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Article

Effect of Copper Doping on the Structural, Electrical, and Magnetic Properties of Mg-Co Ferrites

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Abstract: This research paper focuses on the synthesis and characterization of Cu-doped Mg-Co ferrites with varying compositions ($\text{Mg}_{0.6-x}\text{Co}_{0.4}\text{Cu}_x\text{Fe}_2\text{O}_4$, where $x = 0.0, 0.1, 0.2,$ and 0.3) using a solid-state reaction method. This paper also investigates the structural, morphological, magnetic, and electrical characteristics of synthesized $\text{Mg}_{0.6-x}\text{Co}_{0.4}\text{Cu}_x\text{Fe}_2\text{O}_4$, where $x = 0.0, 0.1, 0.2,$ and 0.3 . The X-ray diffraction (XRD) technique confirms the ferrites spinel structure in the Fd-3m space group, with average crystallite sizes ranging from 57.29 to 48.57 nm. Fourier Transform Infrared (FTIR) spectroscopy verifies chemical and structural changes, while scanning electron microscopy (SEM) reveals cubic crystal growth with an average grain size of 1 to 1.5 μm . DC electrical resistivity decreases with increasing temperature and Cu^{2+} substitution, ranging from $1.4 \times 10^6 \Omega\text{-cm}$ to $6.7 \times 10^5 \Omega\text{-cm}$. The study suggests a correlation between resistivity and Cu^{2+} concentration. Magnetic behavior, studied using a Vibrating Sample Magnetometer (VSM), shows dependence on dopant concentration, with coercivity ranging from 157 Oe to 256 Oe. The results indicate potential applications in magnetic storage devices, antennas, transformers, and high-frequency electronics.

Keywords: Cu-doped Mg-Co ferrites; solid-state reaction method; structural characterization; morphological characterization; magnetic properties; electrical resistivity; magnetic storage devices; high-frequency electronics

1. Introduction

Ferrites are the class of materials having general formula, MFe_2O_4 ($\text{M} =$ divalent metal ions; examples include Mg, Zn, Ni, Co, etc.). Depending on their structure, composition and morphology, ferrites exhibit interesting optical, magnetic, and electrical properties which make them a suitable candidate for applications in various fields [1]. Among various ferrite materials, magnesium ferrite (MgFe_2O_4) is one such important ferrite material having a spinel structure with inversion mode. In normal spinel ferrite, divalent metal ions and trivalent metal ions occupy tetrahedral and octahedral sites respectively [2]. In inverse spinel ferrites, maximum trivalent ions occupy tetrahedral sites, and the remaining trivalent and divalent ions occupy octahedral sites [3]. Ferrites can be doped with additional metal ions, and each additional metal ion affects the chemical formula and physical and chemical properties of the material [4].

Magnesium ferrite due to its high curie temperature, high saturation magnetization, and low coercivity is called soft ferrite and is commonly utilized in high-frequency electronic devices and microwave applications, etc. [5]. Cobalt ferrite (CoFe_2O_4) has an inverse spinel structure and due to its higher coercivity than Mg ferrite, it is also known as a hard ferrite [6]. Cobalt doping improves the coercivity of magnesium ferrite. Additionally, a unit cell of magnesium ferrite shrinks because of cobalt's lower ionic radius [7]. In comparison to other ferrites, Mg-Co ferrite has a high coercivity, moderate saturation magnetization, strong chemical stability, high mechanical hardness, and inexpensive production costs. It is also known as a good candidate for nuclear magnetic resonance and as a photocatalyst. By doping additional metal ions into Mg-Co ferrite, different magnetic materials can be synthesized with unique structures, physical and chemical properties. Metal-doped

magnesium cobalt ferrites have gained attention due to their exceptional electrical and magnetic properties, making them ideal for high-frequency applications [8], such as in the fabrication of transformers where they are integral in reducing signal noise and enhancing signal quality. These materials can also be used in the development of highly sensitive magnetic sensors for applications like automotive and industrial sensing.

Ferrite cores are crucial in Sound Detection and Ranging (SODAR) pre-amplifiers for significantly reducing signal noise, enhancing the accuracy and reliability of environmental monitoring. These cores, made from magnesium and cobalt ferrites, are selected for their high saturation magnetization and custom-tailored coercivity. By doping these ferrites with specific metals like cobalt to boost coercivity or copper to modify electrical properties, their effectiveness in suppressing electromagnetic interference is maximized [9]. This reduction of noise signals is essential for improving the fidelity of sound wave detection and analysis in SODAR systems, demonstrating the unique application of ferrite materials in high-frequency and noise-sensitive technologies, where signal clarity and noise reduction are critical.

The effectiveness of ferrite cores in suppressing electromagnetic interference (EMI) in cables and wires, which is analogous to their role in SODAR systems, has been thoroughly investigated [10,11]. Additionally, advancements in the performance of sense-amplifier circuits through pre-amplification strobing and noise-matched clipping further highlight the technological importance of minimizing noise for enhanced signal clarity [12]. The prediction and analysis of the noise reduction effect of ferrite beads on electromagnetic emission from digital PCBs also provides insight into the broader applications of ferrites in EMI suppression [13].

