

# THE EFFECT OF Mn AND Ti SUBSTITUTED BARIUM FERRITE ON THE ELECTROMAGNETIC MICROWAVE ABSORBER IN THE X-BAND RANGE



HAMID S.AL-JUMAILI\*, KHALID T. RASOUL\*\* AND SAEED N.AL-RASHED\*

\* Dept. of Phy., College of Science University of Anbar  
\*\*Science and Technology Ministry

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## ABSTRACT

The compositional dependence of the electromagnetic wave absorption properties of barium ferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) in which Ti and Mn were substituted for Fe, was investigated. Single phase M-type structure was obtained for  $\text{BaFe}_{12-x}(\text{Ti}_{0.5}\text{Mn}_{0.5})_x\text{O}_{19}$  sintered samples with composition  $0 \leq x \leq 4.5$ . The frequency dependence of reflection loss (R.L) obtained a minimum value of -21dB for the x-band frequency range (8-12GHz), and give a band width greater than 1GHz in the same frequency range. The grain size of the prepared powder were between (7.05-12.06  $\mu\text{m}$ ). Barium ferrite-resin composites with 30,40, and 50 mass% ferrite exhibited a wide band width with  $\text{R.L} < -21$  dB. The composite samples with composition  $x=3.5$  and  $x=4.5$  had wide bandwidth of 2.5 and 1.7 GHz respectively and a R.L of -12.5 and -16 dB respectively it may be concluded that Barium M-type ferrite with substitution by Ti and Mn is a good candidate in the x-band frequency range.

## INTRODUCTION

The Principle of an electromagnetic wave absorber is to utilize the reflection by impedance matching in which the normalized input impedance is calculated with respect to the impedance in free space - [1]. It has been well known that electromagnetic wave propagation in magnetized media can lead to nonreciprocal effect.

The biggest contribution of ferrites to the microwave technology is due to their gyro magnetic properties that enabled reciprocal devices such as the isolators and circulators [2,3]. Interest in barium ferrite as a magnetic material for microwave devices is due to its large the development of compact passive non anisotropy field at room temperature (-17 kOe), which can be readily varied by suitable cation substitutions [1,4-5]. Barium ferrites with a magnetoplumbite structure M-type ( $\text{BaFe}_{12}\text{O}_{19}$ ) exhibit a uniaxial magnetic an

\* Corresponding author at: Dept. of Phy., College of Science University of Anbar, Iraq.  
E-mail address: [Dr\\_Hamid2020@yahoo.com](mailto:Dr_Hamid2020@yahoo.com)

isotropy and so have been used as permanent magnets.

The magnetic loss ( $\mu_r''$ ) of these ferrites increased at the natural resonance frequency [5,6].

The hexagonal ferrite  $\text{Ba Fe}_{12}\text{O}_{19}$  and its substituted derivatives have been considered as candidates with the most potential because of their chemical stability and suitable magnetic characteristics [7]. The issue concerning the substitution of  $\text{Ba}^{2+}$  and / or  $\text{Fe}^{3+}$  ions with mixtures of paramagnetic and diamagnetic ions in the magnetoplumbite structure is to reduce the high magneto crystalline anisotropy of the material [8-11].

The electromagnetic microwave absorption properties of barium M-type ferrite in which  $\text{Fe}^{3+}$  was substituted by M( Ti,Mn,Zn,Sn...) were investigated in the GHz frequency range [1,4-6,8-9]. They found that the anisotropy field ( $H_a$ ) of barium .M-content which enables the natural frequency ( $f_r$ ) to be controlled .

In this paper Mn-Ti substituted barium ferrite was fabricated using solid solution method (ceramic method) with and without PVC polymer then studied the structure of the prepared samples and the absorption of electromagnetic microwave band (X-band) in the range 8-12GHz.

## EXPERIMENTAL PROCEDURE

Ba M-type ferrites with composition  $\text{Ba Fe}_{12-x}\text{Mn}_x\text{O}_{19}$ ,  $\text{Ba Fe}_{12-x}\text{Ti}_x\text{O}_{19}$  and  $\text{Ba Fe}_{12-x}(\text{Mn}_{0.5}\text{Ti}_{0.5})_x\text{O}_{19}$  ( $x=0-4.5$ ) were prepared by solid solution method (ceramic method). The starting materials of  $\text{BaO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{MnO}$  and  $\text{TiO}_2$  powder (purity >99%), were mixed in a planetary ball mill and in to cylindrical shaped sample .There compacts were crushed into powders of size less than  $300\mu\text{m}$  after the primary sintering at  $1000-1100\text{C}^\circ$  for 8-12 hours and then the powder divided in to three portions for different studies . Each type of the powder was compacted in to a cylindrical shape under pressure of  $1-3\text{ tun/cm}^2$  and then sintered at  $1100-1200\text{C}^\circ$  for 1-5 hrs. in air or in nitrogen gas. The fine sintered powder were mixed with polymer risen (PVC).

