

Magnetic Properties of Zn Substituted Cu Ferrite

الخواص المغناطيسية لمركب النحاس - فريت المُطعَم بالزنك

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Abstract

The *Cu-Zn* ferrites samples have the general formula $Cu_{1-s}Zn_sFe_2O_4$, (where *s* is stepped by 0.2 according to $0.0 \leq s \leq 1.0$), The samples are used to measure the magnetization at room temperature. The magnetization increases with the increase of zinc ions up to 60%, and then it decreases with further addition of zinc ions. The increase of the sample magnetization is explained on the basis of Néel's two sub-lattice model, while the decrease in the magnetization beyond $s = 0.6$ was attributed to the presence of a triangular spin arrangement on octahedral O_h sites; and was explained by the three sub-lattice model suggested by Yafet and Kittle. The magnetic moment for tetrahedral T_d sites and octahedral O_h sites were calculated according to the suggested cations distribution $(Zn_s^{2+} Fe_{1-s}^{3+})_{T_d} \{Cu_{1-s}^{2+} Fe_{1+s}^{3+}\}_{O_h} O_4^{2-}$. The relative permeability μ_r is calculated for the above-mentioned samples.

ملخص

يهدف هذا البحث إلى قياس شدة التمغنط لعينات من نحاس-زنك فريت ذات الصيغة العامة cm^{-1} (200 – 1000)، حيث s تتدرج بمقدار 0.2 حسب $(0.0 \leq s \leq 1.0)$ عند درجة حرارة الغرفة ووجد أن شدة التمغنط تزيد مع زيادة أيونات الزنك حتى $s = 0.6$ ، ثم أنها تقل مع زيادة أيونات الزنك. قد نوقشت الزيادة في شدة التمغنط على أساس نموذج نيل (Néel's two sub-lattice)، بينما نوقش النقص في شدة التمغنط بعد $s = 0.6$ خلال وجود ترتيب الزاوي (spin) في الموقع الثماني على أساس نموذج يافت و كيتل (Yafet and Kittle three sub-lattice model). كذلك تم حساب العزم المغناطيسي للموقع الرباعي والثماني حسب التوزيع الكاتيوني المقترح $(Zn_s^{2+} Fe_{1-s}^{3+})_{T_d} \{Cu_{1-s}^{2+} Fe_{1+s}^{3+}\}_{O_h} O_4^{2-}$. وكذلك حساب النفاذية المغناطيسية μ_r للعينات المعطاة.

1. Introduction

The magnetic and electric properties of ferrite materials strongly depend upon their chemical and physical structure. This would help to determine the precise configuration of the atoms and the ions in the ferrimagnetic semiconductor⁽¹⁾.

The magnetic properties are crucial in selecting materials for various applications, such as the transformer cores, low-frequency inductors and deflection systems in TV circuits. Among the technically important ferrites are the mixed Zn-ferrites. Zinc which is substituted in ferrites has been extensively studied by⁽²⁻⁶⁾. It is shown that exceptional properties were obtained when Zinc is substituted Fe_3^+ ions in tetrahedral sites. Zinc as non-magnetic divalent ions that occupy essentially tetrahedral sites when substituted in ferrites. No systematic study has been performed of cations distribution and the magnetization of Cu-Zn ferrites; therefore, in the present work we will study the magnetization M and the relative permeability μ_r of mixed Cu-Zn ferrites. The results of this study are presented in this paper.

2. Experimental

Polycrystalline mixed ferrites have the general formula $Cu_{1-s}Zn_sFe_2O_4$, (where s is stepped by 0.2 according to $0.0 \leq s \leq 1.0$), were prepared by the conventional standard double sintering ceramic method⁽⁷⁾. Ferric oxide (99.8 %pure), Zinc oxide (99.9% pure) and copper oxide (99.9% pure) were weighted and mixed and then grounded to a very fine powder for 5 hours, then the mixed powder oxides presintered at $750^\circ C$ using Laboratory Furnaces for 3 hours soaking time. Then the prefired powder was ground well for 3 hours by using a small quantity of butyl alcohol as a binding material, samples were pressed with hydraulic press under constant pressure of $(3 \times 10^8 Pa)$, in the form of a torodial shape with an external diameter $8.2 mm$, and internal diameter $4.8 mm$, and thickness $3-5 mm$. All samples were sintered at $1100^\circ C$ for soaking time of 5 hours. After sintering, the samples left to be cooled gradually at room temperature. The magnetization (M) was determined as a function of the applied magnetizing current passing through the primary coil which changed in the range $(0-2)A$ at room temperature and an applied frequency $\nu = 10 KHz$. The corresponding applied magnetic field, H , varied in the range $(0-510 A \cdot m^{-1})$. The measurements of the magnetization M and the relative permeability μ_r are based on **Faraday's** law of electromagnetic induction. This concept was applied to the torodial specimen of the Cu – Zn ferrite and used as a core of two coils. This would transfer the induced magnetic field that caused by the current passed throughout primary coil. The field leads to raise the current in the secondary coil.