The present study reports the synthesis of Cu^{2+} ions substituted Mg-Co ferrites ($\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ with $x = 0.0, 0.1, 0.2, \text{ and } 0.3$) by a solid-state reaction method and evaluate the effect of Cu^{2+} substituting on the structural, dc electrical, and magnetic properties of magnesium-cobalt ferrite and to find a correlation between them. The solid-state method, widely used in materials science, involves directly reacting powdered reactants at high temperatures. It offers simplicity, cost-effectiveness, and the production of phase-pure products. This method enables thorough mixing at the atomic level, resulting in homogeneous and finely divided products. Different synthesis processes have specific advantages and considerations, emphasizing the importance of selecting the appropriate method based on desired characteristics, scalability, and application requirements. Solid state synthesis offers precise control over crystallinity and phase purity, making it suitable for applications needing accurate stoichiometry and well-defined crystal structures.

The structural and morphological properties of the synthesized $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ have been investigated by using standard characterization techniques. DC electric resistivity of synthesized ferrite material has been evaluated by a two-probe method and the magnetic measurement has been done using a Vibrating sample magnetometer (VSM). The results of this comprehensive study suggest that the frequency magnetic and electronic device applications, including those in Sound Detection and Ranging (SODAR), offering advancements in signal quality and noise reduction in such sophisticated technologies.

It's pertinent to note that while this investigation seeks to improve ferrites noise reduction capabilities in SODAR applications, it specifically concentrates on the effects of Cu^{2+} ion substitution within a defined range. The findings of this research are primarily applicable to the development and application of ferrite cores in SODAR amplifier systems. Future studies could extend this scope to include other doping elements and investigate their impact on ferrite properties, potentially paving the way for new applications of ferrite materials in high-frequency and noise-sensitive technologies.

2. Materials and Methods

2.1. Chemicals

High-purity analytical grade (A.R) chemicals of transition metal oxides were used for standard ceramic synthesis. Magnesium Oxide ($\text{MgO} \geq 99.99\%$), Ferric oxide ($\text{Fe}_2\text{O}_3 \geq 99.99\%$), Copper oxide ($\text{CuO} \geq 99.99\%$), Cobalt (II) oxide ($\text{CoO} \geq 99.99\%$) were purchased from Himedia. Acetone ($\text{C}_3\text{H}_6\text{O}$),

Potassium Bromide (KBr), Silver (Ag), and Poly Vinyl Alcohol (C_2H_4O)_x were purchased from Merck. All the chemicals were used without further purification.

2.2. Synthesis of Cu^{2+} Substituted Mg-Co Ferrites

Cu^{2+} substituted $Mg_{0.6-x}Cu_xCo_{0.4}Fe_2O_4$ ($x = 0.0, 0.1, 0.2,$ and 0.3) were prepared by the solid-state reaction method. Analytical grade oxides MgO, Fe₂O₃, CuO, and CoO, were taken in stoichiometric proportions and grounded for 6 h in an agate motor and pestle to obtain in powder form. The powder is then calcined at 900 °C for 4 h. After mixing a few drops of PVA as a binder, the calcined samples are pressed to a disk-shaped pellet under a pressure of 7 tons per 5 minutes and sintered at 1200 °C for 4 h in the air at the rate of 5 °C/min. The pellet's surfaces are polished carefully, washed with acetone, and coated with silver paste for use as electrodes. Figure 1 shows the schematic of the synthesis process of Cu-doped magnesium-cobalt ferrites. The prepared samples were used for further characterization.

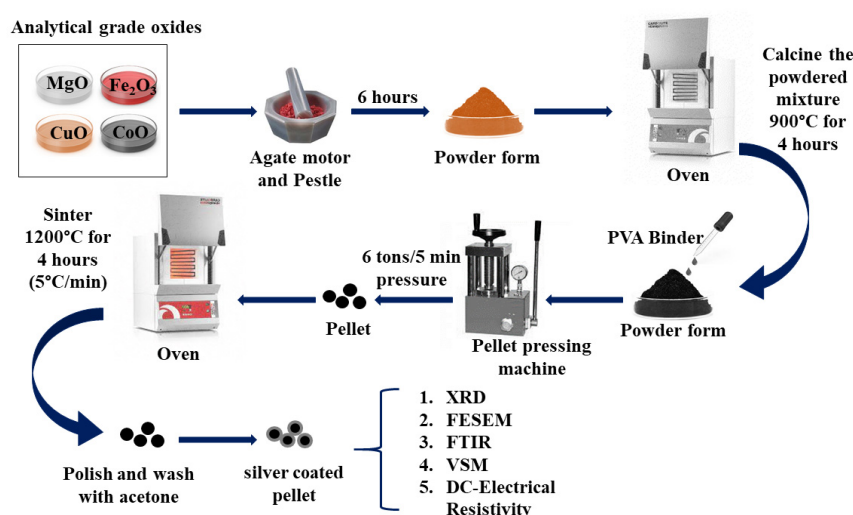


Figure 1. Schematic representation of the synthesis process of Cu-doped Mg-Co Ferrites.

