



This is a repository copy of *To study the structural, electrical, and magnetic properties of M (M = Mg<sup>2+</sup>, Mn<sup>2+</sup>, and Cd<sup>2+</sup>) doped Cu-Ni-Co-La spinel ferrites.*

White Rose Research Online URL for this paper:

<https://eprints.whiterose.ac.uk/193867/>

Version: Accepted Version

---

**Article:**

Aslam, A., Rehman, A.U., Amin, N. et al. (7 more authors) (2023) To study the structural, electrical, and magnetic properties of M (M = Mg<sup>2+</sup>, Mn<sup>2+</sup>, and Cd<sup>2+</sup>) doped Cu-Ni-Co-La spinel ferrites. *Materials Chemistry and Physics*, 294. 127034. ISSN 0254-0584

<https://doi.org/10.1016/j.matchemphys.2022.127034>

---

Article available under the terms of the CC-BY-NC-ND licence  
(<https://creativecommons.org/licenses/by-nc-nd/4.0/>).

**Reuse**

This article is distributed under the terms of the Creative Commons Attribution-NonCommercial-NoDerivs (CC BY-NC-ND) licence. This licence only allows you to download this work and share it with others as long as you credit the authors, but you can't change the article in any way or use it commercially. More information and the full terms of the licence here: <https://creativecommons.org/licenses/>

**Takedown**

If you consider content in White Rose Research Online to be in breach of UK law, please notify us by emailing [eprints@whiterose.ac.uk](mailto:eprints@whiterose.ac.uk) including the URL of the record and the reason for the withdrawal request.



[eprints@whiterose.ac.uk](mailto:eprints@whiterose.ac.uk)  
<https://eprints.whiterose.ac.uk/>

**To study the structural, electrical, and magnetic properties of  $M$  ( $M = \text{Mg}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{Cd}^{2+}$ ) doped Cu-Ni-Co-La spinel ferrites**

Asma Aslam<sup>a</sup>, Atta Ur Rehman<sup>a</sup>, M. Amman<sup>b</sup>, M. Akhtar<sup>a</sup>, N. Amin<sup>a</sup>, N. A. Morley<sup>c</sup>, Mongi Amami<sup>d,e</sup>, Hafiz T Ali<sup>f</sup>, Mohammad Yusuf<sup>g</sup>, Muhammad Imran Arshad<sup>a,\*</sup>

<sup>a</sup>*Department of Physics, Government College University, Faisalabad, Pakistan.*

<sup>b</sup>*Department of Mathematics, University of Management and Technology, Lahore 54770, Pakistan.*

<sup>c</sup>*Department of Materials Science and Engineering, The University of Sheffield, UK, S1 3JD.*

<sup>d</sup>*Department of Chemistry College of Sciences, King Khalid University, P.O. Box 9004, Abha, Saudi Arabia*

<sup>e</sup>*Laboratoire des Matériaux et de l'environnement pour le développement durable LR18ES10. 9 Avenue Dr. Zoheir SAFI, 1006, Tunis, Tunisia*

<sup>f</sup>*Department of Mechanical Engineering, College of Engineering, Taif University, P.O. Box 1109, Taif 21944, Saudi Arabia*

<sup>g</sup>*Department of Clinical Pharmacy, College of Pharmacy, Taif University, P.O. Box 1109, Taif 21944, Saudi Arabia.*

\*Corresponding Author: [miarshadgcuf@gmail.com](mailto:miarshadgcuf@gmail.com)

## **Abstract**

Tertiary  $\text{Cu}_{0.25}\text{Ni}_{0.15}\text{M}_{0.25}\text{Co}_{0.35}\text{La}_{0.15}\text{Fe}_{1.85}\text{O}_4$  ( $M = \text{Mg}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{Cd}^{2+}$ ) spinel ferrites powders were prepared using the cost-effective sol-gel auto combustion route. The lattice parameters, absorption and vibrational bands, energy band gap, AC conductivity, and dielectric loss of the as-prepared ferrites were all measured. The  $\text{Cd}^{2+}$  doped ferrite has a minimum crystallite size ( $D$ ) of 50.9 nm, which is smaller compared to  $\text{Mg}^{2+}$  and  $\text{Mn}^{2+}$  doped ferrites. Furthermore, the X-ray diffraction (XRD), as well as Fourier transform infrared radiation (FTIR) and Raman analysis confirmed the doping of  $\text{Mg}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{Cd}^{2+}$  ions on their respective lattice sites. The resistivity of the divalent ions ( $\text{Mg}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{Cd}^{2+}$ ) doped ferrites decreased in the para region and increased in the ferro region as the temperature increased. Moreover, the resistivity of the  $\text{Cd}^{2+}$  doped ferrite was smaller when compared to the ferrites doped with  $\text{Mg}^{2+}$  and  $\text{Mn}^{2+}$  ions. From Arrhenius plots, the minimum activation energy ( $\Delta E$ ) 0.8671 eV was observed for  $\text{Cd}^{2+}$  doped ferrites. The  $\text{Cd}^{2+}$  substituted ferrite also has the smallest AC conductivity and dielectric loss compared to  $\text{Mg}^{2+}$  and  $\text{Mn}^{2+}$  substituted ferrites. The coercivity and saturation magnetization were 136.41 Oe and 92.29 emu/g for the  $\text{Cd}^{2+}$  doped ferrite, respectively. These results suggest that the  $\text{Cd}^{2+}$  doped ferrite material could be used in high-frequency and high-power applications.

**Keywords:** sol-gel auto combustion; conductivity; activation energies; polaron hopping; high-frequency.

## 1. Introduction

Magnetic materials such as spinel ferrites (SFs) are important due to their outstanding structural [1], magnetic [2], optical [3], morphological [4], and dielectric [4] properties. At the present, soft ferrite magnetic materials have been used in the industrial, electronics, and biomedical fields for their different applications including microwave absorbing materials [5], target drug delivery [6], magnetoelectric switching [7], magnetic resonance imaging [8], antennas [9], oscillators [10], high sensitive sensors [11], filters [12], magnetic ferrofluids [13], and high-frequency devices [14]. Spinel ferrites are generally expressed by the formula ( $AB_2O_4$ ) [15] where  $A$  indicates divalent metal ions including  $Cu^{2+}$ ,  $Co^{2+}$ ,  $Cd^{2+}$ ,  $Ni^{2+}$ ,  $Sn^{2+}$ , etc. and  $B$  represents trivalent metal cations such as  $La^{3+}$ ,  $Ce^{3+}$ , and  $Nd^{3+}$ , etc. Various methods are utilized to prepare SFs including the sol-gel auto combustion (SGAC) route [16], the glass crystallization method [17], the solid-state reaction (SSR) method [18], co-precipitation [19-21], and the microwave method [22]. The SGAC route was found to be better due to its low-temperature preparation, producing better homogeneity of particles, fine surface morphology, and excellent properties [23].

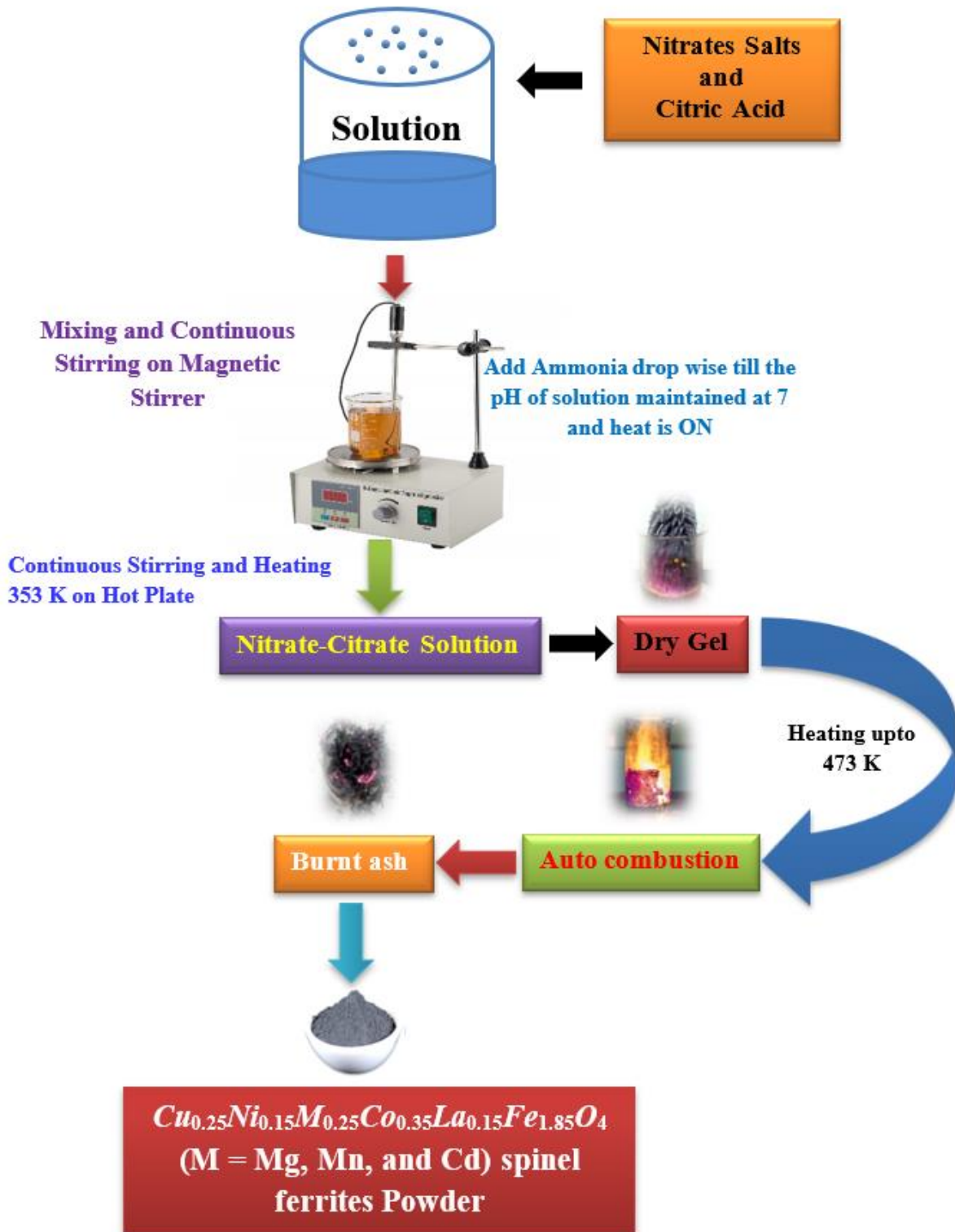
Due to their extraordinary role in different fields, ferrites have gained significant attention from researchers. A small amount of dopant ion within the spinel ferrites is the most appropriate route to modify their structural, dielectric, and electrical characteristics.  $La^{3+}$  is the most suitable rare-earth ion for doping because it is a lighter element with high electric and thermal properties compared to the other rare-earth elements [16]. Bharathi *et al.* [24] studied  $La^{3+}$  doped cobalt ferrites synthesized through a solid-state chemical reaction. The electric parameters of the  $La^{3+}$  doped cobalt ferrites were reduced with a reduction in temperature and the conductivity was  $0.623 \times 10^{-8} \Omega^{-1} \text{ cm}^{-1}$  at room temperature (RT). Kumar *et al.* [25] studied the influence of  $La^{3+}$  substitution on  $CoFe_2O_4$  spinel ferrite synthesized using a co-precipitation route. The electrical resistivity of prepared pure cobalt ferrite at 300 K was  $6.465 \times 10^5 \Omega \text{ cm}$ , while its value for  $La^{3+}$  concentration  $x = 0.15$  was  $26.990 \times 10^6 \Omega \text{ cm}$ . Mulushoa *et al.* [18] prepared Cu-Cr doped  $Mg^{2+}$  nano ferrite samples through the SSR process and reported a significant decrease in lattice parameters with the doping of dopant cation. It was also observed that the electrical resistivity increased from  $0.553 \times 10^5$  to  $0.105 \times 10^8 \Omega \text{ cm}$  due to the substitution of Cu-Cr ions. Hamdaoui *et al.* [26] investigated the properties of Ni-Cd-Mg ferrite synthesized through the sol-gel process, with the lattice parameters increasing with the  $Cd^{2+}$  doping increased. The electric conductivity of the prepared samples reveals their semiconductor nature due to the doping of  $Cd^{2+}$ . Hossen *et al.* [27] reported the sol-gel method

studied  $\text{La}^{3+}$  doped Ni-Cu-Cd ferrites and confirmed the formation of spinel ferrites *via* XRD and FTIR analysis. The AC conductivity improved due to the  $\text{La}^{3+}$  doping. In the current research work, the tertiary  $\text{Cu}_{0.25}\text{Ni}_{0.15}\text{M}_{0.25}\text{Co}_{0.35}\text{La}_{0.15}\text{Fe}_{1.85}\text{O}_4$  ( $\text{M} = \text{Mg}^{2+}, \text{Mn}^{2+}, \text{and Cd}^{2+}$ ) spinel ferrite powders were prepared *via* the SGAC technique and the structural, absorption, optoelectrical, dielectric, and magnetic analysis are reported.

