





Article Effect of Nb³⁺ Substitution on the Structural, Magnetic, and Optical Properties of Co_{0.5}Ni_{0.5}Fe₂O₄ Nanoparticles

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Abstract: $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 $\leq x \leq 0.10$) nanoparticles (NPs) were prepared using the hydrothermal approach. The X-ray powder diffraction (XRD) pattern confirmed the formation of single-phase spinel ferrite. The crystallite size was found to range from 18 to 26 nm. The lattice parameters were found to increase with greater Niobium ion (Nb³⁺) concentration, caused by the variance in the ionic radii between the Nb³⁺ and Fe³⁺. Fourier transform infrared analysis also proved the existence of the spinal ferrite phase. The percent diffuse reflectance (%DR) analysis showed that the value of the band gap increased with growing Nb³⁺ content. Scanning electron microscopy and transmission electron microscopy revealed the cubic morphology. The magnetization analyses at both room (300 K, RT) and low (10 K) temperatures exhibited their ferromagnetic nature. The results showed that the Nb³⁺ substitution affected the magnetization data. We found that Saturation magnetization (M_s), Remanence (M_r), and the Magnetic moment (n_B) decreased with increasing Nb³⁺. The squareness ratio (SQR) values at RT were found to be smaller than 0.5, which postulate a single domain nature with uniaxial anisotropy for all produced ferrites. However, different samples exhibited SQRs within 0.70 to 0.85 at 10 K, which suggests a magnetic multi-domain with cubic anisotropy at a low temperature. The obtained magnetic results were investigated in detail in relation to the structural and microstructural properties.

Keywords: spinel ferrites; Nb substitution; low temperature magnetization; optical properties; TEM analysis

1. Introduction

Spinel ferrite nanoparticle materials are highly preferred in engineering and technology applications like biomedicine, pharmaceuticals, sensors, magnetic resonance imaging, drug delivery,

microwaves, high-frequency devices, information storage, and electronic chips [1–4]. The structure and electromagnetic properties of nano-spinel ferrites can be modified by the substitution of different cations. Rare earth substitutions are highly valuable for reducing the particle size and intensification of the lattice parameter [5]. In this respect, substituting rare earth (RE) cations into the spinel ferrite structure plays an important role in enhancing the dielectric, magnetic, and electric properties due to the Fe–Fe interactions caused by the spin coupling effect of 3d electrons [6]. Therefore, electrical and magnetic behavior may change when rare earth and iron interactions (3d–4f coupling) of the spinel ferrites occur. Different RE substitutions have been proven to have different results on the ferrite structure [7–9]. Many researchers have investigated the effects of RE substitution into cobalt spinel ferrite (CoFe₂O₄) in bulk form, thin film, and nanoparticles [10-12]. Coercivity, anisotropic constant, and uniaxial anisotropy were described as decreasing with the addition of m and Ce ions into nano-cobalt ferrites [13]. Several methods have been used to synthesize nickel-substituted cobalt ferrites, $Co_{1-x}Ni_xFe_2O_4$, such as the auto-combustion method, aerosol route, and co-precipitation method [14–16]. Chen et al. [17] prepared $Ni_{1-x}Co_xFe_2O_4$ nanoparticles (NPs) using the hydrothermal method and studied the increasing trend in saturation magnetization with increasing cobalt content, which occurred due to the substitution of an Ni²⁺ ion to a Co²⁺ ion. Maz et al. [18] synthesized $Co_{1-x}Ni_xFe_2O_4$ nanoparticles in a chemical co-precipitation process and noticed the increasing trend of coercivity (H_c) and saturation magnetization (M_s) with increasing cobalt content.

