



Trade Science Inc.

ISSN : 0974 - 7486

Volume 7 Issue 3

# Materials Science

An Indian Journal

Full Paper

MSAIJ, 7(3), 2011 [152-160]

## Some magnetic properties of Cu-Ge ferrite in low magnetic field

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Received: 15<sup>th</sup> November, 2010 ; Accepted: 25<sup>th</sup> November, 2010

### ABSTRACT

Some magnetic properties such as B-H loops and initial permeability  $\mu_i$  for the polycrystalline soft ferrite,  $Cu_{1-x}Ge_xFe_{2-2x}O_4$ ; with  $x=0.0, 0.05, 0.1, 0.15, 0.2, 0.25$  and  $0.3$  have been studied. The B-H loops were first measured at RT as a function of magnetizing current  $I$  from  $0.5$  to  $6A$  (i.e.  $H=185Am^{-1}$  to  $3250Am^{-1}$ ). It was found that the coercive force  $H_c$  increases with increasing the magnetizing current but the relative magnetization  $m_r$  shows a maximum value for each composition at  $I$  between  $2-3A$ . Also, The B-H loops were measured at elevated temperature and constant magnetizing current ( $I=2.5A$  which is equivalent to  $900Am^{-1}$ ). It was observed that  $H_c$  decreases with increasing the temperature. The initial permeability  $\mu_i$  was measured as a function of temperature for the above investigated ferrite. Utilizing the initial permeability data, the Curie temperature  $T_c$  and the paramagnetic temperature  $T_p$  were estimated. These values of temperature ( $T_c$  &  $T_p$ ) have been found to decrease with increasing  $Ge^{4+}$  ions content.

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### KEYWORDS

Ferrite;  
Magnetization;  
B-H loop;  
Initial permeability.

### INTRODUCTION

Ferrites are still much used in permanent magnets market because of their low price combined with reasonable magnetic performances. Another advantage is the great chemical stability of these oxides which makes in such a context a little improvement of their magnetic properties of great importance<sup>[1]</sup>. Soft ferrites have been extensively used for many kinds of magnetic devices such as transformers, inductors and magnetic heads for high frequency because their electrical resistivity is higher than those of the soft magnetic alloys. Various substitutions have been incorporated to achieve desired electrical and magnetic properties. Ferrites containing cop-

per have shown interesting electrical and magnetic properties<sup>[2-5]</sup>. Several studies have reported addition of divalent, trivalent and tetravalent ions and various parameters have been measured depending on the desired application<sup>[6-9]</sup>. Our previous works have studied X-ray analysis, IR absorption and electrical properties of Cu-Ge and Cu-Ti ferrites<sup>[10-12]</sup>. Also, the influence of the cation distribution on the mössbour spectra of Cu-Ge ferrite has been studied<sup>[13]</sup>. Recently, X-ray diffraction (XRD), scanning electron microscope (SEM) and some magnetic properties such as magnetization and B-H loops were measured in analogous study in low magnetic field between Cu-Ge and Cu-Si ferrites<sup>[14]</sup>.

In the present work, the influence of iron substitu-

tion by tetravalent  $\text{Ge}^{4+}$  ion on the magnetic properties of Cu-ferrite will be study.

## EXPERIMENTAL TECHNIQUE

The mixed copper – germanium ferrite with the chemical formula  $\text{Cu}_{1+x}\text{Ge}_x\text{Fe}_{2-2x}\text{O}_4$  (where,  $x = 0.0, 0.05, 0.1, 0.15, 0.2, 0.25$  and  $0.3$ ) were prepared by using the standard ceramic technique.

The  $B$ - $H$  loops were measured for the toroidal sample where the number of turns of the primary coil  $N_p$  and the secondary coils  $N_s$ , were 16 and 8, respectively. The results were recorded by the aid of a storage oscilloscope (HAMEG, type HM407) connected with pc. The details of the circuit diagram, measurement technique and the calculations were mentioned in our previous work<sup>[14]</sup>.

The initial permeability  $\mu_i$  was measured as a function of temperature at constant frequency  $f = 10 \text{ KHz}$  and low magnetizing current  $i_p = 20 \text{ mA}$ . More details about the measurement techniques and calculations of  $\mu_i$  were explained in our previous work<sup>[15,16]</sup>.

## RESULTS AND DISCUSSION

### Effect of magnetizing current on B-H loops

The hysteresis loops were firstly studied at room temperature at constant frequency ( $\approx 50 \text{ Hz}$ ) for all the above investigated compositions of Cu-Ge ferrite. The measurements were carried out in the range of magnetizing current from  $0.5 \text{ A}$  to  $6 \text{ A}$  (i.e., from  $H = 185 \text{ Am}^{-1}$  to  $2250 \text{ Am}^{-1}$ ). Figure 1 shows the hysteresis loops for  $\text{CuFe}_2\text{O}_4$  at  $I = 1.5$  and  $6 \text{ A}$ , as a representative example. From the  $B$ - $H$  loops, some parameters such as: coercive field  $H_C$  ( $\text{Am}^{-1}$ ), remanence induction  $B_r$  (T), saturation induction  $B_s$  (T), and the relative magnetization ( $m_R = B_r/B_s$ ) were estimated as a function of magnetizing current ( $I$ ) for all compositions. Figure 2 & 3 represent the effect of magnetizing current  $I$  on the coercive field  $H_C$  and the relative magnetization  $m_R$ , respectively.