2.3. Characterizations

2.3.1. Structural Characterizations

The identification of phase formation of synthesized magnesium ferrite powder samples was done on a PANalytical XPERT-PRO diffractometer fitted with Cu $K\alpha$ radiation ($\lambda = 1.54060 \text{ \AA}$) at the scan rate of $2^\circ/\text{min}$ in the 2θ range of 10 to 80 degrees. The observations were recorded at 40 mA and 45 kV. The experimental XRD patterns of the synthesized samples were matched with the standard reference data card from the Joint Committee on Powder Diffraction Standards (JCPDS) or the International Center of Diffraction Data (ICDD).

The chemical and structural changes are verified by FTIR spectrometer (IR Prestige21 Shimadzu). The synthesized ferrite powdered samples were mixed with solid potassium bromide (KBr), grounded, and then pressed in a standard hydraulic press to form a spherical shape pellet for measurements. The spectra of all the synthesized samples were collected in the range of $200\text{-}4000 \text{ cm}^{-1}$ at room temperature.

2.3.2. Morphological and Elemental Characterization

The microstructure and morphology of the synthesized magnesium ferrite samples were measured by Field Emission Scanning Electron Microscope (Carl Zeiss, EVO MA 15, Oxford Instruments, Inca Penta FETx3.JPG equipment) operating at 30 kV. Grain size measurement from the electron micrographs was performed using image processing software ImageJ version 1.53e, which

is a public domain Java-based image processing program developed at the National Institutes of Health, USA [14].

2.3.3. Magnetic Measurements

The Vibrating Sample Magnetometer (Lakeshore 735) was used to measure the magnetization behaviour of the synthesized ferrite samples. Finely powdered samples of synthesized ferrites (approximately 125 mg each) were carefully mounted on the Vibrating Sample Magnetometer (Lakeshore 735) holder. To ensure correct alignment within the magnetic field of the VSM, the samples were isolated from other metallic components to prevent any interference. An external magnetic field at a magnetic field strength of 1 Tesla was applied, and the magnetization of the vibrating samples was recorded, producing a hysteresis loop. All the measurements were conducted at room temperature to maintain stability, as temperature fluctuations could alter magnetization behaviour. Following measurements, data analysis was performed to identify key magnetic properties: coercivity, remanence, and saturation magnetization. Rigorous calibration of the VSM was maintained throughout, with thorough cleaning post-use to prevent contamination.

2.3.4. DC Electrical Resistivity

The DC electrical resistivities of the synthesized magnetic ferrite samples were determined for conductivity investigation by using a two-probe method and measured between the temperatures of 303K and 873 K. The sample in the form of a pellet was held between two electrodes. The silver paste was applied on both surfaces of the pellet to ensure good ohmic contact. The current and voltage measurements were carried out during rising temperatures. The temperature of the sample was measured using a chrome-aluminum thermocouple.

3. Results and Discussion

3.1. Identification and Interpretation of Crystal Structure by X-ray Diffraction (XRD) Analysis

The crystal structure, phase purity, and structural parameters of the synthesized samples were confirmed by analyzing their XRD patterns. Figure 2 shows the X-ray diffraction patterns of the synthesized samples. XRD patterns with the corresponding (hkl) planes; (111), (220), (311), (400), (422), (511), and (440) were observed for all the samples. The XRD patterns were well matched with the standard diffraction patterns of cubic spinel structure with the space group Fd3m for all the samples (JCPDS card no. 73-1720 and 22-1086) [15]. All the reflections are broader than those for samples of ceramic. The presence of broader peaks suggests that the size of the synthesized ferrite particles is small. No impurity peaks were observed. The XRD result confirms the synthesis of Cu²⁺ ions substituted magnesium ferrites with phase purity and small crystallite size [16].

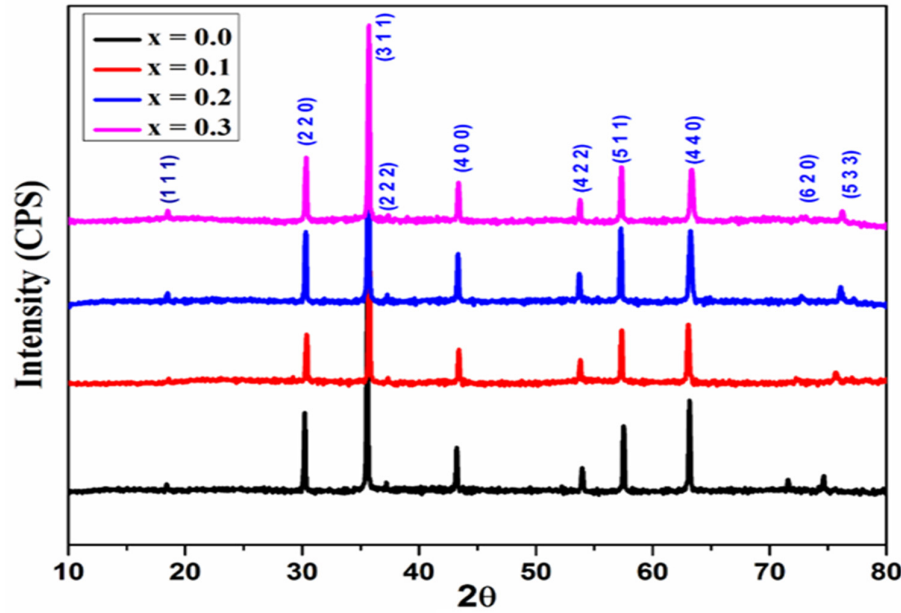


Figure 2. X-ray diffraction patterns of $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ($x = 0, 0.1, 0.2,$ and 0.3).

