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Tuning structural, electrical, dielectric, and magnetic properties of Mg-Cu-Co ferrites via dysprosium (Dy³⁺) doping

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Abstract

In current research work, Dy³⁺ substituted Mg_{0.5}Cu_{0.25}Co_{0.25}Fe_{2-x}Dy_xO₄ (0.0 ≤ x ≤ 0.04 with the step interval of 0.01) soft ferrites synthesized by the sol-gel auto combustion method. The prepared samples were characterized by the techniques using X-Ray diffractometer (XRD), Scanning electron microscopy (SEM), Energy dispersive X-ray spectroscopy (EDS), Fourier transform infrared (FTIR) spectroscopy, Current-Voltage (I-V) measurement, LCR meter, vibrating sample magnetometer (VSM) and Raman. XRD data revealed that the average crystallite size was 49.71 nm and the lattice constant was 8.3703 Å for sample x = 0.03. The non-uniform grain growth was demonstrated by micrographs and impurity-free elemental composition was observed from EDX analysis. The DC resistivity had an increasing and decreasing trend in ferromagnetic and paramagnetic regions with an increase in temperature. Moreover, the high resistivity was observed is of the order of 10¹⁰ Ω cm, and the activation energy was 0.944 eV for samples x = 0.03. The dielectric parameters including dielectric constant, dielectric losses, and impedance gradually decreased with the increase in frequency from 8 Hz – 8 MHz. The minimum dielectric loss at high frequency was found for sample x = 0.03. The coercivity (H_c) and saturation magnetization (M_s) were found in the ranges of 520.82 Oe to 544.02 Oe and 20.5841 emu/g to 21.1473 emu/g, respectively. These observations confirmed that dysprosium (x = 0.03) doped MCC-soft ferrites may be applicable in transformer cores, microwave absorbance, and telecommunication devices.

Keywords: soft ferrites; DC resistivity; dielectric loss; impedance; coercivity.

1. Introduction

Soft ferrites with cubic spinel structures have become a significant area of research. Soft ferrites are applicable in various fields like memory devices, computer components, microwave absorbing materials, cancer treatment, filters and satellite communication, magnetic resonance imaging, magnet recording media, ferrofluids, transformer cores, and antenna rods.^{1,2} The structural, optical, and electrical characteristics of soft ferrites present suitable information for the particular applications of these materials. The general formula representation of spinel ferrite is AB₂O₄ where A and B represent metallic cations. Tetrahedral (A- site) and octahedral (B- site) are different crystallographic sites at which metallic cations are located. Oxygen atoms are coordinated

by the cations of both positions i.e., octahedral and tetrahedral respectively.³ The spinel ferrite unit cell consists of a cubic closed oxygen ions arrangement, which contains sixty-four tetrahedral (*A*-) sites and thirty-two octahedral (*B*-) sites. Metal cations occupy 8 of the *A*-site as well as 16 of the *B*-site. In spinel ferrite, the magnetic and dielectric properties depend on several parameters including the preparation methods, annealing temperature, chemical composition, and tetrahedral octahedral cationic distribution.^{4,5} Amongst the soft ferrites, due to the high Curie temperature, high DC electrical resistivity and environmental permanence the Dy³⁺ substituted ferrites are important magnetic oxides.^{6,7} Generally, microwave devices such as spinel ferrites have an application to control amplitude, frequency, and also transmission path.⁸ Different researchers have explained the rare earth element doped spinel ferrites. Kumar *et al.*,⁹ described the study of Dysprosium doped cobalt ferrite, CoFe_{2-x}Dy_xO₄ ($x = 0.0, 0.05, 0.10, 0.15$) prepared by the chemical method. They found that the dielectric constant for pure cobalt ferrite sintered at high temperatures showed increasing trends. While the DC electrical resistivity was observed in the range of 10⁵ to 10⁹ Ω cm. Kanna *et al.*,¹⁰ developed the series of Dysprosium substituted Mn-Cu ferrites by using the sonochemical route. They observed that both dielectric loss and dielectric constant reduced with the increase of Dy³⁺ concentration. The saturation magnetization showed that the synthesized materials are applicable for microwave-absorbing applications. Bamzai *et al.*,¹¹ studied the MgDy_xFe_{2-x}O₄, where $x = 0.0 - 0.07$ were fabricated by the solid state reaction procedure. They found that magnetization increased in the range of 1.52 – 2.47 emu g⁻¹ and the Curie temperature decreased from 400 – 370 °C with the increase in Dy³⁺ concentration. Junaid *et al.*,¹² fabricated the terbium and dysprosium inserted Li_{0.2}Ni_{0.8}Tb_{0.5x}Dy_{0.5x}Fe_{2-x}O₄ ($x = 0.0 - 0.08$) soft ferrites by using microemulsion process and found that the coercivity increased from 120 to 156 Oe with the increase of terbium and dysprosium doping. They also suggested that such materials are favorite candidates for switching and microwave device applications. Zipare *et al.*,⁶ synthesized the Mn-Zn-Dy soft ferrites *via* the co-precipitation method. They found that the Curie temperature decreased from 124 °C to 84 °C with the addition of dysprosium contents. Almessiere *et al.*,¹³ found an increase in magnetization for Dy³⁺ substituted Ni-Cu-Zn ferrites. Ditta *et al.*,¹⁴ observed that in low-frequency regions the dielectric losses rapidly decreased for Gd³⁺ and Dy³⁺ substituted Co-Ni ferrites. Furthermore, the divalent metals in current composed series like Mg, Cu, and Co also have vital applications in ferrites. Bhukal *et al.*,¹⁵ synthesized the Mg-substituted Co-Zn ferrites by using the sol-gel auto-combustion method. They found that the materials are useful for pulse power applications due to high saturation magnetization. Mammo *et al.*,¹⁶ prepared the Mg-Co soft ferrites through the sol-gel route and concluded that resistivity increased and saturation magnetization varied with the increase of Mg concentration. They also discussed that such materials are suitable

