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Synthetize and Magnetic Properties of Ni Substituted Polycrystalline Zn-spinel Ferrites

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Abstract : The mixed polycrystalline $Ni_{1-s}Zn_sFe_2O_4$ ferrites where s is the percentage increments of Zn ions, were prepared using the standard double sintering by mixing pure metal oxides NiO, ZnO and Fe_2O_3. The netmagnetization (M_{net})was studied at room temperature as a function of applied magnetic field(H)over the range of (0-45) Oe ina constant magnetizing frequency (ν = 50 Hz). M_{net} show increasing with increasing of H. M_{net} is found to increase for the samples of s = 0.0, 0.2, 0.4and 0.6 then decreases thereafter while the concentration of Zn increases in matrix i.e. samples of s = 0.8 and 1.0. The increasing of the magnetization with increasing of Zn^{2+} ions for the samples ofs = 0.0, 0.2 and 0.4 explained by Neel's two-sublattice model. However, for the decreasing of magnetization beyonds> 0.6explained by Yafet and Kittel for the three-sublattice model. The Yafet-Kittel angle (φ_{YK}) for the samples of s = 0.4 to s = 1.0, shows increasing with increasing of the amount of nonmagnetic ions Zn^{2+} in ferrite. The relation between H and relative permeability (μ_r) show an interesting behaviourof the present ferrite samples. μ_r for the samples of s = 0.0, 0.2, 0.8 and 1.0 is found to be smaller than the samples with s = 0.4 and s = 0.6. Substitution of the non-magnetic Zn^{2+} ions in Ni spinel ferrite has a tremendous influence such the magnetic properties. Furthermore, Zn content has significant influence on the magnetic properties for Niferrites, so, the mixed Ni-Zn spinel ferrite is considered a soft ferrite material, which is proved an interesting material for technological and scientific applications.
Keywords—Magnetic Properties, Ni-Zn spinel ferrite, Yafet-Kittel model, relative permeability, Neel's model.

I. INTRODUCTION

Spinel ferrites have a general structure formula DFe_2O_4 (D is divalent metal ions) [1-3]. The magnetic properties of spinel ferrites can be explained according to Wiess domains [4]. The magnetization (M) within the domains are called the intrinsic magnetization per unit mass (M_i/m) at a certain temperature (T) and its value at zero applied magnetic field (H) is the spontaneous magnetization (M_s).As shown in Fig. (1a), for any unmagnetized sample the domains are randomly oriented at $H = 0$; i.e. the net magnetic moment (μ_{net}) is zero. When H increases, the domains become more aligned in the direction of H by rotating slightly until all of them are nearly aligned. At this state, the saturation condition corresponds to the state where all domains are in the same direction parallel to H, which results to magnetize the sample (Fig. (1d)) [4]. The intensity of M_i in spinel ferrite can be explained according to the spins distribution of magnetic ions at tetrahedral (T_d) sites and octahedral (O_h) sites [5]. The superexchange interactions between T_d and O_h cations; via an intermediate O^{2-} ions are greatest, if T_d and O_h cations and O^{2-}ions are collinear and their separations are not too large. The ions arrangement in the spinel lattice is likely to be most important as is shown in Fig. (2) [4]. Suppose that intra-sublattice (I_{TT}, I_{OO}) and inter-sublattice (I_{TO}) correspond to molecular-field constants of cations exchange interaction T_d-T_d, O_h-O_h and T_d-O_h, respectively [6]. Generally, it is thought that the magnetic properties of spinel ferrites depend upon the relative strengths of these types of exchange interactions. When the cations of T_d and O_h sites are totally magnetic, the inter-exchange interactions I_{TO} are much stronger than I_{TT} and I_{OO} interactions i.e.,$|I_{TO}|>>|I_{OO}|>>|I_{TT}|$[7]. So, spins have a collinear structure in which moments on the T_d-sites are anti-parallel to the moments on the O_h-sites. However, when one of the intra-sublattice interactions becomes comparable with the inter-sublattice interaction it leads to a non-collinear spin structure [8]. The result is that the interactions between the sublattices are stronger than those within them. Further, these interactions between the ions within T_d sites are the weakest of all. This result thus; supports the assumption that the sublattices magnetizations are antiparallel.