The lattice constants of the synthesized samples were calculated by using the equation below (Equation (1)) and the values of lattice constant (a) for the synthesized ferrite samples are reported in Table 1 below.

$$a = d \times \sqrt{(h^2 + k^2 + l^2)} \quad (1)$$

Where, d is the inter-planar spacing and $(h\ k\ l)$ are Miller indices of a cubic crystal plane.

Table 1. Lattice constant and Crystallite size of $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ($x = 0, 0.1, 0.2,$ and 0.3).

Parameters	$x = 0.0$	$x = 0.1$	$x = 0.2$	$x = 0.3$
Lattice constant a (Å)	8.3720	8.3828	8.3614	8.3912
V_{cell} (Å ³)	586.79	589.07	584.57	590.84
Crystallite size (nm)	57.29	51.32	49.91	48.57

The value of the lattice constant values in the samples show irregular changes with different levels of substitution x , in the compound. Specifically, the lattice constant grows when x , changes from 0.0 to 0.1, suggesting that the unit cell might be expanding. However, it decreases at $x=0.2$ hinting at a decrease in the space between atoms. Then, it grows again at $x=0.3$. These changes at different substitution levels indicate modification in the distance and interactions between atoms in the crystal, which can significantly affect the material's properties.

The introduction of Cu^{2+} ions, which have a larger ionic radius (0.72 Å) than Mg^{2+} ions (0.65 Å) [17], could be causing these changes in the lattice constant. This might also be related to the presence of Fe^{2+} ions, which have a larger radius (0.78 Å) than Fe^{3+} ions (0.64 Å). The differences in ionic sizes, along with changes in the shape and internal stress in the lattice, are likely causing the lattice constant to increase.

The average crystallite sizes ($D_{(311)}$) of the synthesized Cu doped Magnesium-Cobalt ferrite samples were determined by using the Debye-Scherrer formula (Equation (2)):

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (2)$$

Where D is the average crystallite size, β is the Full width at half maximum of the most substantial reflection in radians, θ is an angle corresponding to the peak position, λ is the wavelength of x-ray radiation which is equal to $\lambda=1.5406 \text{ \AA}$, and K is the shape factor of average crystallite, and its value is ~ 0.9 .

The values of crystallite sizes for the synthesized ferrites samples are shown in Table 1, which are in good agreement with earlier research [18]. The average crystallite sizes were found in the ranges between 57.29 to 48.57 nm, and the trend of this change is consistent with variations in the lattice constant as shown in Table 1. The plot of lattice constant (a) and crystallite size with Co content (x) is shown in Figure 3. The crystallite size has a random value due to the randomness in the cation distribution and sintering condition [19].

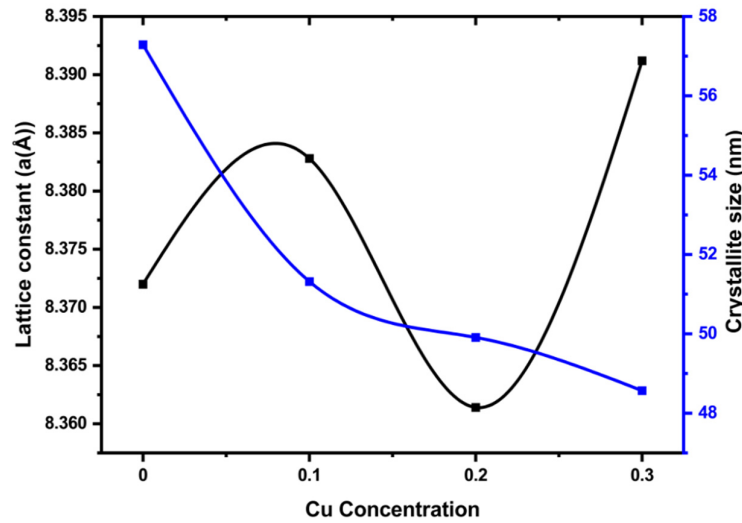


Figure 3. The plot of lattice constant and crystallite size of $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ($x = 0.0, 0.1, 0.2,$ and 0.3).

Average crystallite size is found in the ranges between 57.29 to 48.57 nm calculated using the Debye-Scherrer equation.

3.2. Fourier Transformed Infrared (FTIR) Spectroscopy

The infrared spectra of Cu^{2+} substituted $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ with $x = 0.0, 0.1, 0.2,$ and 0.3 samples are shown in Figure 4. Two absorption bands, ν_1 and ν_2 , were observed which lie between 600 and 400 cm^{-1} . These bands correspond to the lower and higher frequencies of the tetrahedral and octahedral metal-oxygen vibration respectively [20]. These bands were found in the wave number range as listed in Table 2. With increasing Cu^{2+} ion concentration, the value of ν_1 is found within the range of $572.88 - 582.52 \text{ cm}^{-1}$ and values of ν_2 in the range of $401.56 - 407.96 \text{ cm}^{-1}$. The variation in the band positions is due to the cation distribution. These band positions are in good agreement with the previous reports on spinel ferrites [21].