The phases present in the prepared powder were characterized by x-ray diffraction, using a Siemens diffractometer with  $\text{Cu } k_\alpha$  radiation. In order to obtain the morphology of a sample surface, a set of micrographs were taken by an optical microscope type Olomps of 500X

The electromagnetic wave absorption properties were measured using a Hewlett -Packard Hp 8150C

network analyzer in the X-band range.

## RESULTS AND DISCUSSION

### 1. Structure and Morphology

The XRD patterns of particle samples of standard Ba Fe<sub>12</sub>O<sub>19</sub> and Mn-Ti substitution of the Ba Fe<sub>12-x</sub> (Mn<sub>0.5</sub> Ti<sub>0.5</sub>)<sub>x</sub>O<sub>19</sub> ferrite at  $0 \leq x \leq 4.5$  are shown in fig. 1. As can be seen the only crystalline phase that can be detected by XRD is an M-type hexaferrite for all particle samples, no others phases were apparently detectable. With the increase of a mount substitution (x)it can be noted that the peaks height decreased as x increased , which means that the crystallinity is decreased [4]. This means that Mn- Ti substitutions increased the temperature formation of the Ba -M phase. Also from fig.1 the patterns which related to the Ba-ferrite substituted by Mn only are different from that at Ti only. The first one gives a smaller central peak and a wider it, which means that Mn substituted give a bad crystallinity and a lower ferrite structure.

The surface morphology of Ba Fe<sub>12-x</sub> (Mn<sub>0.5</sub> Ti<sub>0.5</sub>)<sub>x</sub>O<sub>19</sub> particles are obtained in the optical microscope picture as shown in fig.2.From this figures

we show that the grain size increased with increasing the amount of substitution (x) which its value between 7.05-7.95  $\mu\text{m}$  when  $x = 2.5$  and 7.57-8.45  $\mu\text{m}$  when  $x = 3.5$  as shown in fig.(2.a)and the porosity was decreased with increasing x. When Ba-ferrite substituted by Mn only at  $x= 2.5$  give a larger grain size (13.6)  $\mu\text{m}$  fig 2-a.3 which agree with the XRD pattern .While fig. 2-a.4 which is for Ba-ferrite substituted by Ti only at  $x= 2.5$  give a similar shape of fig. (2-a.1 and 2-a.2 ) but a larger grain size of it (8.06 - 12.06)  $\mu\text{m}$  . Fig.2-b shows the effect of particles fabricating temperature and time on the surface morphology of the prepared samples, which obtain that the increased sintering time give a larger grain size fig(2-b,2) and lower in the porosity

The density of the prepared samples were calculated from the measuring of the cylindrical size sample volume and from the mass of it. Table 1 obtained the effect of Ba- ferrite substitution and preparation weathering on the density samples. From this table the density of the sample decreased with increasing the substitution of BaF<sub>12</sub>O<sub>19</sub> by Ti and Mn because of the differences of the masses of Fe, Ti and Mn. The density decreased when used a nitrogen gas

instead of air in the sintering which may be related to decreasing the oxygen in the sintering process and then in the Ba-ferrite structure. The density of the samples decreased with increasing an addition sintering time as in table 1. This may be related to be complete the interaction of the mixtures and then complete the substitution, when the Ba-ferrite substituted by Ti only it give a higher density than that substituted by Mn only, which related to the difference between mass and size of Ti & Mn.

## 2. Electromagnetic wave absorption properties

The reflection loss (R.L) and the absorption of the prepared samples were measured in the X-band frequency range (8-12 GHz). The R.L must be greater than 8dB or the absorptivity greater than 84% and the bandwidth must be greater or equal to 1 GHz to become the samples are succeeded for use as microwave absorption materials [6].

Fig. 3 shows the frequency dependence of the R.L and absorptivity in the X-band range at different values of x (2.5, 3.5 and 4.5). The minimum R.L for all samples were less than 15 dB. Therefore it can be said that these samples were acting as electromagnetic microwave absorber, in the range 8-12 GHz. The minimum R.L decreased

with increasing the substituted  $Ti_{0.5}Mn_{0.5}$  contents (x). The bandwidths increased with increasing x, and the value of it for all samples were greater than 1 GHz.