for high-frequency electronic devices. Hammad *et al.*,¹⁷ prepared the Cu-Zn ferrites using a co-precipitation process and observed the enhancement in saturation magnetization and magnetic moment. Jemai *et al.*,¹⁸ fabricated the Mg-Ni-Co ferrites by using the sol-gel technique and suggested that the materials are applicable in devices where the frequency and temperature are stable. Chalisgaonkar *et al.*,¹⁹ synthesized Co-Cu ferrite *via* a sol-gel auto-combustion process and reported that it is useful for the hyperthermia treatment of cancer. Sekhar *et al.*,²⁰ reported that the Co-Cu ferrite is a highly desirable candidate for Magnetostrictive sensing applications due to its mild magnetostriction and magnetization values, large strain derivative values, and significantly reduced coercivity and Curie temperature. Li *et al.*,²¹ reported solid-state synthesized Mg-Cu ferrites as a good candidate for ultra-high frequency anti-electromagnetic interference devices. The Dy-substituted nano ferrites may play a vital role in tuning various properties of Mg-Cu-Co ferrites. The annealing temperature and synthesis process also play a significant role in the cation distribution and other physical characteristics of soft ferrites. Several methods are used to synthesize the nanomaterials such as co-precipitation²²⁻²⁶, flash combustion²⁷, hydrothermal²⁸, sol-gel auto combustion method²⁹⁻³¹, etc. Among these, sol-gel auto combustion is regarded as a suitable and flexible technique, due to its high homogeneity and purity. In current research work, the sol-gel auto-combustion route was adopted to synthesize the $Mg_{0.5}Cu_{0.25}Co_{0.25}Fe_{2-x}Dy_xO_4$ ($0.0 \leq x \leq 0.04$) soft ferrites (*MCCD-soft ferrites*) and studied structural, morphological, compositional, vibrational, optical, electrical, dielectric, and magnetic properties.

2. Experimental details

2.1 Synthesis

The sol-gel auto-combustion method was used for the preparation of $Mg_{0.5}Cu_{0.25}Co_{0.25}Fe_{2-x}Dy_xO_4$ ($x = 0.0, 0.01, 0.02, 0.03, 0.04$) samples. Nitrates of magnesium, copper, cobalt, iron, and dysprosium were used according to stoichiometric calculations. Citric acid was used as a burning agent. Firstly, the nitrates and citric acid were dissolved in distilled water by taking the molar ratio of 1:1. Magnetic stirring was used to dissolve the nitrates and citric acid separately in distilled water. To maintain the pH of the mixture 7, ammonia solution was added dropwise. After continuous stirring of the solution for about 1 hour approximately, the dry gel was obtained at 373 K. After forming the dry gel, stirring was stopped and only the gel was heated until the ending products were obtained by the auto-combustion method. Then grinding was done for half an hour to obtain the refined form of prepared samples. After this, the powders were placed into a furnace for sintering at 773 K for 2 hours to impart strength and integrity to a material as well as reduce the porosity of the material³² and after sintering the samples were again grounded for 15 to 20 minutes. After this, the resulting powder was also pressed into pellets under a pressure of 1500 MPa. Finally,

the powder and pellets were sintered at 1023 K for 8 hours. The whole procedure is shown in Fig.