Coercivity (H_c) and saturation magnetization (M_s) values were shown to decrease with an increase in the Ni content in Co_{1-x}Mn_xFe₂O₄ (0.0 $\le x \le 0.4$) nanoparticles [19]. A few papers have been published on Nd³⁺ ion-substituted cobalt-ferrite in the Fe sites. Almessiere et al. [20] reported the effect of the magnetocrystalline anisotropy constant decreasing with the addition of Nd³⁺ ions in cobalt spinel nano-ferrites Co_{1-2x}Ni_xMn_xFe_{2-y}Nd_yO₄ (0.0 $\le x = y \le 0.3$) synthesized using the sol-gel method. Yadav et al. [21] examined the magnetic and structural properties by incorporating Nd³⁺ ions in cobalt spinel ferrite nanoparticles, CoFe_{2-x}Nd_xO₄ ($x \le 0.1$), synthesized using the sol-gel method. Tahar et al. [22] investigated the RE = La, Ho, Tb, Ce, Gd, Nd, and Sm substitutions in CoFe_{1.9}RE_{0.1}O₄ nanoparticles. Aside from these RE substitutions, Nd³⁺ ions considerably decrease the coercivity and saturation magnetization. Zhao et al. [23] reported that coercivity and saturation magnetization slightly increased with Nd³⁺ substitution in cobalt ferrite nanocrystals CoFe_{2-x}Nd_xO₄ synthesized by the emulsion method. In this study, the effect of Nb³⁺ substituted in the Co-Ni spinel ferrite on the structural, optical, and magnetic properties are discussed in detail.

2. Experimental Materials and Methods

2.1. Materials and Instruments

A Rigaku Benchtop Miniflex X-ray diffraction (XRD) diffractometer (Tokyo, Japan) with Cu K_{α} radiation at room temperature (RT) over the 20 range from 20° to 70° was used for the structural analysis. Scanning electron microscopy (SEM, FEI Titan ST, Hillsboro, OR, USA) along with energy dispersive X-ray spectroscopy (EDX) and transmission electron microscopy (TEM; FEI, Morgagni 268, Prague, Czech Republic) were used for the morphological and composition analyses. Spectral analysis of all products was performed via Fourier transform infrared (FT-IR) spectroscopy (Bruker, Berlin, Germany). Ultraviolet-visible (UV-vis) diffuse reflectance (DR%, Shimadzu, Tokyo, Japan) spectra were recorded in the 200 to 800 nm wavelength range using a DR spectrophotometer. The magnetic properties of the products were measured using a Quantum Design SQUID-PPMS vibrating sample magnetometer (PPMS DynaCool, Quantum Design, San Diego, CA, USA).

Cobalt (II) nitrate $(Co(NO_3)_2)$, nickel (II) nitrate $(Ni(NO_3)_2)$, iron (III) nitrate hexahydrate $(Fe(NO_3)_3.6H_2O)$, and niobium (V) nitrate $(Nb(NO_3)_5)$ were received from Sigma-Aldrich (St. Louis, MO, USA) and used as received.

2.2. Procedure

The hydrothermal method approach was used to synthesize $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 $\leq x \leq 0.10$) nanoparticles. Stoichiometric amounts of iron, nickel, and cobalt nitrates were dissolved in 50 mL deionized (DI) H₂O. Niobium nitrate was dissolved in concentrated HCl in a separate beaker with vigorous stirring. Then, the two solutions were mixed together and stirred for an extra 30 min and the pH of the resulting solution was adjusted to 11 by adding sodium hydroxide (NaOH) with continuous stirring for 30 min. Before transfer to a Teflon-lined vessel, the mixture was pretreated in an ultrasonic water bath for 30 to 40 min. The final solution was transferred to a stainless-steel Teflon autoclave (200 mL) and placed in an oven at 180 °C for 10 h. The resulting product was washed with hot deionized water three times and left to dry overnight at 80 °C for 5 h.

3. Results and Discussion

3.1. XRD Analysis

Figure 1 displays the crystal structure of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ ($0.00 \le x \le 0.10$) nanoparticles. The XRD analysis showed the signature peaks of a single phase of Co spinel ferrite with no presence of any extra secondary phase, which means that the Nb³⁺ was well dissolved in the Co–Ni crystal. Only the x = 0.02 sample had a minor amount of the Fe₂O₃ phase as an impurity. The structural parameters were calculated through Rietveld refinement using the XRD experimental data, as registered in Table 1. We found that the lattice parameters increased with the rise in the Nb³⁺ amount due to the larger ionic radius of Nb³⁺ (0.72 Å) in comparison with Fe³⁺ (0.64 Å), which caused stress in the lattice. The crystallite sizes were estimated to range from 18 to 26 nm.



Figure 1. X-ray diffraction (XRD) powder patterns of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ ($0.0 \le x \le 1.0$) nanoparticles.