Fig. 4 shows the frequency dependence of R.L and absorptivity of  $BaFe_{9.5}Ti_{2.5}O_{19}$  (4-a) and  $BaFe_{9.5}Mn_{2.5}O_{19}$  (4-b), which obtain that the R.L value and the bandwidth of the barium ferrite substituted by Ti greater than that substituted by Mn, were these values are -28dB and 3.2GHz for Ti, while -14.5dB and 1GHz for Mn. This means that Ti give better properties for use as microwave absorption materials, which is good agreement with the results of Sugimoto et al. [1].

Fig.5 obtain the effect of sintering gas on the R.L and absorptivity of  $BaFe_{12-x}(Mn_{0.5}Ti_{0.5})_xO_{19}$ . From Fig.5 we show that the values of bandwidth and the R.L were decreased when used  $N_2$  instead of air and these values are changed from 2.8 to 2GHz for bandwidth and from -25 to -16 dB for R.L. Also when x increased to 3.5 with  $N_2$  Fig.(5-c) the values of bandwidth and R.L are decreased to 1.6 GHz and -13.8dB respectively. This behavior may be related to decreased the oxygen ions in the interaction when used  $N_2$  and the interaction of  $Fe^{3+}-O^{2-}-Fe^{3+}$  in the sintering processes give a good ferrite magnetic

properties [11] . So the decreased of  $O_2$  give a smaller bandwidth and a lower R.L .

Fig.6 shows the frequency dependence of R.L and absorptivity of  $Ba Fe_{9.5} (Mn_{0.5} Ti_{0.5})_{2.5} O_{19}$  with polymer resin (PVC) . We obtain that at increasing the ratio of polymer from 30mass%,40mass% to 60mass%, the minimum value of R.L decreased from -21,-16 to -14 dB respectively, while the values of bandwidth are 2.6,2 and 1.8 dB for each ratio , so the best ratio which give good properties is 30 mass%

Fig.7, obtain the frequency dependence of R.L and absorptivity of  $Ba Fe_{12-x} (Mn_{0.5} Ti_{0.5})_x O_{19}$  with polymer ratio 30mass% . From Fig.7 the value of bandwidth for the barium ferrite of  $x=3.5$  and  $4.5$  equal to 2.5 and 1.7 GHz respectively , while the R.L values are equal to -12.5 and -16 dB for each amount of  $x$ . The frequency range of R.L  $>$  16 dB shifted to lower frequency with increasing  $Ti_{0.5}Mn_{0.5}$  content which due to the decrease of the natural resonance frequency [6].

## CONCLUSION

Ti-Mn substitutions barium M-type ferrite led to material with good candidate for use as an

electromagnetic microwave absorber in the X-band frequency range (8-12 GHz) and have a single phase hexagonal structure .The grain size of the prepared powder were between (7.05-12.06 $\mu$ m) .The reflection loss of the electromagnetic properties give a minimum value which equal to -21dB and wide bandwidth of 3.2 GHz in the X-band range .The substitution with Ti give better properties than with Mn. The composite mixture with polymer resin give a minimum R.L depends on the polymer ratio and on the ferrite composition.

