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Sol-gel auto-combustion preparation of M^{2+} = Mg^{2+}, Mn^{2+}, Cd^{2+} substituted $M_{0.25}Ni_{0.15}Cu_{0.25}Co_{0.35}Fe_2O_4$ ferrites and their characterizations

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Abstract

Cost-effective and controllable synthesis of $M_{0.25}Ni_{0.15}Cu_{0.25}Co_{0.35}Fe_2O_4$ (M^{2+} = Mg^{2+}, Mn^{2+}, and Cd^{2+}) ferrites *via* the sol-gel auto-combustion technique. The impact of divalent cations on the structural, dielectric, optoelectrical properties of ferrites was examined by XRD, FTIR, Raman, LCR, UV-vis, and two probe I-V measurement techniques. The crystallite size was 52.66 nm and the minimum specific surface area was observed 5.1507 m^2/g for Mg^{2+} doped NCCF ferrite. The FTIR and Raman analysis also confirmed the substitution of divalent cations (M^{2+} = Mg^{2+}, Mn^{2+}, and Cd^{2+}) at their respective lattice sites. The maximum energy bandgap was 1.67 eV Mg^{2+} doped NCCF ferrite as compared to other divalent ion-doped ferrites. The dielectric loss decreased while the ac conductivity increased with increasing frequency and the minimum values were observed for Mg^{2+} doped NCCF ferrite. The activation energy was observed maximum for Mg^{2+} doped NCCF ferrite (0.2234 eV). Due to incredible properties including small specific surface area, large energy band gap, high resistivity and loss dielectric loss of Mg^{2+} doped NCCF ferrite have potential applications in different fields.

Keywords: divalent; specific surface area; energy band; Raman; activation energy.

1 Introduction

Nanoparticles of spinel ferrites are widely used in the latest technology as telecommunications [1], usage of wastewater and photocatalytic water splitting [2-4], electronics [5], and magnetic recordings [6]. AB_2O_4 is the general formula of the soft spinel ferrites in which B^{3+} represent trivalent elements occupying octahedral sites while A^{2+} represent divalent elements occupying tetrahedral site [7]. The chemical composition, cation distribution, doping ratio with significant parameters like synthesis technique, and grain morphology affected the optoelectrical, structural, and magnetic parameters of the spinel ferrites [8]. In the electronics growth industry, such remarkable change in the electromagnetic properties of the ferrites makes them the most significant for use in advanced research applications [9, 10].

Warsi *et al.*, [11] prepared Erbium-doped Ni-Co spinel ferrite by using the co-precipitation technique. Two probe IV techniques were used to find the electrical resistivity of the prepared ferrites and it was found 6.02×10^7 Ω cm. Almessiere *et al.*, [12] studied $Ni_{0.3}Cu_{0.3}Zn_{0.4}Fe_2O_4$ synthesized by ultrasonic irradiation method. At 350 K, the maximum ac conductivity for $x = 0.02$. Sadaqat *et al.*, [13] reported Tb^{3+} substituted cobalt ferrite prepared by using the sonochemical procedure in which the lattice constant declined with the addition of Tb^{3+}. Almessiere *et al.*, [14] reported Dy^{3+} replaced $Mn_{0.5}Zn_{0.5}Fe_{2-x}O_4$ ferrites synthesized by the ultrasonic irradiation process in which the energy band gap declined calculated by using the Tauc equation. Slimani *et al.*, [15] synthesized Ni-Cu-Zn nanoparticles by the sonochemical process in which the reaction time declined which minimized the requirement of a large number of organic solvents. At 393 K, the ac conductivity is minimum for $x = 0.02$.

Various techniques are used to synthesize the nanoparticles, co-precipitation [16-21], auto combustion sol-gel techniques [22], reverse micelle technique [23], and solid-state reaction [24]. Because of the fine surface morphology, low-temperature preparation, and greater particle homogeneity, the sol-gel auto combustion approach was shown to be better [22, 25]. In this research work, we synthesized $M_{0.25}Ni_{0.15}Cu_{0.25}Co_{0.35}Fe_2O_4$ ($M^{2+} = Mg^{2+}$, Mn^{2+} and Cd^{2+}) *via* the sol-gel auto-combustion technique. The effects of divalent metal Mg^{2+}, Mn^{2+}, and Cd^{2+} ions on the optoelectrical, dielectric and structural parameters of the $M_{0.25}Ni_{0.15}Cu_{0.25}Co_{0.35}Fe_2O_4$ ferrite ferrites are discussed in this article and observed how it changed the structural parameters and electrical resistivity of the prepared ferrites.

2 Experimental details

2.1 Materials and method

$M_{0.25}Ni_{0.15}Cu_{0.25}Co_{0.35}Fe_2O_4$ ferrites doped with divalent metal Mg^{2+}, Mn^{2+} and Cd^{2+} ions (***M^{2+} doped NCCF ferrites***) were synthesized by auto-combustion sol-gel technique. All the following chemicals [$Cu (NO_3)_2.3H_2O$], [$Cd (NO_3)_2.4H_2O$], [$Mg (NO_3)_2.6H_2O$], [$Mn (NO_3)_2.4H_2O$], [$Ni (NO_3)_2.6H_2O$], [$Co (NO_3)_2.6H_2O$], [$Fe (NO_3)_3.9H_2O$] and citric acid were measured according to stoichiometry and dissolved into distilled water separately on a magnetic stirrer with continuously stirring. All the dissolved chemicals were poured into a beaker to get a homogeneous solution through stirring. Ammonia was added drop-wise into the solution to achieve a pH = 7. After maintaining pH, the solution was stirred at 80 °C to form a gel which was further converted into ash and sintered at 800 °C for 8 h. The fine powder of sintered ash was obtained through grinding and pellets of 7 mm were made by using a hydraulic press.