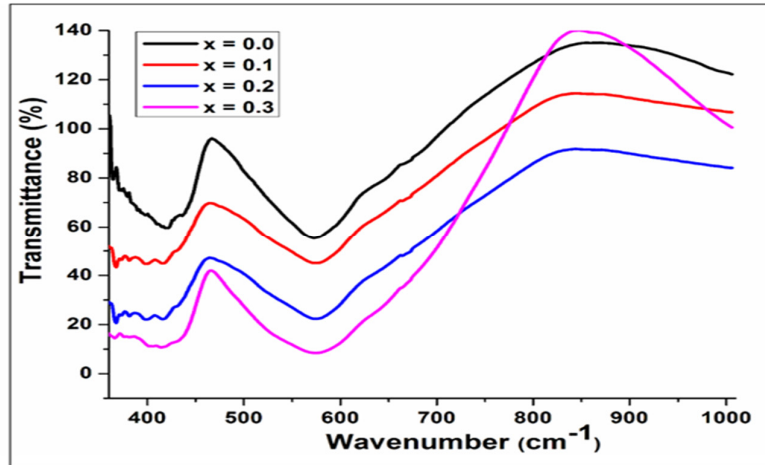


Figure 4. Fourier transform infrared spectra of $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ($x = 0, 0.1, 0.2,$ and 0.3).

Table 2. Wave number value of Cu doped $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ($x = 0, 0.1, 0.2,$ and 0.3).

Composition (x)	x = 0.0	x = 0.1	x = 0.2	x = 0.3
Tetrahedral ν_1 (cm^{-1})	572.88	574.81	579.63	582.52
Octahedral ν_2 (cm^{-1})	410.24	407.96	406.03	401.56

3.3. Assessment of Morphology Using Scanning Electron Microscopy (SEM)

SEM micrographs of the Cu^{2+} substituted $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ with $x = 0.0, 0.1, 0.2,$ and 0.3 are presented in Figure 5. The micrograph shows the growth of regular cubic crystals with the average grain size determined by using Image J software, ranging from 1 to 1.5 μm , depending on the substitution of Cu^{2+} . The microstructure of synthesized ferrite samples is significantly impacted by copper because it makes the liquid phase sintering easier. Moreover, the driving force for grain boundary movement and the restraining force produced by pores can be thought of as competitors in the process of grain growth [22]. The force generated by the thermal energy during sintering causes grain boundaries to cover pores, reducing the volume of the pores and increasing the density of the material. The uniform distribution of grain size occurs if the driving force acting on each grain is homogeneous. The non-homogeneous driving force on the grains in the current investigation is the cause of the non-uniformity of grain size [23]. The observed microstructures of synthesized materials give them suitable electromagnetic properties like permeability and magnetization.

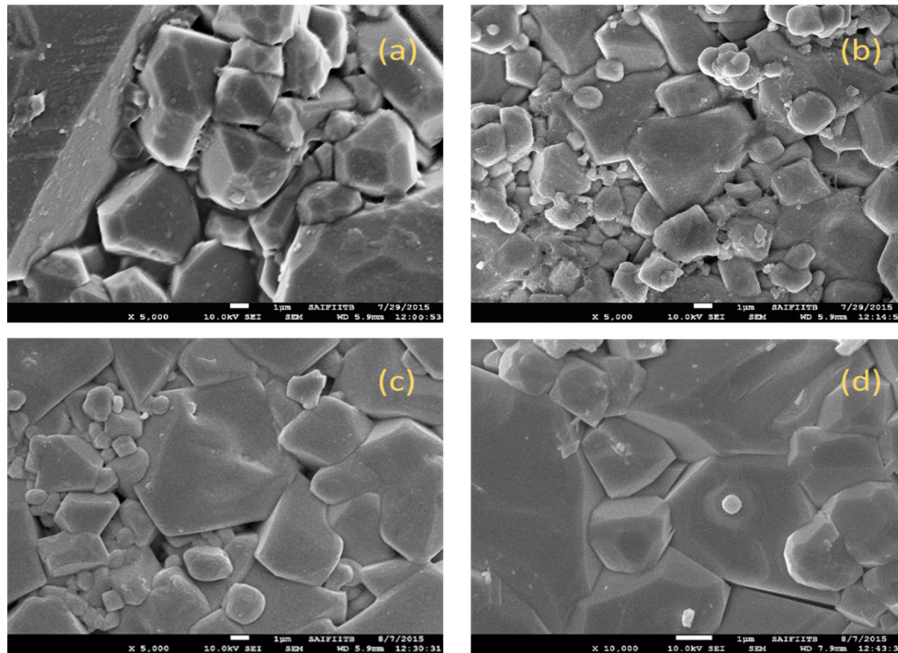


Figure 5. SEM micrographs of $Mg_{0.6-x}Cu_xCo_{0.4}Fe_2O_4$ with (a) $x = 0$, (b) $x = 0.1$, (c) $x = 0.2$, and (d) $x = 0.3$.