## 2. Experimental Part

### 2.1 Samples preparation

The series of  $\text{Cu}_{0.25}\text{Ni}_{0.15}\text{M}_{0.25}\text{Co}_{0.35}\text{La}_{0.15}\text{Fe}_{1.85}\text{O}_4$  ( $\text{M} = \text{Mg}^{2+}, \text{Mn}^{2+}, \text{and Cd}^{2+}$ ) SFs were prepared *via* the SGAC route [ $\text{Cu}_{0.25}\text{Ni}_{0.15}\text{Mg}_{0.25}\text{Co}_{0.35}\text{La}_{0.15}\text{Fe}_{1.85}\text{O}_4$  (**Mg-CNCL Ferrite**),  $\text{Cu}_{0.25}\text{Ni}_{0.15}\text{Mn}_{0.25}\text{Co}_{0.35}\text{La}_{0.15}\text{Fe}_{1.85}\text{O}_4$  (**Mn-CNCL Ferrite**) and  $\text{Cu}_{0.25}\text{Ni}_{0.15}\text{Cd}_{0.25}\text{Co}_{0.35}\text{La}_{0.15}\text{Fe}_{1.85}\text{O}_4$  (**Cd-CNCL Ferrite**)]. All the chemicals were purchased from Sigma-Aldrich. The nitrate salts including  $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$  (99.99%),  $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  (99.99%),  $\text{Mn}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  (98%),  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  (99.95%),  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (97%),  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (98%),  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (99%), and  $\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  (98%) were dissolved into distilled water on a magnetic stirrer for the formation of a homogeneous solution according to the stoichiometric measurements. The citric acid (99.5%) was added into distilled water separately, to keep the solution of citric acid and metal nitrate (C/M) solution at 1:1 ratio. The C/M ratio at 1:1 particularly leads to easy igniting and reaction propagation. Higher C/M ratios (2/1, 3/1) can cause slower combustion with the production of a product that was significantly less foamed and more sintered powder, but lower ratios were unable to spark the reaction and required an additional energy source to continue it [28]. Ammonia was added, to obtain a solution with an acquired pH of  $\sim 7$ . The heat was turned on at 353 K for the formation of gel and after the gel formation, the temperature of the magnetic stirrer increased up to 473 K, while the gel was converted into ash due to continuous heat. After the formation of ash, grinding was done and the ash was converted into fine powder. The fine powder of the as-prepared ferrites was put into the furnace and sintering was done at 1073 K for 8 h. The flow chart of the SGAC process for the preparation of Mg-CNCL Ferrite, Mn-CNCL Ferrite, and Cd-CNCL Ferrite is depicted in Fig. 1.



**Fig. 1** Flow chart of sol-gel auto combustion technique for the as-prepared ferrites powder

## 2.2 Characterization Techniques used

Using a Bruker D8 Advance X-ray diffractometer with Cu  $K\alpha$  radiation ( $\lambda = 0.1542$  nm), X-ray diffraction (XRD) was performed. The FTIR spectrum is used to find the absorption bands. UV-vis spectrum was used to study the optical bandgap energy. The I-V measurements

were taken using Keithley Electrometer Model 2401. Raman spectroscopy was used to study the vibrational characteristics. The dielectric parameters were calculated through an IM3533 LCR Meter. *M-H* loops were measured using a vibrating sample magnetometer (VSM) at RT.

### 3. Experimental Results and Discussion

#### 3.1 XRD study

Fig.2 indicates the XRD pattern for all the as-prepared sample powders. Diffraction peaks at (220), (311), (222), (400), (422), and (511) confirmed the cubic spinel matrix with space group ( $Fd\bar{3}m$ ) for all the prepared samples (JCPDS#75-0541) [16, 29]. The additional peaks confirmed the existence of the orthorhombic secondary phase  $LaFeO_3$  (JCPDS#75-0541) in the as-prepared samples having a space group ( $Pbnm$ ) [16, 30]. The interplanar distance ( $d$ ) was calculated using Bragg's law, which is given as [31];

$$d = \frac{n\lambda}{2\sin\theta} \quad (1)$$

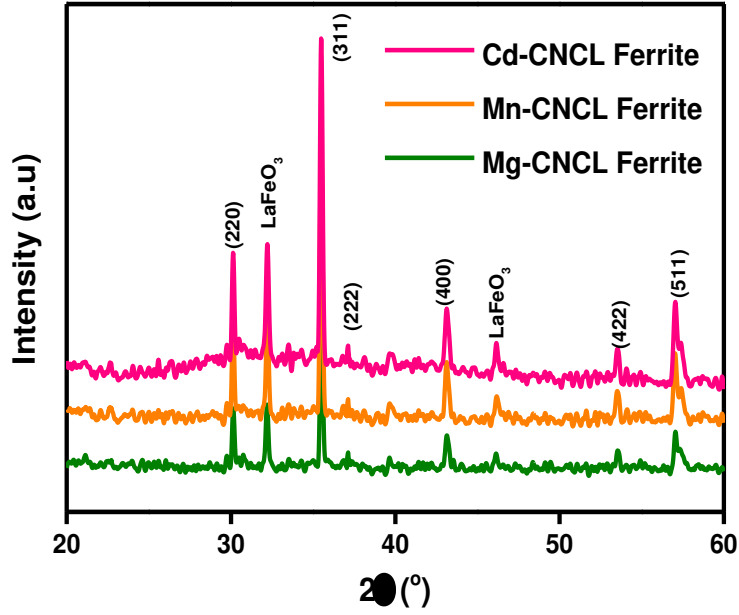
Where  $n = 1$ ,  $\lambda = 1.5046 \text{ \AA}$ , and " $\theta$ " indicate Bragg's angle. Equation (2) was employed to determine the experimental lattice constant ( $a$ ) [31];

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

where ( $hkl$ ) is the Miller indices. The interplanar distance and experimental lattice constant values are given in Table 1 and have the maximum value for the Mn-CNCL Ferrite. The changes in lattice parameters for all the samples is due to the cationic radii difference of the dopant ions, i.e.  $Mg^{2+}$  (0.78 Å) [26],  $Mn^{2+}$  (0.91 Å) [32],  $Cd^{2+}$  (1.03 Å) [26],  $Fe^{3+}$  ion (0.645 Å) [32] and  $La^{3+}$  (1.05 Å) [19]. The crystallite size ( $D$ ) was determined by Scherer's equation [31];

$$D = \frac{0.9\lambda}{\beta\cos\theta} \quad (3)$$

where " $\beta$ " indicates the full width at half maxima. The values of " $D$ " are listed in Table 1. The " $D$ " for sample Mn-CNCL Ferrite was the largest (58.9 nm) as compared to Mg-CNCL Ferrite and Cd CNCL Ferrite because of the greater bond energy of Mn-O when compared to the bond energies of Mg-O, Cd-O, La-O, and Fe-O [20].



**Fig. 2** XRD patterns for as-prepared ferrites

The X-ray ( $\rho_x$ ) and bulk ( $\rho_b$ ) densities were determined *via* the relations [31];

$$\rho_x = \frac{8M}{N_A a^3} \quad (4)$$

$$\rho_b = \frac{Mass}{Volume} = \frac{M}{\pi r^2 \times h} \quad (5)$$

where ‘ $r$ ’ and ‘ $h$ ’ indicate the radius and the thickness of the pellet, respectively. The porosity percentage was determined using equation (6) [31] and is given in Table 1.

$$P (\%) = \left[ 1 - \frac{\rho_b}{\rho_x} \right] \times 100 \quad (6)$$

The calculated values of “ $\rho_x$ ” increased with the insertion of divalent dopant cations while “ $\rho_b$ ” changed due to the existence of a few certain pores, which arise during the preparation and sintering of the prepared samples [20]. The values of “ $\rho_x$ ”, “ $\rho_b$ ” and P (%) are reported in Table 1. The strain ( $\epsilon$ ) [33], specific surface area ( $S$ ) [33], dislocation density ( $\delta$ ) [33], and packing factor ( $p$ ) [33] were determined by using formulas from equations (7), (8), (9) and (10) and are given in Table 1.

$$S = \frac{6000}{\rho_x \times D} \quad (7) \quad p = \frac{D}{d} \quad (8)$$

$$\epsilon = \frac{1}{d^2} \quad (9) \quad \delta = \frac{1}{D^2} \quad (10)$$

The surface area, strain, and dislocation density have a minimum value for the **Mn-CNCL Ferrite** but the packing factor ( $p$ ) was maximum for the **Mn-CNCL Ferrite** (as seen in Table 1). The distance between oxygen ions and tetrahedral ( $A$ -) and octahedral ( $B$ -) cations are given by the hopping lengths  $L_A$  and  $L_B$  respectively. A large value of hopping lengths means a higher potential is required to transfer the charge carriers from one lattice site to another while smaller

hopping lengths means a lower potential is needed for the movement of charge carriers into the sublattice [34]. The hopping lengths  $L_A$  and  $L_B$  were determined via the relations (11) and (12) [34];

$$L_A = \frac{a\sqrt{3}}{4} \quad (11)$$

$$L_B = \frac{a\sqrt{2}}{4} \quad (12)$$

The values of “ $L_A$ ” and “ $L_B$ ” for all the samples are listed in Table 1. The polaron radius ( $\gamma$ ) is calculated using equation (13) [20];

$$\gamma = \frac{1}{2} \sqrt[3]{\frac{\pi a^3}{576}} \quad (13)$$

It was observed that the polaron radius ( $\gamma$ ) changed as the experimental lattice constant changed due to the substitution of dopant  $Mg^{2+}$ ,  $Mn^{2+}$ , and  $Cd^{2+}$  ions into the as-prepared ferrites.