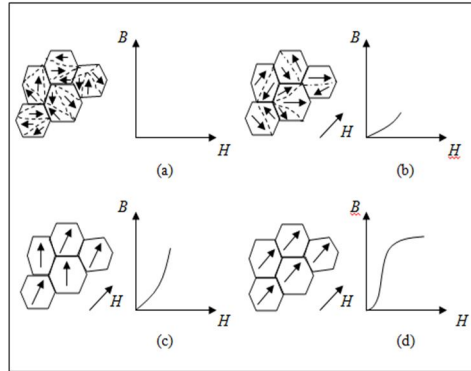# International Journal for Research in Applied Science & Engineering Technology (IJRASET)

Fig. (1): Random orientation spins of an unmagnetized sample of the ferrimagnetic substance alignment of Wiess domains [4].
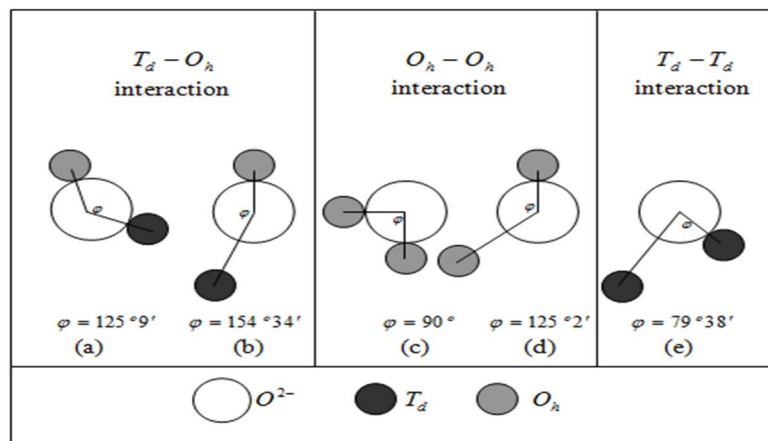

Fig. (2): Superexchange interactions of anions, O^{2-}, with T_d and O_h cations [4].

II. EXPERIMENTAL

The mixed polycrystalline ferrites $Ni_{1-s}Zn_sFe_2O_4$, where s is the percentage increments of Zn ions on the compound which have the value $0.0 \leq s \leq 1.0$, were prepared using the standard double sintering route by mixing pure metal oxides NiO, ZnO and Fe_2O_3 to produce 25.0 grams from $Ni-Zn$ polycrystalline spinel ferrites. The metal oxides were weighted using a sensitive electric balance(ADAM model PW124) with an accuracy $1 \times 10^{-4} gm$. The weighted metal oxides were mixed and then grounded into a very fine powder for 5 hr's. The grounded powder was pre-sintered at $750^o C$ for 3 hr's soaking time using a laboratory Furnace (BIFATHERM model AC62). Then the pre-fired powder was re-grounded for 3 hr's and then was pressed with a hydraulic press under constant pressure of 3×10^8 pa, by using a small quantity of butyl alcohol as a binding material. The samples were pressed intotoroidial shape and were sintered at the 1200 ^{o}C for 5 hr's, then were cooled down gradually to room temperature and the samples were polished to obtain uniform parallel surfaces to measure the magnetic properties. $M_{net}$was measured at room temperature as a function of H in the range (0-45) Oe at a constant magnetizing frequency ($v= 50$ Hz).

III. RESULTS AND DISCUSSION

A. Magnetic Properties

The relation between the net magnetization per gram[$M_{net}(emu/g)$] and the applied magnetic field intensity (H)in the range of[0 – 45 (Oe)] for the prepared ferrite samples of the $Ni_{1-s}Zn_sFe_2O_4$ system were studied at room temperature. The variation of M_{net} and H for the samples of s = 0.0, 0.2, 0.8 and 1.0 were illustrated in Fig. (3) while the samples of s = 0.4 and 0.6 were depicted in Fig. (4). The two Figs. (3 and 4) emphasize that $M_{net}$is directly proportional to H. Richa et. al. reported [9] that the lower value of M_{net} indicates a higher degree of surface disorder and a lower densification stage.Thus, the value of M_{net}increases with increasing of thecrystallinity of ferrite materials.

