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Preparation of Ni-Ferrite Samples for Absorbing Spectrum of X-Band Waves Using Network Analyzer Method

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Abstract

In this work, the spinal ferrite $NiFe_2O_4$ has been prepared. The prepared ferrite material was used as radar absorbing material (RAM). The absorption for microwaves was investigated via the use of network analyzer method. The values of reflectivity and absorption were calculated as a function of frequency at X -band with an $(0.0225)GHz$ increase. The relationship between reflectivity and absorption was plotted as a function of frequency for all prepared samples and their sintering temperature: (1050, 1100, 1150) ^0C. The results were appearance of a numbers of resonance peaks at X -band frequencies. Further, it was shown that the highest values of the absorption and less values of the reflectivity were reached at the sintering temperature 1150 ^0C. Also, testing of $X-ray$ diffraction (XRD) was made for this ferrite, where the results showed compatibility with standard results except for secondary phases with the ferrite samples.

تحضير نماذج من فيرايت النيكل لامتصاص طيف من موجات النطاق السيني باستعمال طريقة المحلل الشبكي

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الخلاصة

تم في هذا البحث تحضير الفيرايت ذو تركيب السبينل $NiFe_2O_4$. استخدمت هذه المادة المحضرة بوصفها مادة ماصة للأشعة الرادارية. تم دراسة امتصاصية الموجات الدقيقة باستخدام طريقة المحلل الشبكي، وحسبت قيم الانعكاسية والامتصاصية دالة للتردد ضمن النطاق السيني $X-band$ بزيادة قدرها (0.0225) GHz، ورسمت العلاقة بين الانعكاسية والامتصاصية دالة للتردد لجميع العينات المحضرة وعددها (3) عينة وملبدة بثلاث درجات حرارية (1150,1100,1050) ^0C . بينت النتائج ظهور عدة قمم رنينية عند ترددات النطاق السيني و أظهرت النتائج ان اعلى قيم للامتصاصية واقل قيم للانعكاسية هي درجة حرارة التلبيد 1150 ^0C. كما أجريت فحوصات حيود الأشعة السينية لهذا الفيرايت النيكل، حيث أظهرت النتائج العملية التي توصلنا اليها تطابقها مع النتائج القياسية مع وجود أطوار ثانوية مع عينات الفيرايت.

1. Introduction

Ferrites, generally, have chemical formula $M^{2+}Fe_2^{3+}O_4^{-2}$ or $MO.Fe_2O_3$ where M is a divalent metal ion and the crystal structure is that possessed by the mineral spinel, $MgAl_2O_4$ [1]. The unit cell contains eight "molecules" and may thus be written as $8(MFe_2O_4)$ or $M_8Fe_{16}O_{32}$. The oxygen anions are physically the greatest and they form, ideally, a face-centered cubic lattice. Within this lattice, two types of interstitial

positions occur and these are occupied by the metallic cations. There are 96 interstitial sites in the unit cell, 64 tetrahedral and 32 octahedral. The divalent metal M, occurring in the formula MFe_2O_4 in ferrimagnetic ferrite is commonly Mn, Fe, Co, Ni, Cu, Zn or Mg .In a simple ferrite, one type of ion only occurs as M, thus $NiFe_2O_4$. However, solid solutions of two or more simple ferrites are called mixed ferrite. Thus, $Ni_{(1-x)}Zn_xFe_2O_4$ represents a mixed ferrite with (x taking values between 0 and 1). According to cations distribution there are two types of spinel ferrits [2].

There are three types of the spinel ferrite according to the metallic ion position: 1) normal spinel ferrite: for Zn and Cd ferrites, in a normal spinel structure, the A ions occupy tetrahedral sites and the B ions occupy octahedral sites, 2) inverse spinel ferrite: most of the simple ferrites, like $NiFe_2O_4$, the B ions occupy all of the tetrahedral sites and the octahedral sites are filled by half the B ions and the A ions, 3) random spinel ferrite: which is intermediate case between both type A and B, such as $Ni-Zn$ ferrite and $Mn-Zn$ ferrite [3,4].