The behaviour of the coercive field  $H_C$  versus the magnetizing current  $I$  could be divided into two regions I & II, where region I for lower than  $3 \text{ A}$  and region II for higher than  $3 \text{ A}$  as shown in figure 2. In region I, the coercive field increases gradually with increasing the magnetizing current. In region II, coercive field  $H_C$  has

stabilization with increasing the magnetizing current for composition of  $x$  up to  $0.2$  as shown in figure 2a. But, for  $x = 0.25$  &  $0.3$ , the increment of  $H_C$  in region II is less than in region I as presented in figure 2b. The stabilization behaviour of  $H_C$  may be related to the samples behave as a single domain.

The effect of the magnetizing current  $I$  on the relative magnetization  $m_R$  were studied as shown in figure 3. It was observed that  $m_R$  shows a peak (max. value) between  $I = 2-3 \text{ A}$ . So that the ratio  $m_R$  vs.  $I$  could be divided into two stages (I & II). The increment of  $m_R$  in stage I and the decrement in stage II depends on the divergence between  $B_r$  &  $B_s$ . The highest value of  $(m_R)_{\text{max}}$  was recorded for  $x = 0.05$  at magnetizing current  $I = 2 \text{ A}$ . This means that loop at this condition can be used for the recording system.

Also, the effect of porosity on the parameters  $H_C$ ,  $B_r$  and  $B_s$  at magnetizing current  $2.5 \text{ A}$  was studied as shown in Fig. 4a-b. It is observed that  $H_C$  is directed to small increasing with increasing porosity. This effect may be caused by the fact that the high-porosity samples contain smaller particles, which have higher coercive forces<sup>[17]</sup>. But two magnetic parameters  $B_r$  and  $B_s$  decrease with increasing porosity as shown in figure 4b. This behaviour may be related to the increase in porosity (i.e. decrease in density) or decreasing of magnetic materials in a specified volume accompanied with the decrease in the two parameters  $B_s$  and  $B_r$ <sup>[18]</sup>.

### Effect of temperature

#### The hysteresis loops

Again, the ( $B$ - $H$ ) loops of  $\text{Cu}_{1+x}\text{Ge}_x\text{Fe}_{2-2x}\text{O}_4$  were studied at elevated temperature (from room temperature up to  $750 \text{ K}$ ) and at constant magnetizing current  $2.5 \text{ A}$  ( $H = 900 \text{ Am}^{-1}$ ). The recorded loops are shown in Fig. 5; for  $x = 0.0$  at  $T = 320$  and  $670 \text{ K}$ . The saturation magnetization  $M_s$  (relative to  $B$ - $H$  loops) was calculated according to the relation:

$$M_s = \left( \frac{B_s}{\mu_0} - H \right) \quad (1)$$

Figure 6 shows the relation between  $M_s$  vs. temperature for  $x = 0.0$ . It was found that  $M_s$  vanishes at a point of temperature (called  $T_s$ ) closed to Curie point  $T_C$ . Below the Curie temperature the presence of non-equivalent sublattices leads to a variety of behaviour of

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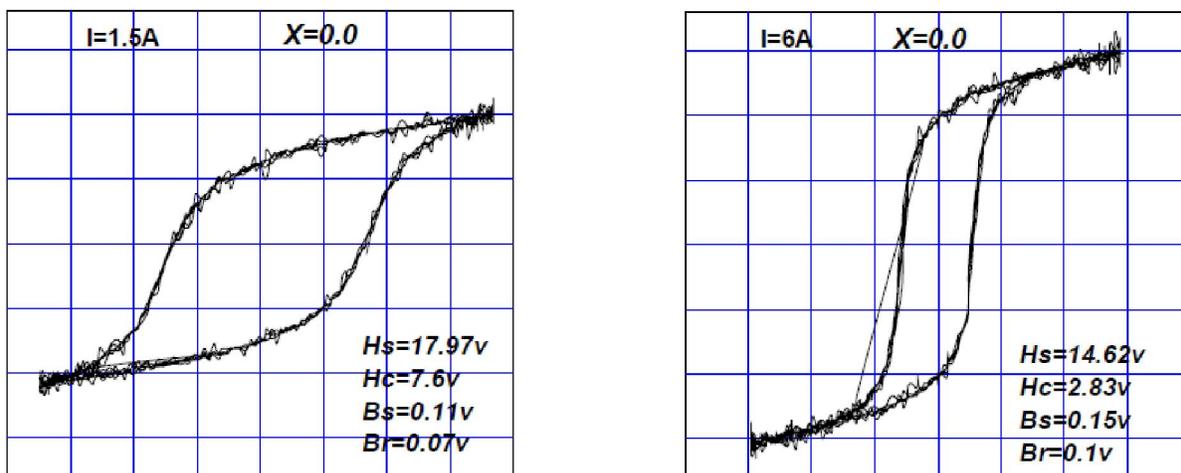