3. Results and Discussion

3.1 Magnetization Study

The relation between the net magnetization $M (A \cdot m^{-1})$ and the applied magnetic field intensity $H (A \cdot m^{-1})$ for the ferrite samples of the system $Cu_{1-s}Zn_sFe_2O_4$ are carried out at room temperature. The

obtained results for the ferrite samples with $s \leq 0.6$ and samples with $s > 0.6$ are illustrated in figures (1 and 2) respectively. In general, the magnetization increases with increase of the applied field for all samples. However, the samples of, $s > 0.6$, of low magnetization are expected to be paramagnetic at room temperature.

In the range of the applied magnetic field, H , under consideration ($0 - 510 \text{ A.m}^{-1}$), the measured magnetization M of the samples do not reach a saturation of magnetization.

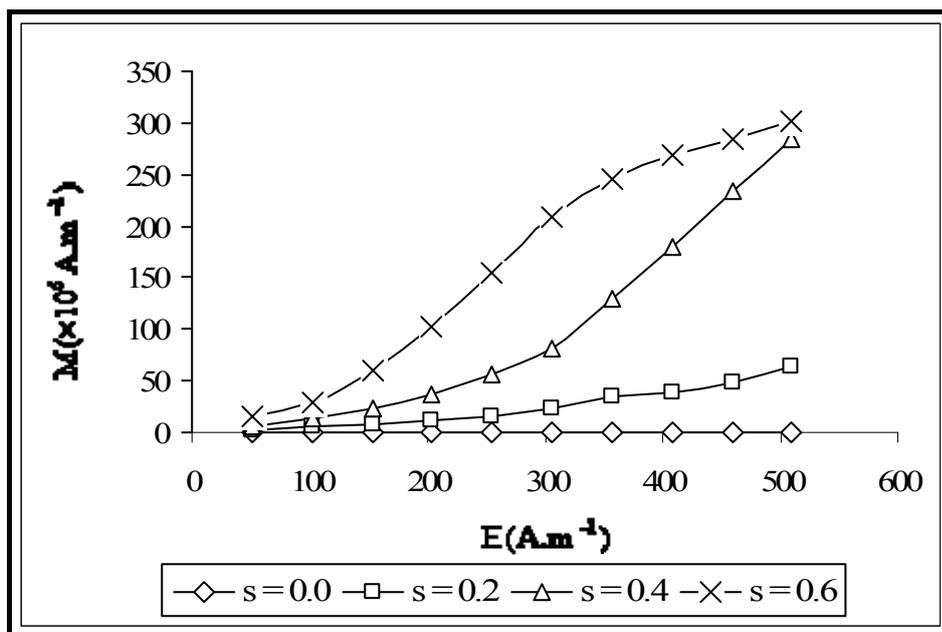


Figure (1): Change of M with H for the samples with $s = 0.0, 0.2, 0.4$ and 0.6 .