1. Different techniques were used to analyze the prepared ferrites.

3. Results and discussions

3.1 Structural analysis

The crystalline structure and formation of spinel structure were investigated by XRD analysis. Fig. 2 shows the XRD pattern of synthesized *MCCD-soft ferrites* and different peaks with planes (220), (311), (400), (422), and (511) confirmed the formation of single-phase spinel ferrite²² and verified by JCPDS file (73-1720).³³ The sharp (311) peaks in all XRD spectra confirmed the formation of cubic spinel ferrite. To determine the crystallite size, the Sherrer's relation ($D = \frac{k}{\beta \cos \theta}$) was used^{34,35}, where λ represents the wavelength of the X-ray source used, k (0.9) defines the shape factor and β represents full width at half maxima.³⁶ The average calculated crystallite size was 43.20 nm, 40.07 nm, 35.81 nm, 49.71 nm, and 28.09 nm for $x = 0.0, 0.01, 0.02, 0.03,$ and 0.04 , respectively. The variations occurring in crystallite size are due to the differences in ionic radii of Dy^{3+} (0.99 Å) and Fe^{3+} (0.64 Å).¹¹ The size of the crystallites decreases as the Dy^{3+} content (x) increases. The fact that the crystallite size has decreased by such a significant number suggests that the Fe^{3+} ions in the spinel-like structure have been replaced by Dy^{3+} ions.³⁷ The lattice constant was determined by using the relation $a = d\sqrt{(h^2 + k^2 + l^2)}$ ³⁵, where d indicates the inter-planar spacing and $(h k l)$ are Miller indices. The lattice constants were found 8.3748 Å, 8.3752 Å, 8.3662 Å, 8.3703 Å, and 8.4019 Å for $x = 0.0, 0.01, 0.02, 0.03,$ and 0.04 , respectively. The increase in lattice constant was due to the fact that the smaller ionic radii ($\text{Fe}^{3+} = 0.64$ Å) was replaced by the larger ionic radii ($\text{Dy}^{3+} = 0.99$ Å).¹¹

3.2 Morphological analysis

The surface morphology of nano ferrites was studied by SEM. Fig. 3 shows the SEM images of dysprosium substituted $x = 0.0 - 0.04$ soft ferrites. It can be observed that most of the particles are in nanometer ranges, with their sizes under 100 nm. The non-uniform grain growth is demonstrated by micrographs with different sizes and the distribution in spherical shapes of particles is enhanced with the addition of Dy^{3+} concentration. Due to the sintering process, the permanent magnetic dipole moments are increased which causes the agglomeration in the nanoparticles³⁸ as can be observed in Fig. 3.

3.3 Elemental compositional analysis

The elemental composition of elements was analyzed by Energy Dispersive X-ray (EDX) study. It confirms the purity and homogeneous mixing of ferrite elements like magnesium, iron, and dysprosium within the 10% range of an expected error.³⁹ Fig. 4 shows the images of dysprosium-substituted soft ferrites. Table 1 illustrates the percentages of elements in the synthesized composition. The percentages show that the chemical reaction is complete and no

contaminations are included in composed nano ferrites. From Table 1 it can be observed that all compositional elements (Mg, Cu, Co, Fe, Dy, and O) are present in the compositions.

3.4 Absorption band analysis

FTIR analysis was accomplished to confirm the presence of several functional groups in *MCCD-soft ferrites*. The purity of nanoparticles was also confirmed by FTIR. The FTIR spectra of synthesized ferrites were noted in the range of 400 – 4000 cm^{-1} as presented in Fig. 5. Two absorption bands were observed in the range of 533.3 – 533.25 cm^{-1} showing the intrinsic vibrations in the tetrahedral site (ν_1) and 416.63 – 417.30 cm^{-1} showing the intrinsic vibrations in octahedral site (ν_2).³⁹ Ferrites containing bands below 1000 cm^{-1} reveal the metal-oxygen ($M - O$) absorption bands and represent the best features of spinel ferrites. The peak around 1744 cm^{-1} represents the asymmetric carboxyl group and around 1359 cm^{-1} represents the symmetric carboxyl group.⁴⁰ The absorption band around 3431 cm^{-1} is due to $O - H$ stretching vibrations.⁴¹