Figure 1 : B-H loops of x=0.0, measured at room temperature (300K) and at I=1.5&6A

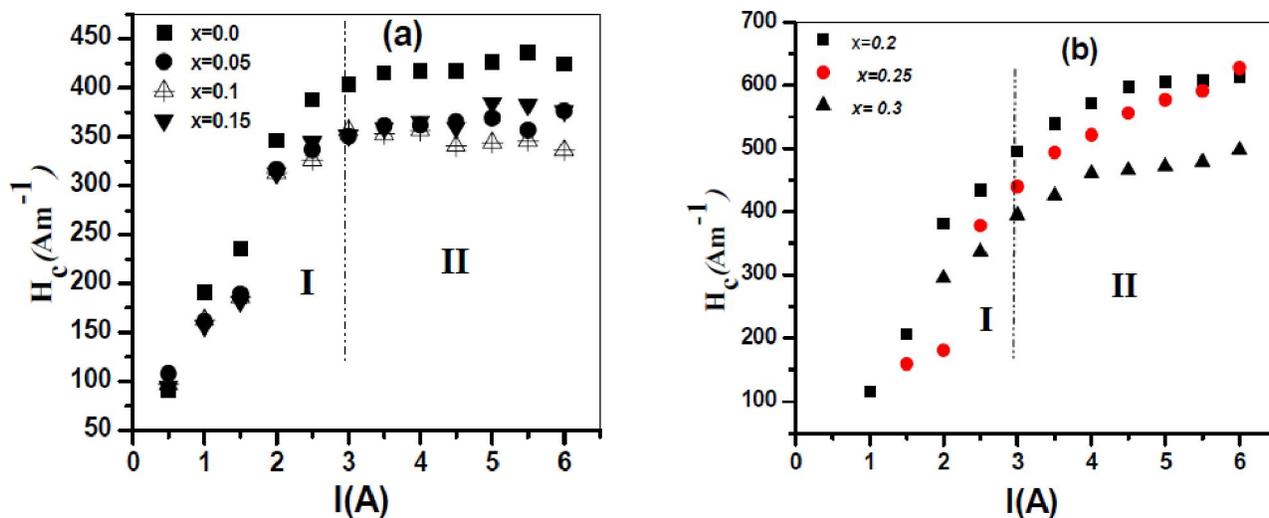


Figure 2 : The effect of magnetizing current on coercive force for  $Cu_{1+x}Ge_xFe_{2-2x}O_4$  ( $0 \leq x \leq 0.3$ )

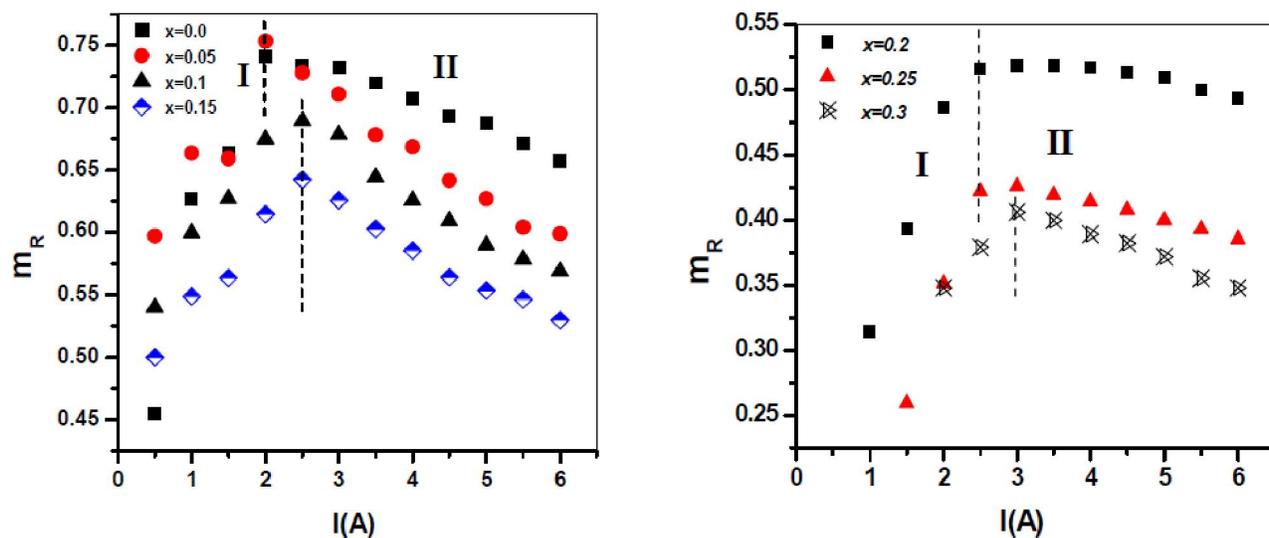


Figure 3 : The effect of magnetizing current on relative magnetization for  $Cu_{1+x}Ge_xFe_{2-2x}O_4$  ( $0 \leq x \leq 0.3$ )

