$ is the relaxation time, $\omega = 2\pi f$.

According to Koops's assumption that $\rho_2 >> \rho_1$, $y << 1$ (where $y$ is the ratio of the grain boundaries thickness to the grain thickness) one can write the total impedance as

$$\rho \approx \rho_1 + \frac{y\rho_2}{1 + \left(\frac{b\rho_1\rho_2\omega^2}{y}\right)}$$

(4.9)
where \( b \) is a constant \([\text{standley} 1962]\). Thus at very high frequency, the impedance \( \rho^\infty \) is given by

\[
\rho^\infty = \rho_1
\]

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 \( \rho^0 \) is given by

\[
\rho^0 = \rho_1 + y\rho_2
\]

According to the assumption that \( y\rho_2 > \rho_1 \), the impedance at low frequency results mainly from the resistivity of the grain boundaries, which have high resistivity. According to the above discussion, it is clear that Koops's model explains satisfactorily the frequency dependence of the AC resistivity.

It is also, obvious that the AC resistivity decreases after \( \gamma \)-irradiation for all concentration. The decrease of \( \rho_{AC} \) for the investigated samples after \( \gamma \)-irradiation could be attributed, as mentioned above also with DC resistivity, to the increase of \( \text{Fe}^{2+}/\text{Fe}^{3+} \) ratio (sec. 4.3.1). The decrease of ferrite AC resistivity after \( \gamma \)-irradiation by gamma ray was reported by different authors \([\text{Ahmed et al.} 1997], [\text{Ahmed et al.} 2003], [\text{Ateia} 2006], [\text{Ahmed et al.} 2007]\).

4.3.4 Frequency dependence of real part of dielectric constant

Figures (4.24), (4.25) and (4.26) show the frequency dependence of real part of the dielectric constant (\( \epsilon \)) at room temperature for the system \( \text{Mg}_x \text{Cu}_{0.5-x} \text{Zn}_{0.5} \text{Fe}_2 \text{O}_4 \) ferrite system,
with $x = 0.0$, 0.2 and 0.4 Mg content respectively, before and after $\gamma$-irradiation. It could be seen that, $\dot{\epsilon}$ 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. Such a normal behavior was reported by [Ravinder et al. 2004], [Rezlescu et al. 1974], [Reddy et al. 1999].

Fig. 4.24 Frequency dependence of the $\dot{\epsilon}$ for Mg$_x$Cu$_{0.5-x}$Zn$_{0.5}$Fe$_2$O$_4$ sample with $x = 0.0$ Mg content before and after $\gamma$-irradiation.
Fig. 4.25 Frequency dependence of the $\varepsilon$ for $\text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ sample with $x = 0.2$ Mg content before and after $\gamma$-irradiation.

Fig. 4.26 Frequency dependence of the $\varepsilon$ for $\text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ sample with $x = 0.4$ Mg content before and after $\gamma$-irradiation.
This behavior of $\epsilon$ could be explained on the basis of the dielectric properties and space charge polarization, which are mainly governed by the conduction mechanism in ferrites \cite{Lipare2004}, wherein the electron hopping takes place. The electron hopping is favorable at low applied field frequency. Therefore, at low frequencies the dielectric constant is large. The electron exchange between Fe$^{3+}$ to Fe$^{2+}$ gives local displacement of electron in the direction of the applied electric field, which induces polarization in ferrites. Beyond a certain frequency, the electron exchange does not follow the alternating field and so the dielectric constant reaches a constant and small value \cite{Wagner1913, Murthy1976, Kharabe2006, Muhammad2007}. In addition, from the figures (4.22), (4.23) and (4.24) there is a slightly increase in the $\epsilon$ is observed after $\gamma$-irradiation. This could be explained in a view of interaction of $\gamma$-rays with the matter, which is summarized as follows, gamma irradiation interacts with ferric ions as

$$Fe^{3+} + \gamma \rightarrow Fe^{2+} + e$$  \hspace{1cm} (4.10)

This interaction creates ferrous ions at the octahedral sites and increases the ratio Fe$^{2+}$/Fe$^{3+}$ at these sites \cite{Hamada2004, Hemeda2005, Ateia2006, Ahmad2007, Mousa1989}. The jumping electrons oriented in the field direction and consequently are given rising in the value of $\epsilon$. 
4.3.5 Frequency dependence of imaginary part of dielectric constant

