

Investigation of Structural Parameters and Magnetic Properties of Mixed Li-Ni Spinel Ferrites

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Abstract

The solid state method was used to synthesize the mixed $Li_{0.5(1-x)}^{1+}Ni_x^{2+}Fe_{2.5-0.5x}^{3+}O_4^{2+}$ spinel ferrites where x is the increments of Ni content. The net magnetization (M_{net}) of the prepared samples was calculated according to the suggested cations distribution $(Fe_{(1)}^{3+})_T \{Li_{0.5(1-x)}^{1+}Ni_x^{2+}Fe_{0.5(1-x)}^{3+}Fe_1^{3+}\}_O O_4^{2+}$. The calculated magnetization of tetrahedral (T_d) site (M_T) unchanged, while the magnetization of octahedral (O_h) site (M_0) and M_{net} decreased due to increasing Ni^{2+} ions. The measured values of M_{net} showed the same behavior as the calculated values, which decreased exponentially with increasing of the Ni concentrations. In addition, M_{net} increased with increasing of applied magnetic field (H), which showed a normal behavior for the ferrites system. The ratio of Fe_T/Fe_O indicated that the relative number of the Fe^{3+} ions on O_h sites decreased with increasing Ni content. The ionic radii of T_d sites (R_T) unchanged where the radii of O_h sites $(\mathbf{R}_{\mathbf{0}})$ increased with increasing of the Ni²⁺ ions. The values of the average ionic radii $R_{av} = (R_T + R_o)/2$ increased with increasing of Ni²⁺ ions. The expected values of lattice parameter (a_{th}) increased, where the values of oxygen parameter $u^{\overline{4}3m}$ and $u^{\overline{3}m}$ decreased with more incorporation of Ni content. All the interionic distances (bond lengths (BLs)) changed due to increasing of Ni contents. The bond angles θ_1 , θ_2 , θ_3 , θ_4 and θ_5 were calculated. The values of θ_1 , θ_2 and θ_5 increased and θ_3 the θ_4 decreased with increasing of Ni^{2+} ions. The relative permeability (μ_r) increased with increasing of the filed H and decreased with the increase in in composition x. Substitution of the Ni^{2+} ions has a tremendous influence on the properties of Li-spinel ferrite such the magnetic properties.

Keywords: Spinel ferrite, relative permeability, cations distributions, Neel's model, jump length, hyperfine magnetic field.

Introduction

Li-ferrites and mixed *Li*-ferrites have become important materials for the microwave applications such as circulators, isolators, and phase shifters due to their high resistivity, low dielectric losses, high Curie temperature, square hysteresis loop, low cost, gyrators memory cores [1,2], high and wide range of saturation magnetization. The diverse interesting properties of spinel *Li*-ferrites originate from their



ability to incorporate a variety of transition metal cations into their lattice, causing a subsequent change in their structural, optical, magnetic, and electrical properties. This is because the degree of inversion in the substituted Li-ferrites and, therefore, their properties are found to be strongly dependent on the preparation conditions (method of preparation, sintering time, sintering temperature, etc.), amount of the substituent and type of substitution [2]. Several studies have been reported with the addition of divalent, trivalent and tetravalent ions in lithium ferrite [3-6]. Besides pure Li ferrites many researchers have extensively studied, Li-Ni [7], Li-Cd [5], Li-Cr [8], Li-Cu [9]. Ferrites. However, reports on the magnetization behaviors of Li-Ni ferrites are still little. Spinel ferrites have a general structure formula $M^{2+}Fe_2^{3+}O_4^{2+}$ (*M* is transition divalent metal ions) [10-14]. The magnetic properties for the spinel ferrite can be understood in terms of cations (M^{2+} and Fe_2^{3+}) distribution between the tetrahedral (T_d) sites and octahedral (O_h) sites and their exchange interactions. These properties are normally dependent on the nature of ions present and their cationic distribution among tetrahedral (T_d) and octahedral (O_b) sites [1]. The observed changes in these properties are brought about by a redistribution of cations among the $T_{\rm d}$ and O_h sites of the ferrites sublattices [2]. Theoretical estimation of magnetic interactions based on cation distribution clearly predicted the weakening in super-exchange interactions with the incorporation of Ni^{2+} ions in the mixed Li-Ni spinel ferrite [15-17]. The current work aims to investigate the effect of Niaddition on the structural and magnetic behaviors of Li-ferrites. Simultaneous substitution of small amount of Ni^{2+} ions in Li-ferrites prepared by solid-state method is expected to produce interesting changes in their properties.