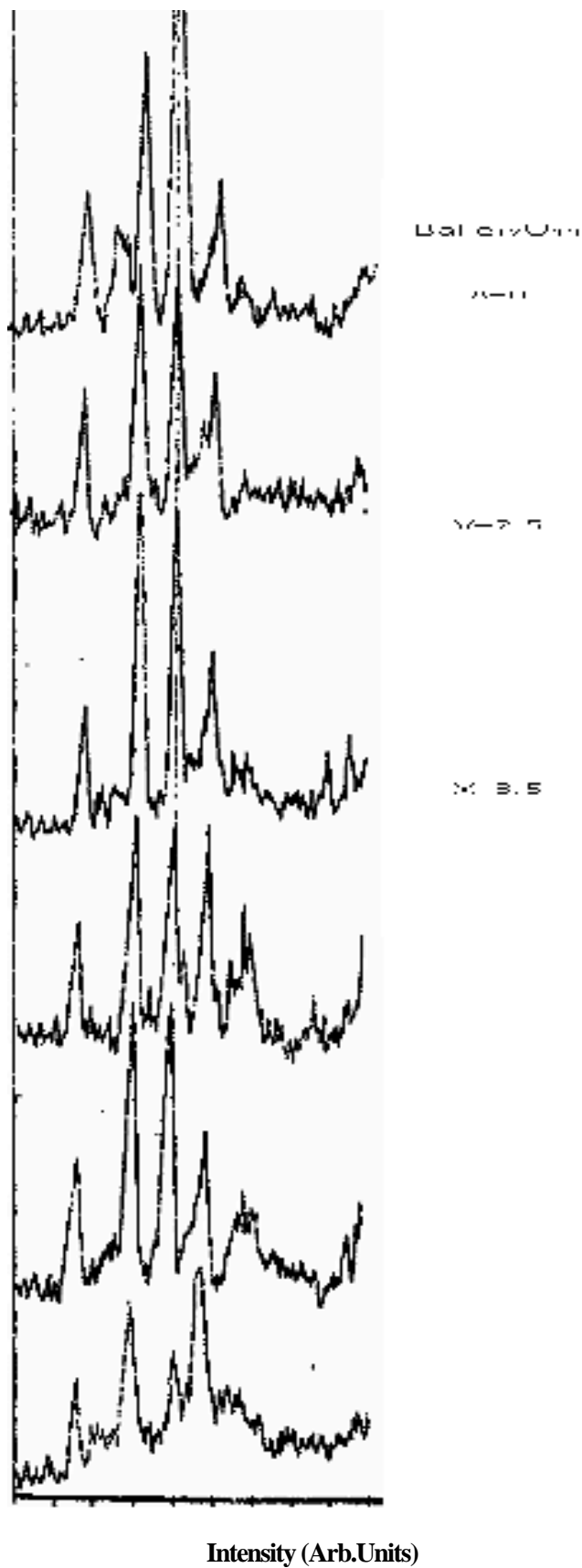
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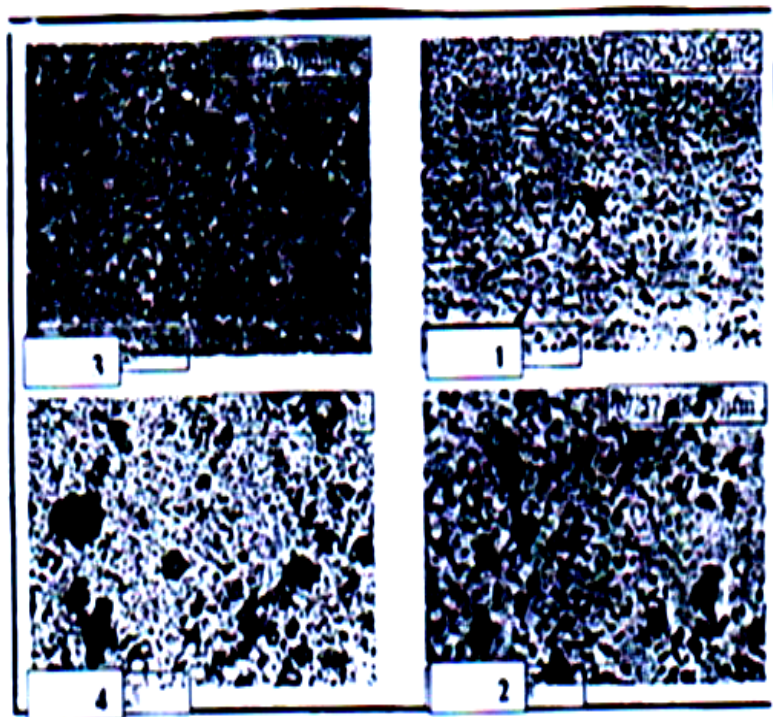
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**Table 1. The effect of barium ferrite substitution on the density of the prepared samples.**