The variation of the imaginary part of the dielectric constant, $\varepsilon''$ (which represents the dielectric loss) with frequency for the investigated samples, before and after $\gamma$-irradiation, with frequency at room temperature are shown in figures (4.27), (4.28) and (4.29). It is observed that, $\varepsilon''$ decreases continuously with increasing the frequency up to certain frequency and then becomes frequency independent. The value of $\varepsilon''$ increases after $\gamma$-irradiation for all investigated samples. A similar decrease of the $\varepsilon''$ with frequency was reported for many other ferrites [Josyulu et al. 1980], [Venugopal et al. 1985], [Mazen 2000], [Ravinder et al. 2001]. The increase of the $\varepsilon''$ after $\gamma$-irradiation, was also reported by [Ateia 2006]. Such a decrease in $\varepsilon''$ could be discussed as follows. The core loss of ferrites consists of many factors: eddy current loss and dipole loss in addition to hysteresis loss. The eddy current loss is inversely proportional to the resistivity of ferrite. At all frequencies, the resistivities of the samples are decreased. Thus, the effect of resistivity is increase the eddy current loss. On the other hand, the dipole loss, which results from the dipole orientation (relaxation) decreases, especially at high frequencies, as the dipole orientation, cannot follow the applied field frequency. Furthermore, it was reported that the hysteresis loss is directly proportional to the frequency [Otsuki et al. 1991]. This means that the effect of both the eddy current and the hysteresis...
losses is to increase the core loss while the dipole loss decreases the core loss.

Fig. 4.27 Frequency dependence of the $\varepsilon''$ for Mg$_x$Cu$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ sample with $x = 0.0$ Mg content before and after $\gamma$-irradiation.
The competition between the different types of losses determines the behavior of $\varepsilon''$ and seems to be responsible for the
constancy of losses at high frequencies. In fact, some ferrites showed a peak in the variation of $\varepsilon''$ with frequency [Murthy et al. 1976]. The condition for observing a peak in the ($\varepsilon''$ vs. $f$) is given by the relation [Mazen 2000].

$$\omega \tau = 1$$ (4.11)

where $\omega = 2\pi f_{\text{max}}$ and $\tau$ is the relaxation time. The relaxation time $\tau$ is related to the jumping probability per unit time, $P$, by the equation $\tau = \frac{1}{2P}$ i.e. $f_{\text{max}} \propto P$ [Ravinder et al. 2002]. This relation shows that $f_{\text{max}}$ is directly proportional to hopping probability. The disappearance of $f_{\text{max}}$ in our samples implies a low hopping probability, which may be due to the high resistivity. This result is in agreement with those previously reported for Li-Mg-Ti ferrites where a maximum in loss was observed only for low resistive composition [Bellad et al. 2000]. Moreover, the low probability of hopping means that the relaxation times for our investigated samples seem to be large at room temperature so the frequency position of the peak is smaller than the range of our measurement (45 Hz – 5 MHz). Hence, the value of $\varepsilon''$ in our results are after the peak position, so $\varepsilon''$ decreases with frequency. The increase of $\varepsilon''$ after $\gamma$-irradiation could be attributed to the resistivities decreased by irradiation process and the effect of resistivity is increase the eddy current loss then $\varepsilon''$ increases which is in a good agreement with the relation [Lipare et al. 2004].

$$\rho_{\text{AC}} = \frac{1}{\varepsilon' \varepsilon_0 \omega}$$ (4.12)

4.3.6 Composition dependence of the dielectric properties

Figure (4.30) represents the composition dependence of $\rho_{\text{AC}}$, $\varepsilon'$ and $\varepsilon''$ (at frequency = 300 Hz) for the $\text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ system, with $x$
= 0.0, 0.2 and 0.4 Mg content respectively. This figure shows that $\rho_{AC}$ increase with increasing Mg content for all investigated samples. This behavior is similar to that for $\rho_{DC}$ and hence could be discussed as mentioned above with $\rho_{DC}$ (sec. 4.3.1). On the other hand, $\varepsilon$ and $\varepsilon''$, before irradiation, have almost a reverse trend to $\rho_{AC}$. The inverse relation between $\rho_{AC}$ and $\varepsilon$ has been reported by [Bellad et al. 1999], [Radha et al. 1995], [Reddy et al. 1991].
Fig. 4.30 Composition dependence of the $\rho_{AC}$, $\varepsilon'$ and $\varepsilon''$ for Mg$_x$Cu$_{0.5-x}$Zn$_{0.5}$Fe$_2$O$_4$ ferrite system $x = 0.0$, $0.2$ and $0.4$ Mg content respectively, before and after $\gamma$-irradiation.
The inverse trend of $\rho_{\text{AC}}$ with $\varepsilon$ could be explained on the basis of the relation between the mobility of the electron hopping and resistivity