**Table 1** Lattice parameters of the as-prepared samples

Parameters	Mg-CNCL	Mn-CNCL	Cd-CNCL
	Ferrite	Ferrite	Ferrite
$d$ (Å)	2.5242	2.5266	2.5241
$a$ (Å)	8.3719	8.3801	8.3715
$D$ (nm)	56.8	58.9	50.9
$\rho_x$ (g/cm <sup>3</sup> )	5.4344	5.5818	5.9454
$\rho_b$ (g/cm <sup>3</sup> )	3.0569	2.9470	2.9315
$P$ (%)	43.75	52.79	50.69
$S$ (cm <sup>2</sup> /g)	0.0194	0.0182	0.0198
$p$	225.15	233.39	201.71
$\varepsilon$	0.0021	0.0020	0.0023
$\delta \times 10^{-4}$ (nm <sup>-2</sup> )	3.10	2.88	3.86
$L_A$ (Å)	3.6250	3.6286	3.6249
$L_B$ (Å)	2.9595	2.9624	2.9593
$\gamma$ (Å)	0.7363	0.7375	0.7359

Numerous methods were used by many researchers to find out the distribution of cations [35-37]. The cations distribution is given in Table 2. This arrangement of cations depends on the hypotheses; a) At the A- site, the sum cationic arrangement is one, while at the B- site, it is two. b) The total charge of the spinel matrix is zero. Therefore, to construct a



neutral structure electrically, the positive and negative charges in the compound are equal. Cadmium ferrite generally has a normal spinel crystal matrix and  $\text{Cd}^{2+}$  occupies only the  $A$ -sites [26]. Cobalt and Copper ferrites have partially inverse spinel crystal structures [38] and  $\text{Co}^{2+}$ ,  $\text{Cu}^{2+}$ , and  $\text{Fe}^{3+}$  cations occupy both the  $A$ - and  $B$ - sites [39].  $\text{La}^{3+}$  ions occupy the  $B$ -site of the lattice [40]. The cationic distribution (Table 2) was used to find out the ionic radii “ $r_A$ ” and “ $r_B$ ” to find at  $A$ - and  $B$ -sites, respectively for all the as-synthesized ferrites by using equations (14) and (15).

$$r_A = (C_{\text{Cu}^{2+}})(r_{\text{Cu}^{2+}}) + (C_{\text{Ni}^{2+}})(r_{\text{Ni}^{2+}}) + (C_{\text{M}^{2+}})(r_{\text{M}^{2+}}) + (C_{\text{Co}^{2+}})(r_{\text{Co}^{2+}}) + (C_{\text{Fe}^{3+}})(r_{\text{Fe}^{3+}}) \quad (14)$$

$$r_B = \frac{1}{2} [(C_{\text{Cu}^{2+}})(r_{\text{Cu}^{2+}}) + (C_{\text{Ni}^{2+}})(r_{\text{Ni}^{2+}}) + (C_{\text{M}^{2+}})(r_{\text{M}^{2+}}) + (C_{\text{Co}^{2+}})(r_{\text{Co}^{2+}}) + (C_{\text{Fe}^{3+}})(r_{\text{Fe}^{3+}}) + (C_{\text{La}^{3+}})(r_{\text{La}^{3+}})] \quad (15)$$

where  $(C_{\text{Cu}^{2+}})$ ,  $(C_{\text{Ni}^{2+}})$ ,  $(C_{\text{M}^{2+}})$ ,  $(C_{\text{Co}^{2+}})$ ,  $(C_{\text{Fe}^{3+}})$  and  $(C_{\text{La}^{3+}})$  represent the concentration of  $\text{Cu}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{M}^{2+}$  ( $\text{Mg}^{2+}$ ,  $\text{Mn}^{2+}$  and  $\text{Cd}^{2+}$ ),  $\text{Co}^{2+}$ ,  $\text{Fe}^{3+}$  and  $\text{La}^{3+}$  taken at the  $A$ -site and  $B$ -site according to cation distribution while  $r_{\text{M}^{2+}}$ :  $r_{\text{Mg}^{2+}} = \text{tet.:}0.57\text{\AA}$  and  $\text{oct.:}0.72\text{\AA}$  [41];  $r_{\text{Mn}^{2+}} = \text{tet.:}0.655\text{\AA}$  and  $\text{oct.:}0.80\text{\AA}$  [42];  $r_{\text{Cd}^{2+}} = 1.03\text{\AA}$  [26],  $r_{\text{Co}^{2+}} = \text{tet.:}0.58\text{\AA}$  and  $\text{oct.:}0.745\text{\AA}$ , and  $r_{\text{Fe}^{3+}} = \text{tet.:}0.49\text{\AA}$  and  $\text{oct.:}0.645\text{\AA}$  [43],  $r_{\text{Cu}^{2+}} = \text{tet.:}0.57\text{\AA}$  and  $\text{oct.:}0.73\text{\AA}$  [38],  $r_{\text{Ni}^{2+}} = \text{tet.:}0.55\text{\AA}$  and  $\text{oct.:}0.69\text{\AA}$  [42], and  $r_{\text{La}^{3+}} = 1.05\text{\AA}$  [44] are the cationic radii of  $\text{Mg}^{2+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Ni}^{2+}$ , and  $\text{La}^{3+}$  cations, respectively. The calculated ionic radii values for octahedral ( $r_B$ ) and tetrahedral ( $r_A$ ) sites are listed in Table 2. The theoretical lattice constant ( $a_{\text{th}}$ ) was determined via following equation;

$$a_{\text{th}} = \frac{8}{\sqrt{3}} [(r_A + R_o) + 1.73 (r_B + R_o)] \quad (16)$$

where  $R_o = 1.32\text{\AA}$  is the ionic radii of oxygen. The values of “ $a_{\text{th}}$ ” are listed in Table 2.

**Table 2** Cationic distribution at sub-lattice  $A$  and  $B$  sites, tetrahedral and octahedral ionic radii ( $r_A$  and  $r_B$ ), the theoretical lattice constant ( $a_{\text{th}}$ )

Samples	Tetrahedral ( $A$ ) site	Octahedral ( $B$ ) site	$r_A$ ( $\text{\AA}$ )	$r_B$ ( $\text{\AA}$ )	$a_{\text{th}}$ ( $\text{\AA}$ )
<b>Mg-CNCL Ferrite</b>	$\text{Cu}_{0.0375}\text{Ni}_{0.015}\text{Mg}_{0.0125}\text{Co}_{0.07}\text{Fe}_{0.865}$	$\text{Cu}_{0.2125}\text{Ni}_{0.135}\text{Mg}_{0.2375}\text{Co}_{0.28}\text{La}_{0.15}\text{Fe}_{0.985}$	0.5984	0.7157	8.3821
<b>Mn-CNCL Ferrite</b>	$\text{Cu}_{0.0375}\text{Ni}_{0.015}\text{Mn}_{0.2}\text{Co}_{0.07}\text{Fe}_{0.6775}$	$\text{Cu}_{0.2125}\text{Ni}_{0.135}\text{Mn}_{0.05}\text{Co}_{0.28}\text{La}_{0.15}\text{Fe}_{1.1725}$	0.6108	0.7072	8.3785
<b>Cd-CNCL Ferrite</b>	$\text{Cu}_{0.0375}\text{Ni}_{0.015}\text{Cd}_{0.25}\text{Co}_{0.07}\text{Fe}_{0.6275}$	$\text{Cu}_{0.2125}\text{Ni}_{0.135}\text{Co}_{0.28}\text{La}_{0.15}\text{Fe}_{1.2225}$	0.6438	0.7076	8.4304

The tolerance factor ( $T$ ) was estimated by using the following relation (17) [20] and values as seen in Table 3.

$$T = \frac{1}{\sqrt{3}} \left( \frac{r_A + R_o}{r_B + R_o} \right) + \frac{1}{\sqrt{2}} \left( \frac{R_o}{r_A + R_o} \right) \quad (17)$$

The tolerance factor must be close to one for the spinel matrix of soft ferrites [20]. The oxygen positional parameter ( $u$ ) was determined via the relation (18) [20];

$$u = (r_A + R_O) \frac{1}{\sqrt{3}a} + \frac{1}{4} \quad (18)$$

It was revealed from Table 3 that the difference in the anion displacement from the ideal structure indicates the small variation of “ $u$ ” from its ideal value of 0.375 Å due to the synthesis and sintering process [20]. At the A-site ( $R_A$ ) and B- site ( $R_B$ ) bond lengths were determined using [33];

$$R_A = (u - 0.25)(a\sqrt{3}) \quad (19)$$

$$R_B = a \sqrt{\left(3u^2 - \frac{11}{4}u + \frac{43}{64}\right)} \quad (20)$$

The values of “ $R_A$ ” and “ $R_B$ ” are listed in Table 3. The shared octahedral edge length ( $d_{BL}$ ), unshared octahedral edge lengths ( $d_{BLU}$ ), and tetrahedral edge length ( $d_{AL}$ ) were estimated using relations [33];

$$d_{BL} = \sqrt{2} \left(2u - \frac{1}{2}\right) a \quad (21)$$

$$d_{BLU} = \left(\sqrt{4u^2 - 3u + \frac{11}{16}}\right) a \quad (22)$$

$$d_{AL} = \sqrt{2} \left(2u - \frac{1}{2}\right) a \quad (23)$$

The bond edge length values are given in Table 3. Inter-ionic lengths are associated with interactions of metal exchange and also reveal the magnetic properties of metal ions. The interionic distance between the ions i.e., Me-O (cation-anion) and Me-Me (cation-cation) interactions are calculated using the following formulas [33];

**Me- O interaction (cation-anion):**

$$p = \left(\frac{5}{8} - u\right) a \quad (24) \quad q = \left(U - \frac{1}{4}\right) \sqrt{3}a \quad (25)$$

$$r = \left(u - \frac{1}{4}\right) \sqrt{11}a \quad (26) \quad s = \left(\frac{1}{3}u + \frac{1}{8}\right) \sqrt{3}a \quad (27)$$

**Metal and metal interaction (cation-cation):**

$$b = \sqrt{2} \left(\frac{a}{4}\right) \quad (28) \quad c = \sqrt{11} \left(\frac{a}{8}\right) \quad (29)$$

$$d = \sqrt{3} \left(\frac{a}{4}\right) \quad (30) \quad e = \sqrt{3} \left(\frac{3a}{8}\right) \quad (31) \quad f = \sqrt{6} \left(\frac{a}{4}\right) \quad (32)$$

The calculated values of Me-O (p-s) and Me-Me (b-f) are reported in Table 4. The variation in the calculated values of inter-ionic distance between metal ions observed due to doping of divalent  $Mg^{2+}$ ,  $Mn^{2+}$ , and  $Cd^{2+}$  ions and interionic distance used to determine the  $\theta_1$ ,  $\theta_2$ ,  $\theta_3$ ,  $\theta_4$ , and  $\theta_5$  bond angles using equations [45];

$$\theta_1 = \left(\frac{p^2 + q^2 - c^2}{2pq}\right) \quad (33) \quad \theta_2 = \left(\frac{p^2 + r^2 - e^2}{2pr}\right) \quad (34)$$