2.2 Characterization used

For structural analysis and phase confirmation, a Bruker D8 Advance X-ray diffractometer (XRD) with a Copper K_α source (λ = 1.54 Å) was employed. The absorption bands were studied using Fourier transforms infrared (FTIR) spectroscopy. UV–visible spectroscopy was used to determine the energy bandgap (E_g) To evaluate electrical resistivity, a Keithley Electrometer Model 2401 was used for current-voltage (I–V) measurements. For dielectric measurements, the IM3536 series LCR Meter was employed.

3 Results and discussion

3.1 XRD analysis

The structural properties of the as-prepared ferrites were studied through the XRD analysis. Fig. 1 indicates the XRD patterns of the as-prepared ferrites and planes (220), (311), (400), (422), and (511) were labeled which established the development of their cubic spinel matrix. Bragg's law was utilized to calculate the inter planer distance of the as-prepared ferrites [26];

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

where θ is Braggs' angle and λ is 1.5046 Å while n = 1. It was observed that the inter planer distance of the M^{2+} doped NCCF ferrites lies in the range of 2.5226 Å – 2.5333 Å (as seen in Table 1). The Scherrer's formula (2) was employed to find the crystallite size of the prepared ferrites and given as [26, 27];

$$D = \frac{n\lambda}{\beta Cos\theta} \qquad (2)$$

where n is a constant which is 0.94, β is FWHM and θ is the angle of diffraction. The following formula (3) [28] was used to find the experimental lattice constant of the M^{2+} doped NCCF ferrites.

$$a_{exp} = d_{hkl}\sqrt{h^2 + k^2 + l^2} \tag{3}$$

where (h k l) represents the miller indices of and 'a' is the experimental lattice constant of the prepared ferrites. Equation (4) was employed to estimate the X-ray density of the prepared ferrites and given as [28];

$$\rho_X = \frac{8M}{N_A a_{exp}^3} \tag{4}$$

where M is the molar mass of the prepared ferrites, N_A are the Avogadro's number (6.022 $\times 10^{23}$ mol^{-1}). It is observed that the lattice constant increased in the range of 8.3664 Å – 8.4021 Å due to the addition of dopant Mg^{2+}, Mn^{2+}, and Cd^{2+} ions in the NCCF ferrites. The crystallite size of the M^{2+} doped NCCF ferrites decreased in the range of 52.66 nm – 22.35 nm. It was found from Table 1 and the minimum crystallite size was 22.35 nm for the Cd^{2+} doped NCCF ferrite. Such remarkable change raised due to substitution of large ionic radii divalent ions Mg^{2+} (0.72 Å), Mn^{2+} (0.66 Å) and Cd^{2+} (0.78 Å) with Fe^{3+} (0.645 Å). The X-ray density of the as-prepared ferrites increased from 5.1507 g/cm^3 – 5.5812 g/cm^3. The bulk density of the prepared ferrites is calculated by using the given formula [29];

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

Here, 'M' is the mass of the pellet while 'h' and 'r' are the thickness and radius of the pellet respectively, and given in Table 1. The porosity percentage was estimated *via* the following formula [29];

$$P\% = [1 - \frac{\rho_b}{\rho_X}] \times 100 \tag{6}$$

Here, ρ_b and ρ_X are the bulk density and X-ray density of the M^{2+} doped NCCF ferrites. The difference in the values of ρ_b and ρ_X observed due to the existence of some pores which develop in the sintering process [30]. X-ray density and the molar mass of the prepared ferrites are directly proportional to each other so ρ_X increased while ρ_b increased due to addition of divalent metal Mg^{2+}, Mn^{2+} and Cd^{2+} content into the prepared ferrites. The porosity percentage of the M^{2+} doped NCCF ferrites increased from 43.03 % to 46.53 % and is tabulated in Table 1.

The experimental lattice constant was confirmed by the Nelson-Riley function which is given as [28];

$$F(\theta) = \frac{1}{2}\left[\frac{Cos^2\theta}{\sin\theta} + \frac{Cos^2\theta}{\theta}\right] \tag{7}$$

The values of the Nelson-Riley function demonstrate it is in good agreement with the calculated values of the experimental lattice constant (as seen in Table 1). Different lattice parameters such as strain (ε), dislocation density (δ), specific surface area (S), and packing factor (p) percentage are calculated through given formulas [28];

$$\varepsilon = \frac{1}{d^2} \tag{8}$$

$$\delta = \frac{15\varepsilon}{a_{exp}D} \tag{9}$$

$$S = \frac{6000}{\rho_{X \times D}} \tag{10}$$

$$p = \frac{D}{d} \tag{11}$$

Here a_{exp}, D, d and ρ_X are the experimental lattice constant, crystallite size, inter planer distance, and X-ray density respectively. The strain and packing factor percentage were reduced while the dislocation density and specific surface area were increased with the substitution of dopant ions as seen in Table 1. Hopping lengths L_A and L_B are calculated by using given formulas [28, 31];

$$L_A = \frac{a_{exp}\sqrt{3}}{4} \tag{12}$$

$$L_B = \frac{a_{exp}\sqrt{2}}{4} \tag{13}$$

where, L_A and L_B are hopping lengths of tetrahedral and octahedral sites respectively. L_A is the distance between cation and oxygen ion at tetrahedral site while L_B is the distance between cation and oxygen ion at octahedral site. The calculated values of L_A and L_B are reported in Table 1. M^{2+} doped NCCF ferrites has mixed spinel structure because Cu^{2+}, Co^{2+}, Mg^{2+}, Ni^{2+} $Fe^{2+/3+}$ ions prefer both A-site (tetrahedral) and B-site (octahedral) while Cd^{2+} ions prefer only tetrahedral sites [32-36]. The cation distribution of the as-prepared ferrites is determined by $[M^{2+}_{1-x}\,T^{3+}_{x}]_A\,[M^{2+}_{x}\,T^{3+}_{2-x}]_B\,O_4$ which are given in Table 2 while the ionic radii are calculated by using following formulas;