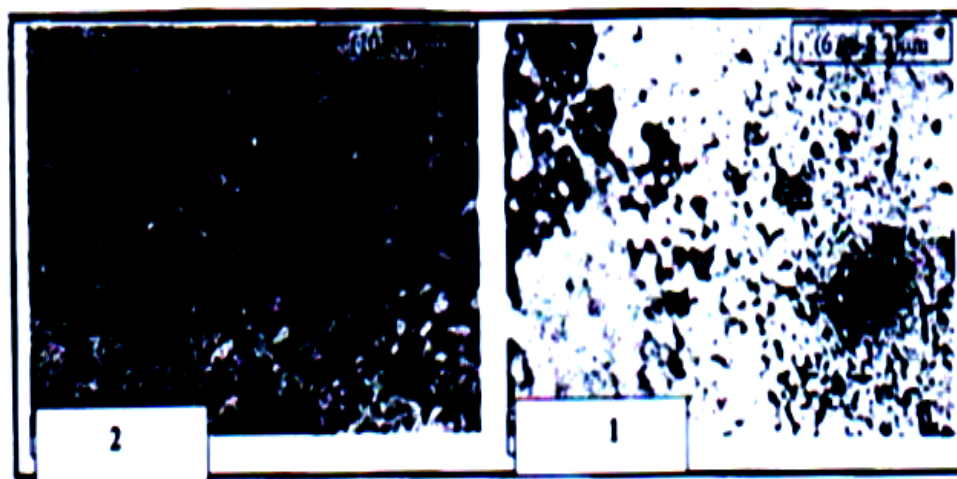
Sample No.	X and another conditions	$\rho$ (gm/cm <sup>3</sup> )
1	2.5	3.787
2	3.5	3.412
3	4.5	3.110
4	2.5+N <sub>2</sub>	3.506
5	3.5+N <sub>2</sub>	3.205
6	2.5 + added sint. 5hr.	3.384
7	3.5 + added sint. 5hr.	3.153
8	4.5 + added sint. 5hr.	2.969
9	2.5 Ti only	3.842
10	2.5 Mn only	3.370







(a)



(b)

**Fig.2** Optical micrograph of  $BaFe_{12-x}(Ti_{0.5}Mn_{0.5})_xO_{19}$  samples (a) with different  $x$  (1)  $x=2.5$  (2)  $x=3$  (3) with  $x=2.5$  Mn (4) with  $x=2.5$  Ti (b) with different sintering time (1)  $1100\text{ }^{\circ}\text{C}$  for 5h (2)  $1100\text{ }^{\circ}\text{C}$  for 6h .