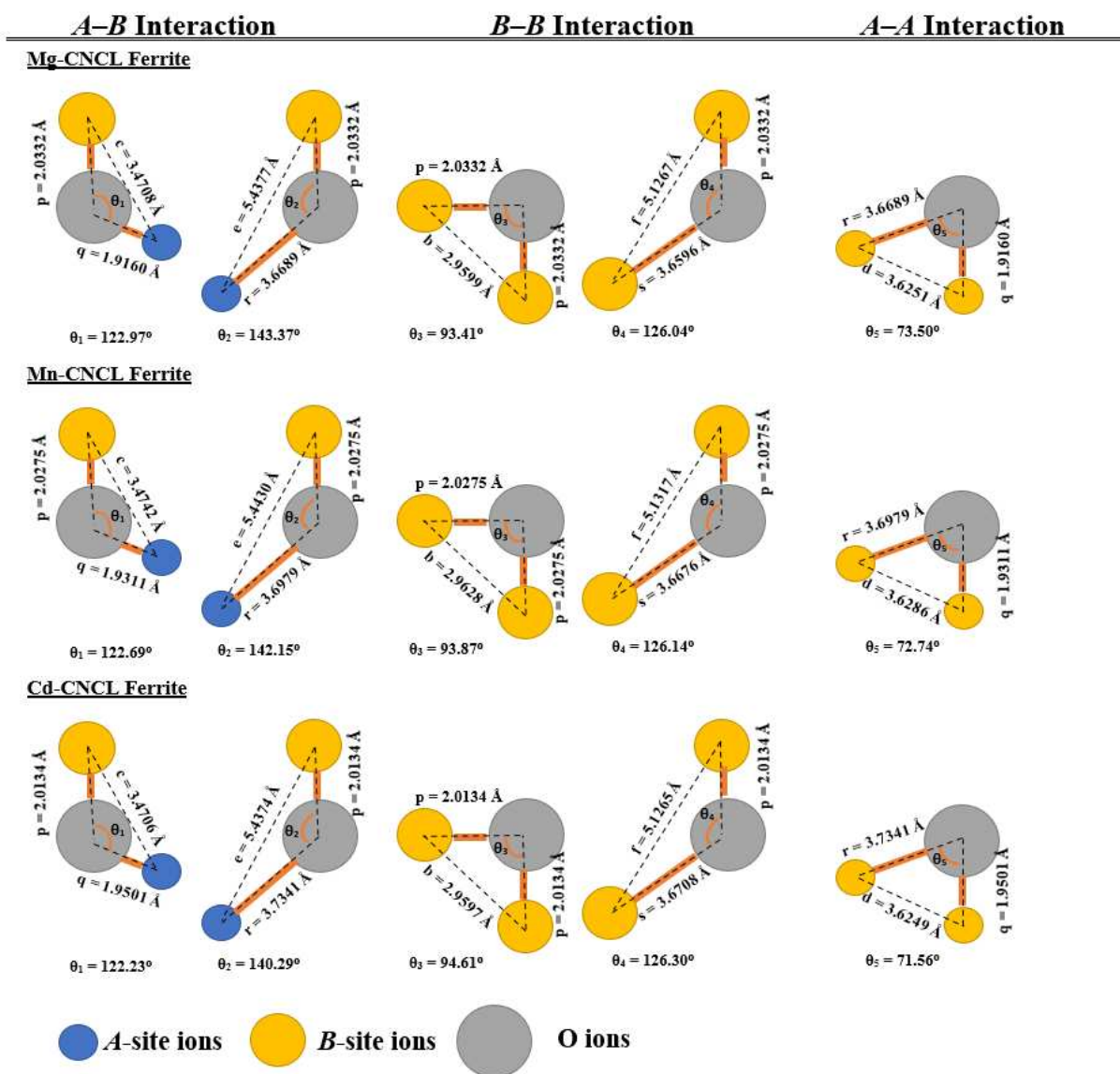
$$\theta_3 = \left( \frac{2p^2 - b^2}{2p^2} \right) \quad (35) \quad \theta_4 = \left( \frac{p^2 + s^2 - f^2}{2ps} \right) \quad (36)$$

$$\theta_5 = \left( \frac{r^2 + q^2 - d^2}{2rq} \right) \quad (37)$$

The bond angles values are reported in Table 4 and the pictorial representation of A–B, A–A, and B–B interactions with the bond angles are depicted in Fig. 3.

**Table 3** Tolerance factor ( $T$ ), oxygen position parameter ( $u$ ), bond lengths ( $R_A$ ,  $R_B$ ), Bond edge lengths ( $d_{AL}$ ,  $d_{BL}$ , and  $d_{BLU}$ ) for all the as-synthesized ferrites

Samples	Tolerance factor	Oxygen position parameter	Bond lengths (Å)		Bond edge lengths (Å)		
	$T$	$U$	$R_A$	$R_B$	$d_{AL}$	$d_{BL}$	$d_{BLU}$
Mg-CNCL Ferrite	1.0306	0.38214	1.9161	2.0332	3.1289	2.7909	2.9623
Mn-CNCL Ferrite	1.0333	0.38305	1.9312	2.0276	3.1536	2.7721	2.9658
Cd-CNCL Ferrite	1.0345	0.38449	1.9501	2.0134	3.1845	2.7351	2.9640



**Fig. 3** *A–B*, *B–B*, and *A–A* interactions along with bond angles

**Table 4** The bond lengths between Me-O (cation-anion), Me-Me (cation-cation) interactions, and bond angles

Parameters		Mg-CNCL Ferrite	Mn-CNCL Ferrite	Cd-CNCL Ferrite
Cation–Anion (Me–O)	p (Å)	2.0332	2.0275	2.0134
	q (Å)	1.9160	1.9311	1.9501
Bond length	r (Å)	3.6689	3.6979	3.7341
	s (Å)	3.6596	3.6676	3.6708
Cation–Cation (Me–Me)	b (Å)	2.9599	2.9628	2.9597
	c (Å)	3.4708	3.4742	3.4706
Bond length	d (Å)	3.6251	3.6286	3.6249
	e (Å)	5.4377	5.4430	5.4374
Bond length Angles	f (Å)	5.1267	5.1317	5.1265
	$\theta_1$	122.97	122.69	122.23
	$\theta_2$	143.37	142.15	140.29
	$\theta_3$	93.41	93.87	94.61
	$\theta_4$	126.04	126.14	126.30
	$\theta_5$	73.50	72.74	71.56

### 3.2 Absorption bands analysis

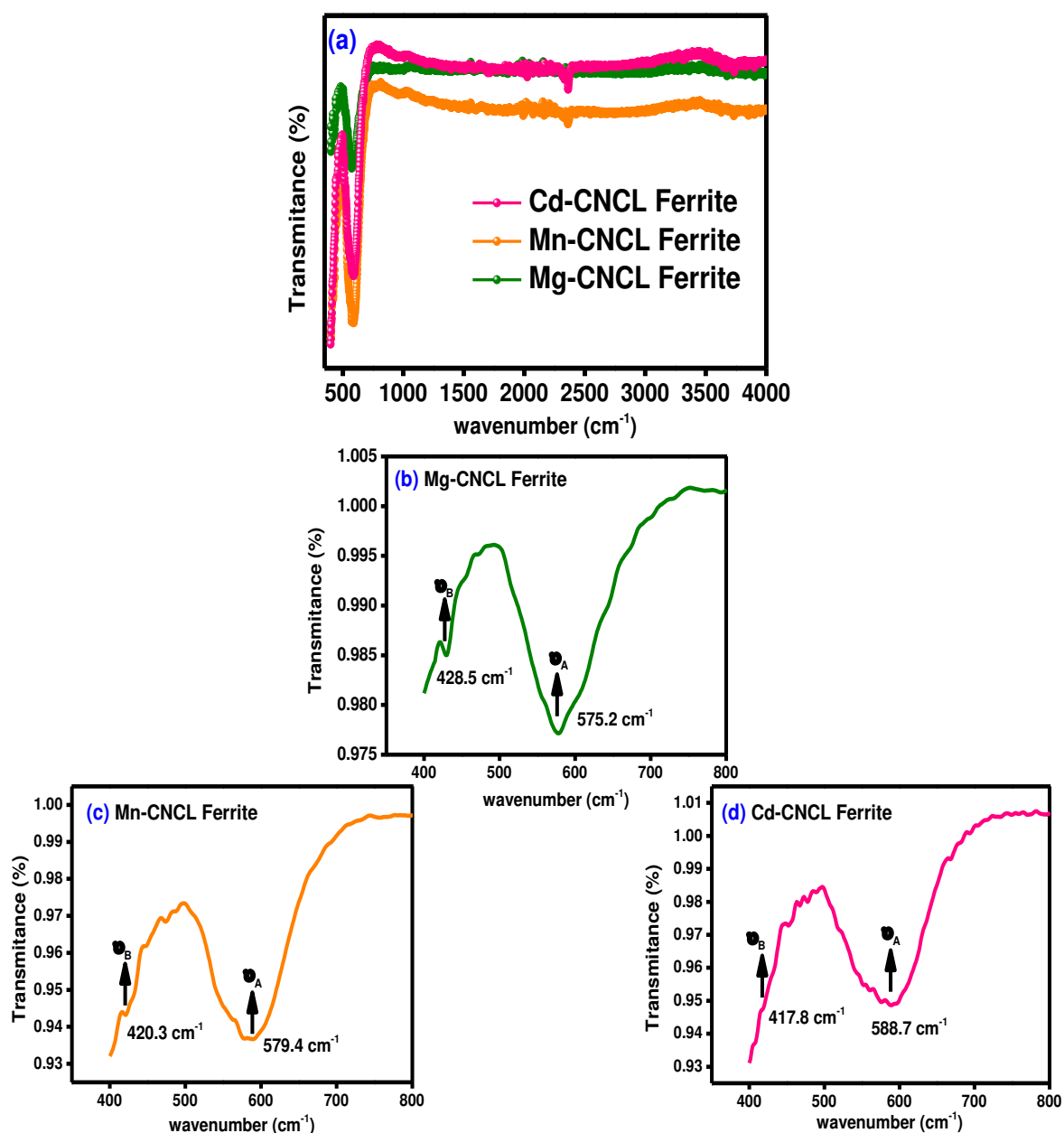
FTIR spectroscopy is a spectroscopic technique that uses the existence of various absorption bands in the lattice sites to gain information about the location of ions within the lattice sites as well as cation distribution [20]. FTIR spectra in the range of 400-4000  $\text{cm}^{-1}$  for the as-prepared ferrites are given in Fig. 4(a). The zoom view of FTIR spectra for all the as-prepared samples is given in Fig. 4(b-c) in the range of 400-800  $\text{cm}^{-1}$ . The two major absorption bands were found i.e., the high-frequency band ( $\nu_T$ ) at the tetrahedral (A-) site around 500-600  $\text{cm}^{-1}$  (Table 5) while the low frequency band ( $\nu_O$ ) at the octahedral (B-) site 400-500  $\text{cm}^{-1}$  (Table 5). The peaks around 2361  $\text{cm}^{-1}$  are from ambient  $\text{CO}_2$  gas [20]. The force constants ( $K$ ) were determined *via* the following relation [31];

$$K = 4\pi^2 \nu^2 c^2 m \quad (38)$$

where  $m$ ,  $c$ , and  $\nu$  represent the reduced mass, speed of light, and wavenumber, respectively. The force constant changed with the addition of the divalent cations  $\text{Mg}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{Cd}^{2+}$  at octahedral ( $K_O$ ) and tetrahedral ( $K_T$ ) sites are given in Table 5.

**Table 5** FTIR parameters of the as-synthesized samples

Samples	$\nu_T$	$\nu_O$	$K_T$	$K_O$
	(cm <sup>-1</sup> )		$\times 10^5$ (dyne/cm <sup>-1</sup> )	
Mg-CNCL Ferrite	575.2	428.5	3.0576	1.6968
Mn-CNCL Ferrite	579.4	420.3	3.1024	1.6325
Cd-CNCL Ferrite	588.7	417.8	3.2028	1.6131



**Fig. 4(a)** Combine FTIR spectra **(b-d)** Zoom view of FTIR spectra for the as-prepared samples

### 3.3 Vibrational bands analysis

Raman spectra as shown in Fig. 5 in the range of 150-800 cm<sup>-1</sup> were recorded and used to investigate the vibrational bands and also structural characteristics associated with the as-synthesized ferrite samples. Fig. 5 revealed the most prominent peaks of Raman modes  $E_g$ ,  $T_{2g}$  (3), and  $A_{1g}$  (2) of all the samples, and their values are reported in Table 6.

The symmetric stretching of the oxygen ion is linked with the  $A_{1g}$  mode, the symmetric bending of the oxygen anion is due to the  $E_g$  mode, and the asymmetric stretching of the oxygen ion is attributed to the  $T_{2g}$  mode with respect to the  $A$ - and  $B$ - sites cations [20]. It was observed

that in Fig. 5 and Table 6 the as-synthesized ferrites have Raman modes at  $\sim 195\text{ cm}^{-1}$ ,  $\sim 320\text{ cm}^{-1}$ ,  $\sim 475\text{ cm}^{-1}$ ,  $\sim 558\text{ cm}^{-1}$ ,  $\sim 609\text{ cm}^{-1}$ ,  $\sim 687\text{ cm}^{-1}$ . At around  $687\text{ cm}^{-1}$ , Raman modes at the lower wavenumber side ( $\sim 609\text{ cm}^{-1}$ ) revealed a shoulder-like peak. At the tetrahedral site, the stretching vibrations of the Fe–O, and M–O bonds were demonstrated by the  $A_{1g}$  (1) and  $A_{1g}$  (2) modes [46, 47]. At the B- site,  $E_g$  and  $T_{2g}$  (3) was due to asymmetric, and symmetric bending of oxygen [46, 47] relative to Fe seemed at  $\sim 320\text{ cm}^{-1}$  and  $\sim 558\text{ cm}^{-1}$ , respectively. Asymmetric stretching of the Fe-O bond also at the B- site is represented by  $T_{2g}$  (2) [46, 47] and observed at  $\sim 475\text{ cm}^{-1}$ . The  $T_{2g}$  (1) mode corresponds to tetrahedron translational motion (Fe-O<sub>4</sub>) [46, 47] and is present at  $\sim 195\text{ cm}^{-1}$ .