Experimental

High purity metals oxides Fe_2O_3 , NiO and Li_2CO_3 were used to prepared the mixed Li-Ni spinel ferrite using the solid state method as described earlier [18-21].

 $0.25(1-x)Li_2CO_3 + xNiO + (1.25 - 0.25x)Fe_2O_3 \rightarrow Li_{0.5(1-x)}Ni_xFe_{2.5-0.5x}O_4 + 0.25(1-x)CO_2$

Net magnetization M_{net} was measured at room temperature as a function of H in the range (200-2650) A.m⁻¹ at a constant magnetizing frequency (v = 50 Hz). Then, the relative permeability (μ_r) was calculated.

Results and Discussion

Cations distribution and the calculated net magnetization

The calculation M_{net} of the given ferrites was done by substituting the value of the respective ion in the Neel's formula $M_{net} = |M_0 - M_T|[5,22]$, with M_T and M_0 of T_d and O_h sub-lattices, respectively. Previous studies [23-25] were reported that, the mixed *Li-Ni* spinel ferrite in the bulk form gives an inverse spinel ferrite. The Ni^{2+} and Li^+ ions show a noticeable stronger favorite for the O_h site than the Fe^{3+} ions [26]. This implies to the suggested cations distribution which is agreed with the ref. [27], $(Fe_{(1)}^{3+})_T \{Li_{0.5(1-x)}^{1+}Ni_x^{2+}Fe_{1.5-0.5x}^{3+}\}_0 O_4^{2+}$,. The cations distribution for each composition are given in the Table 1. One can deduce the M_{net} as follows $M_{net} = 2[xS_{Ni} + (0.5 - 0.5x)S_{Fe}]\mu_B$, with $S_{Ni} = 2/2$ for Ni^{2+} ions and $S_{Fe} = 5/2$ for Fe³⁺ ions. Herein, the Li^{1+} ions are diamagnetic ions, i.e. $S_{Li} = 0$ [28]. The calaculted values of M_0 , M_T and M_{net} for all prepared samples are tabulated in Table 1. In such table, both M_0 and M_{net} decreased, while M_T unchanged with increasing of Ni^{2+} ions. The decreasing of M_{net} with increasing Ni^{2+} ions can be attributed to the relative number of the Fe^{3+} ions occupied T_d and O_h sites.



Consequently, the ratio of Fe^{3+} ions between the T_d and O_h sites (Fe_T/Fe_O) for each sample are shown in Table 1. Based on the values of Fe_T/Fe_O , it is evident that relative number of the Fe^{3+} ions on O_h sites decreases with increasing *Ni* content. It is worthwhile to note that the occupation of Fe^{3+} on T_d and O_h sites range from 66.7% to 100.0 % within the range studied.

Table (1): The calculated values of Fe_T/Fe_0 , M_0 , M_T and M_{net} according to the cations distribution of	t the							
mixed Li-Ni spinel								

x	Cations distrubution	Fe _T /Fe _O	$M_{\rm O}\left(\mu_{B}\right)$	$M_{\mathrm{T}}\left(\mathbf{\mu}_{B}\right)$	$M_{\rm net} \left(\mu_{B} \right)$
0.0	$(Fe_1^{3+})_T \{Li_{0.5}^{1+}Fe_{1.5}^{3+}\}_0 O_4^{2+}$	66.7	7.5	5	2.5
0.2	$(Fe_1^{3+})_T \{Li_{0.4}^{1+} Ni_{0.2} Fe_{1.4}^{3+}\}_0 O_4^{2+}$	71.4	7.4	5	2.4
0.4	$(Fe_1^{3+})_T \{Li_{0,3}^{1+} Ni_{0,4} Fe_{1,3}^{3+}\}_0 O_4^{2+}$	79.9	7.3	5	2.3
0.6	$(Fe_1^{3+})_T \{Li_{0.2}^{1+} Ni_{06} Fe_{1.2}^{3+}\}_0 O_4^{2+}$	83.3	7.2	5	2.2
0.8	$(Fe_1^{3+})_T \{Li_{0,1}^{1+} Ni_{0,8} Fe_{1,1}^{3+}\}_0 O_4^{2+}$	90.9	7.1	5	2.1
1.0	$(Fe_1^{3+})_T \{Ni_{1.0}Fe_1^{3+}\}_0 O_4^{2+}$	100.0	7.0	5	2.0