3.5 Molecular Vibrational Analysis

Molecular vibrational analysis for dysprosium inserted *MCCD-soft ferrites* was carried out with the help of Raman spectroscopy as well as Lorentz fit spectra with 200 to 800 cm^{-1} range as illustrated in Fig. 6(a – e). A_{1g} , E_g , and $3T_{2g}$ are the first five active Raman modes⁴² of the spectra at room temperature. The symbols A , E , and T are symbolized in single, two, and three-dimensional depictions, correspondingly and g denotes the center of inversion symmetry.⁴³ The quasi-molecular characterization of cubic structure associated with normal mode movements of a tetrahedron ($Fe - O_4$) in the procedure as; A_{1g} (1) is because of symmetric stretching of oxygen atoms on ($Fe - O$) tetrahedral bonds appearing at 687.97 – 690.40 cm^{-1} with respect to metal ions lying at the tetrahedral site.⁴⁴ T_{2g} (3) is associated with asymmetric bending of oxygen at the tetrahedral site and E_g is associated with symmetric bending of oxygen also at the tetrahedral site⁴⁴, both are relative to iron appearing at 553.98 – 533.54 cm^{-1} and 357.36 – 379.93 cm^{-1} , correspondingly. T_{2g} (2) is associated with symmetric stretching of the ($Fe - O$) bond lying at the tetrahedral site⁴⁴ and found at 463.70 – 461.56 cm^{-1} . T_{2g} (1) is associated with translational movements of a complete tetrahedron ($Fe - O_4$)⁴⁴ and is situated at 321.81 – 319.74 cm^{-1} . Table 2 illustrates the detected values of Raman modes from Raman spectra. Some additional vibrational bands also existed along with Raman active modes in composed Dy^{3+} substituted for *MCCD-soft ferrites*. These modes are due to the existence of some contaminations in the matrix. The variation in polarizability-molecular vibrations in crystal structure caused the occurrence of Raman active modes. This factor originates from Raman mode activation and symmetry loss. The insertion of doping ions may cause variations in modes due to the location of ions in the crystal structure.²²

3.6 Electrical analysis

For this purpose, the DC electrical resistivity was calculated by increasing the temperature of nano ferrite pellets from room temperature to 823 K. The DC electrical resistivity was estimated by using the following relation $\rho = \frac{RA}{L}$, where ρ , A , and L represent the resistivity, the area, and the thickness of pellets, correspondingly. Fig. 7 illustrates the trend of DC electrical resistivity (ρ) with the inverse of temperature. It can be observed that initially, the “ ρ ” increases and then starts decreasing. The kink point where the trend change is called Curie or transition temperature (T_C). The regions before and after the T_C point are called ferromagnetic and paramagnetic regions⁴⁵, respectively as can be observed in Fig. 7. The resistivity shows increasing and decreasing trends in ferromagnetic and paramagnetic regions, correspondingly with the increase in temperature. The reduction in resistivity with the increase in temperature confirms the semiconducting nature of materials. The values of DC resistivities at respective Curie temperatures for $x = 0.0$ to 0.04 soft ferrites are demonstrated in Table 3. The increase and decrease in resistivity are associated with the fall and rise in drift mobility of charge carriers, respectively, and depending upon substitution, crystallite size, and density.²² Fe^{2+} and Fe^{3+} ions are sources of charge carriers’ hopping between the octahedral and tetrahedral sites, respectively, and hence are also responsible for the conduction mechanism.⁴⁶ So, the increase in resistivity is due to the difference in DC resistivities of dopant ions i.e., $\text{Dy}^{3+} = 92.6 \text{ } \Omega \text{ cm}$ and $\text{Fe}^{3+} = 9.71 \text{ } \Omega \text{ cm}$.⁴⁷ Hence these materials are suitable for transformers to decrease eddy current losses.⁴⁵ From Fig. 8 (a-c), it is clear that the DC electrical resistivity had a minimum value at $x = 0.01$ and a maximum at $x = 0.04$. The Arrhenius relation⁴⁸ was used to express the activation energies of dysprosium-doped *MCCD-soft ferrites*. According to the Irkhin and Turov principle, the ferromagnetic regions have lower activation energies than the paramagnetic due to the ordering and disordering states of atoms in ferromagnetic and paramagnetic regions, respectively. In paramagnetic states, it requires a large amount of energy to shift the charge carriers because of the disordering states of atoms.⁴⁹ In soft ferrites activation energy is associated with the substitution of Dy^{3+} with Fe^{3+} and spin disordering.²² The conductivity in soft ferrites is associated with activation energy and spin disordering may be the fact of non-linearity shown by the activation energy. The reduction in activation energy for samples $x = 0.01$ (0.052 eV) and 0.04 (0.553 eV) with the addition of dysprosium contents might be due to the reduction in resistivity. It is because resistivity has similar behaviour to the activation energy. In the same way, the activation energy for samples $x = 0.02$ (0.479 eV) and $x = 0.03$ (0.944 eV) increases with the addition of dysprosium concentration. Such trends might be attributed to an increase in DC resistivity. Fe^{3+} ions are partially replaced by Dy^{3+} ions and Dy^{3+} ions prefer to reside on an octahedral site. Hence, with the increase of dysprosium concentration the relocation