**Table 6** Raman modes of as-synthesized ferrite samples

Samples	$T_{2g}$ (1)	$E_g$	$T_{2g}$ (2)	$T_{2g}$ (3)	$A_{1g}$ (2)	$A_{1g}$ (1)
	Tetrahedral site	Octahedral site	Tetrahedral site			
$\text{cm}^{-1}$						
<b>Mg-CNCL Ferrite</b>	195	320	475	558	609	687
<b>Mn-CNCL Ferrite</b>	197	324	464	559	618	686
<b>Cd-CNCL Ferrite</b>	199	325	463	560	619	685

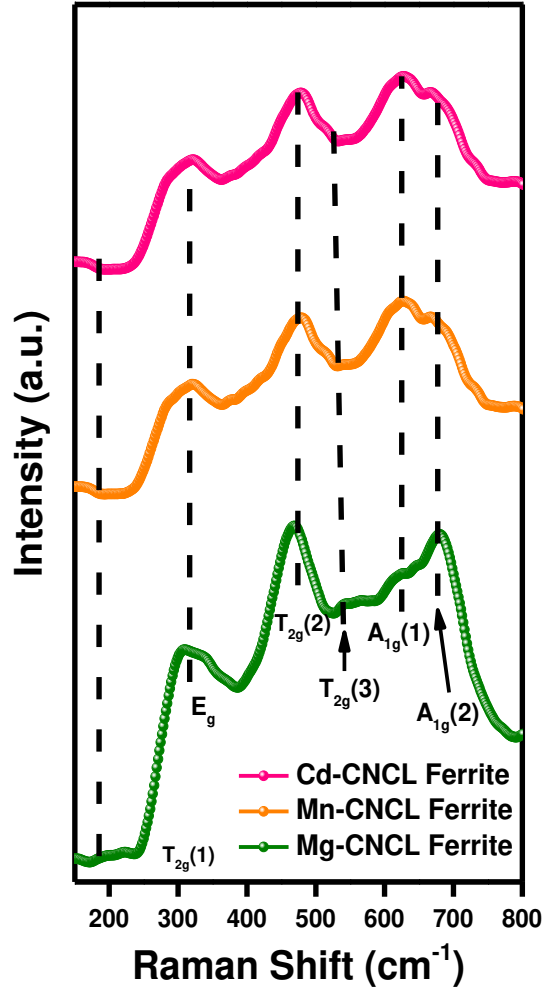


Fig. 5 Raman spectra of the as-prepared ferrites

### 3.4 Optical bandgap energy analysis

The optical bandgap energy ( $E_g$ ) was studied using UV-vis spectroscopy for the as-synthesized ferrite samples. Equation (39) is used to calculate the coefficient of absorption given as [48];

$$\alpha = 2.303 \frac{\log \log A}{t} \quad (39)$$

where “A” is the absorbance and “t” indicates the length in which the sample was placed. Tauc’s relation was utilized to determine “ $E_g$ ” which is given as [48];

$$\alpha h\nu = B(h\nu - E_g)^m \quad (40)$$

where, “m” is a constant which is 2 and  $\frac{1}{2}$  for an indirect and direct bandgap of the samples, respectively. Fig. 6(a-c) shows the plots of “ $h\nu$ ” versus  $(\alpha h\nu)^2$  used to calculate “ $E_g$ ” of the as-synthesized samples. The calculated optical bandgap energy ( $E_g$ ) for all the samples is given



in Fig. 6(a-c) and the maximum value was 1.95 eV for the Cd-CNCL Ferrite. The as-prepared samples optical bandgap energy suggest that the materials are a good candidate for optical applications including visible light absorption application.

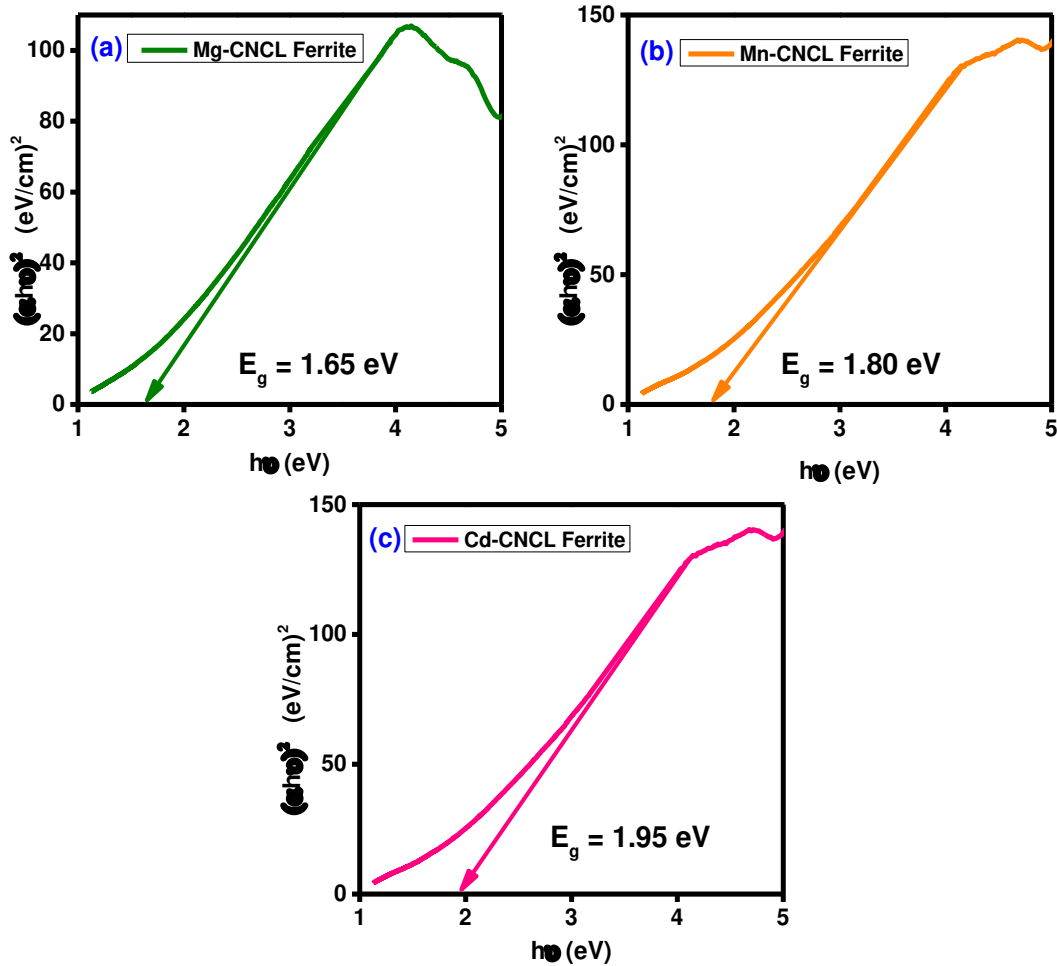


Fig. 6(a-c) Tauc's plots for the as-prepared samples

### 3.5 I-V Characterization analysis

The electrical properties of the spinel ferrites were altered by the partial replacement of  $\text{Fe}^{3+}$  with divalent and trivalent cations, which caused lattice distortion and strain. The electrical resistivity ( $\rho$ ) depends on the synthesized process and composition which varies with the distribution of cations at the A- and B- sites [16]. The electrical resistivity of the as-prepared samples was determined via the following relation [49];

$$\rho = \frac{RA}{L} \quad (41)$$

where,  $L$  = thickness of pellet,  $A$  = Area of the pellet. Fig 7 shows the Arrhenius plots of the as-prepared samples. The activation energy ( $\Delta E$ ) was estimated by taking the slope of Arrhenius plots using the following relation [49];

$$\Delta E = 2.303 \times k_B \times 1000 \times slope (eV) \quad (42)$$

where  $k_B$  represents Boltzmann constant. The Curie temperature ( $T_C$ ) is the temperature at which a material transitions from one state of electrical resistivity to another state (as seen in Fig 7). At " $T_C$ " magnetic disorder occurs within the as-prepared ferrites. Also, the ferrite's material behavior changes from ferromagnetic to paramagnetic. The region below the " $T_C$ " is known as the ferro region and above the " $T_C$ " is known as the para region. The calculated values of activation energy for the as-synthesized ferrites are given in Fig 8.

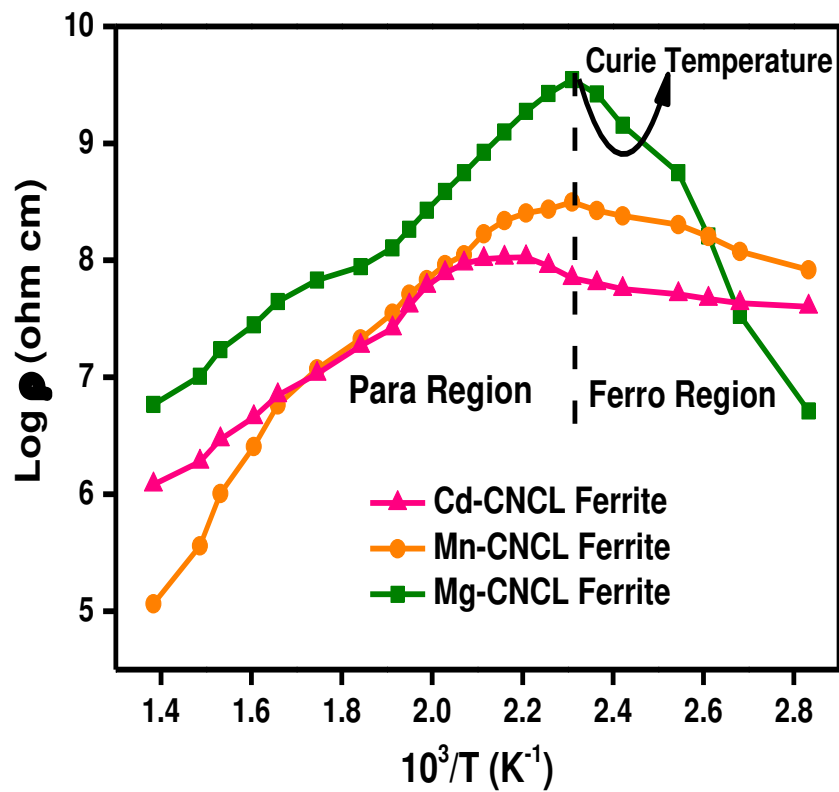
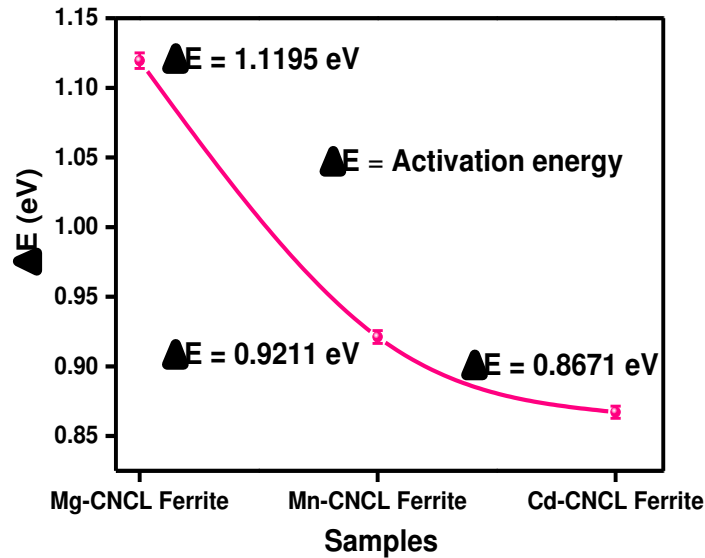
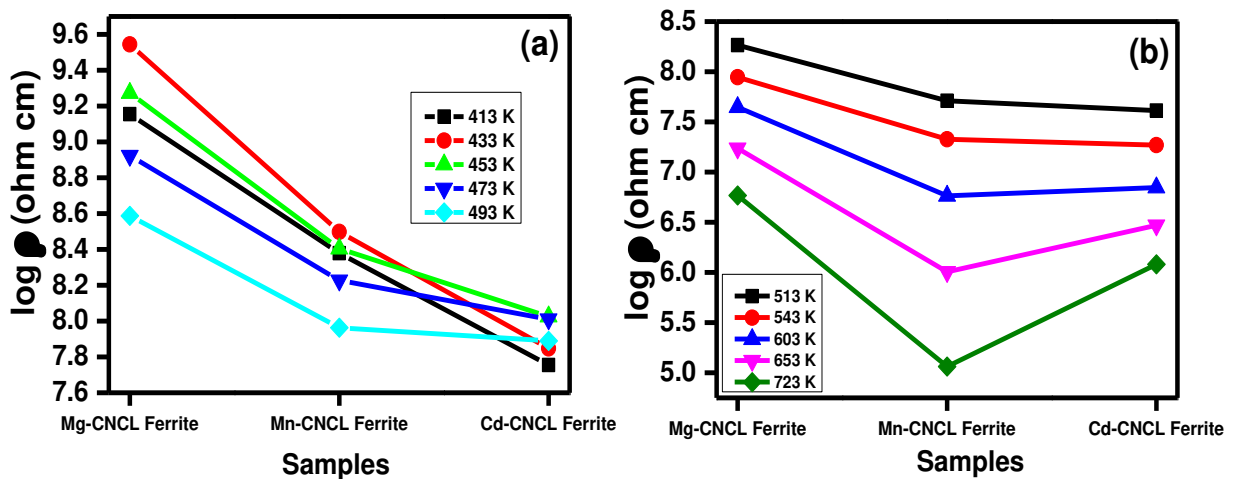