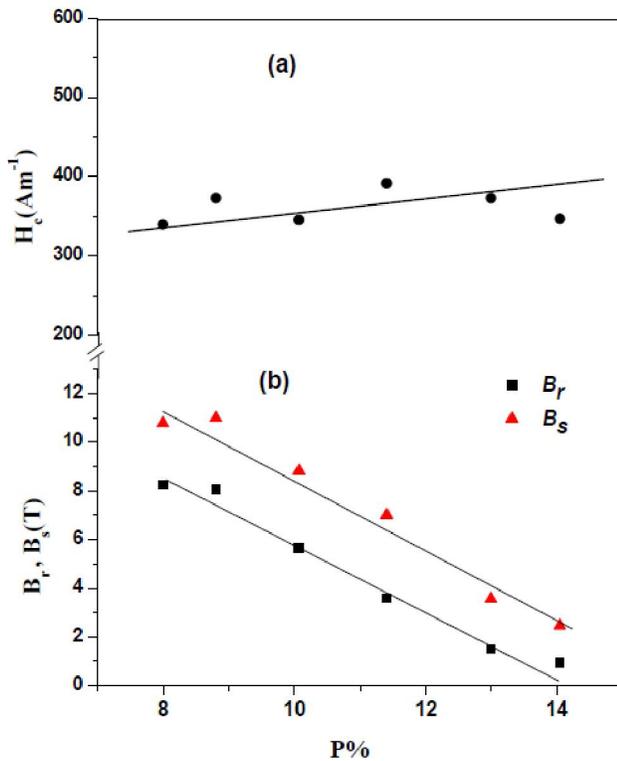


Figure 4a & 4b : The effect of porosity on: a) coercive force and b) remanance and saturation magnetizations

the net magnetization ( $M = M_B - M_A$ ) as a function of temperature, since each sublattice has a magnetic moment with its own temperature dependence. The form of these curves has been explained by Néel in terms of an internal field for each sublattice. The behaviour of  $M_s$  vs.  $T$  is similar to the behaviour of the magnetization of type R<sup>[18]</sup>, where one sublattice is not saturated at

OK; a finite value of  $\left(\frac{dM}{dT}\right)_H$  is predicted. This may be

shown to conflict the third law of thermodynamics, in which the entropy is independent of the field  $H$  when  $T$

$\rightarrow 0$ , so  $\left(\frac{\delta M}{\delta T}\right)_H \rightarrow 0$ . Therefore,  $M_s(0)$  can be estimated from the extrapolation curves of figure 6. figure 7 shows the variation of  $M_s(0)$  with the addition of Ge content. It observed that it increases up to  $x=0.05$  and then decreases with further increases of Ge content. This behaviour can be discussed on the basis of the statistical distribution of the various cations over the A and B-sites using Néel's theorem of two sublattices model<sup>[18]</sup>. The individual magnetization,  $M_A$  and  $M_B$  of the two sublattices cannot be observed, but the calculated magnetic moment for each site  $\mu_A$ ,  $\mu_B$  and  $\mu_{eff}$  (i.e.

TABLE 1 : The calculated values of anisotropy constant  $K$ , exchange constant  $A$ , wall energy  $\sigma_w$  and wall thickness  $\delta_w$

X	D ( $\mu\text{m}$ )	$\mu_i$	$T_c$ (K)	$T_p$ (K)	K ( $10^4$ J/m <sup>3</sup> )	A(0) ( $10^{-12}$ J/m)	A(0)	$\sigma_w$ ( $10^{-3}$ J/m <sup>2</sup> )	$\delta_w$ ( $10^{-8}$ m)	
							Theoretically			By extra-polation
0.0	3.97	137.32	718	723	5.13	11.81	12.24	8.98	1.36	2.65
0.05	9.35	195.94	693	716	54.60	11.39	11.81	8.66	4.35	0.79
0.1	23.95	284.78	643	666	44.30	11.6	11.15	7.71	3.70	0.83
0.15	21.24	195.90	613	633	29.30	10.09	10.71	7.18	2.90	0.99
0.2	19.49	131.70	598	623	16.90	9.85	10.45	6.92	2.16	1.28
0.25	16.59	94.01	573	585	1.67	9.44	10.12	6.48	0.66	3.94
0.3	14.93	57.43	553	574	0.92	9.1	9.86	6.12	0.47	5.17

the effective magnetic moment) can be calculated according to:

$$|\vec{\mu}_{eff}| = |\vec{\mu}_B| - |\vec{\mu}_A| \quad (2)$$

The magnetization of each composition depend on the distribution of  $\text{Fe}^{3+}$  ions among the two sites A and B, where  $\text{Ge}^{4+}$  ion is non magnetic. On the basis of the above results for the two series, the cation distribution can be suggested as follow<sup>[14]</sup>:

$(\text{Cu}_t^{2+} \text{Ge}_x^{4+} \text{Fe}_{1-x-t}^{3+})^A [\text{Cu}_{1+x-t}^{2+} \text{Fe}_{1-x+t}^{3+}]^B \text{O}_4^{2-}$ ; for  $x=0.0$  &  $0.05$ .