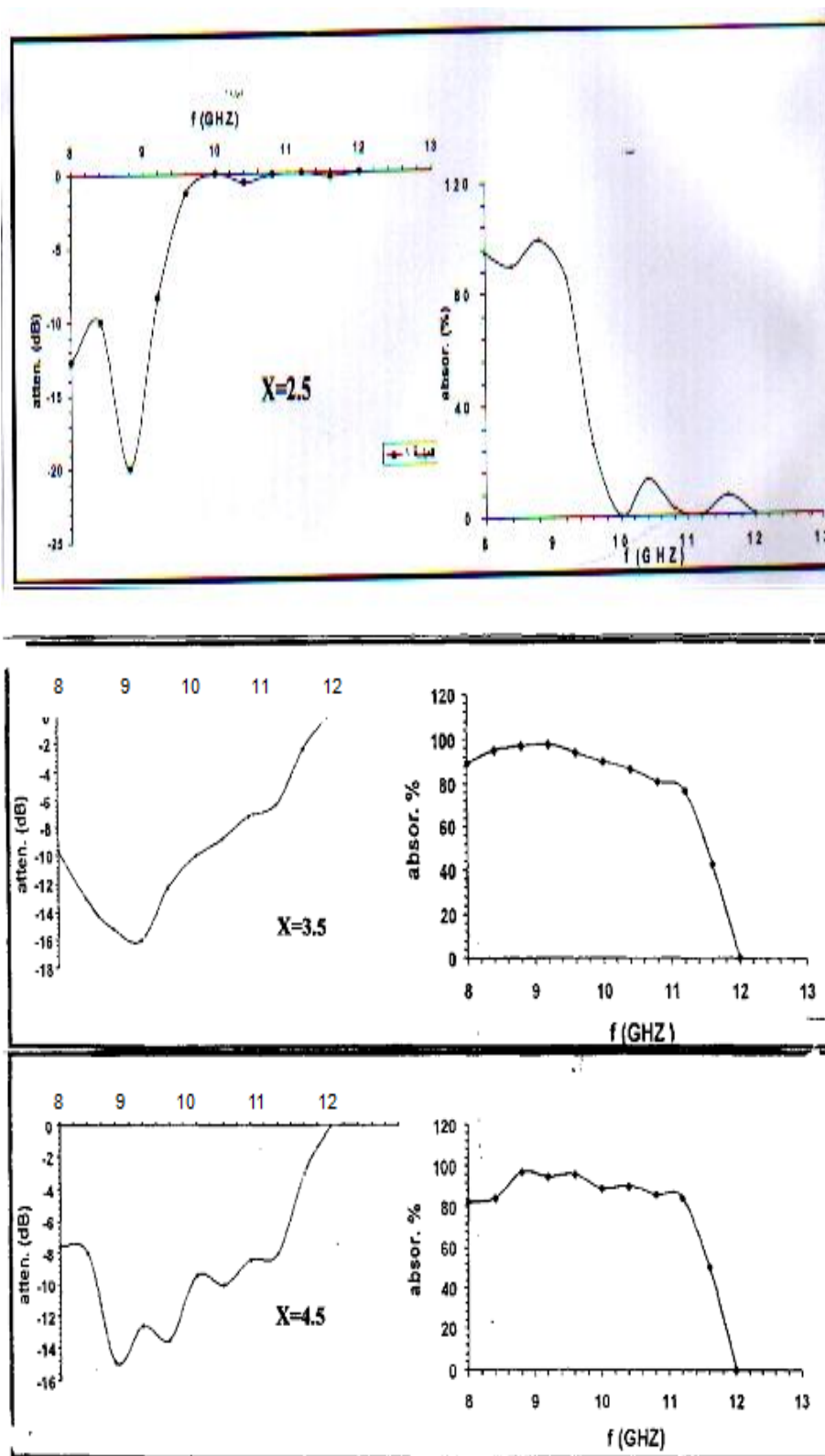


Fig. 3 The frequency dependence of reflection loss (dB) and absorptivity of  $\text{BaFe}_{12-x}(\text{Ti}_{0.5}\text{Mn}_{0.5})_x\text{O}_{19}$

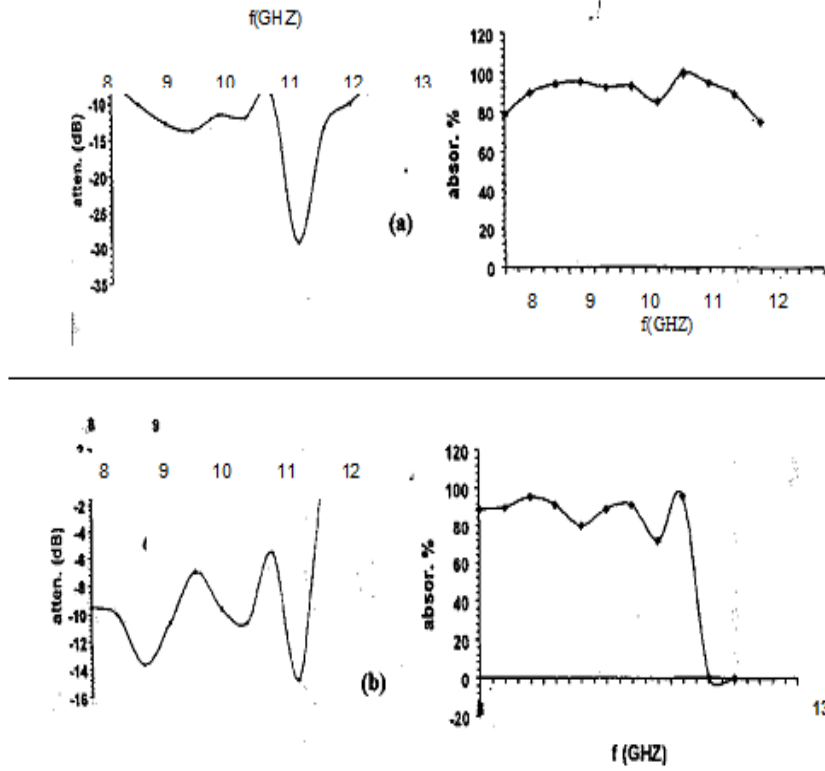


Fig.4 The frequency dependence of reflection loss (dB) and absorptivity of (a)  $\text{BaFe}_{9.5}\text{Ti}_{2.5}\text{O}_{19}$  (b)  $\text{BaFe}_{9.5}\text{Mn}_{2.5}\text{O}_{19}$ .