Synthesized ferrites show the growth of regular cubic crystals with average grain size ranges from 1 to 1.5 μm , depending on the substitution of Cu^{2+}

3.4. Magnetic Measurements

The magnetic measurements were done using a vibrating sample magnetometer (VSM), to determine the values of saturation magnetization, and coercivity of the Cu^{2+} substituted $Mg_{0.6-x}Cu_xCo_{0.4}Fe_2O_4$ ($x = 0.0, 0.1, 0.2$, and 0.3) at a magnetic field strength of 1T at room temperature. Figure 6 shows the plotted M-H curve of saturation magnetization against the applied magnetic field for different concentrations of synthesized ferrites samples. It is clear from the figure that magnetization increases with the applied magnetic field.

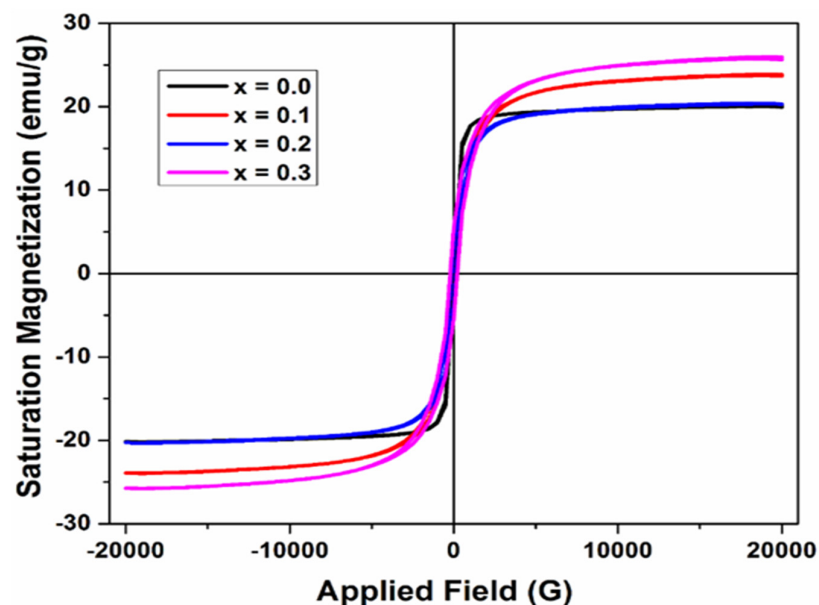
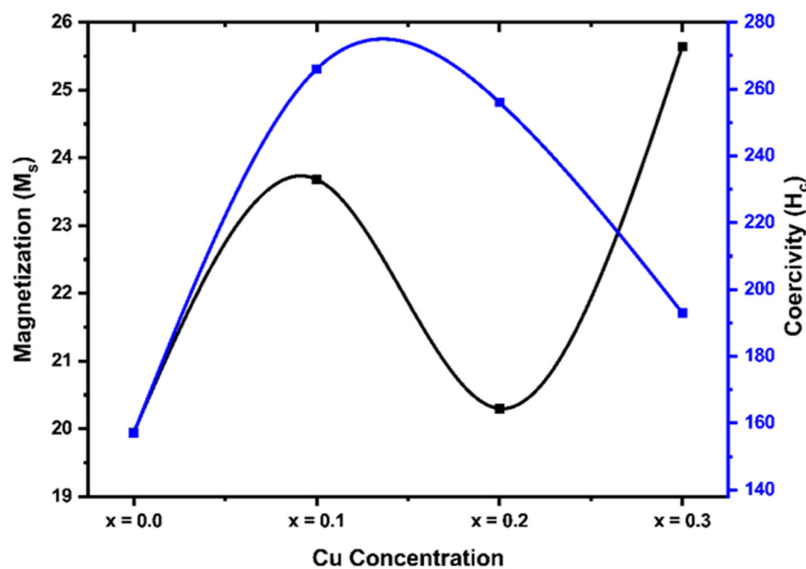


Figure 6. Magnetic Hysteresis loops of $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ($x = 0, 0.1, 0.2, \text{ and } 0.3$).

The saturation magnetization (M_s) and coercivity (H_c) of synthesized ferrite samples were obtained from the hysteresis loops as shown in Figure 6 and their values are listed in Table 3 below. It was observed that the value of saturation magnetization (M_s) first increased and then decreased with the increasing dopant (Cu^{2+}) concentration. The coercivity was found to depend on Cu^{2+} concentration and ranges from 157 Oe to 256 Oe. This result is consistent with the previous studies, which showed the magnetic properties are enhanced due to the cation substitution. In a study, Hoyos-Sifuentes et al. have reported the synthesis of magnesium ferrite material (MgFe_2O_4) and a M_s value of 17 emu/g [24]. Balavijayalakshmi et al., in their work synthesized the cobalt substituted magnesium ferrites and reported an increase in saturation magnetization (M_s), remanent magnetization (M_r) and coercivity (H_c) as the concentration of cobalt substitution increases, attributed to the change in the concentration of cation distribution [25]. Thus, the excellent magnetic behavior of the synthesized Cu-doped Mg-Co ferrites can be used for high frequency magnetic storage device applications.