of a few iron ions from tetrahedral to the octahedral site to replace the reduction in iron ions at the octahedral site cause a decrease in activation energy. The such consequence is due to the increase in electron exchange level amongst divalent and trivalent iron ions.⁵⁰

3.8 Dielectric properties

The dielectric properties of soft ferrites are related to the synthesis process and the cations distribution. For effective properties of spinel ferrites, high resistivity and low dielectric losses are required.⁵¹ Fig. 9 shows the relation between dielectric constant and frequency. It can be seen that the dielectric constant has a linear behaviour for $x = 0.0, 0.02$ to 0.04 soft ferrites. For $\text{Mg}_{0.5}\text{Cu}_{0.25}\text{Co}_{0.25}\text{Fe}_{1.99}\text{Dy}_{0.01}\text{O}_4$ soft ferrite, the dielectric constant reduces gradually, then obtains a linear trend like the rest of the nanoparticles after 5 MHz frequency as given in Fig. 9. Hopping of electrons between divalent and trivalent iron ions is responsible for such behaviour and causes polarization. Also, the substitution of Dy^{3+} ions declines the polarization and causes a decrease in the dielectric constant. The spinel ferrites consist of well-conducting grain and highly resistive grain boundary layers named primary and secondary layers, respectively. In the presence of an applied field, the charge localization causes surface polarization. Due to the reduction in surface polarization, the applied field does not disturb the existence of the exchange of ions between Fe^{2+} and Fe^{3+} .⁵² The inset of Fig. 9 shows the dielectric constant in low-frequency regions. The inset Fig. 9 revealed the sample ($x = 0.01$) has the highest dielectric constant nearly 8.5×10^4 . The majority of divalent Fe ions are responsible for the maximum dielectric constant at low frequencies. Such a majority of ions are because of charge polarization at the grain boundaries.¹⁴ In dielectric materials, the space charge transporters need a limited time to align the direction of axes parallel to the applied electric field. With the increase in frequency, a point will come where the space charge transporter will not cope up with the applied field. Such behaviour causes a reduction in the dielectric constant of nanomaterials. The highest dielectric constant shown by the sample $x = 0.01$ may be due to the resonance effect.⁹ The increase in resonance occurs because of alike frequency of charge transport amongst divalent and trivalent iron ions with the applied electric field. Currently, the dielectric constant is influenced by the oxidation conditions of metal ions, the synthesis process, and the solubility of dysprosium ions. Furthermore, the higher ionic radii (Dy^{3+}) have restricted solubility in the lattice. The phases which may not be detected by the XRD like the existence of ions on the grain boundaries may have the ability to play role in polarization and are one of the roots to increase the dielectric constant of the sample.⁹ Moreover, due to the facilitation of additional charge carriers, defects including lattice imperfections, oxygen vacancies in ferrite lattice are reduced, conduction is increased, and hopping between ferrous and ferric ions is enhanced. This is responsible for high dielectric constant values.⁵³ The plot between dielectric losses ($\text{Tan } \delta$) and frequency is shown in Fig. 10. It can be seen that dielectric losses are reduced