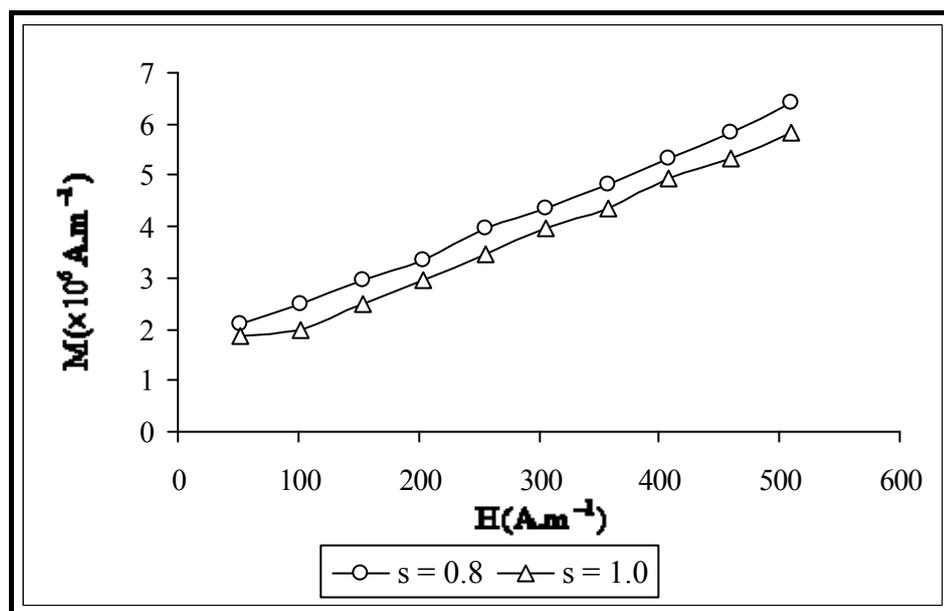


Figure (2): Change of M with H for the samples with $s = 0.8$ and 1.0 .

The saturation of magnetization ⁽⁸⁾ of mixed Cu-Zn spinel ferrites at the absolute zero point would be expected to increase with increase of the Zn^{2+} ions. Thus, the remarkable fact appears that, the substitution of the magnetic ions in a ferrimagnetic materials by the non-magnetic ions such as Zn^{2+} can lead to an increase in the saturation of magnetization ⁽¹⁾.

Fig. (3) shows the variation of M with the Zn content for different fields. From this figure, it is noticed that, the net magnetization M increases with the addition of the Zn^{2+} ions. This behavior was for all samples with $s \leq 0.6$, while the samples with $s > 0.6$ showed a very low net magnetization. The present results agreed well with the results obtained earlier by other groups ⁽⁸⁻¹⁴⁾.

Increasing the net magnetization when the Zn^{2+} ions increased can be explained by Néel's two-sublattice model of the magnetism theory of the ferrimagnetic materials ⁽¹⁵⁻¹⁶⁾, in this model Néel considered that a

ferrimagnetic crystal lattice could be divided into two magnetic sublattices or groups, i.e. T_d and O_h sites, in the spinel lattice structure, where their magnetic moments are not equal, as shown in the figure (4), so that, a net magnetic moment is found. This happens either because they are made from elements in different ionic state, e.g. Fe^{2+} and Fe^{3+} , or from different elements in the same or different ionic states, e.g. Cu^{2+} and Fe^{3+} ⁽¹⁷⁾. Néel's type of spin arrangements is favored up to 50% of zinc ions existing in the samples ⁽¹⁸⁻²⁰⁾.

The drop in the net magnetization for samples with $s > 0.6$ is due to the occurrence of the non-collinear spin structure, as a result, some sort of triangular, antiferromagnetic, or other spin arrangements occur within the O_h sublattice and hence decreasing the net moment. This could not be explained on the basis of Néel's two-sublattice model, but by the three-sublattices model that was suggested by Yafet and Kittel ⁽¹⁸⁾. According to this model