2. Materials

In this work, one compound of spinel ferrite, which is: Ni-ferrite, with formula $NiFe_2O_4$, as bulk sample with one thickness, were prepared using the conventional ceramic method "the solid state reaction method" [5,6]. It is very important to choose the raw materials with very high purity, to avoid any influence on the compound properties. The weight of the used raw materials are accurately calculated from its atomic weights. The raw materials made in England and Germany. For instance, to

prepare one mole of $NiFe_2O_4$ we calculated the amount of its raw materials as shown [7]:

$$Fe_2O_3 = 2\times 55.845 + 3\times 16 = 159.69 \quad g$$
$$NiCO_3 = 58.693 + 12 + (3\times 16) = 118.6934 \quad g$$
$$total\,(\mathrm{NiFe_2O_4}) = 118.6934 + 159.699 = 278.3834 \quad g$$

3. The Network Analyzer

The complex permittivity and complex permeability can be defined as [6]:

$$\varepsilon = \varepsilon' - j\varepsilon'' \qquad (1)$$
$$\mu = \mu' - j\mu'' \qquad (2)$$

where ε', ε'' are the real and imaginary parts of permittivity, and μ', μ'' are the real and imaginary parts permeability. The permittivity and permeability of medium may be written as

$$\varepsilon = \varepsilon_0 \varepsilon_r \qquad (3)$$
$$\mu = \mu_0 \mu_r \qquad (4)$$

where ε_r and μ_r are the relative permittivity and the relative permeability, respectively, the values of ε_r and μ_r changes with temperature and frequencies. Here, ε_0 and μ_0 are the permittivity and permeability in free space [5].

Network analyzers are widely used to measure the four elements in a scattering matrix: S_{11}, S_{12}, S_{21}, and S_{22}. A network analyzer mainly consists of a source, signal separation devices, and detectors. Basically, a network analyzer can measure the four waves independently: two forward traveling waves a_1 and a_2; and two reverse traveling waves b_1 and b_2 [8,9]. The responses of a network to external circuits can also be described by the input and output microwave waves. The input waves at

port 1 and port 2 are denoted as a_1 and a_2, respectively, and the output waves from port 1 and port 2 are denoted as b_1 and b_2, respectively. These parameters (a_1, a_2, b_1, and b_2) may be voltage or current, and in most cases, we do not distinguish whether they are voltage or current. The relationships between the input wave (a) and output wave (b) are often described by scattering parameters (S) [10]. Scattering parameters or scattering coefficients, in short it is abbreviated as S-parameters. These S-parameters are complex numbers and generally related to familiar measurements such as gain, loss, reflection/transmission coefficient, and impedance/ admittance. The number of S-parameters for a given device is equal to the square of the number of ports. For example, a two-port device has four S-parameters. The numbering convention for S-parameters is that the first number following the S is the port at which energy emerges, and the second number is the port at which energy enters [9]. The S-parameters are defined by the following equations [8,10]

$$b_1 = S_{11}a_1 + S_{12}a_2$$
$$b_2 = S_{21}a_1 + S_{22}a_2 \qquad (5)$$

Here, the travelling wave variables a_1, b_1 at port 1 and a_2, b_2 at port 2 of the two-port network are defined in terms of total voltage and current (U_1, I_1 and U_2, I_2) and reference impedance Z_0 as follows [9]:

$$a_1 = \frac{U_1 + I_1 Z_0}{2\sqrt{Z_0}}\ ,\ a_2 = \frac{U_2 + I_2 Z_0}{2\sqrt{Z_0}}\ ,\ b_1 = \frac{U_1 - I_1 Z_0}{2\sqrt{Z_0}}\ ,\ b_2 = \frac{U_2 - I_2 Z_0}{2\sqrt{Z_0}} \qquad (6)$$

There are various approaches for obtaining the permittivity and permeability from S-parameters. Each of the conversion method has different advantages and limitations. The selection of the method depends on several factors such as the measured S-parameters, sample length, the desired dielectric properties, speed of conversion and accuracies in the

converted results. In the work, we will focus on the Nicholson-Ross-Weir (*NRW*). Nicolson and Ross and Weir combined S_{11} and S_{21} and derived explicit formulas for the calculation of permittivity and permeability. The algorithm is usually called *NRW* algorithm [8] that will be used to calculate the results. This method provides a direct calculation of both the permittivity and permeability from the S-parameters. It is the most commonly used method for performing such conversion. Measurement of reflection coefficient and transmission coefficient requires all four ($S_{11}, S_{21}, S_{12}, S_{11}$) or a pair ($S_{11}$, S_{21}) of S-parameters of the material under test to be measured. The procedure proposed by *NRW* is deduced from the following equations [11]:

$$S_{11} = \frac{\Gamma\left(1 - T^2\right)}{\left(1 - \Gamma^2 T^2\right)}, \qquad S_{21} = \frac{T\left(1 - \Gamma^2\right)}{\left(1 - \Gamma^2 T^2\right)} \tag{7}$$

where the T is transmission coefficient in a two port network and Γ is reflection coefficient.