The Structural study

The study of the structural properties of the ferrites displays an important role to illustrate the changing of their properties. For the given ferrites samples, the structure of each sample changes since the ions with different ionic radius distributes into the T_d and O_h sites. The cations radii R_T and R_o are the mean ionic radii per molecule for the T_d and the O_h sites, respectively, can be calculated according to the suggested cations distribution, using equations [29]

$$R_T = R_{Fe^{3+}} \tag{1}$$

$$R_{0} = \frac{1}{2} \left[0.5(1-x)R_{Li^{1+}} + xR_{Ni^{2+}} + (1.5 - 0.5x)R_{Fe^{3+}} \right]$$
(2)

where $R_{Li^{2+}} = 0.090 \ nm$ is the ionic radius of the Li^{1+} ion in the O_h site, $R_{Ni^{2+}} = 0.083 \ nm$ is the ionic radius of the Ni^{2+} ion in the O_h site, $R_{Fe^{3+}}$ is the ionic radius of the Fe^{3+} ion which is 0.063 nm and 0.069 nm in the T_d site and in the O_h site, respectively. The calculated values of the R_T , R_O and R_T/R_O for the mixed Li-Ni spinel ferrite are tabulated in Table 2. From this table, it is clear that, with increasing of the Ni^{2+} ions the R_T remain constant while R_O increased. This behavior is attributed to replacement of the small ionic radius of the Fe^{3+} ions with the large ionic radius of Ni^{2+} ions in the O_h sites. The change in the R_T and R_O definitely causes deviations in the arrangement of the nearest neighboring ions and the interactions between them [30]. A similar result has been found in our earlier work spinel ferrite system [29]. Furthermore, the values of the average ionic radius $R_{av} = (R_T + R_O)/2$ increase with increasing in composition x as in Table 2. Also, the same table shows that, the value of R_T/R_O decreases with increasing of the Ni^{2+} ions. The substitution of Ni^{2+} ions, in the given ferrites system implies to expand/contract of the O_h cations. Thus, the increase in the R_O indicate that there was a slight movement of anions surrounding the cations the movement O^{2-} ions can be explained by means of O^{2-} ions positional

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parameter or anion parameter (*u*) [31,32]. The detailed positions for the O^{2^-} ions can be determined by *u*, which reflects alteration of the structure to accommodate differences in the R_T/R_0 [15,16].

In the spinel structure each unit cell contains eight formula units of $M^{2+}Fe_2^{3+}O_4^{2+}$ [10-14]. The position of the atoms in the spinel depends on the choice of origin in Fd3mO₇^h, which is either T_d site cations with $\overline{4}3m$ or an O_h site vacancy with $\overline{3}m$. The values of $u^{\overline{4}3m}$ for each composite were calculated using the formula [33-36]

$$u^{\overline{4}3m} = (1/a_{th}\sqrt{3})(R_T + R_{ox}) + (0.25)$$
(3)

where $u^{\overline{4}3m}$ is the center of symmetry (0.375, 0.375, 0.375), which relates to the $u^{\overline{3}m}$ with the center of symmetry (0.25, 0.25, 0.25) by $u^{\overline{3}m} = u^{\overline{4}3m} - (0.125)$. $a_{th} = (8/3\sqrt{3})[R_T + \sqrt{3}R_0 + R_{0x}(1 + \sqrt{3})]$ is the lattice parameter with $R_{0x} = 0.138 \ nm$ is the ionic radius of O^2 ion. This imples that