$$r_A = C_{AM}r(M^{2+}) + C_{ANi}r(Ni^{2+}) + C_{ACu}r(Cu^{2+}) + C_{ACo}r(Co^{2+}) + C_{AFe}r(Fe^{2+}) \tag{14}$$

$$r_B = \frac{1}{2}[C_{BM}r(M^{2+}) + C_{BNi}r(Ni^{2+}) + C_{BCu}r(Cu^{2+}) + C_{BCo}r(Co^{2+}) + C_{BLa}r(La^{3+}) + C_{BFe}r(Fe^{2+})] \tag{15}$$

where, $r(Cd^{2+})$, $r(Ni^{2+})$, $r(Cu^{2+})$, $r(Co^{2+})$ and $r(Fe^{2+})$ are the ionic radii while C_{Cd}, C_{Ni}, C_{Cu}, C_{Co} and $C_{Fe}r$ are the concentration of Cd^{2+}, Ni^{2+}, Cu^{2+}, Co^{2+} and Fe^{3+} respectively. The calculated values of r_A and r_B changed due to addition of divalent metal Mg^{2+}, $Mn^{2+}$and Cd^{2+} in the prepared ferrites which are given in Table 2.

The theoretical lattice constant of the M^{2+} doped NCCF ferrites are calculated by using the formula [28];

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

where, r_A, r_B is the ionic radii of A and B-site of the sub-lattice while $R_o = 1.32$ Å is the oxygen ion radius [28]. The theoretical lattice constant is tabulated in Table 2 lies in the range 8.2975 Å – 8.3496 Å due to doping of Mg^{2+} (0.72 Å), Mn^{2+} (0.66 Å), and Cd^{2+} (0.78 Å) ions into the NCCF lattice. A small difference between theoretical and experimental lattice constant is because of the cation distribution of the anions and cations in the spinel structure. The tolerance factor is calculated using the given formula [28];

$$T = \frac{1}{\sqrt{3}}\left(\frac{rA + Ro}{rB + Ro}\right) + \frac{1}{\sqrt{2}}\left(\frac{Ro}{rA + Ro}\right) \quad (17)$$

where, r_A, r_B, R_o (1.32 Å) is the ionic radii of A-site, B-site, and oxygen ion respectively. The tolerance factor is close to one which indicates the development of the spinel matrix because of tolerance factor is 1 for a perfect spinel matrix [16]. The oxygen ion parameter of the prepared ferrites is calculated by using the given formula [28];

$$U = (r_A + R_o)\,\frac{1}{\sqrt{3a}} + \frac{1}{4} \quad (18)$$

The oxygen ion parameter has an ideal value of 0.375 [16]. The chemical composition of the material, synthesis technique and sintering of the prepared ferrites may affect the oxygen ion parameter from its ideal value [16]. The calculated values of “U” are tabulated in Table 3. The smallest distance between A-site (tetrahedral) and oxygen ion is known as tetrahedral bond length (R_A) while the shortest distance between B-site and oxygen ion is known as octahedral bond length (R_B). Equations (19) and (20) were utilized to determine the tetrahedral and octahedral bond lengths of the prepared ferrites respectively and reported in Table 3.

$$R_A = a_{exp}\sqrt{3}\,\left(\delta + \frac{1}{8}\right) \quad (19)$$

$$R_B = a_{exp}\sqrt{\left(\frac{1}{16} + \frac{\delta}{2} + 3\delta^2\right)} \quad (20)$$

Here, a_{exp} is the experimental lattice constant and δ is known as the inversion parameter which is equal to $U\text{-}U_{ideal}$. It is observed that both bond lengths increased due to the addition of dopant divalent metal (Mg^{2+}, $Mn^{2+,}$ and Cd^{2+}). Equations (21), (22), and (23) are used to determine the octahedral shared edge length d_{BL}, octahedral unshared edge length $d_{BLU,}$ and tetrahedral edge length d_{AL} of the prepared ferrites and reported in Table 3.

$$d_{BL} = \sqrt{2}\left(2U - \frac{1}{2}\right) a_{exp} \quad (21)$$

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

$$d_{AL} = \sqrt{2}\left(2U - \frac{1}{2}\right) a_{exp} \quad (23)$$

Here, U is the oxygen ion parameter and a_{exp} is the experimental lattice constant. The interionic distance was determined using relations (24) and (25) [28];

Metal and metal interaction (cation- cation):

$$b = \left(\frac{a_{exp}}{4}\right)\sqrt{2},\ c = \left(\frac{a_{exp}}{8}\right)\sqrt{11},\ d = \left(\frac{a_{exp}}{4}\right)\sqrt{3},\ e = \left(\frac{3a_{exp}}{8}\right)\sqrt{3},\ f = \left(\frac{a_{exp}}{4}\right)\sqrt{6}, \tag{24}$$

M -O interaction (cation- anion):

$$p = a_{exp}\left(\frac{5}{8} - U\right),\ q = \left(U - \frac{1}{8}\right)\sqrt{3}\ ,\ r = a_{exp}\left(U - \frac{1}{8}\right)\sqrt{11},\ s = \frac{a_{exp}}{3}\left(U + \frac{1}{2}\right)\sqrt{3} \tag{25}$$

The interionic distance explains the magnetic interaction between M–O (p-s) and M–M (b-f) due to the addition of dopant Mg^{2+}, Mn^{2+}, and Cd^{2+} ions in the prepared ferrites. The trend of interionic distances increased and decreased between M–M and M–O interaction is given in Table 4.

3.2 FTIR analysis

The absorption bands of M^{2+} doped NCCF ferrites were studied by the FTIR spectra. Fig 2 indicates the spectra of FTIR which was observed in the range of 400 – 4000 cm^{-1}. The lower frequency absorption band (v_O) at octahedral site recorded in the range of 421.26 – 433.43 cm^{-1} (Table 5) which also endorses the development of the single-phase spinel cubic structure while high-frequency absorption band (v_T) at tetrahedral site recorded in the range of 575.48 – 585.21 cm^{-1} (Table 5) for all the as-prepared ferrites which also confirm the existence of M-O bond at the tetrahedral site [16].