These parameters can be obtained directly from the network analyzer. In the *NRW* algorithm, the reflection and transmission are expressed by the scattering parameters S_{11} and S_{21}. The reflection coefficient is given by [12]

$$\Gamma = X \pm \sqrt{X^2 - 1} \tag{8}$$

The root of the above equation can be found out from the modulus of Γ (< 1) in terms of S-parameter [8,11]

$$X = \frac{S_{11}^2 - S_{21}^2 + 1}{2S_{11}} \tag{9}$$

and also [8]

$$T = \frac{S_{11} + S_{21} - \Gamma}{1 - (S_{11} + S_{21})\Gamma} \tag{10}$$

The permeability and permittivity are defined as [13,14]

$$\mu_r = \frac{1+\Gamma}{\Lambda(1-\Gamma)\sqrt{\frac{1}{\lambda_0^2}-\frac{1}{\lambda_c^2}}} \tag{11}$$

$$\varepsilon_r = \frac{\lambda_0^2}{\mu_r}\left(\frac{1}{\lambda_c^2}-\left(\frac{1}{2\pi l}\ln\left(\frac{1}{T}\right)\right)^2\right) \tag{12}$$

where λ_o is free space wavelength and λ_c is the cut off wavelength of the guide and Λ is equal to [11]

$$\frac{1}{\Lambda^2} = -\left(\frac{1}{2\pi l}\ln\left(\frac{1}{T}\right)\right)^2 \tag{13}$$

Here l is the thickness of the sample. Reflection loss from the coating layer is given by [15]

$$R(dB) = -20\log_{10}\left|\frac{Z_{in}-1}{Z_{in}+1}\right| \tag{14}$$

where

$$Z_{in} = \sqrt{\frac{\mu_r}{\varepsilon_r}}\tanh\left(-i\frac{2\pi f\,\ell}{c\sqrt{\varepsilon_r\mu_r}}\right)$$

and f is microwave frequency. After calculating the reflection coefficient in Eq.(8), it can be used to obtain absorbance [7] as follows**:**

$$\Gamma^2 + A^2 = 1 \tag{15}$$

where A is the absorption coefficient.

4. Results and Discussion

All samples have been tested at the range of X-band (8-12.5) GHz due to its importance in most application of industry. The parameter are estimated using Eqs.(8) to (15). In Fig.(1), the first row represents μ, where the continuous line represents the real part and the discrete represents the imaginary part. The second row represents ε, where the

continuous represents the real part and the discrete represents the imaginary part. The third row represents the Γ^2 and A^2, where the continuous line represents the Γ^2 and the discrete represents the A^2. In all these figures the columns represent the temperatures (1050, 1100, 1150) ^{0}C listed. It is noticed from Fig.(1) that the increases in the value of the A^2 and decreases in the values of Γ^2, this means that the best desired results when the values of Γ^2 are at the minimum value, and all the values of parameters in graphs depended upon the values of sintering temperature of the samples. The figure shows that there are resonance peaks for the all ferrites samples, these peaks are formed when there is matching between the ε_r and μ_r of ferrite, and due to domain rotation, and domain wall movement for ferrites samples. The ferrite materials have a high relative magnetic permeability values and dielectric constant in low frequencies, and these values decrease in high frequencies for microwave. The best values for the absorption is at 1150C^0 and 1100C^0 and then 1050, due to the completion of ferrite in this class, as well as increased density which reduces the porosity. The highest values of the absorption at 1100°C are (0.66,0.75,0.6,0.80.7,0.99) at frequencies (8.4,8.6, 8.8,9.7,10.2,11.5) GHz respectively. and the corresponding respectively values at 1100°C are (0.09,0.26,0.175,0.26,0.173,0.26) at frequencies (8.4,8.6, 8.8,9.7, 10.2,11.5) GHz respectively, and the range of absorption (8-10.7), and the highest values of the absorbance at 1150°C are (0.75,0.9,0.98, 0.66,0.7) at frequencies (8.3,8.8,9.3,10.1,10.8) GHz respectively, and the corresponding respectively values at 1150°C are (0.173,0.1,0.095,0.3,0.2) at frequencies (8.3, 8.8,9.3,10.1,10.8) GHz respectively, and the range of absorption (8.8-11.4).