Fig. 7 Graph between  $10^3/T$  and log of resistivity



**Fig. 8** Activation energy for all the samples

The electrical resistivity was decreased as the temperature in the para region increased as represented in Fig 9(a-b). The maximum electrical resistivity was observed for the Mg-CNCL Ferrite. The change in electrical resistivity may be due to charge hopping caused by different conducting channels including  $\text{Fe}^{3+}\text{-Fe}^{3+}$  and  $\text{Fe}^{3+}\text{-Me}$  (Me is  $\text{Cd}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{La}^{3+}$ ) [16]. The high resistivity suggests that the ferrites are useful for computer memories, electrical and electronic devices, high-density information storage, high-quality filters, high-frequency transformers, magnetic recording media, magnetic resonance imaging, magnetic sensors, microwave absorption, multi-layered chip inductors, power applications, radio frequency circuits, transformer cores, microwave devices, and many more [50].



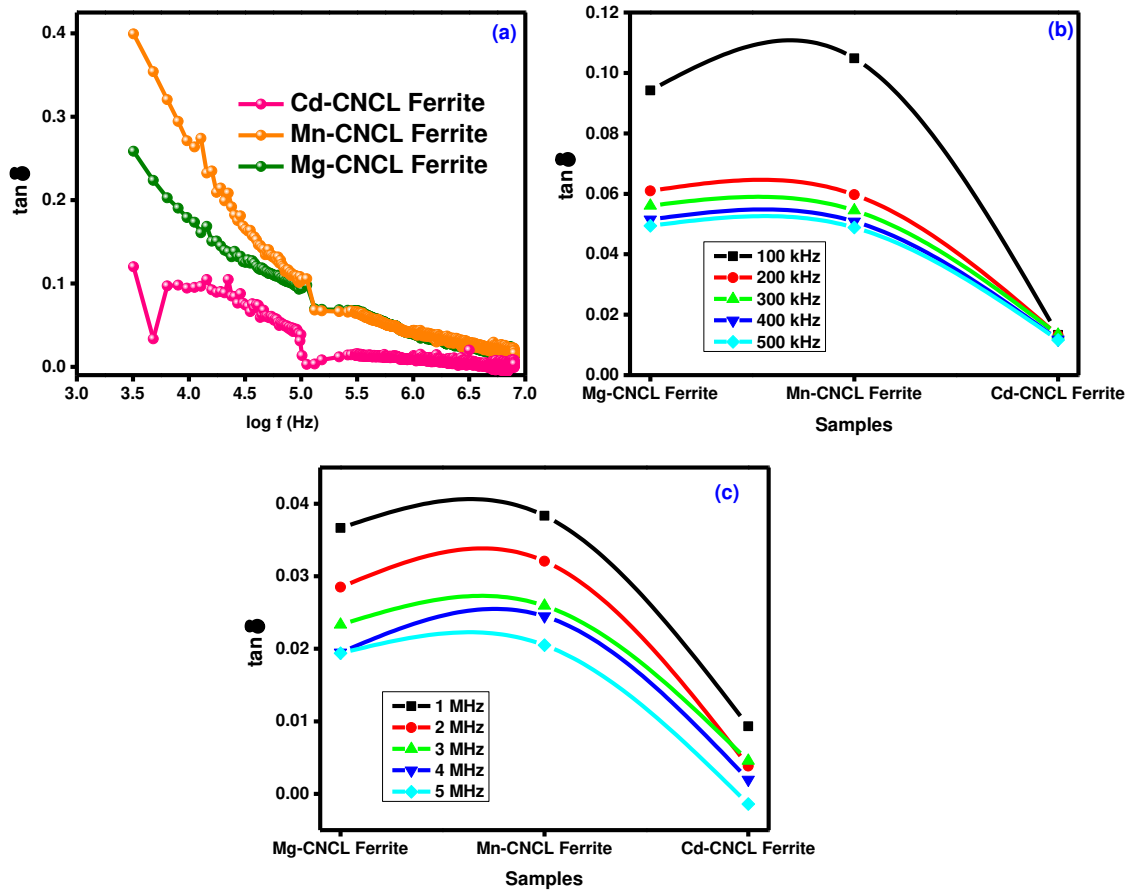
**Fig. 9(a)** Electrical resistivity in ferromagnetic region **(b)** Electrical resistivity in paramagnetic region

### 3.6 Dielectric analysis:

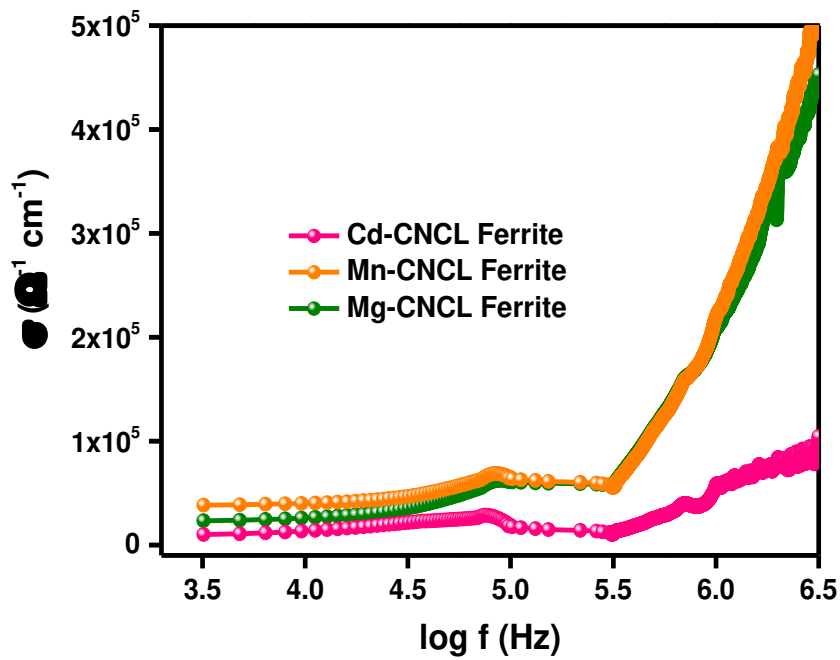
The particle composition, distribution of cations, preparation technique, temperature of sintering, and the ratio between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  can change the dielectric characteristics of the ferrites [51]. The low value of the dielectric tangent loss for they could be used in microwave applications [52]. The dielectric tangent loss is reduced with rising frequency and at a certain frequency limit, the dielectric tangent loss ( $\tan\delta$ ) becomes almost constant (as depicted in Fig. 10(a)). In the presence of an applied AC field, dielectric tangent loss ( $\tan\delta$ ) is the electrical energy lost in the form of heat. At lower frequencies (Fig. 10(b)) the  $\tan\delta$  is higher because at lower frequency values there is a high resistivity in the polarization process. At high frequencies (Fig. 10(c)) the resistivity magnitudes are low in the polarization process, therefore the dielectric tangent loss values were small. So, in the present study, it is noted that for the sample CD the dielectric tangent loss was a minimum. The AC conductivity ( $\sigma$ ) was determined by using equation [53];

$$\sigma = 2\pi f \tan\delta \quad (43)$$

Fig. 11 revealed that the log of frequency versus the AC conductivity ( $\sigma$ ) and minimum AC conductivity ( $\sigma$ ) was observed for the CD sample. The values of “ $\sigma$ ” were increased as the frequency increased due to the low conductivity of the grain boundaries. This change in AC conductivity ( $\sigma$ ) is due to grain boundaries possessing low conductivity and the grains are highly conductive which is well-matched with Koop's theory of polarization [53].



**Fig. 10** (a) Log of frequency *versus* dielectric tangent loss (b-c) dielectric tangent loss at different frequencies



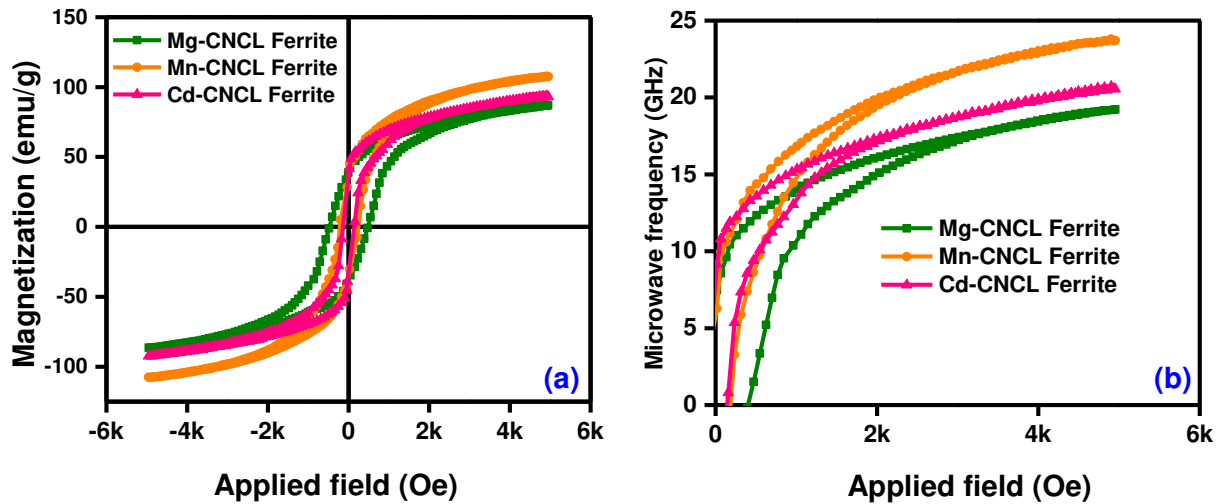
**Fig. 11** log of frequency *versus* AC conductivity of the as-prepared samples