3.3 Raman analysis

Raman spectroscopy was used to investigate the vibrational and structural characteristics of M^{2+} doped NCCF ferrites. Fig 3 elaborates the five Raman modes of the as-prepared ferrites which changed with the addition of divalent metal ions (Mg^{2+}, Mn^{2+}, and Cd^{2+}). All the peaks of Raman mods are listed in Table 6 which are observed in the range of 200- 850 cm^{-1}. Raman modes observed at low frequency (less than 500 cm^{-1}) and high-frequency mode (more than 500 cm^{-1}) belong to the octahedral site and tetrahedral site of phonon vibrations respectively which also confirm the cation distribution due to the addition of the dopant divalent metal Mg^{2+}, Mn^{2+}, and Cd^{2+} ions into the prepared ferrites [37, 38].

3.4 Dielectric analysis

The dielectric characteristics of the prepared ferrites were analyzed by LCR meter in the pellet form having a diameter of 7 mm. The effect of dopant divalent (Mg^{2+}, Mn^{2+}, Cd^{2+}) metal ions on the dielectric characteristic of the prepared ferrites were recorded in the frequency range of 4 Hz to 8 MHz. Fig. 4 indicates the dielectric constant as a frequency function of M^{2+}

doped NCCF ferrites. It is observed that the dielectric constant has an inverse relation with frequency as frequency increased, the dielectric constant decreases. This result is ascribed by a slowdown in electron hopping which reduced polarization as frequency increased because the prepared ferrites show no response to the applied external field at a high-frequency range [39, 40]. Koops and Maxwell-Wagner model [41] was used to elaborate the slowdown of hoping electrons at grain boundaries which decreased the dielectric constant at high frequencies. Fig. 5 indicates the decrease in dielectric loss of M^{2+} doped NCCF ferrites due to the addition of divalent metal ions into the M^{2+} doped NCCF ferrites. Fig. 6 revealed the ac conductivity of M^{2+} doped NCCF ferrites at a frequency range from 4 Hz to 8MHz. The ac conductivity has a direct relation with frequency as frequency increased and ac conductivity also increased (as seen in Fig. 6) which is a good match with Koops and Maxwell- Wagner's model [42].

3.5 UV-vis spectroscopy:

UV-vis spectra were used to investigate the optical characteristics of M^{2+} doped NCCF ferrites in the range of 200 nm to 800 nm. Equation (26) was used to find the absorption coefficient of the prepared ferrites as given [28];

$$\alpha = 1.303 \log A \qquad (26)$$

Here A and α indicate the absorbance and absorbance coefficient respectively. The following Tauc's equation (27) [28] was used to determine the energy bandgap of the prepared ferrites.

$$\alpha h\nu = B\,(h\nu - E_g)^m \qquad (27)$$

Here, m and B are the constants. h, ν and E_g represent the Plank's constant, frequency of the incident photon, and the energy bandgap of the prepared ferrites. Fig 7 shows the optical energy band gap by plotting between $(\alpha h\nu)^2$ and $h\nu$ of the M^{2+} doped NCCF ferrites and observed in the range of 1.67 eV to 0.85 eV as listed in Table 5. The outcomes of the prepared ferrites revealed that the Cd^{2+} doped ferrite has a low energy band gap as compared to Mg^{2+}, and Mn^{2+} doped ferrites.

3.6 Current-Voltage measurements

DC resistivity depends on the composition and synthesized technique of the prepared ferrites which varied the distribution of cations at the tetrahedral and octahedral sites. The relation was used to find out resistivity [28];

$$R = \frac{\rho L}{A} \qquad (28)$$

Here, L is the thickness of the pellet; A is the area of the pellet and ρ is the electric resistivity. Fig. 8 shows the electric resistivity of the M^{2+} doped NCCF ferrites with the addition of divalent metal ions at various temperatures. The higher exchange of charger carriers between Fe^{2+} and Fe^{3+} at the octahedral site reduced the electric resistivity. The calculated activation energy of the M^{2+} doped NCCF ferrites reveals the resistivity nature of the ferrites. Equation (29) was used to the electric resistivity of the prepared ferrites as given [28];

$$\rho = \rho_o \exp(\frac{\Delta E}{k_B T}) \qquad (29)$$

Here, ρ, ρ_o and ΔE is the electrical resistivity, temperature constant, and activation energy of the prepared ferrites respectively. k_B is the Boltzmann's constant. Fig 8 shows the electric resistivity measurements with 1000/T of the prepared ferrites. The obtained results have two regions in which the first region has the temperature range of 343 K to 413 K, while the second region has the range above 413 K. It can be seen that the Mn^{2+} doped ferrite has a high electrical resistivity while the Cd^{2+} doped ferrite has a low value of electrical resistivity in the para region. The bending curves were divided into two portions (ferromagnetic and paramagnetic) regions by kink points which indicated the transition temperature. These kink points revealed the variation of conductivity due to changing magnetic order from ferromagnetic region to paramagnetic region. The region below transition temperature is known as paramagnetic while the region above transition temperature is known as the ferromagnetic region. Fig 9 (a, b, and c) shows the electrical resistivity of the Mn^{2+} doped ferrite is high as compared to Mg^{2+} and Cd^{2+} doped ferrites. Fig 10 shows the trend of activation energy for the as-prepared ferrites and values are listed in Table 7. It was observed that the Mn^{2+} has a small value of activation energy as compared to Mg^{2+} and Cd^{2+} doped ferrites.