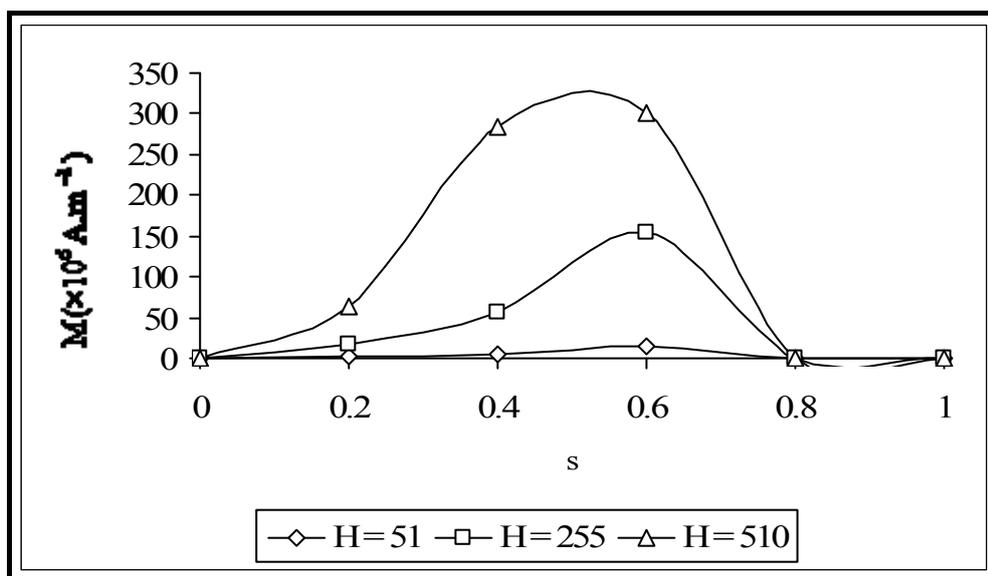


Figure (3): Variation of M with the Zn composition, s , for different values of $H(A.m^{-1})$

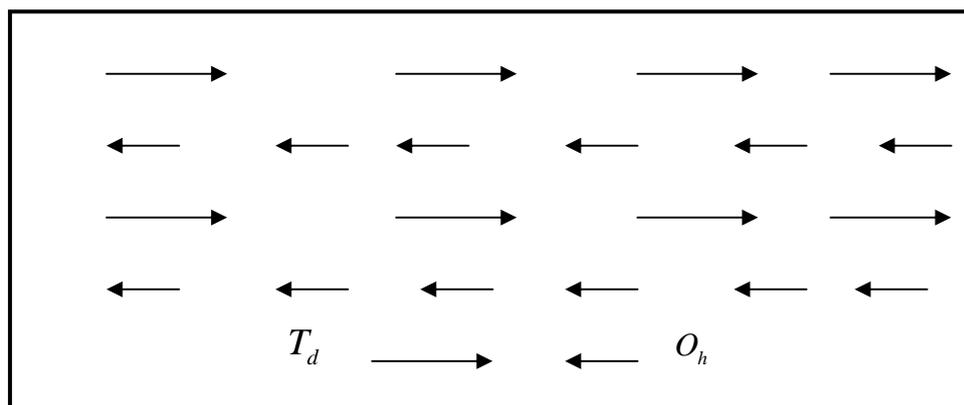


Figure (4): Distribution of magnetic moments at the T_d sites and the O_h sites.

the spin arrangements are the triangular arrangement and can be developed by supposing that, O_h sublattices are subdivided magnetizations M_{O_1} and M_{O_2} respectively. As shown in figure (5), M_{O_1} and M_{O_2} canted at an angle θ that changes from 0° to 180° with respect to M_T . The magnitude of the angle θ is given by:

$$\theta = \cos^{-1}\left(-\frac{N_{TO}}{2N_{OO}}\right)$$

where

M_{O_1} is the intensity of the intrinsic magnetization within the O_1 sites. M_{O_2} is the intensity of the intrinsic magnetization within the O_2 sites. M_T is the intensity of the intrinsic magnetization within the T_d sites. N_{TO} is the molecular magnetization field constant for the next nearest neighbor interactions between the T_d sites and the O_h sites, but N_{OO} is the molecular magnetization field constant for the next neighbor interactions within the O_h sites. Certainly the above equation provided that, $N_{TO} < 2N_{OO}$, however, when $N_{TO} > 2N_{OO}$ the result is that, the angle

θ will be very small. Then T_d and O_h sublattices magnetizations are antiparallel tend to make the net magnetization to be weaker.