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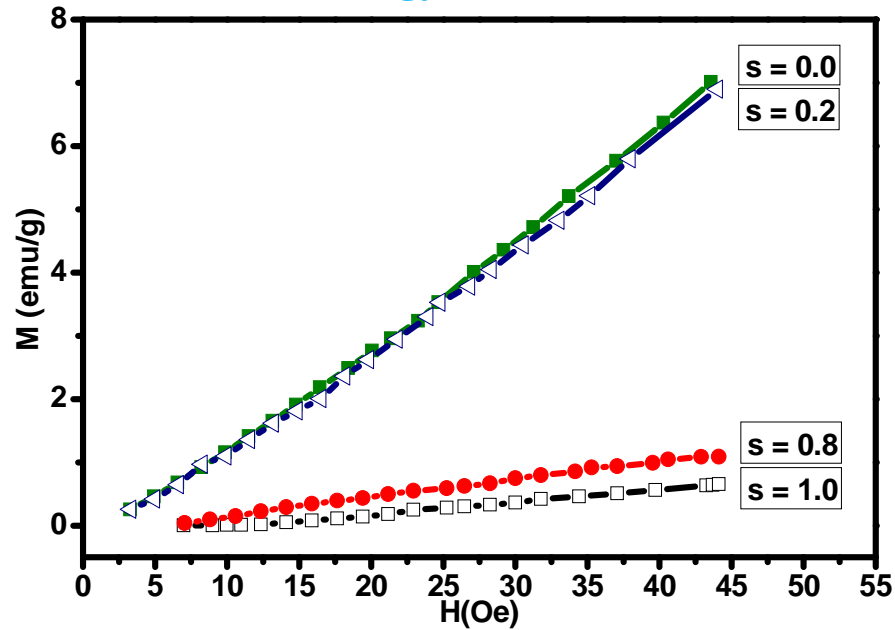

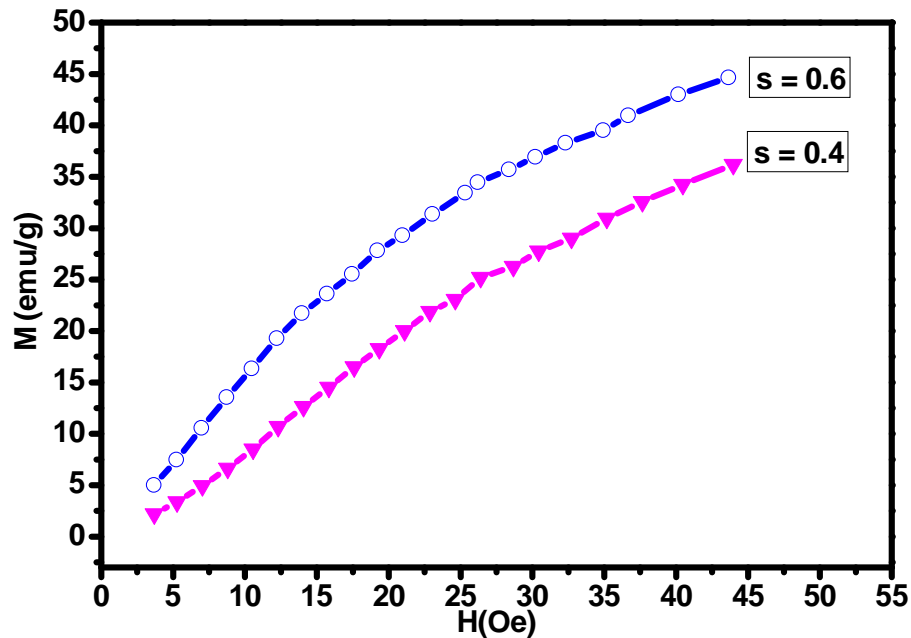Fig. (3): Variation of M_{net} with H for the samples of s = 0.0, s = 0.2, s = 0.8 and s = 1.0.

Fig. (4): Variation of M_{net} with H for samples of s = 0.4 and s = 0.6.

Based on the results in Figs (3 and 4), the dependence of M_{net} on the ratio of Zn in matrix $Ni_{1-s}Zn_sFe_2O_4$ for different values of H is represented in Fig. (5). It can be noted from Fig. (5), that initially, M_{net} increases for the samples of s = 0.0, 0.2, 0.4 and 0.6 then decreases thereafter while the concentration of Zn^{2+} ions increases in matrix i.e. samples of s = 0.8 and 1.0. The same behavior was seen by many researchers earlier [6,10-15], but most of them reported that M_{net} decreased when the concentration of Zn ranged from 40-60%. Depending on the particle sizes, which increased with increasing Zn concentrations and the theoretical magnetization foreseen for the spinel lattice, the magnetization was expected to increase. However, this increase was observed only at Zn concentrations of samples have $s \leq 0.6$.