Thus it can be concluded the following: the appearance resonance peaks of relative magnetic permeability and dielectric constant for the real and imaginary parts in the same frequency which is appeared in the measured reflectivity and absorbance for all types of the prepared ferrite samples, this is results because of depend the measures of Γ^2, A^2, ε_r and μ_r on the reading S_{11} and S_{21}. The real part of ε_r, μ_r for all prepared samples is bigger than its imaginary part, this means that the samples have a high A^2, thus the value of A^2 direct proportion to the resistivity and inversely with the imaginary part of the dielectric constant and μ_r. The real parts of ε_r, μ_r indicate the possibility of penetrating the microwave surface of ferrite, while the imaginary parts indicate has the capacity to A^2 ferrite for these waves.

The density measurement of the prepared samples has been measured for some compounds after a process of sintering in which the completely dry samples have been weighted, and the size of the sample which is measured by using micrometer. The density of the sample ρ is given by $\rho = m/V$ where the mass m is measured by the unit of gram and V is measured by unit of cm^3 [12]. To investigate the crystal structure of the prepared samples after sintering, phase analysis was done by XRD using $Cu-K_\alpha$ radiation, and wavelength λ=1.54060 A^0; the range of the Braggs angles 2θ are taken as (20^0-60^0) for the samples, the type of this devise is (XRD -6000) and made in Japan by Shimadzu. The interplaner distance d can be calculated using Braggs law $2dSin\theta = n\lambda$ [13]. We are calculated the density of each sample.

Figs.(2) to (4) show the *XRD* pattern of $NiFe_2O_4$, and then comparing the resultant XRD patterens with international standerd,

international center for diffraction data (ICDL) which is the ASTM, as shown in Table (1). After XRD tests can be calculate the grain size ($G.S$) from Debye-Scherrer formula [14,15] $G.S = K\lambda/\beta\cos\theta$, where K is a constant taken the range 0.89 ~ 0.9, λ is the wavelength of the XRD, β is the full width at half maximum ($FWHM$). The XRD results shown that demonstrate the completion of the $NiFe_2O_4$ phase of the spinel structure at this temperature, obviously it is a polycrystalline. We are calculated the grain size for Ni-ferrite samples. Fig.(5) explains the density as a function of sintering temperature for all ferrites. It is clear that the increasing of sintering temperature will increase the density, note that, the increase of density will reduce the porosity. Fig.(6) explains the grain size as a function of sintering temperature for Ni-ferrite samples. It is clear that the increasing of sintering temperature will increase the grain size.

4. Conclusions

The sintering temperature has an important influence in forming the ferrite materials and the absorption ability. It has an effect on amount of density, grain size and the number of secondary phases. The best value of sintering temperatures is at 1150 ^{0}C for spinel ferrite this indicates that spinel ferrite needs to sintering temperature of more than 1100 ^{0}C for the complete formation of ferrite and getting best absorbance because of high sintering temperature cancels all secondary phases that made up with ferrite. The density of samples and grain size and porosity greatly depends on the sintering temperature, where increases sintering temperature led to increases density and grain size and decreases porosity. The factors that lead to high value of absorption for ferrite materials is the relative magnetic permeability and dielectric constant for

these material, specially the imaginary parts that cause material absorption for microwave, where the absorption as caused by the domain rotation, or domain magnetic wall movement, with noticing of incomplete hysterics magnetic loop in microwave frequencies. From the results of the structure testing by of the prepared samples one can find that the structure is polycrystalline for all samples, and the phase has been completed at sintering temperature 1150 ^{0}C .