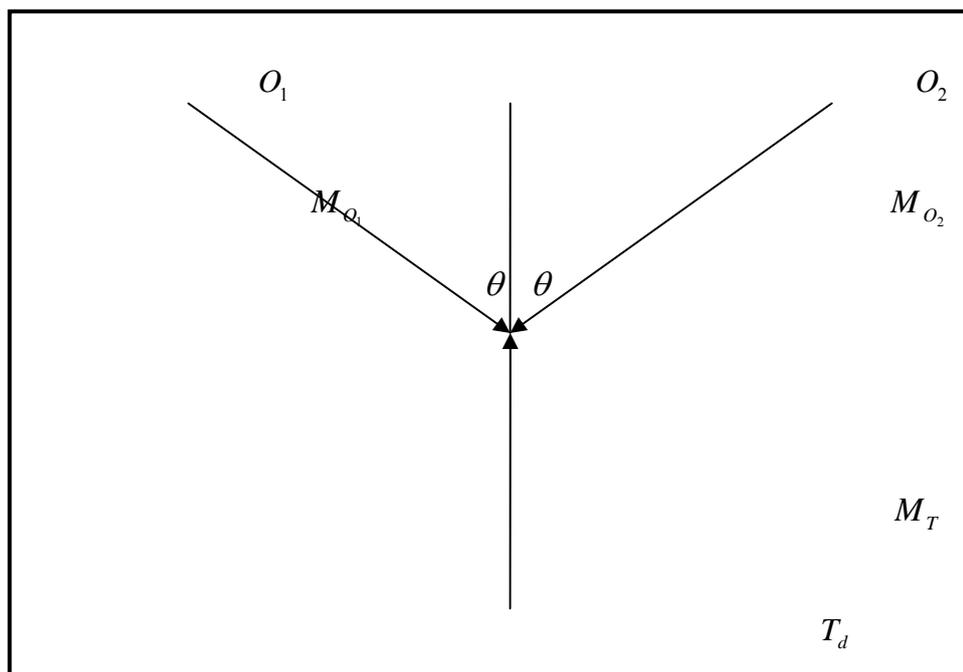


Figure (5): Triangular spin arrangements.

Owing to the presence of the non-magnetic Zn^{2+} ions at the T_d sites, the net magnetization of the T_d lattice will be smaller than in the simple ferrite, where, the Fe^{3+} ions have the largest magnetic moment that positioned at the O_h sites. When the Zn^{2+} ions increase the cations distribution is not altered, but the interaction between the T_d and the O_h sublattices will be weak. The interaction between O_h and O_h sublattices undergo a change in its tendency from ferrimagnetic state to antiferromagnetic state⁽¹⁹⁻²⁰⁾. This behavior is related to the increase of the non-magnetic ions (Zn^{2+}) in the ferrite compositions⁽²¹⁻²³⁾ that the

Zn spinel ferrite in the bulk form gives a normal spinel ferrite with the Zn^{2+} ions at the T_d sites. Also the Fe^{3+} ions at the O_h sites behave antiferrimagnetic below 9.5K and paramagnetic above this temperature. Thus, the change of the net magnetic moment μ_{net} with the Zn^{2+} ions will then correspond to the triangular of spins on the O_h sites. Similar behavior is also confirmed both experimentally and theoretically for the mixed Cu-Zn spinle ferrite by ^(10,19).

The net magnetization for the ferrimagnetic materials cannot be observed and defined by the μ_{net} per unit volume for each sample in **Bohr** magnetons unit μ_B . The μ_{net} in **Bohr** magnetons could be calculated according to the suggested cations distribution as follows (19).



According to the above form, the magnetic moment for the T_d sites, μ_T , and the O_h sites, μ_O , were calculated using the following equations (17,24)

$$\mu_T = 2(1-s)S_m\mu_B \tag{1}$$

$$\mu_O = [(1-s)S'_m + (1+s)S_m]2\mu_B \tag{2}$$

with $S_m = 5/2$ and $S'_m = 1/2$ are the spin quantum number of the Fe^{3+} ions and the Cu^{2+} ions, respectively, where the Zn^{2+} ions are diamagnetic ions which have a zero spin quantum number.

Therefore the net magnetic moment μ_{net} is given by ^(17,24)

$$|\bar{\mu}_{net}| = |\bar{\mu}_O| - |\bar{\mu}_T| \tag{3}$$

The calculated values of μ_T , μ_O and μ_{net} are listed in table (1) and their variation with the composition s is depicted in figure (6). The

variation of μ_{net} with the composition s could be explained by assuming that, when s increases the relative number of the Fe^{3+} ions are increased on the O_h sites, and shows a decrease on the T_d sites. This tends to increase μ_O , however, μ_T will be decreased. Therefore, the μ_{net} should rise linearly with the Zn^{2+} ions up to a value of $10\mu_B$ per molecule for $s=1.0$, in this sample all the divalent magnetic ions are replaced by the Zn^{2+} ions. Table (1) illustrates that, as the composition s is increased the μ_T will be reduced by 100%, while μ_O and the μ_{net} is increased by 66.67 % and 90 %, respectively.