$(\text{Ge}_x^{4+} \text{Fe}_{1-0.6x}^{3+})^A [\text{Cu}_{1+x}^{2+} \text{Fe}_{1-1.4x}^{3+}]^B \text{O}_4^{2-}$ ; for  $(0.1 \leq x \leq 0.3)$ .

For the basic ferrite  $\text{CuFe}_2\text{O}_4$  ( $x=0.0$ ) is known as inverse ferrite, where  $\text{Cu}^{2+}$  are located on B-sites. However, there is a probability of migration of a small fraction ( $t$ ) of  $\text{Cu}^{2+}$  ions to A-sites<sup>[19]</sup>; in this case it may be considered as a partially inverse ferrite. For Cu-Ge ferrite, the ionic magnetic moment of  $\text{Cu}^{2+}$  is 1 and the magnetic moment of  $\text{Fe}^{3+}$  is 5. The replacement of high spin quantum number ion  $\text{Fe}^{3+}$  by  $x\text{Ge}^{4+}$  ions; where  $\text{Ge}^{4+}$  ions prefer to occupy A-site<sup>[20]</sup>, this gives  $(1-x-t)\text{Fe}^{3+}$  ions on the A-site and  $(1-x+t)\text{Fe}^{3+}$  ions on the B-site. This substitution will lead to increase  $\text{Fe}^{3+}$  ions on the B-site and consequently the magnetization of the B-site will increase. At the same time the magnetization of the A-site will decrease according to decrease the  $\text{Fe}^{3+}$  ions on A-site. Accordingly, the net magnetization will increase for  $x=0.05$ . As the germanium ions increase (in the range of  $0.1 \leq x \leq 0.3$ ), the magnetization decrease with increasing  $x$ . This behaviour may be related to the migration of  $\text{Cu}^{2+}$  ions to B-site. The increase of Ge content will prevent the existence of  $\text{Cu}^{2+}$  ions on A-site. Also, the

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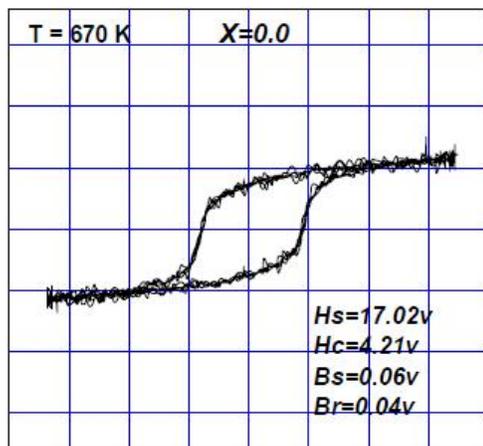
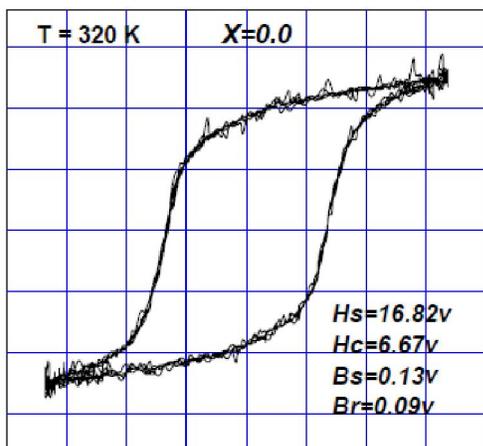


Figure 5 : B-H loops of  $x=0.0$ , measured at  $T=320$  and  $670\text{K}$  and constant magnetizing current  $I=2.5\text{A}$