$$u^{\overline{3}m} = (1/a_{th}\sqrt{3})(R_T + R_{ox}) - (0.125)$$
⁽⁴⁾

The calculated values of a_{th} , $u^{\overline{4}3m}$ and $u^{\overline{3}m}$ are shown in Table 2. In such Table, the values of a_{th} increased with increasing of Ni content. The small values of a_{th} are attributed to the possible creation of Fe^{2+} ions in samples and the approximation is based on Fe^{3+} ions [32]. As such, it can be concluded that the O_h sites plays dominant role in influencing the value of a_{th} [17]. As shown in Table 2 the values of the $u^{\overline{4}3m}$ and the $u^{\overline{3}m}$ decreased with the incorporation of Ni content. This results agreed with the other worker [37]. The observed trend is suggesting that with the increasing substitution of Ni^{2+} ions, the anions are moving toward the cations at T_d interstices due to the expansion of the T_d interstices [15,16]. For spinel compounds, the anion sublattice expands or reduces by varying u, until the T_d site and O_h site volumes match the radii of the consistent cations [17].

X	$R_{\rm T}$ (nm)	$R_{\rm O}$ (nm)	R _{av} (nm)	$R_{\rm T}/R_{\rm O}$	$a_{\rm th}~({\rm nm})$	$V(nm)^3$	u^{43m}	u^{3m}
0.0	0.0630	0.0743	0.0686	0.8485	0.8755	0.6710	0.3826	0.2576
0.2	0.0630	0.0746	0.0688	0.8445	0.8764	0.6731	0.3824	0.2574
0.4	0.0630	0.0750	0.0690	0.8406	0.8773	0.6753	0.3823	0.2573
0.6	0.0630	0.0753	0.0691	0.8367	0.8783	0.6774	0.3821	0.2571
0.8	0.0630	0.0757	0.0693	0.8328	0.8792	0.6796	0.3820	0.2570
1.0	0.0630	0.0760	0.0695	0.8290	0.8801	0.6818	0.3819	0.2569

Table 2: Variation of T_d site (R_T) , O_h site (R_O) , R_{av} , R_T / R_O , a_{th} , $u^{\overline{4}3m}$ and $u^{\overline{3}m}$ with the Zn content.

As shown in table 2 the calculated values of $u^{\overline{4}3m}$ (3826 to 0.3819) and $u^{\overline{3}m}$ (0.2576 to 0.2669), which are larger than the ideal values of $u_{ideal}^{\overline{4}3m} = 0.375$ and $u_{ideal}^{\overline{3}m} = 0.25$, decrease with increasing of *Ni* content. This demonstrates that the O^{2-} ions are displaced in such a way that weakens the T_d -O_h interaction, the distance between T_d and O_h ions and between O_h and O^{2-} ions decreases, leading to

strengthening the interaction of T_d - T_d and O_h - O_h as was also observed earlier and lead to changes in magnetic properties [38].

Interionic distances (bond lengths (BLs)) can be defined as a function of the a_{th} and the u [31]. The BLs between the cation-cation (*C*-*C*) and cations-anion (*C*-*A*) play a crucial role on the overall strength of different magnetic interactions and super exchange interaction. Therefore, in present work we tried to evaluate the same to get information about the effect of Ni^{2+} ions on the super-exchange interactions [15,16].

The defined BLs between the C-C are b,c,d,e and f, also between the C-A are p,q,r and s [29] can be calculated using (Eqs. 5-13 first column in the Table 3) [33-36]. In such Table, all the BLs of both C-C and C-A increased except p and r unchanged due to increasing of Ni contents, which result to increase V (Table 2). The increase in C-C and C-A distances should result in the undermining of strength of magnetic interactions [32]. Consequently, changes in BLs of C-C and C-A would strongly influence superexchange interactions O_h -A- O_h , T_d -A- O_h and T_d -A- T_d , which in-turn will have effect on magnetic properties. The increase in C-C and C-A distances can be clarified on basis of reordering of cations or generation of defects that may be lead to the weakening of the strength of inter-atomic bonding in system [17]. It is of value note that the existence of a specific ions Fe, Ni and Li on T_d or O_h sites will have considerable influence on C-C and C-A distances so the magnetic properties would get affected [38].