Table (1): Calculated values of μ_T , μ_O and μ_{net} according to the cations distribution of the mixed *Cu-Zn* spinel ferrite.

s	$(\mu_O)\mu_B$	$(\mu_T)\mu_B$	$(\mu_{net})\mu_B$
0.0	6	5	1
0.2	6.8	4	2.8
0.4	7.6	3	4.6
0.6	8.4	2	6.4
0.8	9.2	1	8.2
1.0	10	0	10

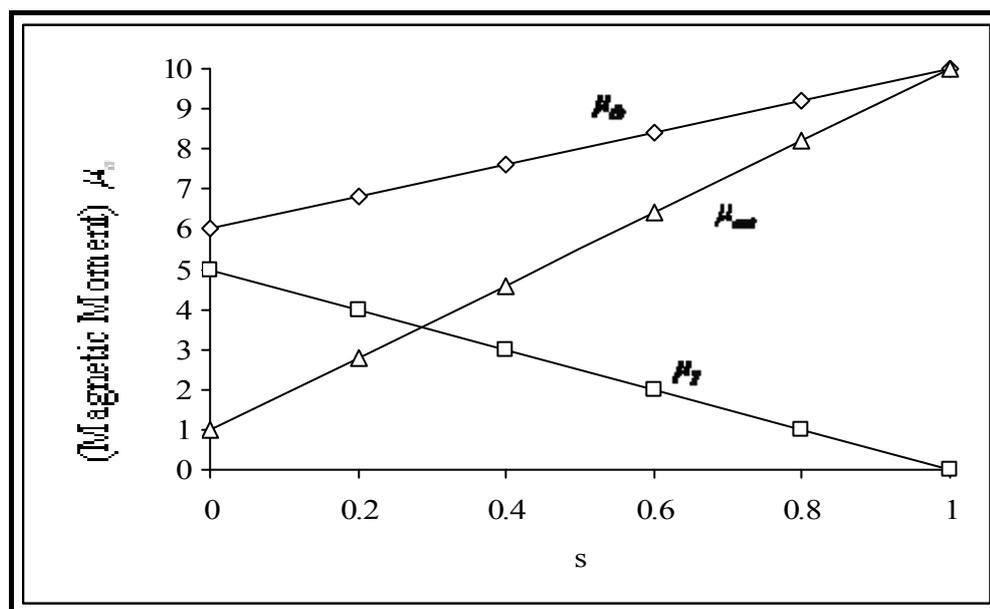


Figure (6): Variation of μ_T , μ_O and μ_{net} with compositions.

3.2 Relative Permeability

The relative permeability μ_r for all samples gives the description of the magnetization behavior during the change of an external applied magnetic field H . The ratio between the induction field B and the intensity of the applied magnetic field H , is defined as $\mu_r = B / \mu_o H$. The relation between H and μ_r for the present ferrite samples is illustrated in figures (7 and 8). Figure (7) indicates that for the samples with $s \leq 0.6$, the μ_r increased with increase of the H and the Zn^{2+} ions. The increment of μ_r could be related to the alignment effect of H on the ionic spins. However, increasing H causes a rapid increase of B , which in turn a pronounced increase of μ_r will take place.

However, for a sample with $s = 0.6$, a significant hump was found at $H = 350(A.m^{-1})$ and then decreases for an excess of magnetization. This

behavior of decrement is due to the further increase of H that causes a slight increase of B giving rise to distinct decrease in μ_r . This behavior is also observed for samples with $s > 0.6$ as shown in figure (8), where the samples have very low magnetization. Similar behavior was also found for Li – Cu and Cu – Ti samples ^(8,25).

4. Conclusions

The prepared samples were used to investigate the magnetization at room temperature for the applied magnetic field in the range (0 - 510 $A.m^{-1}$).