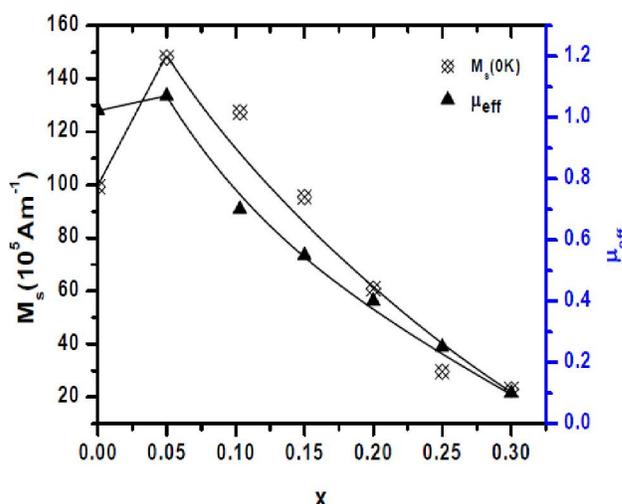
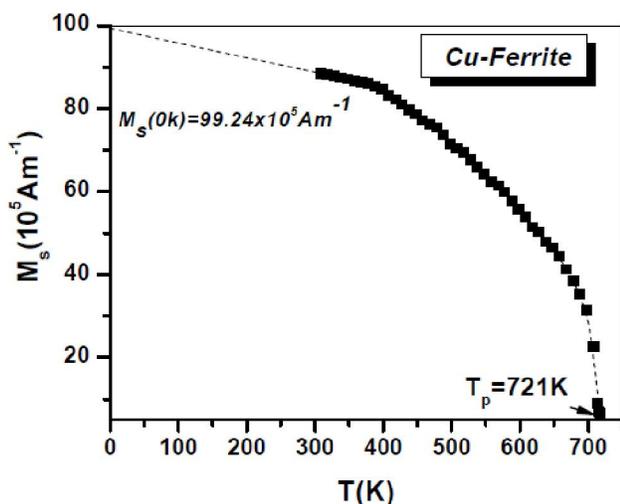


Figure 6 : The effect of temperature on the variation of saturation magnetization for  $\text{CuFe}_2\text{O}_4$

Figure 7 : The variation of  $M_s$  at  $0\text{K}$  and the effective magnetic moment  $\mu_{\text{eff}}$  with the composition  $x$

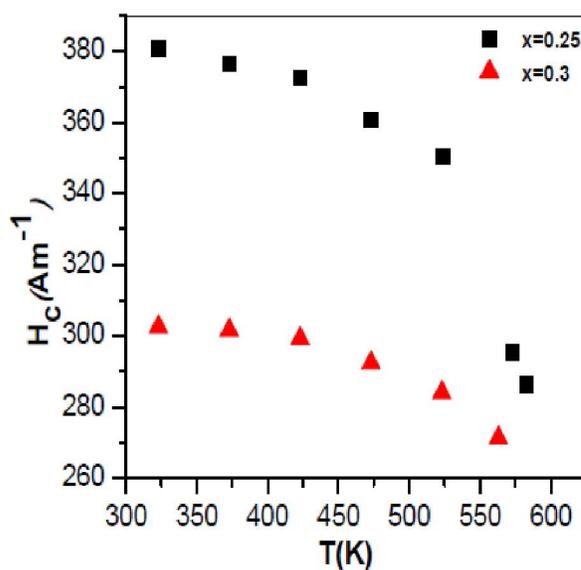
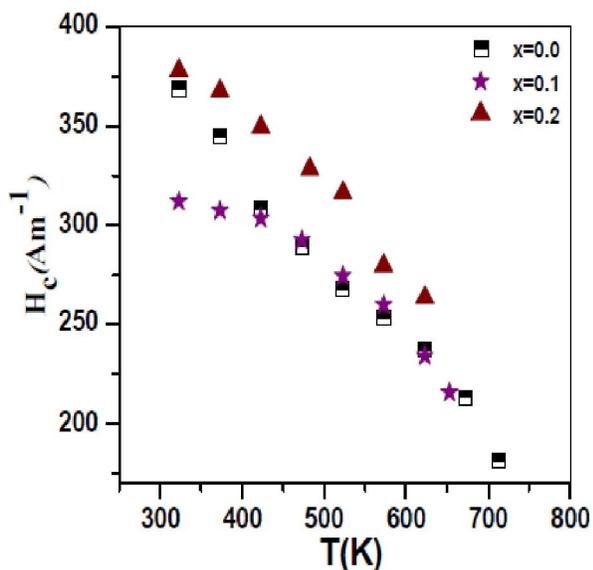


Figure 8 : The effect of temperature on coercive force for  $\text{Cu}_{1+x}\text{Ge}_x\text{Fe}_{2-2x}\text{O}_4$  ( $0 = x = 0.3$ )