with the increase of Dy³⁺ concentration. It may be attributed to a decrease in conductance with the enhancement of dysprosium contents. In spinel ferrites, the soft conducting grains are separated by low-energy conducting grains. These conducting grains become less active at low-frequency states and this leads to an increase in energy requirements for the hopping process. Consequently, dielectric losses enhance in low-frequency regions. In high-frequency areas, the conducting grains turn more efficient. Thus, low energy is required to move the charge carriers for hopping as a result in higher frequency regions dielectric losses declines.⁴⁵ Also, it can be noticed that dielectric losses have linear nature with the increase in frequency. Further, Fig. 10 shows that Mg_{0.5}Cu_{0.25}Co_{0.25}Fe_{1.97}Dy_{0.03}O₄ nano ferrites have minimum dielectric losses. Such nano ferrites show minimum dielectric losses after the 2×10^4 Hz frequency. It is because energy losses are reduced. Hence, this material is a potential applicant with minimum energy losses than others. It can be noticed that the sample ($x = 0.01$) has the highest dielectric losses in low-frequency regions. The dielectric loss is associated with different factors like stoichiometric ratios, Fe²⁺ ions, structural homogeneity, and synthesis procedure.⁹ For sample $x = 0.01$ in the low-frequency regions the hopping frequency of charge transporters pursue the applied field thus dielectric loss is relatively higher. Also, at a low-frequency state, the hopping between divalent and trivalent iron ions follows the applied field and leads to an increase in dielectric loss. It can also be found that with the increase in the addition of Dy³⁺ concentration the dielectric loss decreases for the rest of the ferrites. Because when the doping increase beyond a certain stage then the resonance frequency decreases.¹² The nature of dielectric losses with the increase of Dy³⁺ concentration at different frequencies is given in Fig. 11 (a – b). Fig. 11 (a) reveals that dielectric losses increase for $x = 0.0 - 0.01$ and shows as maximum value for Mg_{0.5}Cu_{0.25}Co_{0.25}Fe_{1.99}Dy_{0.01}O₄ and then decreases for $x = 0.02$. For $x = 0.03 - 0.04$, in the low frequency region dielectric loss again decreases. In the high-frequency region (Fig. 11 (b)), $x = 0.01$ again has the highest dielectric losses similar to the lower low-frequency region. While $x = 0.0$ and $x = 0.02 - 0.04$ have lower dielectric losses. Such behaviour is based on the polarization theory stated by Murthy and Sobhandri. According to them the exchange of ions between divalent and trivalent iron ions causes the polarization in spinel ferrites.⁵⁴ Furthermore, Mg_{0.5}Cu_{0.25}Co_{0.25}Fe_{1.97}Dy_{0.03}O₄ soft ferrites have shown minimum dielectric losses with the increase in frequency. Such behaviour is in agreement with the DC electrical resistivity outcomes as given in Table 3. Fig. 12 shows the relation between impedance and frequency for $x = 0.0 - 0.04$. It can be observed that impedance is reduced with the increase in frequency. Maximum impedance is shown by Mg_{0.5}Cu_{0.25}Co_{0.25}Fe_{1.96}Dy_{0.04}O₄ nano ferrites. Such a trend is attributed to the interaction between substituted and replaced ions.