Bond lengths (nm)		Composition x							
		0.0	0.2	0.4	0.6	0.8	1.0		
(C-A)									
$p = a(0.625 - u^{\overline{4}3m})$	(5)	0.2123	0.2126	0.2130	0.2133	0.2137	0.2140		
$q = a(u^{43m} - 0.25)\sqrt{3}$	(6)	0.2010	0.2010	0.2010	0.2010	0.2010	0.2010		
$r = a(u^{43m} - 0.25)\sqrt{11}$	(7)	0.3849	0.3849	0.3849	0.3849	0.3849	0.3849		
$s = \alpha(\frac{u^{\bar{4}3m}}{3} + 0.125)\sqrt{3}$	(8)	0.3829	0.3832	0.3836	0.3839	0.3843	0.3846		
(<i>C</i> - <i>C</i>)									
$\boldsymbol{b} = (\boldsymbol{0}. \boldsymbol{25u}^{\mathtt{I}3m}) \sqrt{2}$	(9)	0.3095	0.3099	0.3102	0.3105	0.3108	0.3112		
$c = (0.125u^{43m})\sqrt{11}$	(10)	0.3629	0.3633	0.3637	0.3641	0.3645	0.3649		
$d=(0.25u^{\overline{4}3m})\sqrt{3}$	(11)	0.3791	0.3795	0.3799	0.3803	0.3807	0.3811		
$e = (0.375u^{43m})\sqrt{3}$	(12)	0.5686	0.5692	0.5698	0.5704	0.5711	0.5717		
$f = (0.25u^{43m})\sqrt{6}$	(13)	0.5361	0.5367	0.5373	0.5378	0.5384	0.5390		

Table 3: The values of interionic distances (bond lengths) of cation-cation (*C*-*C*) (b, c, d, e, f) and cationanion (*C*-*A*) (p, q, r, s) as a function of *Ni* concentration (x).

Another parameter can be affected to the strength/magnitude of super-exchange interactions is the bond angles (θ_1 , θ_2 , θ_3 , θ_4 and θ_5) [32]. The θ_1 , θ_2 , θ_3 , θ_4 and θ_5 were calculated by simple trigonometric principle using the BLs with the help of the given formula (Eqs. 14-18 first column in the Table (4)) [15-16]. The favorable bond angles of the configurations of ion pairs in spinel ferrites for



effective magnetic interaction are $\theta_1 = 126^\circ$, $\theta_2 = 154^\circ$, $\theta_3 = 90^\circ$, $\theta_4 = 125^\circ$ and $\theta_5 = 79^\circ$ [39]. As shown in Table 4 with increasing of Ni^{2+} ions, the values of θ_1 , θ_2 , and θ_5 increased and θ_3 the θ_4 decreased. The increasing trend in θ_1 , θ_2 , and θ_5 is again suggesting the strengthening of T_d - O_h and T_d - T_d interactions and the decreasing in θ_3 as well θ_4 indicating the weakening of O_h - O_h interactions.

Bond angles		Composition x						
		0.0	0.2	0.4	0.6	0.8	1.0	
$\theta_1 = \cos^{-1}[(p^2 + q^2 - c^2)/2pq]$	(14)	122.86	122.89	122.94	122.98	123.02	123.07	
$\theta_2 = cos^{-1}[(p^2 + r^2 - e^2)/2pr]$	(15)	142.81	143.00	143.19	143.38	143.57	143.76	
$\theta_3 = cos^{-1}[(2p^2-b^2)/2p^2]$	(16)	93.63	93.56	93.49	93.41	93.35	93.28	
$\theta_4 = cos^{-1}[(p^2+s^2-f^2)/2ps]$	(17)	126.09	126.07	126.06	126.04	126.03	126.01	
$\theta_5 = cos^{-1}[(r^2+q^2-d^2)/2rq]$	(18)	73.16	73.28	73.39	73.51	73.63	73.75	

Table 4: The values of bond angles (in degrees) as a function of *Ni* concentration (*x*).