Table 1. Nb content and refined structural parameters (a is the lattice parameter, V is the volume of the cell, D_{XRD} is the crystallite size obtained from the broadening of the highest peak by using the Scherrer method, χ^2 (chi²) is the squared ratio, R_{Bragg} is the Bragg factor) for the Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O₄ (0 ≤ x ≤ 1.0) nanoparticles.

x	a (Å)	V (Å) ³	$\mathrm{D}_{\mathrm{XRD}}$ (nm) \pm 0.09	χ^2 (chi ²)	R _{Bragg}
0.00	8.345 (1)	581.15	22.86	1.75	12.67
0.02	8.345 (9)	581.33	18.71	1.32	3.12
0.04	8.352 (3)	582.67	24.18	1.52	3.99
0.06	8.356 (6)	583.57	27.59	1.32	2.48
0.08	8.362 (3)	584.76	26.86	1.77	7.88
0.10	8.362 (4)	584.77	24.70	1.32	8.77

3.2. Spectral Analysis

Figure 2 highlights the FT-IR spectra of $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 $\leq x \leq$ 0.10). The strongest stretching bands at 584.36 and 410.7 cm⁻¹ represent the Co spinel ferrite structure. These bands can be attributed to the stretching of the vibration band between F and O. The vibration bands shifted toward higher wavenumbers when the Nb³⁺ content increased, which was due to the variation in the bond length as a result of the larger ionic radii of Nb³⁺ [24–26].



Figure 2. Fourier transform infrared (FT-IR) spectra of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.0 $\leq x \leq$ 1.0) nanoparticles.

3.3. Morphological Analysis

The Field emission scanning electron microscope (FE-SEM) microstructure of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ ($0.00 \le x \le 0.10$) nanoparticles is depicted in Figure 3. The images exhibited a higher agglomeration cubic shape with an average grain size of less than 27 nm, which agreed with the crystal size estimated by XRD. The EDX spectrum and elemental mapping confirmed the existence of Co, Ni, Nb, and O in compositions x = 0.02 and 0.06, as shown in Figure 4. The quantitative analysis of selected samples that recorded the atomic weigh of x = 0.02 and 0.06 showed that the estimated values were close to the expected values for the samples. The TEM and SAED (Selected area electron diffraction) pattern of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (x = 0.06) nanoparticles are shown in Figure 5. The images confirmed the cubic spinel structure and aggregate.



Figure 3. Scanning electron microscope (SEM) micrographs of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ ($0.0 \le x \le 0.10$) nanoparticles.



Figure 4. Cont.



Figure 4. Energy dispersive X-ray (EDX) spectrographs and elemental mapping images of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ nanoparticles for x = 0.02 and 0.06.



Figure 5. Transmission electron diffraction (TEM) images (with different magnetization) and selected area electron diffraction (SAED) pattern of $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ for x = 0.06 nanoparticles.

3.4. Optical Analysis

The optical properties of $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 $\le x \le 0.10$) nanoparticles were studied using a DR-UV-visible spectrophotometer ranging from 200 to 800 nm. Figure 6 shows that the compositions at different concentrations exhibited spectra in the visible range. The Kubelka–Munk model was used to compute the optical band gap energy (E_g) [27]. The band gap energy was calculated using a plot of $(\alpha hv)^2$ vs. photon energy (hv), which is also called the Tauc plot, of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 \le $x \le 0.10$) nanoparticles (Figure 7).

The band gap values were 0.25, 0.35, 0.51, 0.72, 0.76, and 0.77 eV for x = 0.00, 0.02, 0.04, 0.06, 0.08, and 0.1, respectively. When the value of x increased, the band gap value increased. The increase in the band gap value was ascribed to the development of the energy level or interface defects in M. Almessiere et al. [28]. The increase in the band gap could also be due to the synergistic effect of nano-ferrite with niobium, which decreases the electron hole recombination [29].



Figure 6. DR% spectra of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ ($0.0 \le x \le 0.10$) nanoparticles.



Figure 7. $[\alpha hv]^2$ versus *hv* graphs of the Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O₄ ($0.0 \le x \le 0.10$) nanoparticles.