### 3.7 Magnetic properties

Data storage, magnetization switching devices, spintronic devices, and functional materials could all benefit from the magnetic performance of conventional spinel ferrites [54]. The Mg-CNCL, Mn-CNCL, and Cd-CNCL sample's  $M$ - $H$  loops are depicted in Fig. 12(a), while Fig. 12(b) gives the applied field *versus* microwave frequency ( $\omega_m$ ) plots for all the samples. The S-shaped hysteresis loops were seen in all samples, which are typical loops for soft magnetic materials, indicating that the material has soft ferromagnetic properties. The anisotropic density,  $A$ - $B$  interactions, and grain growth are all factors that influence magnetic parameters [55]. The magnetic characteristics of CNCL ferrite are considerably influenced by the addition of  $Mg^{2+}$ ,  $Mn^{2+}$ , and  $Cd^{2+}$  ions. The remanence ( $M_r$ ), coercivity ( $H_c$ ), saturation ( $M_s$ ) magnetization, anisotropy constant ( $K$ ), squareness ratio ( $SQ$ ), initial permeability ( $\mu_i$ ), and microwave frequency ( $\omega_m$ ) of all the samples are seen in Table 7. The minimum remanence and maximum saturation magnetization were observed for the Mn-CNCL sample (as seen in Table 7) and the squareness ratio was also minimum for the Mn-CNCL sample (as seen in Table 7). The variation in saturation magnetization may be due to different magnetic moment of the divalent ion such as  $Mg^{2+}$  ( $0 \mu_B$ ) [56],  $Mn^{2+}$  ( $4.5 \mu_B$ ) [57], and  $Cd^{2+}$  ( $0 \mu_B$ ) [56]. The reduction in coerciveness (as seen in Table 7) with the addition of divalent ions ( $Mg^{2+}$ ,  $Mn^{2+}$ , and  $Cd^{2+}$ ) was due to a reduction in anisotropic fields, which reduced the domain wall energy coercive. The small coercivity indicates that the as-synthesized ferrites can be easily demagnetized for electromagnetic applications. The anisotropy constant ( $K = \frac{H_c \times M_s}{0.96}$ ) [33], and initial permeability ( $\mu_i = \frac{M_s^2 \times D}{K}$ ) [33] were determined and both showed an opposite trend with the doping of  $Mg^{2+}$ ,  $Mn^{2+}$ , and  $Cd^{2+}$  ions as reported in Table 7. The microwave frequency ( $\omega_m = 8\pi^2 M_s \gamma$ ) was calculated [16, 48, 58-60], where gyromagnetic fraction ( $\gamma$ ) is 2.8 MHz/Oe. The microwave frequency is proportional to the saturation magnetization, and the smaller the microwave frequency means the saturation magnetization is small. The microwave frequency was maximum for the Mn-CNCL sample.

**Table 7** Magnetic parameters of all the as-prepared samples

Samples	$H_c$ (Oe)	$SQ$	$M_r$ (emu/g)	$M_s$ (emu/g)	$K$ (erg/cm <sup>3</sup> )	$\mu_i$	$\omega_m$ (GHz)
<b>Mg-CNCL Ferrite</b>	470.81	0.449	38.79	86.24	42294.43	9.98	19.06
<b>Mn-CNCL Ferrite</b>	214.69	0.335	35.62	106.29	23770.20	27.99	23.49
<b>Cd-CNCL Ferrite</b>	136.41	0.395	36.52	92.29	13113.83	33.05	20.40



**Fig. 12** (a) Hysteresis ( $M-H$ ) loops (b) Applied field *versus* microwave frequency of the as-prepared samples

## Conclusions

In this present study, we report on the investigation of the structural, absorption, vibrational, optoelectrical, and dielectric properties of  $Cu_{0.25}Ni_{0.15}M_{0.25}Co_{0.35}La_{0.15}Fe_{1.85}O_4$  ( $M = Mg^{2+}$ ,  $Mn^{2+}$ , and  $Cd^{2+}$ ) ferrites prepared using SGAC process. The smallest crystallite size was observed for  $Cd^{2+}$  ferrite compared with the  $Mg^{2+}$  and  $Mn^{2+}$  doped ferrites. The lattice parameters variation was determined from XRD, and absorption bands from FTIR and Raman modes at the  $A$ - and  $B$ - sites confirmed the substitution divalent ( $Mg^{2+}$ ,  $Mn^{2+}$ , and  $Cd^{2+}$ ) cations at the lattice sites. The maximum optical bandgap energy was 1.95 eV for  $Cd^{2+}$  substituted CNCL ferrite when compared with  $Mg^{2+}$  and  $Mn^{2+}$  substituted CNCL ferrites. The resistivity was increased in the ferromagnetic region and reduced in the paramagnetic region as the temperature was increased. Furthermore, the minimum activation energy was 0.8671 eV for  $Cd^{2+}$  doped CNCL ferrite. The AC conductivities of the as-prepared ferrites increase as the frequency rises and minimum dielectric loss was found for  $Cd^{2+}$  doped CNCL ferrite compared with substituted  $Mg^{2+}$  and  $Mn^{2+}$  ions in CNCL ferrites. The smallest coercivity was observed and the microwave frequency was 20.40 GHz for  $Cd^{2+}$  doped CNCL ferrite. Therefore, the increase in conductivity may be linked to an increase in charge carrier thermally activated drift mobility and our finding suggests that such a promising function of  $Cd^{2+}$  doped CNCL ferrite, means it could be used in high-frequency and high-power applications.

## Authorship contribution statement

**Asma Aslam:** Writing – original draft, **Atta Ur Rehman:** Conceptualization, Investigation, **M. Amman:** Validation, **Maria Akhtar:** Methodology, **Nasir Amin:** Visualization, **N.A. Morley:** Writing – review & editing, **Mongi Amami:** Resources, **Hafiz T Ali:** Formal analysis

## References

- [1] K. Hussain, N. Amin, M.I. Arshad, Evaluation of structural, optical, dielectric, electrical, and magnetic properties of  $Ce^{3+}$  doped  $Cu_{0.5}Cd_{0.25}Co_{0.25}Fe_{2-x}O_4$  spinel nano-ferrites, *Ceramics International*, 47 (2021) 3401-3410.
- [2] R.S. Yadav, J. Havlica, J. Masilko, L. Kalina, J. Wasserbauer, M. Hajdúchová, V. Enev, I. Kuřitka, Z. Kožáková, Impact of  $Nd^{3+}$  in  $CoFe_2O_4$  spinel ferrite nanoparticles on cation distribution, structural and magnetic properties, *Journal of Magnetism and Magnetic materials*, 399 (2016) 109-117.
- [3] C. Kumari, H.K. Dubey, F. Naaz, P. Lahiri, Structural and optical properties of nanosized Co substituted Ni ferrites by coprecipitation method, *Phase Transitions*, (2020) 1-10.
- [4] T. Tatarchuk, I. Mironyuk, V. Kotsyubynsky, A. Shyichuk, M. Myslin, V. Boychuk, Structure, morphology and adsorption properties of titania shell immobilized onto cobalt ferrite nanoparticle core, *Journal of Molecular Liquids*, 297 (2020) 111757.
- [5] A.R. Bueno, M.L. Gregori, M.C. Nóbrega, Microwave-absorbing properties of  $Ni_{0.50-x}Zn_{0.50-x}Me_xFe_2O_4$  (Me = Cu, Mn, Mg) ferrite-wax composite in X-band frequencies, *Journal of Magnetism and Magnetic Materials*, 320 (2008) 864-870.
- [6] N. Sulaiman, M.J. Ghazali, B.Y. Majlis, J. Yunas, M. Razali, Superparamagnetic calcium ferrite nanoparticles synthesized using a simple sol-gel method for targeted drug delivery, *Bio-medical materials and engineering*, 26 (2015) S103-S110.
- [7] M. Bichurin, R. Petrov, Y.V. Kiliba, Magnetolectric microwave phase shifters, *Ferroelectrics*, 204 (1997) 311-319.
- [8] Z. Ghasemian, D. Shahbazi-Gahrouei, S. Manouchehri, Cobalt zinc ferrite nanoparticles as a potential magnetic resonance imaging agent: An in vitro study, *Avicenna journal of medical biotechnology*, 7 (2015) 64.
- [9] F. Reggia, E. Spencer, A new technique in ferrite phase shifting for beam scanning of microwave antennas, *Proceedings of the IRE*, 45 (1957) 1510-1517.
- [10] M.T. Weiss, A solid-state microwave amplifier and oscillator using ferrites, *Physical Review*, 107 (1957) 317.
- [11] J. Paulsen, A. Ring, C. Lo, J.E. Snyder, D.C. Jiles, Manganese-substituted cobalt ferrite magnetostrictive materials for magnetic stress sensor applications, *Journal of Applied Physics*, 97 (2005) 044502.
- [12] H. Tanbakuchi, D. Nicholson, B. Kunz, W. Ishak, Magnetically tunable oscillators and filters, *IEEE Transactions on Magnetics*, 25 (1989) 3248-3253.
- [13] I. Sharifi, H. Shokrollahi, S. Amiri, Ferrite-based magnetic nanofluids used in hyperthermia applications, *Journal of Magnetism and Magnetic Materials*, 324 (2012) 903-915.
- [14] N. Hiratsuka, Soft magnetic hexagonal ferrites for high frequency devices, *Journal of the Magnetics Society of Japan*, 37 (2013) 141-146.
- [15] N.M. Astik, P.K. Jha, Investigation of structural and electrical properties of mixed ferrite system, in: *AIP Conference Proceedings*, AIP Publishing LLC, 2015, pp. 070001.
- [16] A.U. Rehman, N. Morley, N. Amin, M.I. Arshad, M.A. un Nabi, K. Mahmood, A. Ali, A. Aslam, A. Bibi, M.Z. Iqbal, Controllable synthesis of  $La^{3+}$  doped  $Zn_{0.5}Co_{0.25}Cu_{0.25}Fe_{2-x}La_xO_4$  ( $x = 0.0, 0.0125, 0.025, 0.0375, 0.05$ ) nano-ferrites by sol-gel auto-combustion route, *Ceramics International*, 46 (2020) 29297-29308.