In spinel ferrite, S the BLs between the C-A at T_d -site (d_{TL}) , and O_h -site (d_{OL}) , together with the length of closest A-A approach, T_d edge (d_{TE}) , shared and unshared O_h edges (d_{OE}) and (d_{OEu}) , respectively, can be estimated by the following expressions [35,36]

$$d_{TL} = a_{th} \sqrt{3} \left(u^{\overline{4}3m} - 1/4 \right) \tag{19}$$

$$d_{OL} = a_{th} \sqrt{3 \left(u^{\overline{4}3m} \right)^2 - (11/4) u^{\overline{4}3m} + (43/64)}$$
(20)

$$d_{TE} = a_{th} \sqrt{2} \left(2u^{\overline{4}3m} - 1/2 \right) \tag{21}$$

$$d_{OE} = a_{th} \sqrt{2} \left(1 - 2u^{\overline{4}3m} \right) \tag{22}$$

$$d_{OEu} = a_{th} \sqrt{4(u^{\overline{4}3m})^2 - 3u^{\overline{4}3m} + (11/16)}$$
(23)

Figures 1 and 2 illustrate the d_{TL} , the d_{OL} , the d_{TE} , the d_{OE} and the d_{OEu} as a function in Ni content. It can be seen that, the d_{OE} , the d_{OEu} , the d_{TE} and the d_{OL} increased, where the d_{TL} unchanged with increasing of Ni^{2+} ions. This may be due to the expansion of the unit cell arising from the substitution of relatively larger Ni^{2+} ions instead of Fe^{3+} and Li^{1+} ions. It is reported that the hyperfine magnetic field (H_T and H_O) of T_d and O_h sites, respectively, increase with increasing of d_{TL} and d_{OL} , which reduces the d electron covalent character of the $Fe^{3+}-O^{2-}$ bond at these sites. This increase causes the increase of the s-d orbital overlap and, hence, the hyperfine fields H_T and H_O decrease. This covalent character is higher at the T_d sites, thus H_T is lower than H_O [40].





Fig. 1: Allied parameters d_{TE} , d_{OE} and d_{OEu} with concentration 'x' for the mixed *Li-Ni* spinel ferrites.



Fig. 2:Allied parameters d_{TL} and d_{OL} with concentration 'x' for the mixed *Li-Ni* spinel ferrites.

The jump or hopping length (HL) is the separation or distance between the centers of the adjacent magnetic ions occupied two sites T_d and O_h , which depends on different factors such as ionic radius [40]. Therefore, the introduction of larger ions results in an increase of the HL between the magnetic ions. The incorporation of Ni^{2+} ions in place of Li^+ ions and Fe^{3+} ions caused to an expansion of the O_h site. The expansion of O_h site is observed in terms of R_o and d_{OE} . This expansion of O_h site was attributed to the shift of O^{2-} ions without changing the structure symmetry. For the spinel structure, the T_d - T_d sites separation $(HL_{T-T} = (a_{th}/4)\sqrt{3})$, $O_{\rm h}-O_{\rm h}$ sites separation $(HL_{O-O} = (a_{th}/4)\sqrt{2})$ and $T_{\rm d}-O_{\rm h}$ sites separation $(HL_{T-0} = (a_{th}/8)\sqrt{11})$ as reported by [41,42]. The variation of the HL_{T-T} , HL_{0-0} and HL_{T-0} are shown in Figure 3. It was clear that HL_{T-T} , HL_{0-0} and HL_{T-0} have the same behavior as that of a_{th} , where they increase with increasing of Ni^{2+} ions concentration. This may be due to the expansion of unit cell arising from the substitution of relatively larger Ni^{2+} ions instead of smaller Fe^{3+} ions. The reducing of the Fe^{3+} ions in the O_h -sublattices and introduction of the Ni^{2+} ions in the O_h -sublattices make the magnetic ions move away for each other and decrease the HLs between them [36]. It is reported that $H_{\rm T}$ and H_0 depend on the HLs between cations [36], which reflects the same behavior with changing them. It is reported that the longest hopping i.e. the longest T_d - T_d distance corresponds to highest H_T , while the shortest hopping i.e. $O_h - O_h$ distances correspond to lowest H_0 . This confirms $T_d - O_h$ magnetic exchange interaction exceeds Oh-Oh magnetic interactions.



Fig. 3: Variation of hopping lengths with concentration 'x' for the mixed Li-Ni spinel ferrites.