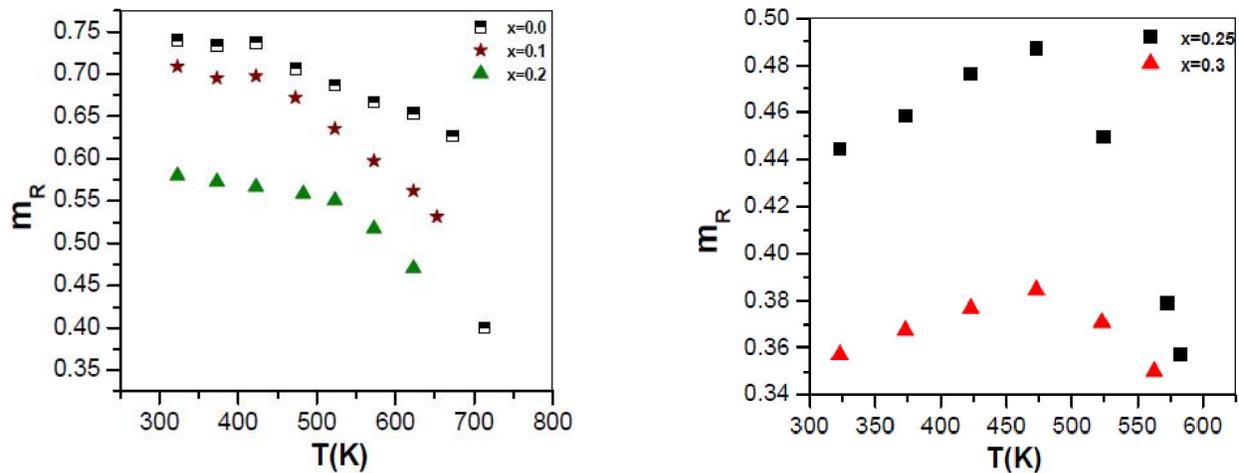


Figure 9 : The effect of magnetizing current on relative magnetization for  $\text{Cu}_{1+x}\text{Ge}_x\text{Fe}_{2-2x}\text{O}_4$  ( $0 \leq x \leq 0.3$ )

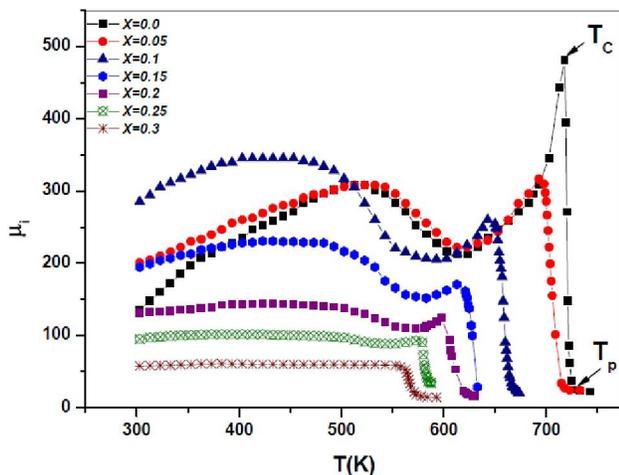


Figure 10 : Thermal spectra of the initial permeability of  $\text{Cu}_{1+x}\text{Ge}_x\text{Fe}_{2-2x}\text{O}_4$  ( $0.0 \leq x \leq 0.3$ )

number of  $\text{Fe}^{3+}$  ions will decrease on the B-site and increase on A-site. This replacement will weak the magnetization of the whole lattice.

The calculated (effective) magnetic moment  $\mu_{\text{eff}}$  has been determined for Cu-Ge ferrite from the magnetic moment for each site  $\mu_A$  and  $\mu_B$  as explained in our previous work<sup>[15]</sup> and in the reference of Oxide Magnetic Materials<sup>[18]</sup>. It is noticed that the addition of  $\text{Ge}^{4+}$  ions lead to decrease the magnetic moment in each site (A and B sites). According to Néel's molecular field model, the A-B interaction will be reduced. Therefore,  $\mu_{\text{eff}}$  decreases with increasing the non – magnetic ions of  $\text{Ge}^{4+}$  in the composition as shown in figure 7. From this figure,  $\mu_{\text{eff}}$  is similar to the behaviour of  $M_s(0\text{K})$  versus  $x$ . This means the calculation values of  $\mu_{\text{eff}}$  are proportional to the experimental data of saturation magnetization.

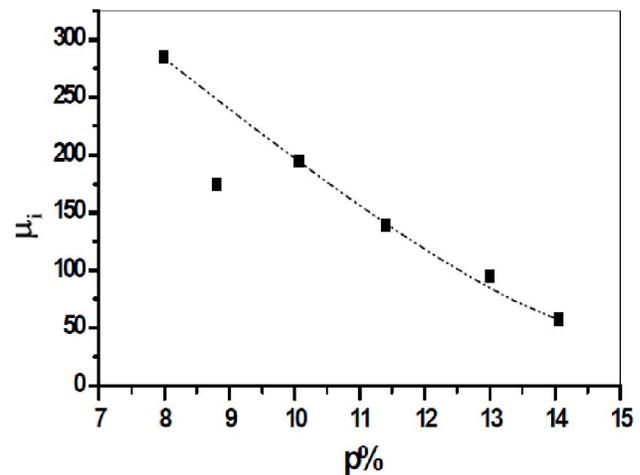


Figure 11 : The variation of initial permeability with percent-age porosity for Cu-Ge ferrite