Table 3. Magnetization and coercivity values for $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$.

Parameters	$x = 0.0$	$x = 0.1$	$x = 0.2$	$x = 0.3$
Magnetization M_s [emu/g]	19.95	23.68	20.30	25.64
Coercivity H_c (Oe)	157	266	256	193

**Figure 7.** Variation of saturation magnetization (M_s) and coercivity (H_c) with Cu content for $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$.

Neel's model can explain the observed magnetic behaviour of the Cu-substituted Mg-Co ferrites samples. The saturation magnetization, and coercivity, of a ferrite depends on its cation occupancy, composition, microstructure, and density. Neel's two-sublattice model states that the magnetic moment per formula unit is given by the difference between the magnetic moments of each sublattice. The total magnetization value can be obtained by their differences:

$$M = |M_B - M_A|$$

Where M_A and M_B are the A and B sublattice's magnetic moments per formula unit respectively in μ_B (Bohr magneton).

From Table 3, the observed variation in saturation magnetization, i.e., first increase and then decrease with the increasing dopant (Cu^{2+}) concentration, indicates the fluctuation in the cationic distribution in the two interstitial sites; tetrahedral (A) and octahedral (B) [26]. With the substitution of Cu^{2+} on the B site, the reduction in B-site magnetization causes a decrease in the ferrite's magnetization value (M_s), and this substitution of Cu^{2+} ions into the B-sites will promote the migration of Fe^{3+} ions into the A-site, which then increase magnetization of A-site.

The different exchange interactions, such as A-B, A-A, and B-B, which depend on the distribution of magnetic and non-magnetic ions at the A and B sites, can also be used to explain the variations in M_s value. The B-B and A-A interactions are known to be subordinate to the A-B interaction, which is the strongest. As the Cu level increased, the iron ions moved to the A site, exhibiting less A-B interaction with iron on the B site and thus the ferromagnetic behaviour decreased with increasing Cu^{2+} concentration. Moreover, the coercivity first increased and then decreased due to the greater anisotropic constant of the Cu^{2+} substituted Mg-Co ferrites, which is completely related to the bulk density of the samples. This result indicates that samples with high coercivity can be utilized for applications in microwave absorption systems, switch-mode power supplies, MLCIs, and dc-dc converters.

3.5. DC Electrical Resistivity Studies

The prepared pellets of the synthesized samples were examined by two probe methods to determine the resistivity of Cu^{2+} substituted $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ with $x = 0.0, 0.1, 0.2,$ and 0.3 samples in the range $300\text{K} - 873\text{K}$. The temperature of the sample was measured using a chrome-alumini thermocouple. The resistivity of the sample is obtained by using Equation (3) below:

$$\rho = R_b A / L \quad (3)$$

R_b is the resistance of the sample, A is the surface area of the sample and given by πr^2 , r is the radius of the sample pellet, and L is the thickness of the sample.

The plot of $\log \rho$ against $1/T$ is shown in Figure 8. The graph obeys the Arrhenius relation (Equation (4)), showing the semiconducting behavior of synthesized samples [27].

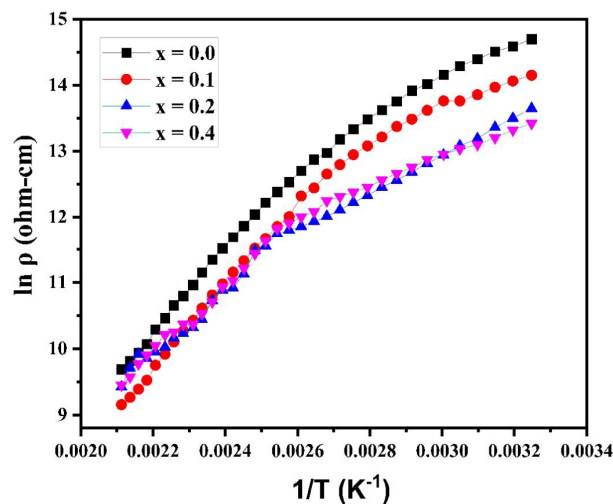


Figure 8. Plots of DC electrical resistivity of various $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$.

$$\rho = \rho_0 \exp(\Delta E/kT) \quad (4)$$

Where k is the Boltzmann constant, T is the absolute temperature, ΔE is the activation energy for conduction, ρ_0 is resistivity at 0K , and ρ is the resistivity of the material at $T\text{K}$.