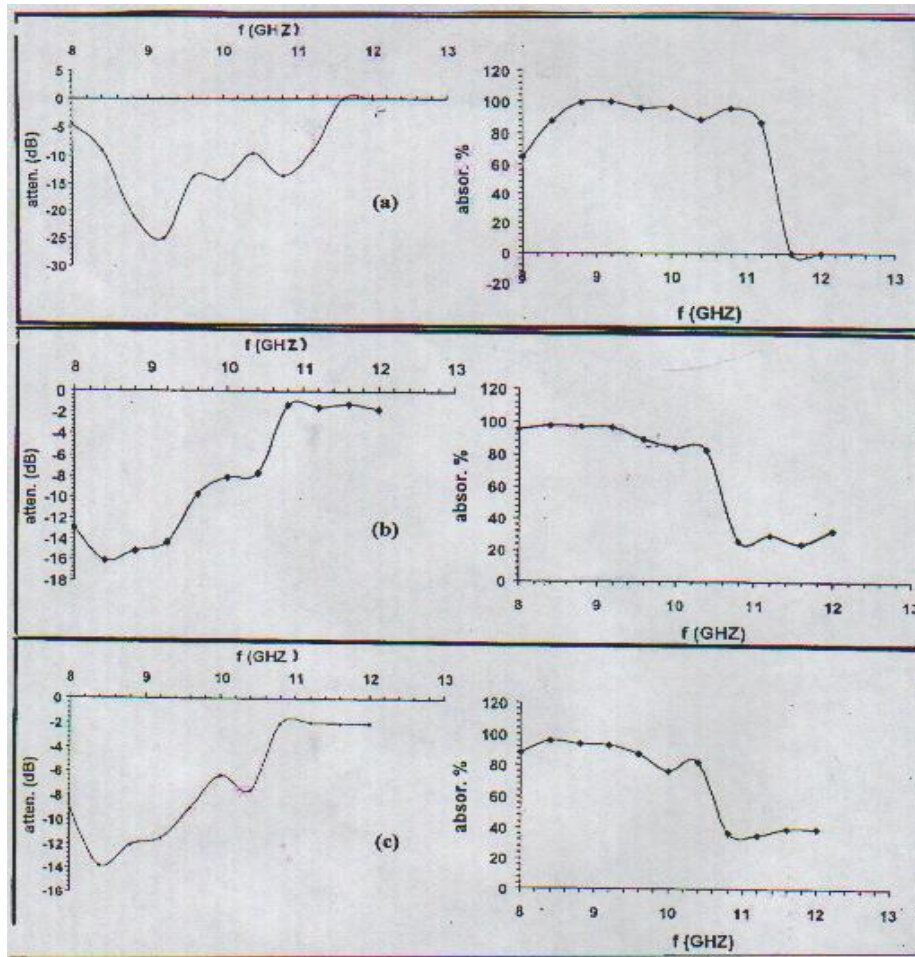


Fig. 5 The frequency dependence of reflection loss (dB) and absorptivity of  $\text{BaFe}_{12-x}(\text{Ti}_{0.5}\text{Mn}_{0.5})_x\text{O}_{19}$  (a)  $x=2.5$  with air (b)  $x=2.5$  with  $\text{N}_2$  and (c)  $x=3.5$  with  $\text{N}_2$ .

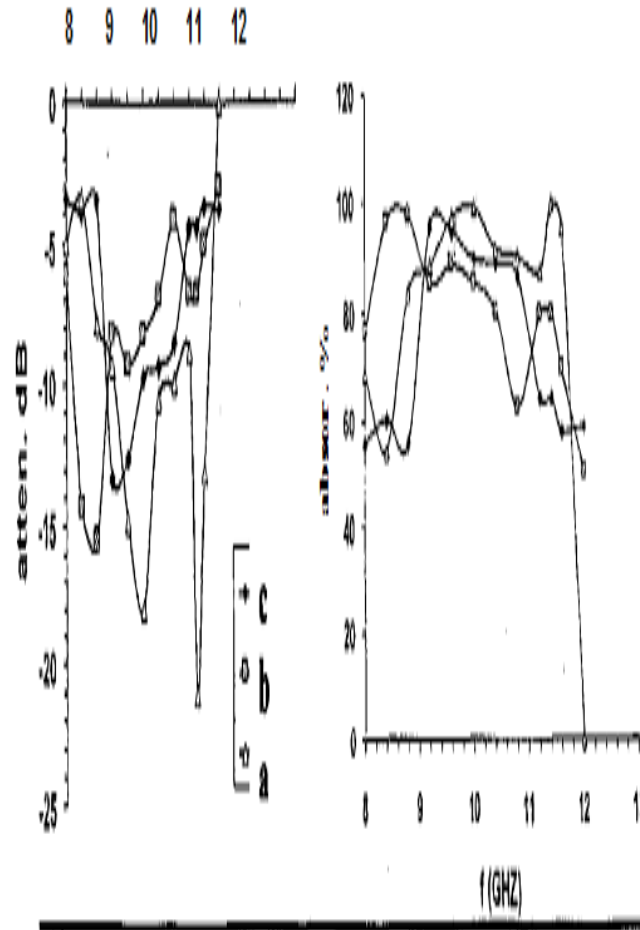


Fig.6 The frequency dependence of reflection loss (dB) and absorptivity of  $BaFe_{9.5}(Ti_{0.5}Mn_{0.5})_{2.5}O_{19}$  With (a) 30 mass % (b) 40mass% (c) 50 mass % polymer

