

# Study of the Electric, Magnetic Properties of $Ca_{1-x}Cu_xFe_2O_4$ - Ferrite Samples as Radar Absorbing Materials Using Network Analyzer Method

Russell Thamer<sup>1</sup>, Hashim Ammar<sup>1</sup>, Hassan Yasser<sup>2</sup>

<sup>1</sup>Wasit University, College of Science, Department of Physics

<sup>2</sup>Thi-Qar University, College of Science, Department of Physics

**Abstract:** In this work, the spinel ferrite  $Ca_{1-x}Cu_xFe_2O_4$  has been prepared where  $X = (0, 0.2, 0.4)$ , used as radar absorbing materials (RAM). Conventional ceramic techniques method was used to prepare these materials at  $X$  - band microwaves with the range of frequencies (8-12) GHz. The absorption for microwaves were examined by the utilized the network analyzer. The relationship between reflectivity, absorption and attenuation coefficient were plotted as a function of frequency, and sintered at a temperature  $1100^\circ C$ . The results showed the emergence of several resonance peaks at  $X$  - band, samples showed high Absorbency in contrast low reflectivity and high values of attenuation coefficient at the same frequencies, this is due to domain rotation as well domain wall motion of the ferrite. The values of the real and imaginary parts of relative magnetic Permeability and relative electric permittivity were measure for all samples. The results revealed the apparent resonance peaks at same frequencies that apparent at absorption and reflectivity curves of the same samples. X-ray diffraction tests were made for one sample and result showed the structure of the sample is polycrystalline.

**Keywords:** Ferrite, RAM, X-ray diffraction, Network Analyzer

## 1. Introduction

Spinel 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 ferrites [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:  $Ca-Cu$ -ferrite, with formula  $Ca_{1-x}Cu_xFe_2O_4$ , as bulk sample, were prepared using the conventional ceramic 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 weights 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  $Ca_{0.8}Cu_{0.2}Fe_2O_3$  we calculated the amount of its raw materials as shown [7]:

$$Fe_2O_3 = 2 \times 55.845 + 3 \times 16 = 159.69g$$

$$CaO = 40.078 + 16 = 56.087g$$

$$CuO = 63.546 + 16 = 79.546g$$

$$total\ Ca_{0.8}Cu_{0.2}Fe_2O_4 = 220.4688g$$

## 3. The Network Analyzer

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

$$\epsilon = \epsilon' - j\epsilon'' \quad (1)$$

$$\mu = \mu' - j\mu'' \quad (2)$$

Where  $\epsilon'$ ,  $\epsilon''$  are the real and imaginary parts of permittivity, and  $\mu'$ ,  $\mu''$  are the real and imaginary parts permeability. The permittivity and permeability of medium may be written as

$$\epsilon = \epsilon_0 \epsilon_r \quad (3)$$

$$\mu = \mu_0 \mu_r \quad (4)$$

Where  $\epsilon_r$  and  $\mu_r$  are the relative permittivity and the relative permeability, respectively, the values of  $\epsilon_r$  and  $\mu_r$  changes with temperature and frequencies. Here,  $\epsilon_0$  and  $\mu_0$  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]

$$\begin{aligned} b_1 &= S_{11}a_1 + S_{12}a_2 \\ b_2 &= S_{21}a_1 + S_{22}a_2 \end{aligned} \quad (5)$$

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]:

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

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  $S_{21}$  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]:

$$S_{11} = \frac{\Gamma(1-T^2)}{(1-\Gamma^2 T^2)}, \quad S_{21} = \frac{T(1-\Gamma^2)}{(1-\Gamma^2 T^2)} \quad (7)$$

Where the  $T$  is transmission coefficient in a two port network and  $\Gamma$  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]

$$\Gamma = X \pm \sqrt{X^2 - 1} \quad (8)$$

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

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

and also [8]

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

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

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

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

Where  $\lambda_0$  is free space wavelength and  $\lambda_c$  is the cut off wavelength of the guide and  $\Lambda$  is equal to [11]

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

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

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

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:

$$\Gamma^2 + A^2 = 1 \quad (15)$$

where  $A$  is the absorption coefficient

## 4. Results and Discussion

All samples were tested at the range of X-band (8-12) Because of its importance in industrial applications. The parameters were calculated using Eqs. (8) to (15). In Fig.(1), the first column represents, the second column represents where and the third column represents the attenuation coefficient measured by (dB), where the blue colour represents the real part and the red colour represents the imaginary part. Fig. (2), the first column represents and, the second column represents, where the blue colour represents the real part and the red colour represents the imaginary part. Fig.(3), the first column represents where the blue colour represents and the red colour represents, and the second column represents refractive index where the blue colour represents the real part and red colour represents the imaginary part. From fig. (1) noticed that the values of vary with frequency, as a result to that change values of  $\Gamma^2$ ,  $A^2$ ,  $\varepsilon_r$ , and  $\mu_r$ , Because of the absorption of ferrite of waves dependent on frequency. From fig. (2) noticed increases in the values of the on the other hand decreases in the values of, this means that the better desired results when the values of are at the maximum value. Fig. (2), the emergence of several resonance peaks for the ferrites samples, these peaks are composed when there is matching between  $\mu_r$  and  $\varepsilon_r$  of ferrite, and this is due to domain rotation and domain wall movement for ferrites. The ferrite materials have a high values relative permeability and permittivity in low frequencies, and in high frequencies for microwave these values decrease.