It is clear from the variation of electrical resistivity ($\log \rho$) with temperature ($1/T$) curves (at different Cu^{2+} concentrations) that each plot adheres to the Arrhenius relation. The resistivity of ferrite samples (at different Cu^{2+} concentrations) reduces as the temperature rises, showing that the samples are semiconducting. The DC electrical resistivity was maximum with a value of $\sim 2.4 \times 10^6$ without copper substitution in magnesium-cobalt ferrite samples. With the copper substitution, the electrical resistivity was calculated and found to decline from $1.4 \times 10^6 \text{ } \Omega\text{-cm}$ (for $x = 0.1$) to $6.7 \times 10^5 \text{ } \Omega\text{-cm}$ (for $x = 0.3$). It shows that with the increase in Cu cation concentrations synthesized ferrite samples show a decrease in the resistivity. The smaller grain size is responsible for the greater resistivity value. Large numbers of grain boundaries found in smaller grains serve as sites for scattering the flow of electrons, thus raising the resistivity. Bharathi et al. in a study, reported the temperature dependent dc electrical resistivity and a semiconducting behaviour of synthesized Cu^{2+} substituted Mg-Co ferrite [28]. This result is consistent with our study, which showed the decrease in electrical resistivity dependent on temperature and cation substitution and semiconducting nature of synthesized copper-doped Mg-Co ferrites.

The activation energy plot of samples against Cu^{2+} concentration is shown in Figure 9, where the concentration content decreases the activation energy and samples' resistivity due to the hopping mechanism [29]. The activation energy values for $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ($x = 0, 0.1, 0.2, \text{ and } 0.3$) is shown in Table 4.

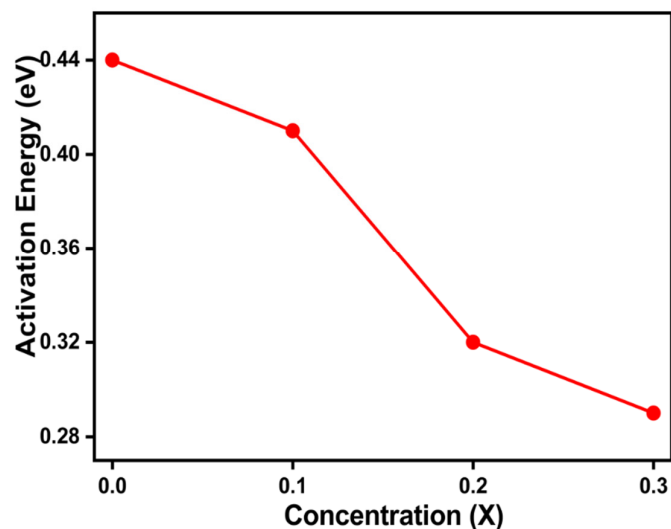


Figure 9. Various activation energy with Cu content for $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$.

Table 4. Activation energy values for $\text{Mg}_{0.6-x}\text{Cu}_x\text{Co}_{0.4}\text{Fe}_2\text{O}_4$ ($x = 0, 0.1, 0.2, \text{ and } 0.3$).

Parameters	$x = 0.0$	$x = 0.1$	$x = 0.2$	$x = 0.3$
Activation Energy (eV)	0.430	0.408	0.386	0.362

The electrical conduction in ferrites with Cu^{2+} ions is due to the electron jumping (or) hopping between Fe^{2+} to Fe^{3+} on the adjacent octahedral sites (B-sites) of the ferrite structure. When the sintering of samples is done at high temperatures, the conversion of Fe^{3+} ions to Fe^{2+} ions create oxygen vacancies for maintaining the spinel lattice's neutrality. The result of this study is consistent with a previous report, where Parajuli et al. synthesized magnesium substituted copper-cobalt

ferrites and reported their electrical properties and the activation energy based on the hopping mechanism [30].

4. Conclusion

The Cu²⁺ substituted Mg_{0.6-x}Cu_xCo_{0.4}Fe₂O₄ with x = 0.0, 0.1, 0.2, and 0.3 were successfully prepared by a solid-state reaction method. XRD pattern of all the samples shows the single-phase cubic spinel structure. Larger ionic radii of Cu²⁺ than that of the Mg²⁺ ions increase the lattice constant. SEM micrographs exhibit uniform size cubic crystals with an average grain size range between 1-1.5 μm. In the FTIR spectra of all the synthesized samples, two absorption bands were found in the range of 600 and 400 cm⁻¹. It was corresponding to the lower and higher frequencies of the tetrahedral and octahedral metal-oxygen vibration respectively, which confirms the spinel structure of synthesized ferrites. The band position variation with concentration was due to their ionic radii and cation redistribution. For synthesized ferrites, the plots of log ρ against the reciprocal of temperature show their semiconducting behavior. The activation energy of synthesized ferrites was found to decrease with Cu²⁺ substitution and the electrical resistivity was found to decline from 1.4 × 10⁶ Ω-cm (for x = 0.1) to 6.7 × 10⁵ Ω-cm (for x = 0.3). It shows that with the increase in Cu²⁺ concentrations, the synthesized ferrite samples show a decrease in the resistivity of synthesized ferrite samples. The magnetic saturation and coercivity of synthesized ferrites were found to increase with Cu²⁺ concentration and the coercivity ranges from 157 Oe to 256 Oe. Thus, the addition of Cu²⁺ ions in Mg-Co ferrites has powerfully affected the magnesium ferrite's structural, dc electrical resistivity, and magnetic properties. It suggests that the synthesized Cu-doped Mg-Co ferrites, with high DC electrical resistivity and coercivity, can be utilized for magnetic storage devices, antennas, transformers, and high-frequency electronic applications.

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