Magnetic Properties

The magnetic properties for the spinel ferrite can be understood in term of cations distribution and exchange interactions between the two sub lattices, $T_{\rm d}$ and $O_{\rm h}$ of spinel structure. The relation between the $M_{net}(A/m)$ and the applied magnetic field intensity (H) in the range of [200-2650 (A/m)] for the prepared spinel ferrite samples of the system $Li_{5-.5x}Ni_xFe_{(2.5-.5x)}O_4$ were studied at room temperature of 299 K. The obtained results for the ferrite samples were illustrated in Figure (4). In such Figure emphasizes that M_{net} increases with increasing of H, which is the normal behavior for the ferrites. Depending on our pervious discussion of the ionic radii, which increased with increasing of Ni concentrations and the M_{net} foreseen for the spinel lattice, the magnetization was expected to decrease. Figure 5 shows the variation of M_{net} for H = 500, 1500 and 2500 A.m⁻¹. From the Figure (5), it can be seen that the values of the M decrease exponentially with increasing of the Ni concentrations. This behavior can be explained by Neel's two-sublattice model of the magnetism theory of the ferrimagnetic materials [43]. According to this model, the magnetic ordering in the simple spinel ferrites is based on $T_{\rm d}$ and $O_{\rm h}$ sites and the resultant M_{net} are the difference between two of them provided that they are collinear and anti-parallel to each other. In mixed Li-Ni spinel ferrite, Ni^{2+} ions preferentially in $O_{\rm h}$ site in the cubic spinel lattice. Therefore, by adding of the Ni^{2+} ions (low spin quantum number) the concentration of Fe^{3+} ions (high spin quantum number) in the O_h site, which implies to decrease the M_{net} . Thus; the decrease of M_{net} is, also, attributed to that $T_d - O_h$ exchange interaction becomes weaker or comparable with $O_h - O_h$ exchange interactions.



Fig. 4: Variation of *M* with *H* for all samples.

Fig. 5- Variation of M_{net} with the composition x for different values of H.

Relative permeability

The relative permeability (μ_r) for all samples gives the description of the magnetic behavior during the change of the exposed external H. It is defined as the ratio between the induction field (*B*) and the intensity of the *H*, i.e. $\mu_r = B/\mu_o H$ with μ_o is the free space permeability. The relation between *H* and μ_r has an interesting behavior for the present ferrite samples as shown in Figure (6). As indicated in this Figure, μ_r increased with increasing of the *H* and decreasing with the increase of Ni^{2+} ions. The



increment of μ_r could be related to the alignment effect of *H* on the ionic spins. In such away, the increasing of *H* causes rapid increasing of *B*, which causing a pronounced increasing of μ_r . The behavior of μ_r versus *H* for samples of x = 0.0 was divided into two stages I and II as in Figure (6). In stage I, μ_r increases with increasing of *H* up to H = 750 A/m then μ_r decreases. The increasing of *H* causes a very rapid increase in *B*, which imply to increase μ_r . This behavior can be related to the aligning effect of *H* on the ionic spins. In stage II, μ_r decreasing with increasing of *H* might be cause a slight increase of B, which can be explained as that this sample have the highest spin ordering. This means that, the ionic ordering of this sample is closer to the saturation state. The same behavior was observed by H. Dawoud [10].



Fig.6: Variation of μ_r with H for the samples with composition x =0.0, 0.2, 0.4, 0.6, 0.8 and 1.0.

Conclusions

Substitution of the non-magnetic Ni^{2+} ions in *Li*-spinel ferrite has a tremendous influence such the magnetic properties. From this study, we concluded that:

- > The calculated and measured values of the net magnetization (M_{net}) of the mixed *Li-Ni* spinel ferrite are decreased as increasing of Ni^{2+} ions.
- > The structural properties of the mixed *Li-Ni* spinel ferrite are changed as increasing of Ni^{2+} ions.
- The change in the structural properties showed an influence on the magnetic interactions between cations.
- > The ionic radii of tetrahedral (T_d) sites and octahedral (O_h) sites are found to change linearly with increasing of the Ni^{2+} ions.
- > The caluclated magnetic permeability was decreased with increasing of the Ni^{2+} ions.

Moreover, Ni^{2+} content has important influence on the magnetic properties of *Li*-ferrites, so, the mixed *Li-Ni* spinel ferrite is subjected as a soft ferrite material, which is evidenced an interest material for technological and scientific applications.

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