It is noticed that as the Zn^{2+} ions added for the samples with the composition $s \leq 0.6$ the magnetization will be increased. This increment of the magnetization was explained on the basis of Néel's two-sublattices-model, whereas for samples with $s > 0.6$ the magnetization has low values in that range. This is due to the presence of a triangular spin arrangements on the O_h sites according to the three – sublatic – model which was suggested by Yafet and Kittel.

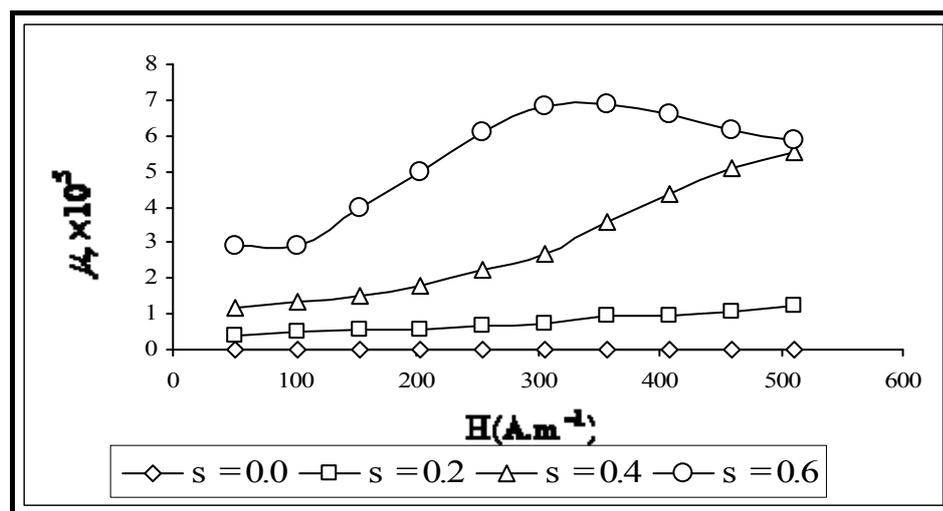


Figure (7): Variation of μ_r with H for the samples with $s = 0.0, 0.2, 0.4$ and 0.6 .

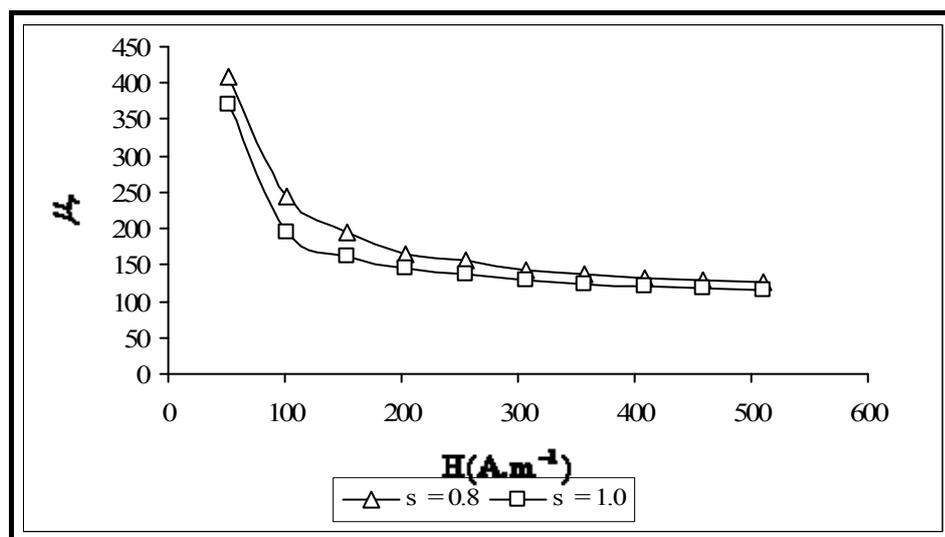


Figure (8): Variation of μ_r with H for the samples with $s = 0.8$ and 1.0 .

In general, the present work would give a basic understanding for the magnetic properties of the selected samples. we would also suggest to perform further research in order to study the A.C. conductivity, initial permeability, dielectric properties and different parameters for samples of the present work. Certainly, these parameters of the samples are basically vital important for an electronic industrial production point views.

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