The increasing of the magnetization with increasing of Zn^{2+} ions for the samples of $s \leq 0.6$ can be explained by Neel's two-sublattice model of the magnetic theory of the ferrimagnetic materials [1,16]. According to the this model, the magnetic ordering in the simple

spinel ferrites is based on T_d and O_h sites and the resultant magnetization are the difference between two of them provided that they are collinear and anti-parallel to each other. But Neel's model failed to explainthe decreasing of magnetization beyond$s >0.6$which can be explained by Yafet and Kittelforthe three-sublattices model [17,18].

In mixed *Ni-Zn* ferrite, Zn^{2+}ions concentrate preferentially in T_d site and Ni^{2+} ions in O_h site in the cubic spinel lattice. It is reported that, when*Ni-Zn*ferrites are substantially diluted with higher content of non-magneticZn^{2+}, spin canting and spin frustration takes places, giving rise to antiferromagnetic/anomalous behavior [8]. As*Zn* content increases more, the exchange interactions are weakened and spins ofO_h site are no longer held rigidly parallel to the few remaining spins of T_d site. Thus; the decrease of magnetization beyond$s >0.6$is attributed to that$T_d - O_h$ exchange interactionbecomes weaker or comparable with $O_h - O_h$ exchange interactions. This disturbs the parallel arrangement of spin magnetic moments on O_h site paving way for canted spins. The decrease in O_h site sublattice moment,interpreted as spin departure from co-linearity causes theeffect known as canting, this effect also described in samples of *Cu-Zn* ferrite [19,20].
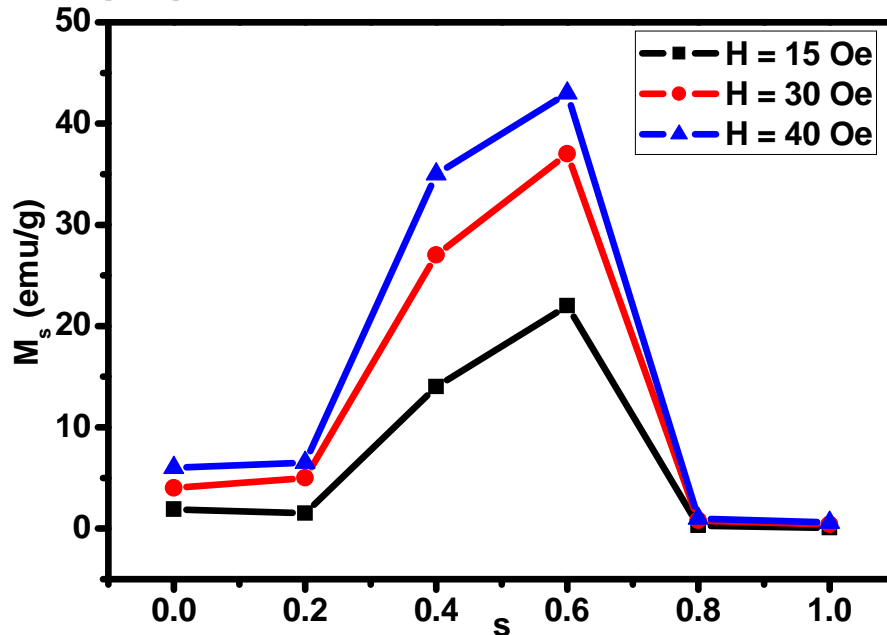

Fig. (5) : Variation of $M_{net}$with the composition *s*for differentvalues of*H* (Oe).

B.   Magnetic Moment

The net magnetization (M_{net}) for the ferrimagnetic materials can be calculated from individual magnetization,M_T andM_O of T_d and O_h sub-lattices, respectively. Furthermore, according to Neel's theory M_{net} at $T = 0 K$ is given by [1,21]

$$M_{net} = |M_O - M_T| \qquad (1)$$

where$M_O(0) = \sum_j N_j g\mu_B S_{mj}$ and $M_T(0) = \sum_i N_i g\mu_B S_{mi}$, with N_j and N_i are the numbers of the magnetic ions,S_{mj} and S_{mi} are spin quantum numbers of each ion in O_h and T_d sub-lattices, respectively, and g is thelande' splitting factor or spectroscopy splitting factor, which is approximately equal to 2.0003 for the free electron. M_O and M_T are calculated for the mixed *Ni-Zn* spinel ferriteaccording to the cations distribution formula$(Zn_s^{2+}Fe_{1-s}^{3+})\{Ni_{1-s}^{2+}Fe_{1+s}^{3+}\}O_4^{2-}$as in refs [9,17,22-27]