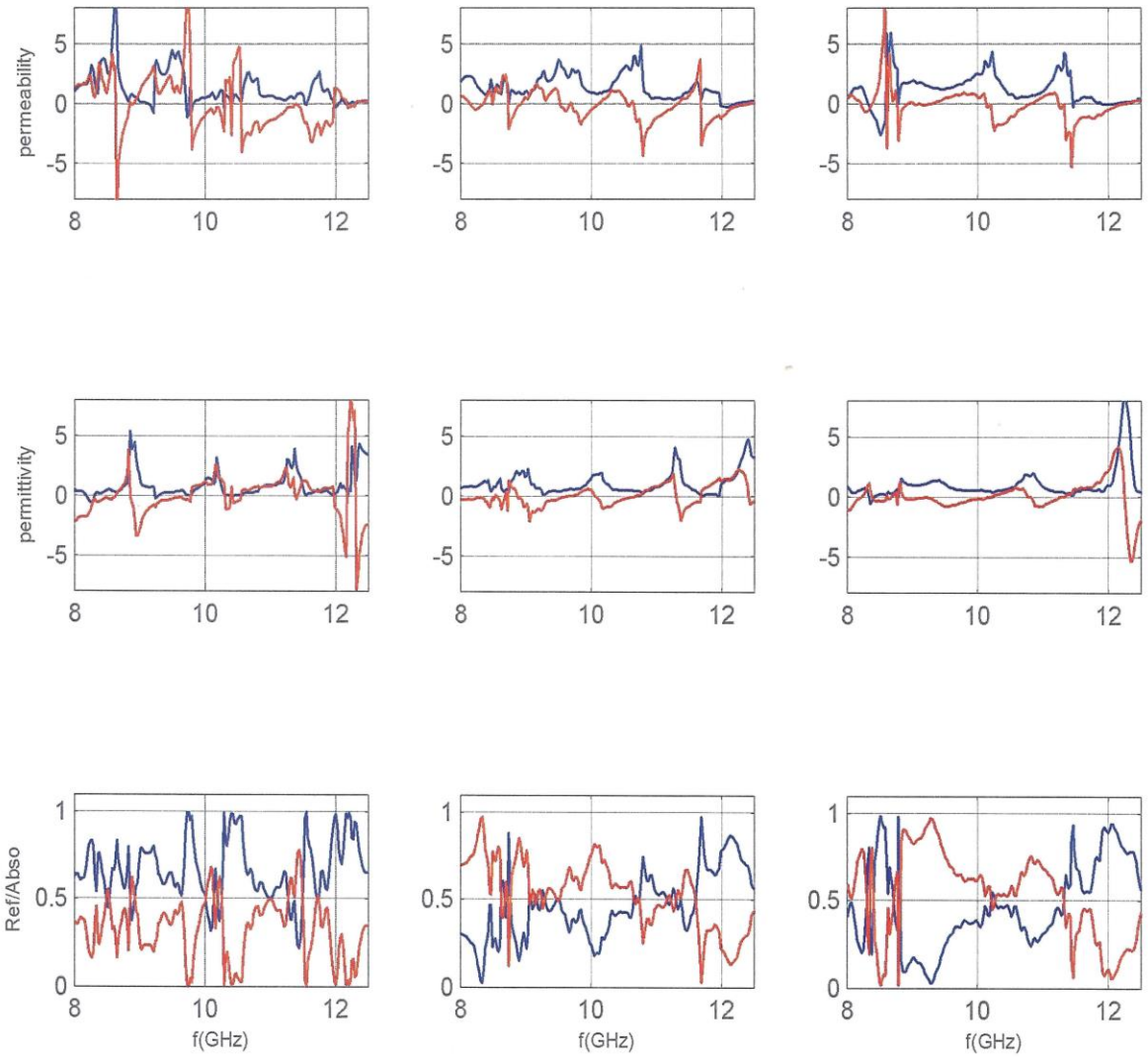

Fig.(1): The actual results for *Ni* -ferrites.