The magnetization plots against an applied magnetic field of ±20 kOe, M(H) for all the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 $\le x \le 0.10$) nanoparticles performed at RT are illustrated in Figure 8. Table 2 summarizes the various deduced magnetic parameters for all the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 $\le x \le 0.10$) nanoparticles at RT. The Nb³⁺ substitution in the Fe³⁺ sites altered the magnetic properties of the CoNi ferrite. The different CoNi ferrites displayed remanence magnetization (M_r) ranging from 13.00 to 23.66 emu/g. H_c ranged from 207.31 to 1129.92 Oe. The $M_{max,20}$ (magnetization at a maximum field of 20 kOe) was found to be between 42.36 and 49.77 emu/g. The Stoner–Wohlfarth (S–W) theory was used to extract the saturation magnetization (M_s) [30–32]. An example of the estimation of M_s for the x = 0.00 sample is shown in Figure 9. The extrapolation of this plot at high magnetic fields approaching zero produces the M_s value. The M_s values of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 $\le x \le 0.10$) nanoparticles ranged from 43.15 to 50.62 emu/g at RT. According to the obtained findings, we confirmed that the differently produced $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ (0.00 $\le x \le 0.10$) nanoparticles ranged from 43.15 to 50.62 emu/g at RT.

Table 2. Magnetic parameters of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ ($0 \le x \le 1.0$) nanoparticles at room temperature (RT) ($M_{max,20}$: max magnetization at 20 kOe, M_s : saturation magnetization, M_r : remanence magnetization, K_a : magnetic anisotropy constant, SQR: squareness ratio, H_c : coercivity and n_B : magnetic moment).

x	M _{max,20} (emu/g)	M_s (emu/g)	<i>M_r</i> (emu/g)	<i>K_a</i> (Erg/g)	SQR	<i>Н</i> _с (Ое)	n _B (μ _B)
0.00	49.77	50.62	18.47	1.96×10^5	0.365	648.11	2.13
0.02	47.98	48.81	20.9	$1.74 imes 10^5$	0.428	509.9	2.06
0.04	46.57	47.36	13	$1.78 imes 10^5$	0.274	286.11	2.00
0.06	45.68	46.52	23.66	$2.16 imes10^5$	0.509	1129.92	1.97
0.08	43.93	44.71	18.5	1.77×10^5	0.414	775.67	1.90
0.10	42.36	43.15	13.48	$1.63 imes 10^5$	0.312	207.31	1.84

The M(H) were also performed for all of the Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O₄ (0.00 \leq x \leq 0.10) nanoparticles at 10 K (Figure 10). The deduced magnetic parameters at 10 K are listed in Table 3. *H_c* ranged from 708.93 to 5882.24 Oe. *M_r* ranged from 34.11 to 48.09 emu/g. The *M_{max,20}* values ranged from 45.36 to 57.22 emu/g. *M_s* varied from 45.71 to 57.96 emu/g. Spinel ferrite nanoparticles have been reported to have a superparamagnetic threshold below 10 nm [33]. Since our nanoparticles had dimensions larger than 10 nm, the effect of the superparamagnetic state nanoparticles on lower magnetization was neglected. The obtained magnetic results at 10 K revealed the semi-hard FM nature of all products. Compared to 300 K, the *M_s*, *M_r*, and *H_c* showed a remarkable increase at 10 K. This increase was due to reduced thermal fluctuations of the magnetic moments [34,35]. In the literature, the anisotropy contribution of RE ions has been reported via spin-orbit coupling when they occupy the B sites of spinel ferrites [31,36]. This is one of the reasons for the observation of higher coercivities at CoMn ferrites doped with rare-earth ions.

At both measurement temperatures, the x = 0.00 sample had the highest magnitudes of M_s , which were about 49.77 and 57.22 emu/g at 300 and 10 K, respectively. Likewise, the x = 0.00 product exhibited the maximum M_r values with magnitudes of 18.47 and 48.09 emu/g at 300 and 10 K, respectively. The magnetization magnitudes found in the present study for the non-substituted sample were comparable to those of the CoFe₂O₄ and NiFe₂O₄ inverse spinel ferrites [37,38], but they are larger than those reported in the literature for Co_{0.5}Ni_{0.5}Fe₂O₄ [39]. The obtained magnitudes were smaller than those reported for both the bulk CoFe₂O₄ and NiFe₂O₄ inverse spinel ferrites [33,40]. The lower M_s and M_r magnitudes, in comparison to that of the bulk samples, were largely attributed to the smaller crystallite size, which leads to a structural disorder on the surface since the spin

disorder will be significant when the volume and surface ratio are important [41]. Spin canting as a result of antiferromagnetic interaction competition, the construction of a magnetic inactive layer, the non-collinear arrangement of the magnetic moments of Fe³⁺ ions, and the disordered cations distributions on the surface could all explain the lowered magnetization magnitudes [42,43].