4. Conclusions

Sol-gel technique was employed to synthesize Mg^{2+}, Mn^{2+}, and Cd^{2+} doped NCCF nanoparticles, and their cubic spinel structure was confirmed by the XRD analysis. The minimum lattice constant was 8.3664 Å while crystallite size decreased from 52.66 to 22.35 Å and Mg^{2+} doped NCCF ferrite has a maximum value of crystallite size. The variation in the upper and lower frequency bands of the M^{2+} doped NCCF ferrites indicates the distribution of cations at tetrahedral and octahedral sites which are also confirmed by Raman spectra. The minimum energy bandgap was 0.85 eV for Cd^{2+} doped NCCF ferrite and the maximum was 1.67 eV for Mg^{2+} doped NCCF ferrite. The dielectric loss decreased with the substitution of divalent ions and minimum dielectric loss was observed for Mg^{2+} doped NCCF ferrite. Also, ac conductivity increased with an increasing applied frequency may be due to the large impedance. Mg^{2+} doped NCCF ferrite has 0.2234 eV activation energy while Mn^{2+} doped NCCF ferrite has 0.1046 eV activation energy. Hence, due to their incredible properties, Mg^{2+} doped NCCF ferrite has potential applications in different fields.

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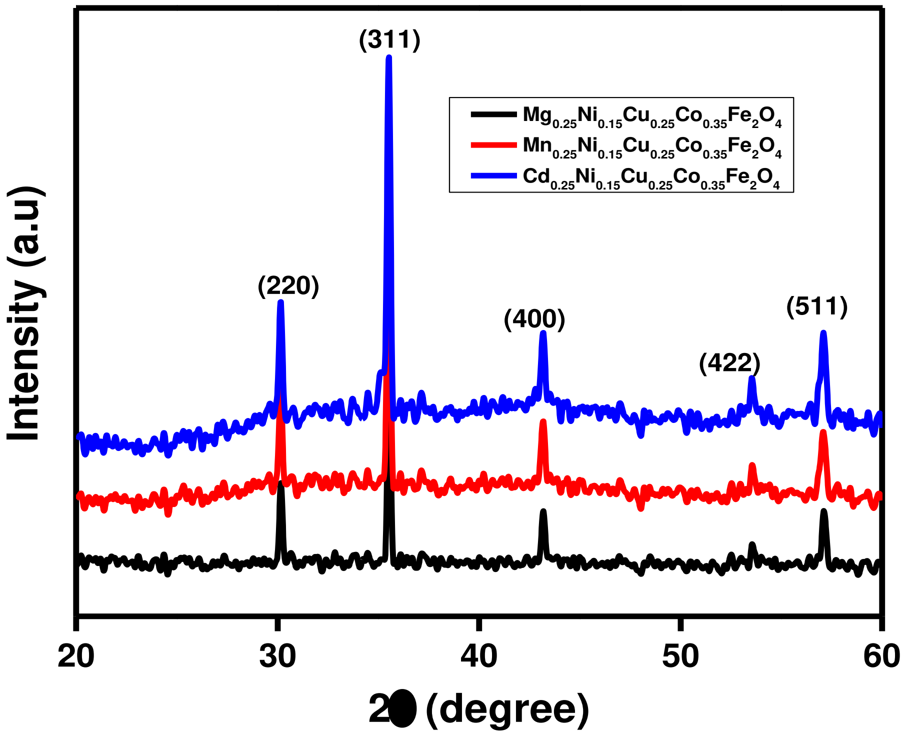