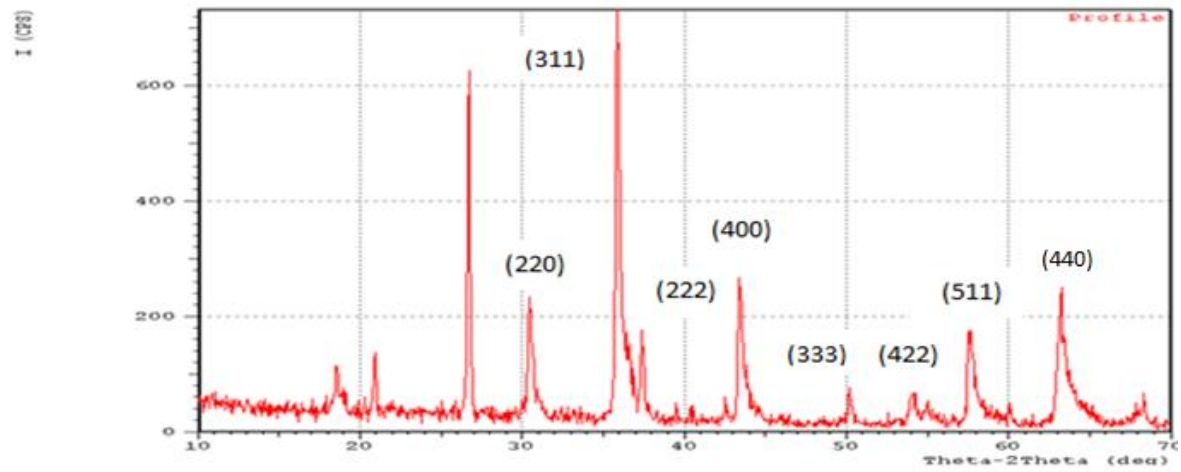

Fig.(2): *XRD* pattern of $NiFe_2O_4$ at T=1050 ^{0}C**.**

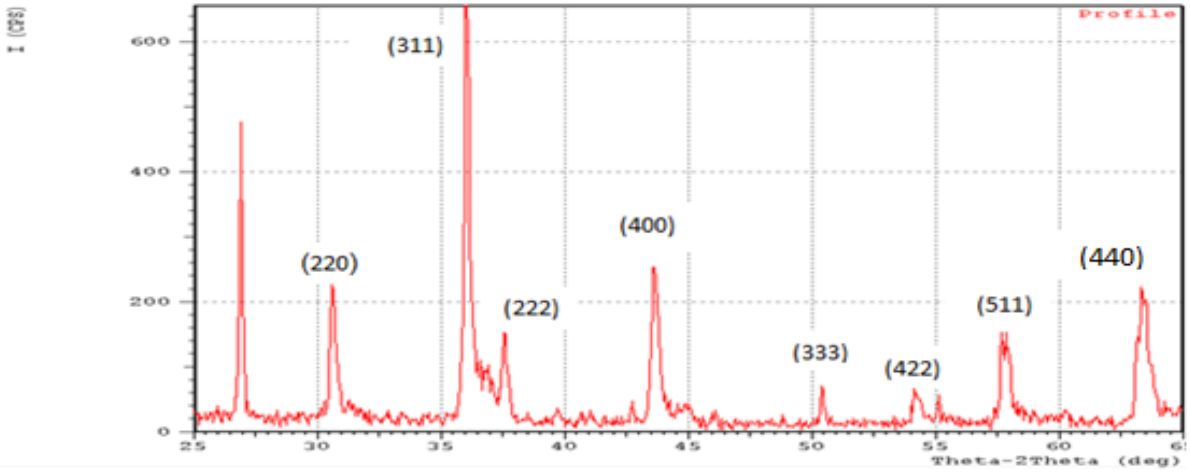

Fig.(3): *XRD* pattern of $NiFe_2O_4$ at T=1100 ^{0}C**.**

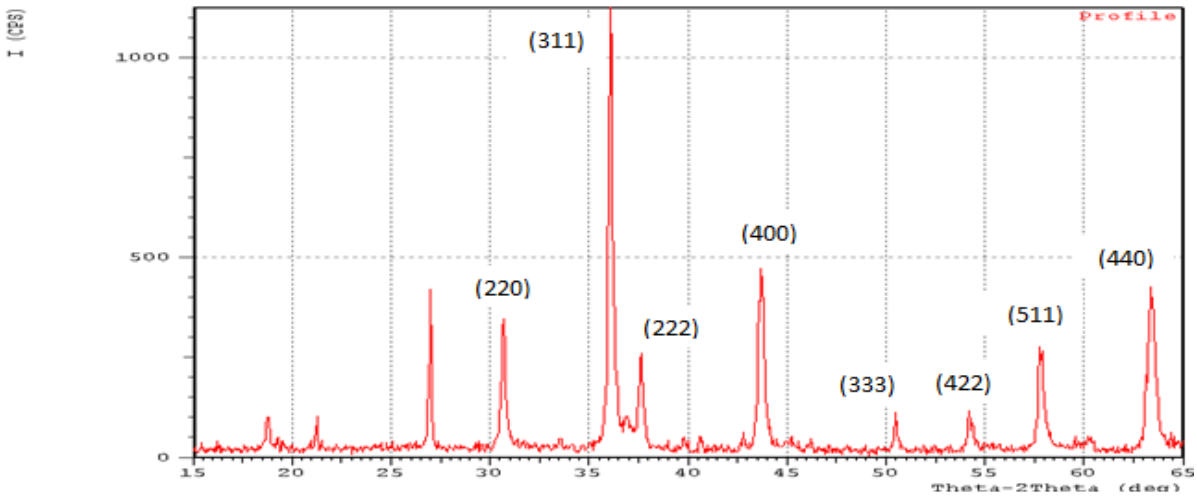

Fig.(4): *XRD* pattern of $NiFe_2O_4$ at T=1150 ^{0}C**.**

Table (1): the interplaner distances *d* and $2\theta^0$ of *XRD* pattern of $NiFe_2O_4$ comparing with the *ASTM* card.