3.9 Magnetic properties

Fig. 13 shows the hysteresis loops for pure and Dysprosium inserted *MCCD-soft ferrites* at room temperature. Loops demonstrate the ferromagnetic nature of composed soft ferrites. Fig. 13 shows that all specimens have the same behaviour and different parameters can be determined from this information i.e., saturation magnetization (M_S), remanent magnetization (M_r), and coercivity (H_c) as given in Table 4. The saturation magnetization for $\text{MgDy}_{0.01}\text{Fe}_{1.99}\text{O}_4$ has been previously reported as 17.74 emu g^{-1} .⁵⁵ In current research work, for $x = 0.01$, $M_S = 19.0649 \text{ emu g}^{-1}$. M_S is showing a variable trend with the increase of Dy^{3+} substitution as given in Table 4. The magnetic properties of soft ferrites depend on the cation distribution. The magnesium ions are distributed randomly and some magnesium ions are forced to the *A*- site as a result Fe^{3+} ions are shifted from *A*- to *B*- site. Such a factor increases the magnetization on *B*-site and then it reduces due to the decrease in Fe^{3+} ions.⁵⁵ The increase in magnetization for the rest of the nano ferrites i.e., $x = 0.0$, 0.02 , and 0.03 might be because of the substitution of Fe with effective magnetic moment $\mu_{\text{eff}} = 5.85$ by the replacement of Dy^{3+} with effective magnetic moment $\mu_{\text{eff}} = 10.50$.⁵⁶ Table 4 also illustrates that coercivity increases for $x = 0.0$ to 0.02 , then shows minimum value of 491.65 Oe for $x = 0.03$ and then again increases to 544.02 Oe for $x = 0.04$. Such behaviour is due to the crystallite size of nanoparticles because crystallite size and coercivity are inversely proportional to each other.²² Furthermore, the lowest coercivity is shown by the $\text{Mg}_{0.5}\text{Cu}_{0.25}\text{Co}_{0.25}\text{Fe}_{1.97}\text{Dy}_{0.03}\text{O}_4$ sample. Table 4 also indicates the squareness ratio ($SQ = M_r/M_S$) for $x = 0.0 - 0.04$ in the range of $0.6545 - 0.6243$, respectively. Such values of squareness showed that the nano ferrites consisted of homogeneous mixtures. The values of M_S , H_c , and M_r/M_S revealed that high spin-orbital coupling is produced at the octahedral site due to dysprosium substitution. The following relations were used to study the magneto crystalline anisotropy constant (K), initial permeability (μ_i), and Bohr's magnetron (n_B):⁵⁷

$$K = \frac{H_c \times M_S}{0.96} \quad (1) \quad \mu_i = \frac{M_S^2 \times D}{K} \quad (2)$$

$$n_B = \frac{M \times M_S}{5585d_x} \quad (3)$$

From Table 4 it can be seen that both factors K and μ_i are increasing with the addition of Dy^{3+} contents. The Bohr's magnetron (n_B) is showing a variable trend with the addition of Dy^{3+} contents. From Table 4 it can be found that n_B decreased from 0.1629 ($x = 0.0$) to 0.1509 ($x = 0.01$) then increased from $0.1634 - 0.1832$ for $x = 0.02$ to 0.03 and then again decreased to 0.1690 for $x = 0.04$. Such a variable trend is because of cation distribution.¹¹ The increase in n_B indicates that the superexchange interactions amongst the tetrahedral and octahedral sites are strengthening while the decrease in n_B means superexchange interactions are becoming weaker.¹³ The spin canting angle or Yafet and Kittel ($Y - K$) angle²² is also given in Table 4 and confirms the enhancement in

triangular spin configuration at the octahedral site. The increase or decrease in *A*- and *B*-interactions are because of an increase or decrease in triangular spin configuration at the octahedral site.⁵⁸ The effective magnetic moment of dysprosium ($\mu_{\text{eff}} = 10.50$) is responsible for a reduction in *A*- and *B*- interactions. The magnetic moments demonstrate the variable trends due to the non-linear spin of magnetic ions or canting caused by spin irritations. The increase in trivalent iron ions on *B*-site because of site preference is responsible for the increase in spin canting and anti-parallel spin coupling. Such behaviour causes the reduction in net magnetic moments, as well as decreases the super exchange interactions between *A*- and *B*- sites. Furthermore, the variable trend shown by the M_s is attributed to the decrease and increase in magnetic moments (n_B) and also because of the magnetic exchange interactions between tetrahedral and octahedral sites.¹³ Microwave frequency⁵⁹ was also calculated and illustrated in Table 4. According to Arshad *et al.*,²² in such a frequency range the synthesized materials are applicable in longitudinal media and microwave absorbance devices.

Conclusions

In current research work, the single-phase cubic spinel structure along with the substitution of Dy^{3+} ions in *MCC-soft ferrites* was confirmed by XRD. The lattice constant showed an increasing trend in the range between 8.3748 – 8.4019 Å for Dy^{3+} concentration $x = 0.0 - 0.04$. The replacement of dysprosium metal ions with ferric ions was confirmed by FTIR and Raman analysis. The homogeneous mixing of elements with a 10% expected error was confirmed by EDX. The dielectric losses had a minimum value at high frequencies for sample $x = 0.03$. The lowest coercivity ($H_c = 491.65$ Oe), maximum saturation magnetization ($M_s = 23.1787$ emu/g), and maximum frequency ($\omega_m = 5.11$ GHz) were examined for sample $x = 0.03$. The fabricated samples were my favorite candidates for telecommunication devices and transformer cores.

Supplementary file

In supplementary file we have added some additional information about structural, and optical properties.

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