$$\sigma = \frac{1}{\rho} = ne\mu$$  \hspace{1cm} (4.13)

where, $\mu$ and $n$ are the mobility and the concentration 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. $\varepsilon$ increases. Moreover, the reverse behavior of the $\rho_{\text{AC}}$ and $\varepsilon''$ is expected as the increase of resistivity decreases the loss $\varepsilon''$ and vice versa which is in a good agreement with the eq. (4.12).
Conclusion

In this study, the physical, electrical and magnetic properties of the system $\text{Mg}_x\text{Cu}_{x-0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$, $x = 0.0$, $0.2$ and $0.4$ Mg content were investigated before and after $\gamma$-irradiation. According to the obtained results, the following conclusion could be drawn.

- All the investigated samples revealed spinel single phase structure before and after $\gamma$-irradiation.
- The XRDPs spectra of the samples after $\gamma$-irradiation showed peaks shift and conversions in the relative intensity of the peaks. These results were attributed to the distortion occurred in the cubic lattice by irradiation.
- The crystallite size was found to be decreased with Mg content and after $\gamma$-irradiation process.
- Both the lattice parameter and porosity decreased with Mg content while they increased after $\gamma$-irradiation in each composition.
- Magnetization, initial permeability and Curie temperature decreased with Mg-content and after $\gamma$-irradiation.
- The results of initial permeability and Curie temperature before and after $\gamma$-irradiation were explained according to domain theory and pair model respectively.
- Curie temperature was found to be decreased from $195$ °C to $165$ °C for the sample of $x=0.0$ and significant decreasing in permeability was noticed for all investigated samples.
- The homogeneity of the samples increased with Mg-content while it was decreasing after $\gamma$-irradiation.
Conclusion

- The DC resistivity increased as Mg-concentration increase while it was decreasing after $\gamma$-irradiation for all investigated samples. The decrease in $\rho_{DC}$ i.e. conductivity increase, after $\gamma$-irradiation is attributed to the increase in the ratio of $\text{Fe}^{2+}/\text{Fe}^{3+}$ on B-sites as consequence of hopping reaction.

- The temperature dependence of $\rho_{DC}$ showed two conduction mechanisms, band mechanism for $T < T_c$ and hopping conduction mechanism for $T > T_c$. It is also noticed that the values of activation energy decreased after $\gamma$-irradiation in every region.

- Dispersion curves of the dielectric constant $\varepsilon$, dielectric loss $\varepsilon''$ and $\rho_{AC}$ with frequency were found to be decreased with frequency for each composition. The values of $\varepsilon$ and $\varepsilon''$ decreased also after $\gamma$-irradiation. These results were explained in the light of two layers model (Koops’s model).

- The increase of the conductivity, dielectric constant and dielectric loss after $\gamma$-irradiation is related to the increase of the ratio $\text{Fe}^{2+}/\text{Fe}^{3+}$.

- This result reveals that the structure, the magnetic and the electrical properties of Mg-Cu-Zn ferrites are highly affected by irradiation.
References


References


Globus, Proceeding of ICF2, September (1976).


Hak P.J., Ho K.J., Hee C.S., in: Proceedings of the Seventh
International Conference on Ferrites, Bordeaux, France, (1996)
348.


63.


Jilies D. "Introduction to Magnetism and Magnetic Materials",

1694.


415.


Karim Asif, Shirsath Sagar E., Shukla S. J., Jadhav K. M., Nucl.

Kharabe R.G., Devan R.S., Kanamadi C.M., Chougule B.K.,

Koops C.G., Phys. Rev. 121 (1951) 83.
Kramers H., physica, 1 (1934) 182.


Maxwell J.C., Electricity and Magnetism, vol. 1, Oxford University Press, Oxford, 1929 (Section 328) p 752.


References


References

Tareev(1) B., “Physics of Dielectric Materials”, Mir Publisher, Moscow (1979) 34.


Zemansky C. Phys. Rev. 82(1952) 403.