Fig.1 XRD patterns of M^{2+} doped NCCF ferrites

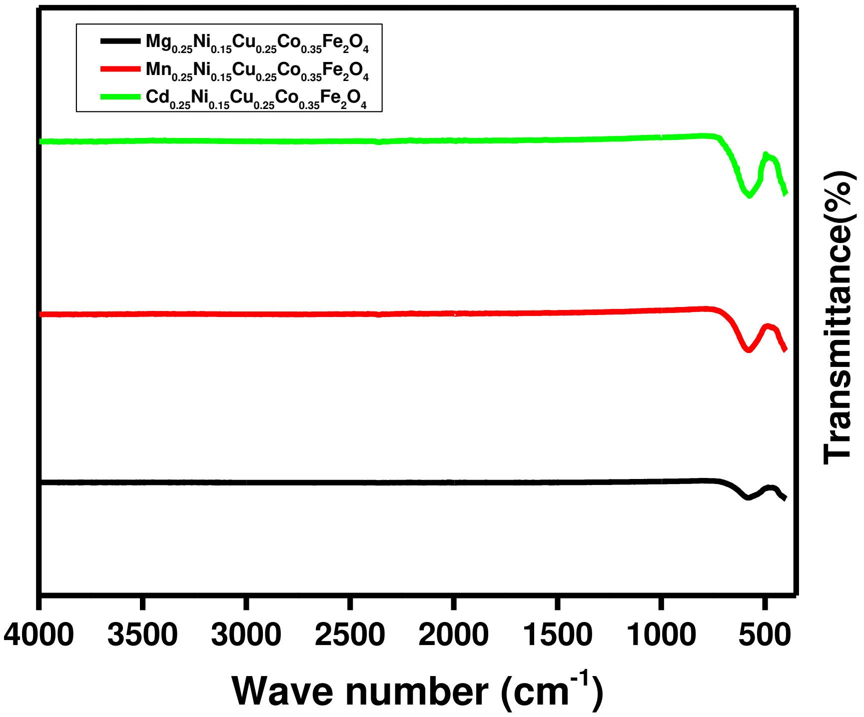

Fig. 2 FTIR spectra of M^{2+} doped NCCF ferrites

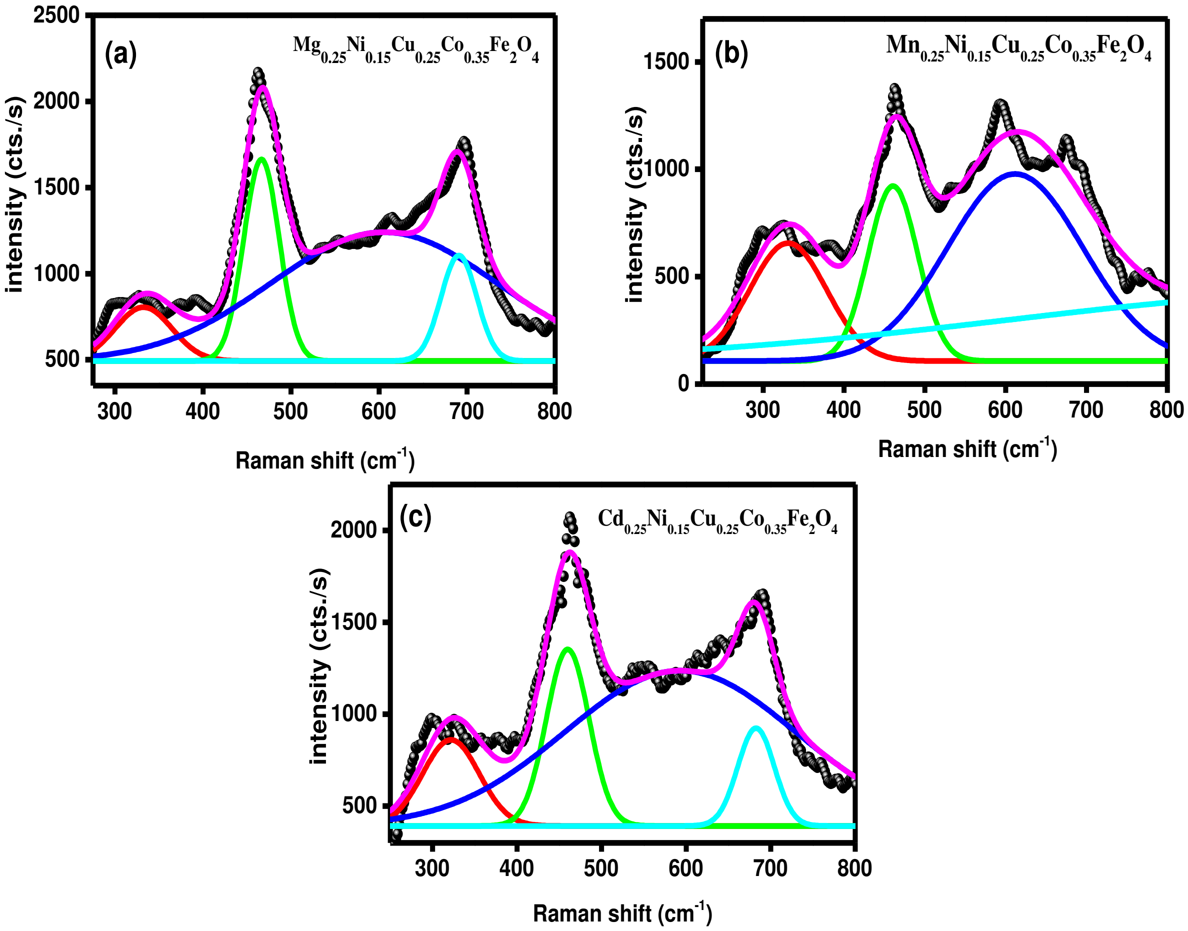

Fig. 3 Raman peaks of M^{2+} doped NCCF ferrites

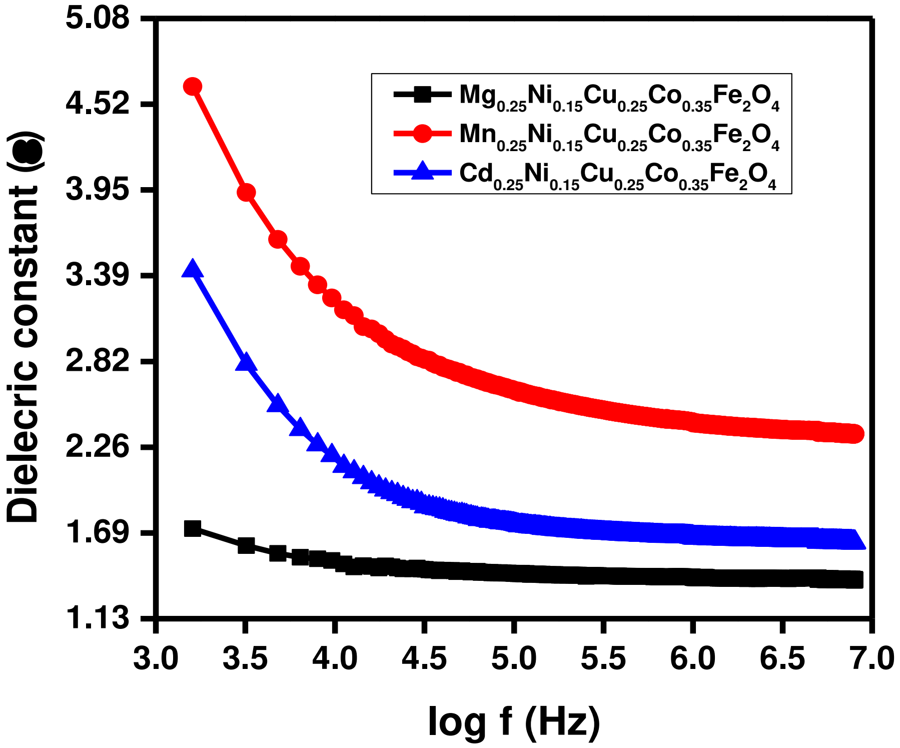

Fig. 4 Dielectric constant and frequency function of M^{2+} doped NCCF ferrites