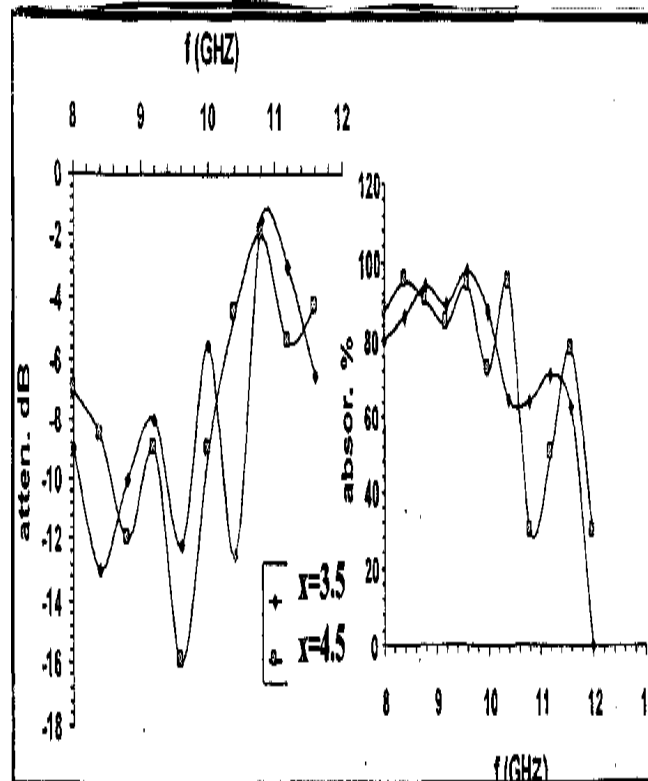


Fig.7 The frequenc dependence of reflection loss (dB) and absorptivity of  $BaFe_{12}(Ti_{0.5}Mn_{0.5})_xO_{19}$  with 30 mass % polymer

## تأثير إستبدال المنغنيز والتيتانيوم لفرأيت الباريوم على امتصاص الموجات الكهرومغناطيسية الدقيقة ضمن مدى الحزمة السينية.

حامد صالح الجميلي ، خالد طه رسول، سعيد نايف الراشد

E-mail: [Dr\\_Hamid2020@yahoo.com](mailto:Dr_Hamid2020@yahoo.com)

### الخلاصة :

تم دراسة خصائص امتصاص الموجات الكهرومغناطيسية بالاعتماد على مكونات الباريوم فرأيت للتملج الملبدة، حيث  $BaFe_{12x}(Ti_{0.5}Mn_{0.5})_xO_{19}$  استبدل فيه الحديد بالتيتانيوم والمنغنيز . لقد حصل على تركيب احادي الطور للباريوم فرأيت ضمن مدى تردد الحزمة -21dB بين 0 و 4.5. إن اعتمادية التردد لخسارة الانعكاس (التوهين) اظهرت اقل قيمة وهي X تراوحت قيم بنفس مدى التردد. الحجم الحبيبي للمسحوق المحضر تراوح بين 1GHz واعطت عرض حزمة اكبر من 8-12 GHz) السينية (قد اعطى 30 and 50 mass% بنسب خلط PVC . اما مركب مسحوق الباريوم فرأيت المخلوط مع البوليمر (7.05-12.06  $\mu m$ ) اعطت (3.5 and 4.5) فيها تساوي X . النماذج المخلوطة مع البوليمر والتي كانت قيم -21dB عرض حزمة اوسع مع فقد انعكاس اقل من على التوالي ايضا. -16dB و -12dB على التوالي واعطت توهين مقداره 1.7GHz و 2.5GHz عرض حزمه مقداره لقد اوضحت النتائج بان الباريوم فرأيت المستبدل بالتيتانيوم والمنغنيز يعطي قيم امتصاص جيدة ضمن مدى تردد الحزمة السينية.