2θº	d(Å) EXP.	d(Å) ASTM	hkl
30.665	2.9132	2.948	220
36.061	2.489	2.513	311
37.593	2.391	2.408	222
43.662	2.071	2.085	400
51.13	1.853	1.87	333
53.73	1.705	1.69	422
57.725	1.596	1.6051	511
62.756	1.483	1.478	440

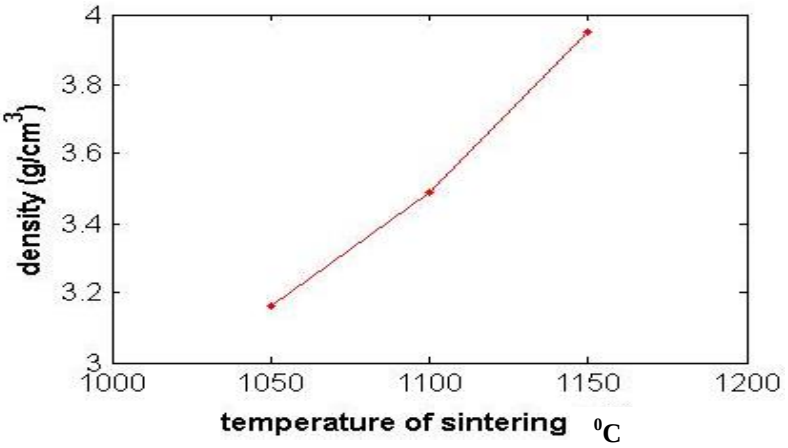

Fig.(5): the measured density of $NiFe_2O_4$

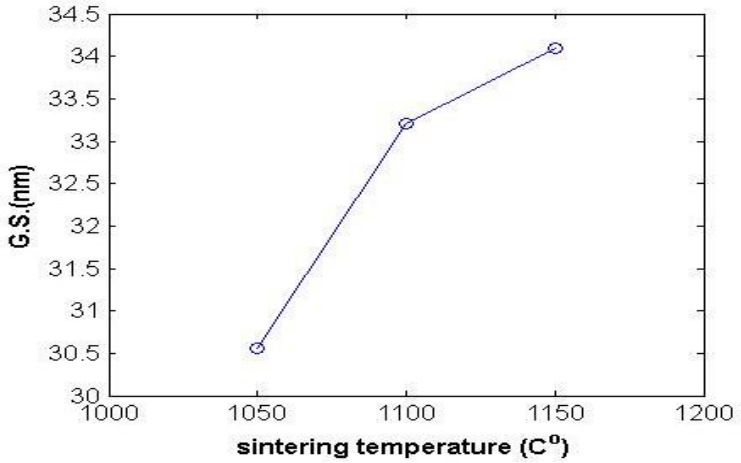

Fig.(6): the *G.S* of samples $NiFe_2O_4$ prepared**.**

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