From fig.(3), see these samples have high values of the absorption and the corresponding low values of the reflectivity, when at 1100°C the highest values of the absorption are (0.99, 0.98, 0.98, 0.97, 0.96) at frequencies (8, 8.7, 9.5, 10.6, 12)GHz respectively and the corresponding respectively values of the attenuation coefficient (-25, -20, -18, -22, -15) at the same frequencies. When  $X = 0.4$  at 1100°C it achieved a very high absorbency over 0.95 where it appears in the form of an almost straight line it includes all

frequencies, In contrast, very few reflectivity and high attenuation coefficient at the same frequencies. When at 1100°C the highest values of the absorption are (0.98, 0.97, 0.99, 0.94, 0.97) at frequencies (8, 8.7, 9.7, 10.7, 11.5) GHz respectively and the corresponding respectively values of the attenuation coefficient (-22, -25, -18, -21, -24).

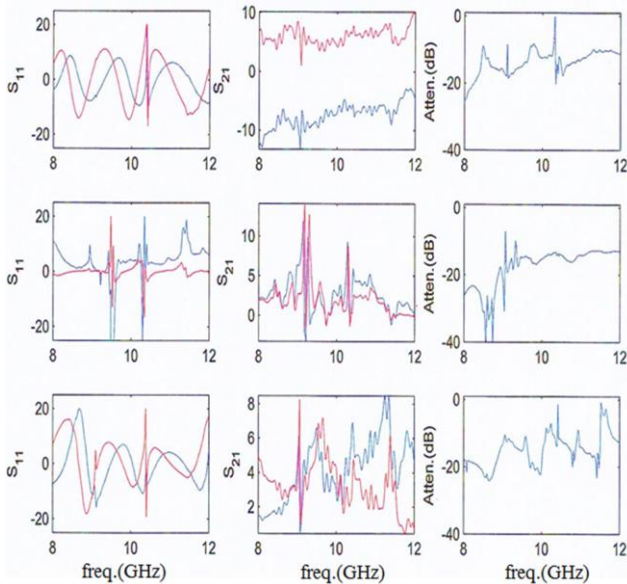
To examine the crystal structure of the prepared ferrite samples, phase analysis was done by X-ray diffraction (XRD) using devise (XRD -6000) made in Japan by Shimadzu, radiation was used, wavelength =1.54060 and the Braggs angles are taken the range as for the prepared samples. By using Braggs law can be calculated the interplaner distance [13].

Figs. (4) show the XRD pattern of, and after that compare the results of XRD patterns with ASTM card, as described in the table (1). The results shown that demonstrate the completion of the phase of the spinel structure at 1100°C, obviously it is a polycrystalline.

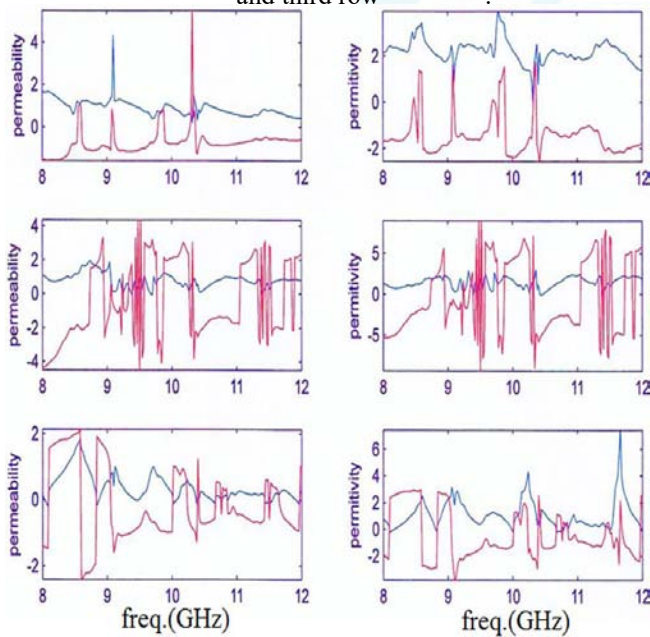
## 5. Conclusions

the emergence resonance peaks of relative permeability and relative permittivity for the real parts and imaginary parts in the same frequency which is emerged in the measured absorbance and reflectivity for all the prepared samples, this is results due to depend the measures of  $\Gamma^2$ ,  $A^2$ ,  $\varepsilon_r$  and  $\mu_r$  on the reading  $S_{11}$  and  $S_{21}$ , The real parts of  $\mu_r$ ,  $\varepsilon_r$  for all prepared samples are greater than its imaginary parts, that means that the samples have a high absorbance, thus the value of absorbance Inversely proportional to the imaginary parts of the dielectric constant and direct proportion with resistivity. The real parts of, indicate the possibility to penetrate the microwave ferrite surface, while the imaginary parts refer to has the capacity to absorbance ferrite for these waves.