- [17] S. Woltz, R. Hiergeist, P. Görnert, C. Rüssel, Magnetite nanoparticles prepared by the glass crystallization method and their physical properties, *Journal of Magnetism and Magnetic Materials*, 298 (2006) 7-13.
- [18] S.Y. Mulushoa, N. Murali, M.T. Wegayehu, S. Margarete, K. Samatha, Influence of Cu-Cr substitution on structural, morphological, electrical and magnetic properties of magnesium ferrite, *Results in physics*, 8 (2018) 772-779.
- [19] I. ALIa, N. Amin, A. REHMAN, M. Akhtar, M. Fatima, K. Mahmood, A. ALIa, G. Mustafa, M. Hasan, A. Bibi, ELECTRICAL AND MAGNETIC PROPERTIES OF  $\text{BaCo}_x\text{Cd}_{2-x}\text{Fe}_{16}\text{O}_{27}$  W-TYPE HEXAFERRITES ( $0 \leq x \leq 0.5$ ), *Digest Journal of Nanomaterials Biostructures*, 15 (2020), 67-73.
- [20] A. Aslam, A.U. Rehman, N. Amin, M.A. un Nabi, Q. ul ain Abdullah, N. Morley, M.I. Arshad, H.T. Ali, M. Yusuf, Z. Latif, Lanthanum doped  $\text{Zn}_{0.5}\text{Co}_{0.5}\text{La}_x\text{Fe}_{2-x}\text{O}_4$  spinel ferrites synthesized via co-precipitation route to evaluate structural, vibrational, electrical, optical, dielectric, and thermoelectric properties, *Journal of Physics and Chemistry of Solids*, 154 (2021) 110080.
- [21] N. Amin, M. Akhtar, M. Sabir, K. Mahmood, A. ALIa, G. Mustafa, M. Hasan, A. Bibi, M. Iqbal, F. Iqbal, SYNTHESIS, STRUCTURAL AND OPTICAL PROPERTIES OF Zn-SUBSTITUTED Co W-FERRITES BY COPRECIPITATION METHOD, *Journal of Ovonic Research*, 16 (2020) 11-19.
- [22] R. Melo, F. Silva, K. Moura, A. De Menezes, F. Sinfrônio, Magnetic ferrites synthesised using the microwave-hydrothermal method, *Journal of Magnetism and Magnetic Materials*, 381 (2015) 109-115.
- [23] M.N. Akhtar, A.B. Sulong, M. Akhtar, M.A. Khan, Systematic study of  $\text{Ce}^{3+}$  on the structural and magnetic properties of Cu nanosized ferrites for potential applications, *Journal of Rare earths*, 36 (2018) 156-164.
- [24] K.K. Bharathi, C. Ramana, Improved electrical and dielectric properties of La-doped Co ferrite, *Journal of Materials Research*, 26 (2011) 584-591.
- [25] P. Kumar, S. Sharma, M. Knobel, M. Singh, Effect of  $\text{La}^{3+}$  doping on the electric, dielectric and magnetic properties of cobalt ferrite processed by co-precipitation technique, *Journal of Alloys Compounds*, 508 (2010) 115-118.
- [26] N. Hamdaoui, Y. Azizian-Kalendaragh, M. Khlifi, L. Beji, Cd-doping effect on morphologic, structural, magnetic and electrical properties of  $\text{Ni}_{0.6-x}\text{Cd}_x\text{Mg}_{0.4}\text{Fe}_2\text{O}_4$  spinel ferrite ( $0 \leq x \leq 0.4$ ), *Journal of Alloys Compounds*, 803 (2019) 964-970.
- [27] M.M. Hossen, M.B. Hossen, Structural, electrical and magnetic properties of  $\text{Ni}_{0.5}\text{Cu}_{0.2}\text{Cd}_{0.3}\text{La}_x\text{Fe}_{2-x}\text{O}_4$  nano-ferrites due to lanthanum doping in the place of trivalent iron, *Physica B: Condensed Matter*, 585 (2020) 412116.
- [28] S. Lorentzou, K. Karadimitra, C. Agrafiotis, A. Konstandopoulos, New routes for ferrite powders synthesis, in: *PARTEC 2004, International Conference for Particle Technology, Nuremberg, Germany, Citeseer, 2004*, pp. 16-18.
- [29] E.C. Devi, P. Soibam, Effect of Zn doping on the structural, electrical and magnetic properties of  $\text{MnFe}_2\text{O}_4$  nanoparticles, *Indian Journal of Physics*, 91 (2017) 861-867.
- [30] S. Deepapriya, S.L. Devi, P.A. Vinosha, J.D. Rodney, C.J. Raj, J.E. Jose, Das, Estimating the ionicity of an inverse spinel ferrite and the cation distribution of La-doped  $\text{NiFe}_2\text{O}_4$  nanocrystals for gas sensing properties, *Applied Physics A*, 125 (2019) 1-13.
- [31] A. Aslam, A. Razzaq, S. Naz, N. Amin, M.I. Arshad, M.A.U. Nabi, A. Nawaz, K. Mahmood, A. Bibi, F. Iqbal, Impact of Lanthanum-Doping on the Physical and Electrical Properties of Cobalt Ferrites, *Journal of Superconductivity and Novel Magnetism*, (2021) 1-10.

- [32] L. George, C. Viji, M. Maheen, E. Mohammed, Enhanced magnetic properties at low temperature of Mn substituted Ni-Zn mixed ferrite doped with Gd ions for magnetoresistive applications, *Materials Research Bulletin*, 126 (2020) 110833.
- [33] K. Hussain, A. Bibi, F. Jabeen, N. Amin, K. Mahmood, A. Ali, M.Z. Iqbal, M. Arshad, Study of structural, optical, electrical and magnetic properties of Cu<sup>2+</sup> doped Zn<sub>0.4</sub>Co<sub>0.6-x</sub>Ce<sub>0.1</sub>Fe<sub>1.9</sub>O<sub>4</sub> spinel ferrites, *Physica B: Condensed Matter*, 584 (2020) 412078.
- [34] M.A.U. Nabi, M. Moin, M. Hasan, M. Arshad, A. Bibi, N. Amin, K. Mahmood, S. Ali, Study of Electrical Transport Properties of Cadmium-Doped Zn–Mn Soft Ferrites by Co-precipitation Method, *Journal of Superconductivity and Novel Magnetism*, (2020) 1-10.
- [35] R. Kershi, Rare-earth ions as a key influencer on the magnetic, spectroscopic and elastic properties of Er<sub>γ</sub>Zn<sub>0.2</sub>Co<sub>0.8</sub>Fe<sub>2-γ</sub>O<sub>4</sub> nanoparticles, *Journal of Alloys and Compound*, (2020) 158114.
- [36] B.P. Rao, B. Dhanalakshmi, S. Ramesh, P.M. Rao, M. Materials, Cation distribution of Ni-Zn-Mn ferrite nanoparticles, *Journal of Magnetism and Magnetic Materials*, 456 (2018) 444-450.
- [37] C.A.P. Gómez, C.A.B. Meneses, S. Matute, Structural parameters and cation distributions in solid state synthesized Ni-Zn ferrites, *Material Science and Engineering: B*, 236 (2018) 48-55.
- [38] I. Ahmad, T. Abbas, M. Islam, A. Maqsood, Study of cation distribution for Cu–Co nanoferrites synthesized by the sol–gel method, *Ceramics International*, 39 (2013) 6735-6741.
- [39] S. Masoudpanah, S.S. Ebrahimi, M. Derakhshani, S. Mirkazemi, Structure and magnetic properties of La substituted ZnFe<sub>2</sub>O<sub>4</sub> nanoparticles synthesized by sol–gel autocombustion method, *Journal of magnetism and magnetic materials*, 370 (2014) 122-126.
- [40] E.E. Ateia, M. Ahmed, L. Salah, A. El-Gamal, Effect of rare earth oxides and La<sup>3+</sup> ion concentration on some properties of Ni–Zn ferrites, *Physica B: Condensed Matter*, 445 (2014) 60-67.
- [41] B.V. Prasad, B.R. Babu, M.S.R. Prasad, Structural and dielectric studies of Mg<sup>2+</sup> substituted Ni–Zn ferrite, *Materials Science-Poland*, 33 (2015) 806-815.
- [42] Q.-M. Wei, J.-B. Li, Y.-J. Chen, Cation distribution and infrared properties of Ni<sub>x</sub>Mn<sub>1-x</sub>Fe<sub>2</sub>O<sub>4</sub> ferrites, *Journal of Materials Science*, 36 (2001) 5115-5118.
- [43] M.B. Mohamed, M. Yehia, Cation distribution and magnetic properties of nanocrystalline gallium substituted cobalt ferrite, *Journal of Alloys and Compounds*, 615 (2014) 181-187.
- [44] K.A. Ganure, L.A. Dhale, S.E. Shirsat, K.S. Lohar, Morphological Study of Lanthanum-Doped Nano Spinel Ferrite via Normal Micelles Method, *Journal of Inorganic and Organometallic Polymers and Materials*, 28 (2018) 1821-1828.
- [45] P. Thakur, R. Sharma, V. Sharma, P. Barman, M. Kumar, D. Barman, S. Katyay, P. Sharma, Gd<sup>3+</sup> doped Mn-Zn soft ferrite nanoparticles: Superparamagnetism and its correlation with other physical properties, *Journal of Magnetism and Magnetic Materials*, 432 (2017) 208-217.
- [46] J. Massoudi, M. Smari, K. Nouri, E. Dhahri, K. Khirouni, S. Bertaina, L. Bessais, Magnetic and spectroscopic properties of Ni–Zn–Al ferrite spinel: from the nanoscale to microscale, *RSC Advances*, 10 (2020) 34556-34580.
- [47] G. Datt, M.S. Bishwas, M.M. Raja, A. Abhyankar, Observation of magnetic anomalies in one-step solvothermally synthesized nickel–cobalt ferrite nanoparticles, *Nanoscale*, 8 (2016) 5200-5213.
- [48] M.I. Arshad, M. Hasan, A.U. Rehman, M. Akhtar, N. Amin, K. Mahmood, A. Ali, T. Trakoolwilaiwan, N.T.K. Thanh, Structural, optical, electrical, dielectric, molecular vibrational and magnetic properties of La<sup>3+</sup> doped Mg–Cd–Cu ferrites prepared by Co-precipitation technique, *Ceramics International*, 48 (2022) 14246-14260.

- [49] M. Raghasudha, D. Ravinder, P. Veerasomaiah, Electrical resistivity studies of Cr doped Mg nano-ferrites, *Materials Discovery*, 2 (2015) 50-54.
- [50] S.B. Narang, K. Pubby, Nickel spinel ferrites: a review, *Journal of Magnetism and Magnetic Materials*, 519 (2021) 167163.
- [51] M.Y. Lodhi, K. Mahmood, A. Mahmood, H. Malik, M.F. Warsi, I. Shakir, M. Asghar, M.A. Khan, New  $Mg_{0.5}Co_xZn_{0.5-x}Fe_2O_4$  nano-ferrites: structural elucidation and electromagnetic behavior evaluation, *Current Applied Physics*, 14 (2014) 716-720.
- [52] S.P. Dalawai, S. Kumar, M.A.S. Aly, M. Khan, H. Zaved, R. Xing, P.N. Vasambekar, S. Liu, A review of spinel-type of ferrite thick film technology: fabrication and application, *Journal of Materials Science: Materials in Electronics*, 30 (2019) 7752-7779.
- [53] M. Arshad, S. Ikram, K. Mahmood, A. Ali, A. Nabi, N. Amin, F. Jabeen, Effect of La ions on nickel-cadmium spinel ferrites synthesized by co-precipitation method, *Journal of Ovonic Research*, 14 (2018) 27-34.
- [54] A. Hao, X. Ning, Recent advances in spinel ferrite-based thin films: synthesis, performances, applications, and beyond, *Frontiers in Materials*, (2021) 501.
- [55] H. Anwar, A. Maqsood, Enhancement of electrical and magnetic properties of  $Cd^{2+}$  doped Mn-Zn soft nanoferrites prepared by the sol-gel autocombustion method, *Journal of magnetism and magnetic materials*, 333 (2013) 46-52.
- [56] M. Eltabey, A. Massoud, C. Radu, Microstructure and superparamagnetic properties of Mg-Ni-Cd ferrites nanoparticles, *Journal of Nanomaterials*, 2014 (2014) 492832.
- [57] C. Y. Tsay, Y. C. Chiu, C. M. Lei, Hydrothermally synthesized Mg-based spinel nanoferrites: Phase formation and study on magnetic features and microwave characteristics, *Materials*, 11 (2018) 2274.
- [58] A. Majeed, M.A. Khan, F. ur Raheem, A. Hussain, F. Iqbal, G. Murtaza, M.N. Akhtar, I. Shakir, M.F. Warsi, Structural elucidation and magnetic behavior evaluation of rare earth (La, Nd, Gd, Tb, Dy) doped  $BaCoNi-X$  hexagonal nano-sized ferrites, *Journal of magnetism and magnetic materials*, 408 (2016) 147-151.
- [59] S. Gaba, A. Kumar, P.S. Rana, M. Arora, Influence of  $La^{3+}$  ion doping on physical properties of magnesium nanoferrites for microwave absorption application, *Journal of Magnetism and Magnetic Materials*, 460 (2018) 69-77.
- [60] A.U. Rehman, N. Amin, M.B. Tahir, M.A. un Nabi, N. Morley, M. Alzaid, M. Amami, M. Akhtar, M.I. Arshad, Evaluation of spectral, optoelectrical, dielectric, magnetic, and morphological properties of  $RE^{3+}$  ( $La^{3+}$ , and  $Ce^{3+}$ ) and  $Co^{2+}$  co-doped  $Zn_{0.75}Cu_{0.25}Fe_2O_4$  ferrites, *Materials Chemistry and Physics*, 275 (2022) 125301.