Figure 8. Magnetization versus applied field of ± 20 kOe, M(H), for the Co_{0.5}Ni_{0.5}Fe_{2-x}O₄ (0.0 $\leq x \leq$ 0.10) nanoparticles at RT.

Table 3. The deduced magnetic parameters of the $Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O_4$ ($0 \le x \le 1.0$) nanoparticles at 10 K ($M_{max,20}$: max magnetization at 20 kOe, Ms: saturation magnetization, Mr: remanence magnetization, Ka: magnetic anisotropy constant, SQR: squareness ratio, H_c : coercivity, and n_B : magnetic moment).

x	<i>M_{max,20}</i> (emu/g)	<i>M_s</i> (emu/g)	<i>M_r</i> (emu/g)	<i>K_a</i> (Erg/g)	SQR	<i>Н</i> _с (Ое)	n _B (μ _B)
0.00	57.22	57.96	48.09	$4.30 imes 10^5$	0.830	5335.84	2.43
0.02	54.44	55.25	44.47	$3.98 imes 10^5$	0.805	4760.39	2.33
0.04	53.53	54.26	44.24	$3.78 imes 10^5$	0.815	3440.9	2.29
0.06	51.23	52.05	44.08	$5.24 imes 10^5$	0.847	5882.24	2.21
0.08	48.39	48.62	34.11	$3.57 imes 10^5$	0.702	2848	2.07
0.10	45.36	45.71	34.94	$2.67 imes 10^5$	0.764	708.93	1.95



Figure 9. A schematic example of the M vs. $1/H^2$ plot for the Co_{0.5}Ni_{0.5}Fe₂O₄ nanoparticles at RT.



Figure 10. Magnetization against an applied field of ± 20 kOe, M (H), for all of the $Co_{0.5}Ni_{0.5}Fe_{2-x}O_4$ ($0.0 \le x \le 0.10$) nanoparticles at 10 K.

At both temperatures, the highest H_c was observed for x = 0.06 and the lowest for x = 0.10. Various parameters governed the coercivity, like grain size, magnetic particle morphology, magnetocrystalline anisotropy, strains and exchange coupling between the collinear spins in the core, and the canted spins on the surface [34,35]. The improvement in coercive field can be principally attributed to the increase of magnetocrystalline anisotropy [44,45]. Equation (1) describes the proportionality between the coercivity H_c and magnetic anisotropy constant K_a [44].

$$H_c \propto \frac{2K_a}{\mu_o M_s} \tag{1}$$

where μ_o is the permeability constant. When the magnetic anisotropy increases with increasing substitution content, coercivity grows. To determine the anisotropy constant K_a , the expression used to estimate the values of M_s according to the S–W fit is shown below [31,32].

$$M = M_s \left(1 - \frac{\beta}{H^2} \right) \tag{2}$$

Consequently, the slope of the linear fitting provides the constant β , which is related to the magnetocrystalline anisotropy constant K_a . Once the values of the β constant are determined, the magnetic anisotropy constant (K_a) can be estimated by using the equation below [31,32].

$$K_a = M_s \left(\frac{15\,\beta}{4}\right)^{\frac{1}{2}} \tag{3}$$

The deduced K_a values at 300 and 10 K are listed in Tables 2 and 3, respectively. The magnetocrystalline anisotropy was the maximum for the x = 0.06 product and was the minimum for x = 0.10. This result explains the highest coercivity in the x = 0.06 sample.

The Nb³⁺ substitution led to a gradual reduction in the M_s and M_r values. The lowest magnitudes were observed for the Co_{0.5}Ni_{0.5}Fe_{1.9}Nb_{0.1}O₄ (x = 0.1) sample. The minimum M_s values belonging to the x = 0.1 product were about 43.15 and 45.71 emu/g at 300 and 10 K, respectively. The minimum M_r for x = 0.1 were 13.00 and 34.11 emu/g at 300 and 10 K, respectively. The evolutions in the M_r values showed a similar trend to M_s with respect to the Nb³⁺ concentration. It has been reported previously that evolutions in the M_r values depend principally on evolutions in M_s and on the net alignment of magnetization grains derived from super-exchange interactions between the magnetic particles [32].