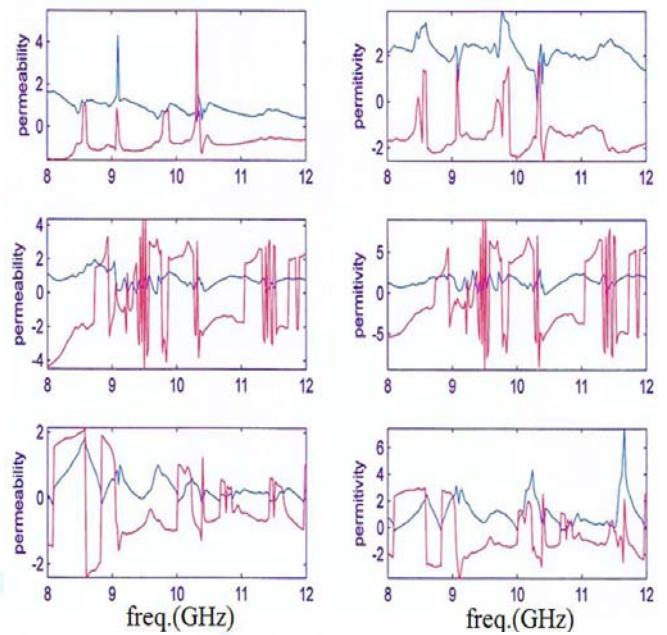
Factors causing the high values of absorption for ferrite are the relative magnetic permeability and permittivity for these material. The absorption due to the domain rotation and domain wall movement, with note of incomplete hysteresis magnetic loop in microwave frequencies. X-ray diffraction tests were made for one sample and result showed the structure of the sample is polycrystalline, and the phase of the sample has been completed at temperature 1100°C.



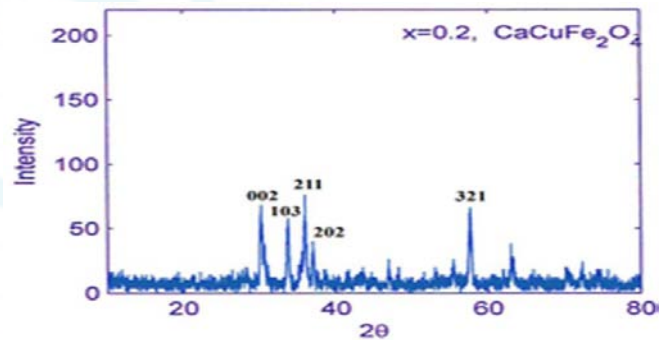
**Figure 1:**  $S_{11}$ ,  $S_{21}$  and Attenuation coefficient as a function of the frequency type of  $Ca_{1-x}Cu_xFe_2O_4$  where  $x = 0, 0.2, 0.4$  and the first row  $x = 0$ , second row  $x = 0.2$  and third row  $x = 0.4$ .



**Figure 2:**  $\mu_r$  and  $\epsilon_r$  as a function of the frequency type of  $Ca_{1-x}Cu_xFe_2O_4$  where  $X = 0, 0.2, 0.4$  and the first row  $X = 0$ , second row  $X = 0.2$  and third row  $X = 0.4$ .



**Figure 3:** The first column represents the absorbance and reflectivity as a function of the frequency and the second column represents the refractive index as a function of the frequency where  $X = 0, 0.2, 0.4$  and the first row  $X = 0$ , second row  $X = 0.2$  and third row  $X = 0.4$ .



**Figure 4:** XRD pattern of  $Ca_{0.8}Cu_{0.2}Fe_2O_4$  at  $T = 1100 C^0$

**Table 1:** The interplaner distances  $d$  and  $2\theta^0$  of XRD Pattern of  $Ca_{0.8}Cu_{0.2}Fe_2O_4$  comparing with the ASTM card.

$2\theta^\circ$	$d(A^\circ)$ ASTM	$d(A^\circ)$ EXP	$hkl$
30.5	2.45	2.928	002
33.8	2.704	2.649	103
35.8	2.502	2.506	211
37	2.419	2.247	202
47	1.92	1.931	112

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