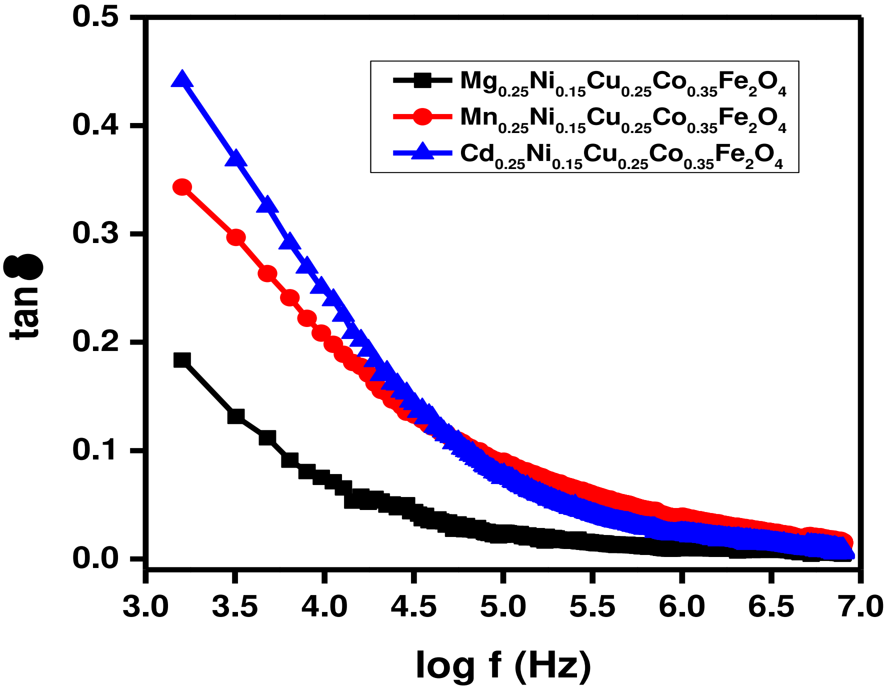

Fig. 5 Dielectric loss and frequency function of M^{2+} doped NCCF ferrites

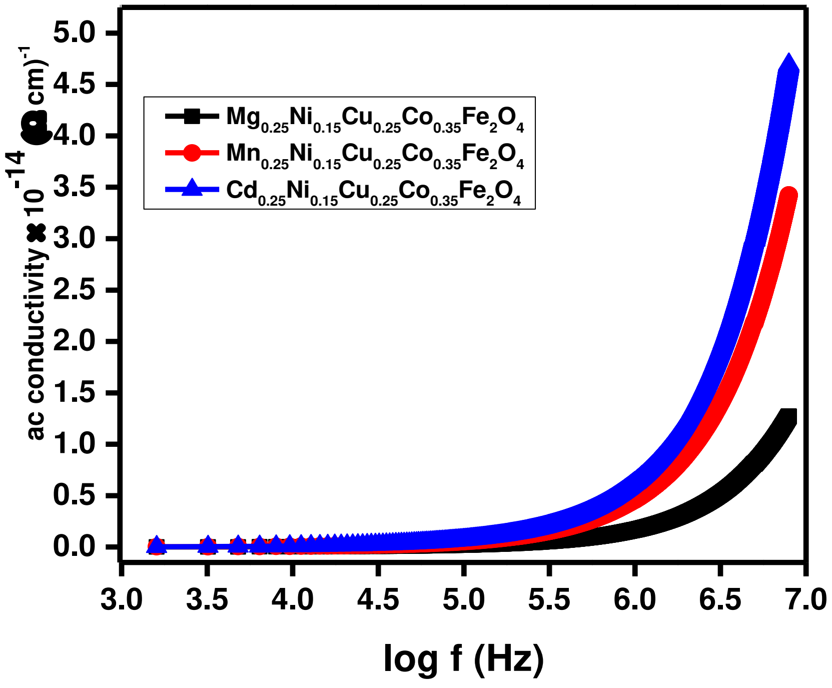

Fig. 6 ac conductivity and frequency function of M^{2+} doped NCCF ferrites

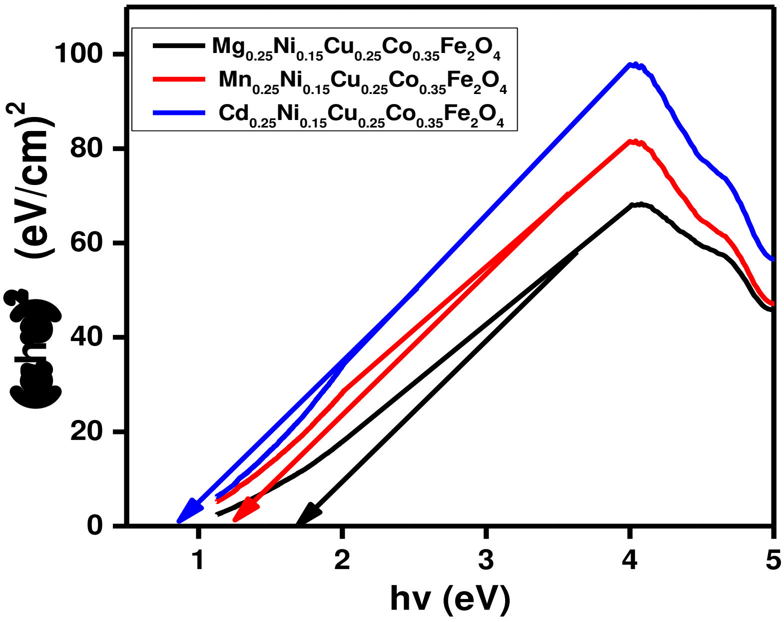

Fig. 7 Optical band gap of M^{2+} doped NCCF ferrites