$$M_T = [sS_{Zn} + (1-s)S_{Fe}]2\mu_B \qquad (2)$$

$$M_O = [(1-s)S_{Ni} + (1+s)S_{Fe}]2\mu_B \qquad (3)$$

Substituting equations (2) and (3) into the equation (1), which follows

$$M_{net} = [(1-s)S_{Ni} + 2sS_{Fe}]2\mu_B \qquad (4)$$

Where $S_{Fe} = 5/2$ and $S_{Ni} = 1$for Fe^{3+} and Ni^{2+} ions, respectively. It is well known, that Zn^{2+} ion is a diamagnetic, i.e. $S_{Zn} = 0$. The

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variation of M_O, M_T and M_{net} with the composition s ispoltted in Fig. (6). From Fig. (6),it is clear that, with increasing of composition s, both M_O and M_{net} increase, while M_T decreases. The variation of M_{net} with Zn^{2+} ions could be explained by assuming that, by increasing of Zn^{2+}ions in the samples the relative number of the Fe^{3+} ions will increase in O_h sites, and will decrease in T_d sites. This tends to increase M_O and decrease M_T. Therefore, M_{net} would rise linearly with increasing of Zn^{2+} ions and would reach a value $10\,\mu_B$ per molecule might be expected on$s$ =1.0. The reason for this behavior isthe number of Fe^{3+} ions i.e. spins occupying the T_d site, causing M_{net} increases due to the introduction of Fe^{3+} ions having $5/2\mu_B$spin magnetic moment [8]. It can be concluded that, as Zn^{2+}replaces Fe^{3+} magnetic ions in T_d sites, the magnitude of $M_O$decreases but M_{net} increases, as a result M_{net} increase. It is reported that, when the concentration of Fe^{3+} ions in T_d site isdiluted by low concentration of diamagnetic substancessuch as Zn^{2+} and Cd^{2+} ions, implies to increase of M_{net} [8].
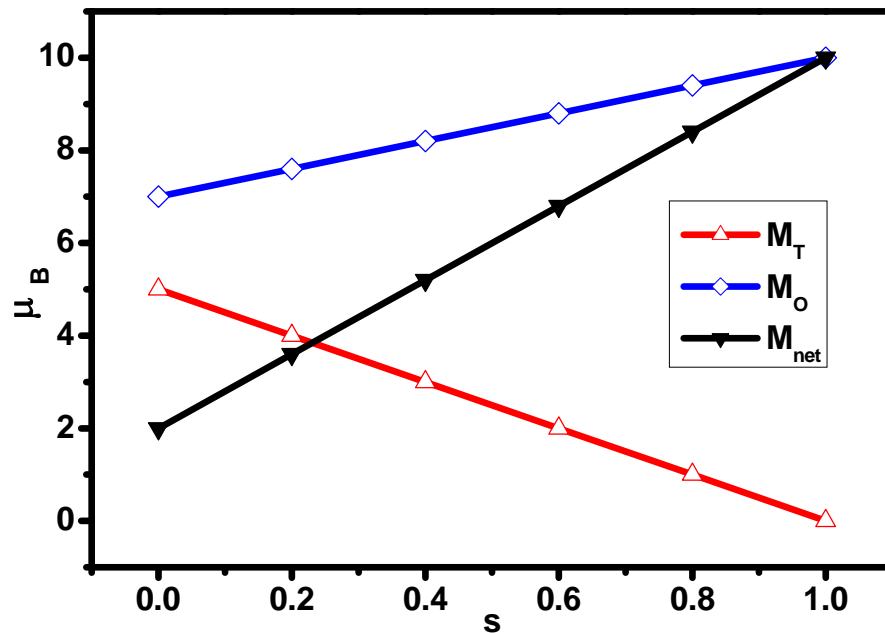

Fig. (6): Variation of M_O, M_T and M_{net} with Zn ratio"s".