Numerous factors can affect the magnetic properties of spinel ferrites, including the crystallite size change, variations in magnetic moments (n_B) , variations in the nature and concentration of different sites, and the preferred site occupancy of different ions [36]. The local strains and the super-exchange interactions between different ions might influence the magnetic parameters [34,35]. Principally, the magnetic moment of spinel ferrites is derived from the iron ions and their distribution in the crystal sites. The A–A and B–B interactions were unimportant. However, the A–B exchange interactions were dominant. Consequently, any factors that affect the strength of various exchange interactions will modify the magnetization. In our case, the observed decrease in M_s and M_r values with Nb³⁺ substitution was attributable to the weakening of the exchange interactions in the Fe sites. The ions of the host Fe³⁺ (0.62 Å) displayed a slightly smaller ionic radius when compared to that of Nb³⁺ (0.72 Å). The contrast of magnetic moments and ionic radii of the host and substituted ions might produce a non-collinear ferromagnetic arrangement and local strains that cause the disorder and variations in electronic states in the hexaferrite systems [32,34,35]. The substitution of Fe³⁺ ions with Nb³⁺ ions resulted in increasing the distance separating the magnetic ions and, therefore, decreasing the strength of the A–B super-exchange interactions. The relation between the magnetic moment (n_B) and M_s is given by the formula below [36].

$$n_B = rac{Molecular \ Weight imes M_s}{5585}$$

The estimated n_B values of all of the Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O₄ (0.00 $\leq x \leq$ 0.10) nanoparticles at 300 and 10 K are summarized in Tables 2 and 3, respectively. The decrease in n_B values resulting from the weakening of the super-exchange interactions among the various sites led to a decrease in the n_B values. In our case, n_B was found to decrease with increasing Nb³⁺ content. The x = 0.00 sample where the M_s value was the highest displayed the greatest n_B . The x = 0.10 sample where the M_s value was the lowest n_B . This indicates a weakening of the super-exchange interactions.

The squareness ratios ($SQR = M_r/M_s$) were calculated for all the Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O₄ (0.00 $\leq x \leq 0.10$) nanoparticles at 300 and 10 K. According to the S–W theory, the SQR can take two values including one around 0.83 associated with the cubic anisotropy, and another around 0.5 that corresponds to uniaxial anisotropy [30,32]. The findings of *SQR* equal to or above 0.5 indicated that the particles were in the single magnetic domain, and those below 0.5 could be attributed to the formation of a multi-domain structure [46]. As can be seen from the tables, the *SQR* at RT was found to be around 0.509 for the x = 0.06 sample, which suggests a single magnetic domain with uniaxial anisotropy. However, the other samples displayed *SQRs* that ranged from 0.31 to 0.42, which were less than 0.50 and can be attributed to surface spin disorder effects. This *SQR* was lower than 0.5, which indicates the formation of a multi-domain structure with uniaxial anisotropy. At 10 K, the different samples were found to have *SQRs* ranging between 0.70 and 0.85, which are greater than 0.5. This suggests a single magnetic domain with cubic anisotropy.

4. Conclusions

A series of Nd³⁺-substituted Co-Ni ferrite was synthesized via the hydrothermal approach. The XRD and FT-IR analyses proved the existence of single-phase spinel Co-ferrite. TEM analyses showed the hexagonal morphology of the products with minor agglomeration. The optical results showed that the band gap of the Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O₄ (0.00 $\le x \le 0.10$) nanoparticles were 0.25, 0.35, 0.51, 0.72, 0.76, and 0.77 eV, respectively. The M(H) analyses showed an FM comportment at both RT and 10 K for the Co_{0.5}Ni_{0.5}Nb_xFe_{2-x}O₄ (0.00 $\le x \le 0.10$) nanoparticles. The magnetic parameters strongly depend on temperature and Nb substitution content. The deduced M_s , M_r , and n_B values were the highest for the x = 0.00 sample and decreased with increasing Nb substitution. This effect is due to the weakening of super-exchange interactions, the creation of local strains, the preferred site

occupancy, and the decrease in the magnetic moments (n_B) with respect to the Nb content. The SQR values at RT were found to be smaller than 0.5, which postulates a single domain nature with uniaxial anisotropy for all the produced ferrites. However, the different samples exhibited SQRs in the 0.70–0.85 range at 10 K, which suggests a magnetic multi-domain with cubic anisotropy at a low temperature.

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