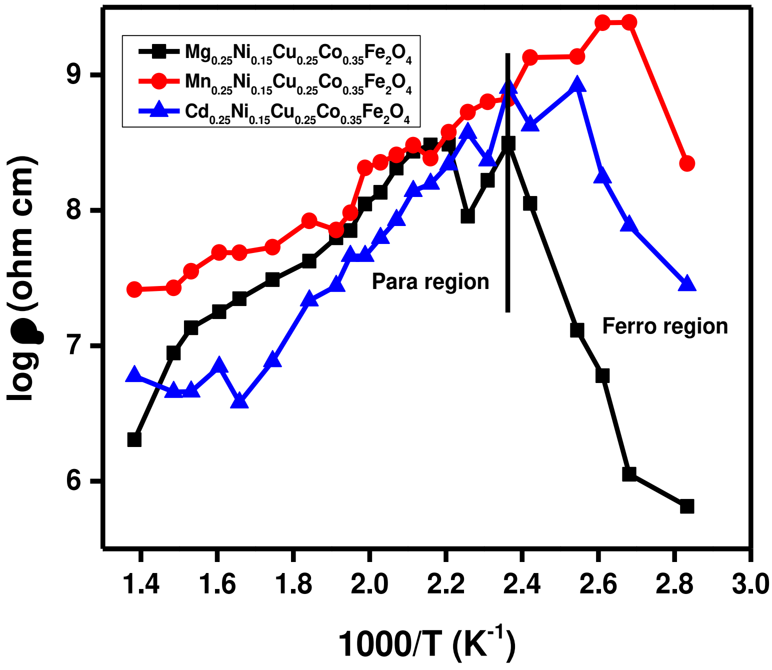

Fig. 8 log of Resistivity and 1000/T of M^{2+} doped NCCF ferrites

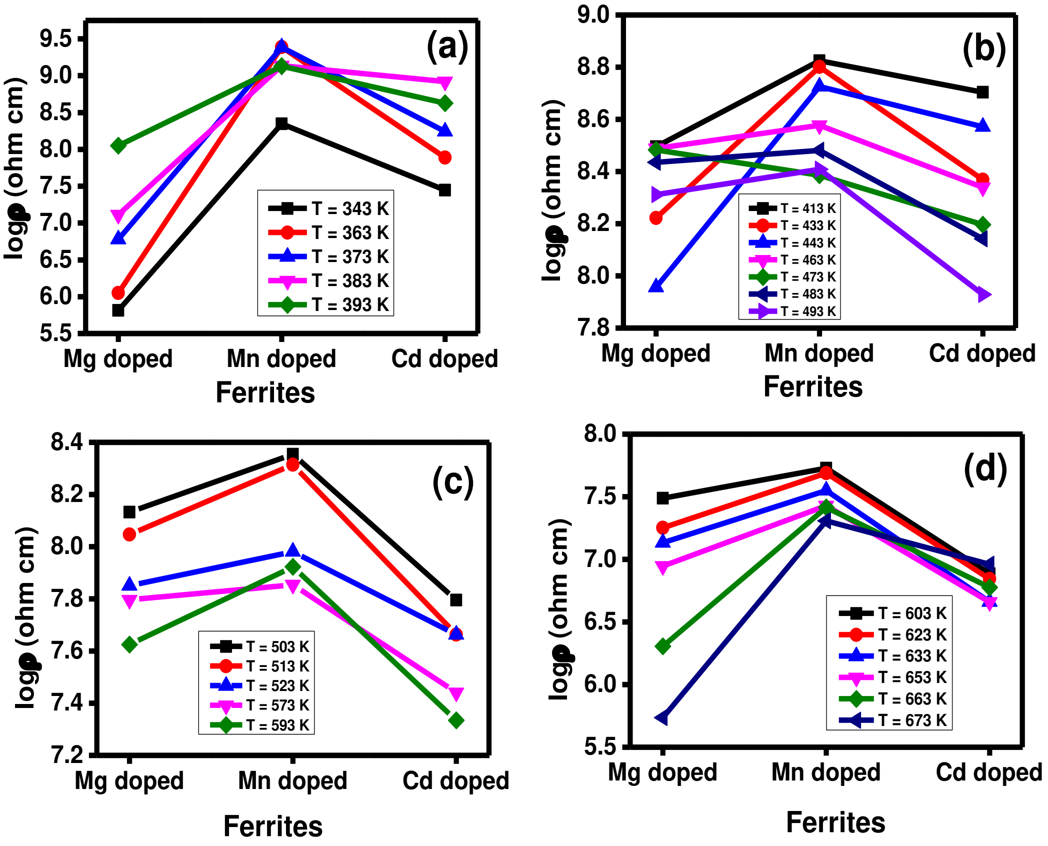

Fig. 9 Ferrites *versus* the log of resistivity of M^{2+} doped NCCF ferrites

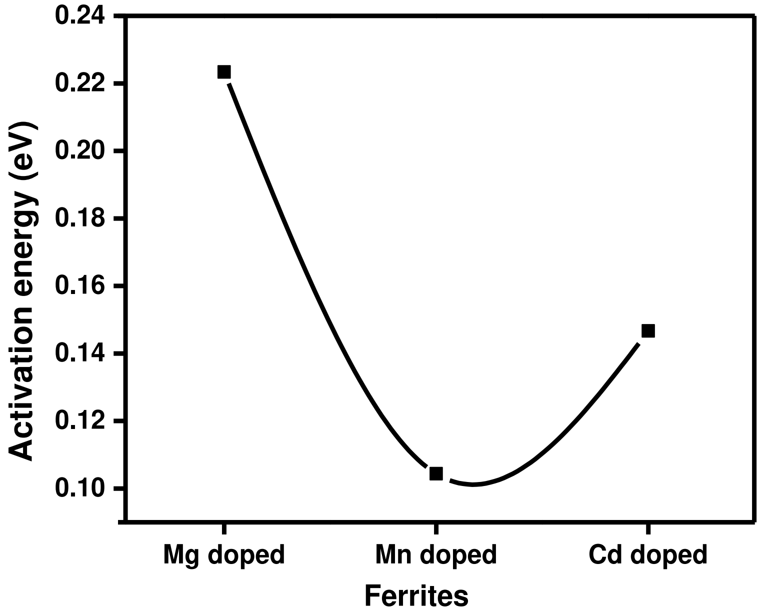

Fig. 10 Ferrites *versus* activation energy