According to the Yafet-Kittel model, M_{net} is given by [15,28,29]

$$M_{net} = \left| M_O \cos \varphi_{YF} - M_T \right| \quad (5)$$

Fig. 7 shows Yafet-Kittel angle (φ_{YK}) for the samples of s = 0.6upto s = 1.0, which isbetween the direction of the magnetic moment of T_d ions and magnetic moments of O_{h1} and O_{h2} ions. From Fig. (7), it can be noted that, the canting angle increases with increasing of the amount of nonmagnetic ions Zn^{2+} in ferrite, which causes weakening exchange interactions. The same results were obtained as in [15,28,30,31].The exchange interactions between magnetic ions through oxygen ion are superexchange interaction with antiferromagnetic nature. These interactions depend on bond distance and bond length. Normally, $|I_{TO}| \gg |I_{OO}| \gg |I_{TT}|$, therefore, the magnetic moments of T_d sublatticeare antiparallel oriented with spins in O_h sublattice. The increase of Zn content in Ni-Zn ferrites make weakening I_{TO} and could lead to canting structure in O_h site. The existence of canted spin gives rise to φ_{YK} which compares the strength of T_d-O_h and O_h-O_h exchange interaction. The physical origin of this spin arrangement is from the canting of the spins in the O_h-sublattice due to weakening of I_{TO} and enhancing of I_{OO}on nonmagnetic Zn substitution in the T_d-sublattice. As in the case of spinels, $I_{OO}$is antiferromagnetic, the O_h-sublattice splits into two sublattices forming φ_{YK} between the directions of the spins resulting in a decrease of M_O [8].

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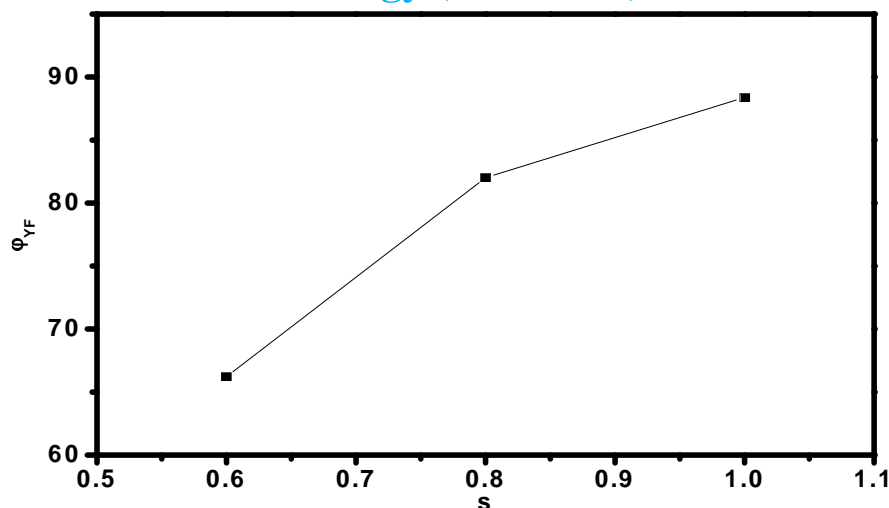

Fig. (7): Variation of φ_{YK} with composition s = 0.6 , 0.8 and 1.0.

C. Relative Permeability

The relative permeability (μ_r)describes the magnetization behavior of the material during the change of the exposed H. μ_r is defined as the ratio between the induction field (B) and H, which is given by $\mu_r = B/\mu_o H$, where μ_o is the permeability of free space. The relation between H and μ_r has an interesting behavior of the present ferritesamples as shown in Fig. (8) and Fig. (9).For the samples of s = 0.0, 0.2, 0.8 and 1.0 that have the smaller values of μ_r are shown in Fig. (8). This behavior is attributed to that these samples have a very low magnetization. While Fig. (9) shows μ_r for the samples with $s = 0.4$ and $s = 0.6$, whichincreases with increasing of H till a maximum value, after this value it decreases. This means that these two samples have the highest spin ordering (highest intrinsic magnetization) compared with the other samples. It is also noticed that, the ionic ordering of these two samples is closer to the saturation state than the others. Accordingly, the increase of H might cause a slight increase of B gives rise to a distinct decrease in μ_r.

The increase of H causes a pronounced increase of μ_r which decrease with increasing of Zn^{2+} content (low spin ion). The increment of μ_r couldbe related to the alignment effect of H on the ionic spins. In such away, the increase of H causes rapid increasing of B, which causes increasing of μ_r,the maximum value of μ_r is found in the sample of $s =0.6$ with composition $Ni_{0.4}Zn_{0.6}Fe_2O_4$.
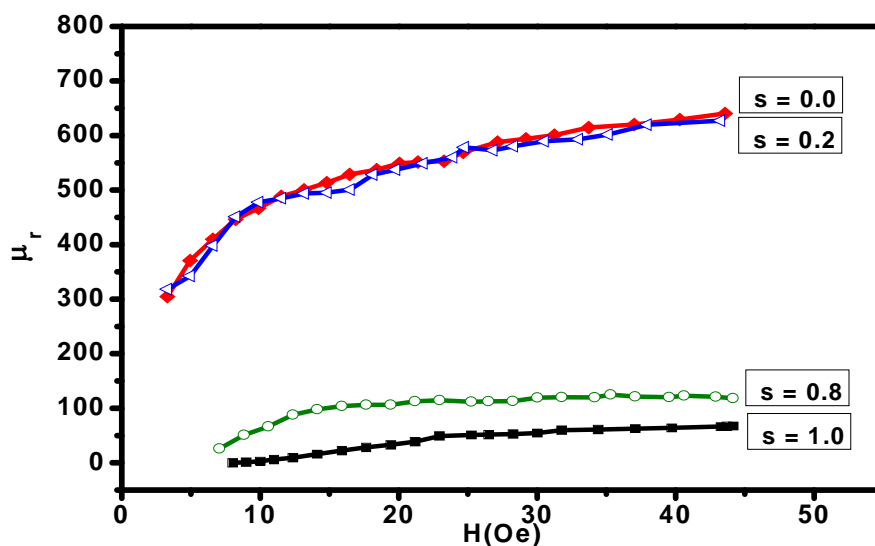

Fig. (8): Variation of μ_r with H for samples with composition s=0.0,0.2,0.8and1.0.

116

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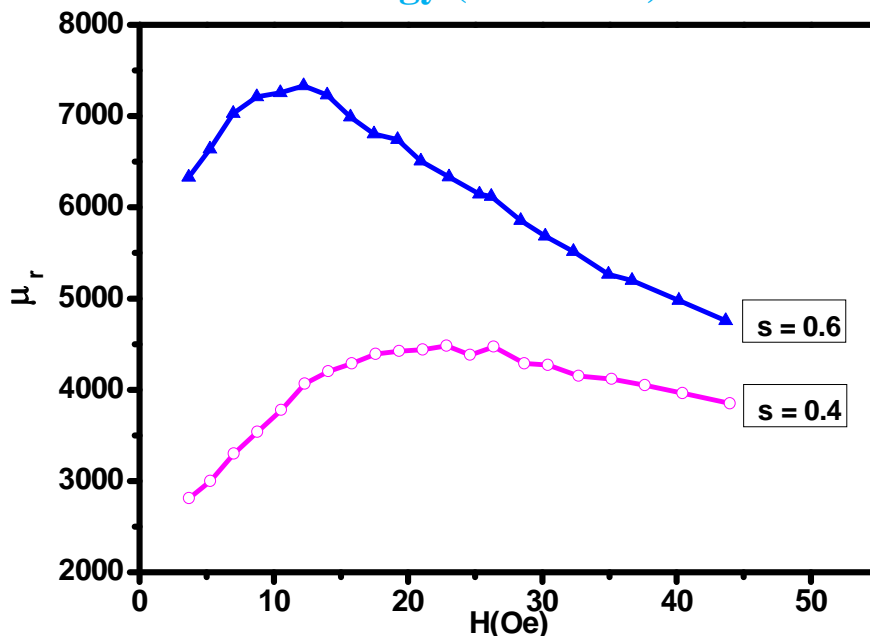

Fig. (9): Variation of μ_r with H for samples with composition s = 0.4 and 0.6.

IV. CONCLUSION

Substitution of the non-magnetic Zn^{2+} ions in Ni spinel ferrite has a tremendous influence such the magnetic properties. From this study, we concluded from the obtained results that The magnetization increased with the increasing of the Zn^{2+} ions in the samples with s = 0.0 to s = 0.6, then decreased for samples of s >0.6 while Zncontent increase .The increasing of the magnetization was explained on the basis of Neel's two sublattices model. Furthermore, Zn content has significant influence on the magnetic properties for Ni ferrites, so, the mixed Ni-Zn spinel ferrite is considered a soft ferrite material, which is proved an interesting material for technological and scientific applications

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