The variation of coercive field  $H_c$  and the relative magnetization  $m_R$  with temperature are shown in figure 8 & 9, respectively for all values of  $x$ . It can be seen that  $H_c$  decreases gradually with increasing the temperature from  $x = 0.0$  to  $0.2$  as shown in figure 8a, but for  $x = 0.25$  and  $0.3$  there is sharp decreasing in  $H_c$  for  $T \geq 470\text{K}$  (Figure 8b). The relative magnetization  $m_R$  decreases with increasing  $T$  for  $x = 0 \rightarrow 0.2$ , but for  $x = 0.25$  and  $0.3$   $m_R$  increases with  $T$  for  $T \leq 470\text{K}$  and then decreases with further increasing the temperature. This means that  $m_R$  shows a maximum value at  $T = 470\text{K}$ . The behaviour of  $H_c$  (and also  $m_R$ ) could be explained through Brown's relation<sup>[21]</sup> which is given by:

$$H_c = \frac{2K_1}{\mu_0 M_s} \quad (3)$$

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where  $K_1$  is the anisotropy constant which varies with temperature tending to zero at the transition temperature  $T_c$ . It is known that the presence of  $Fe^{2+}$  ions in ferrites will affect on the crystal field and consequently on the anisotropy field<sup>[22]</sup>. Therefore, the decreasing in  $K_1$  and  $B_s$  (i.e.  $M_s$ ) with temperature will reflect on the decreasing behaviour of  $H_c$  and  $m_R (=B_r/B_s)$ .

### The initial permeability ( $\mu_i$ )

The initial permeability  $\mu_i$  of a ferromagnetic substance can be due either to a simultaneous rotation of the spins in each Weiss domain, or to a reversible displacement or bluging of domain walls. The initial permeability due to rotations is determined by the anisotropies<sup>[17]</sup>.

The initial permeability  $\mu_i$  and saturation magnetization  $M_s$  are related to the ionic structure and sensitive to the magnetic properties. It is well known that both quantities are complicated functions of temperature.

The thermal spectra of  $\mu_i$  versus temperature of  $Cu_{1-x}Ge_xFe_{2-2x}O_4$  are shown in Fig. 10. It is found that at room temperature, the initial permeability increases till  $x=0.1$  and then decreases with further increase of Ge concentration as tabulated in TABLE 1. The sample of  $x=0.1$  has the highest value of  $\mu_i$  while the lowest value of  $\mu_i$  at  $x=0.3$ .

It is noticed that  $\mu_i$  increases with increasing the temperature and falling abruptly close to Curie temperature,  $T_c$ . The Curie point  $T_c$  "the transition point from ferromagnetic to paramagnetic state" and the paramagnetic point  $T_p$  "when the sample becomes completely paramagnetic" were determined from the thermal variation of  $\mu_i$  vs.  $T$  as shown in figure 10. The transition temperature  $T_c$  and  $T_p$  are tabulated in TABLE 1. It was found that  $T_p$  is bit higher than  $T_c$  by about 15K. The two temperatures decrease with increasing Ge content. The presence of  $Ge^{4+}$  ion in the A-site is weakening the A-B interaction and causes a reduction in the molecular fields ( $H_m$ ) at the two sites. This leads to a decrease in various magnetic linkages, which mainly reduces  $H_m$  and the net result is a decrease in magnetization and consequently a decrease in the Curie temperature.

From the curves of initial permeability versus temperature, (Figure 10), it can be seen that, the specimens of  $x = 0.0$  up to 0.15 show a secondary peak

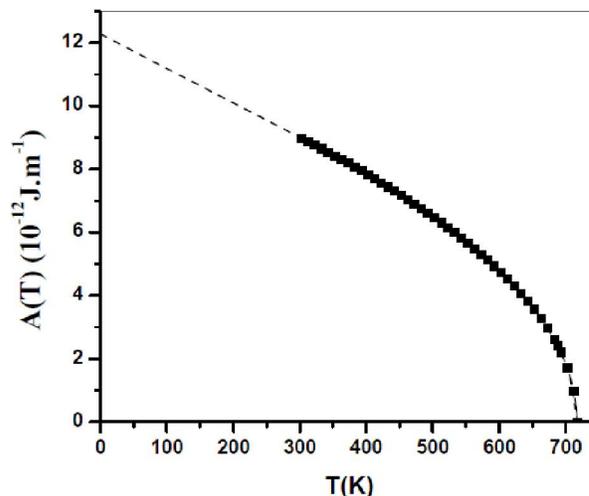


Figure 12 : The effect of temperature on the exchange constant for  $CuFe_2O_4$

(hump shape) superimposed upon a general rise, as the Curie point is approached. While the other samples of  $x = 0.2 - 0.3$ , show a monotonic stabilization of  $\mu_i$  with temperature and this behaviour is similar to the behaviour of a single domain<sup>[23]</sup>. The secondary permeability maximum (SPM) is occur due to the anisotropy constant  $K_1$ <sup>[24]</sup>, which varies with temperature tending to zero at the transition temperature  $T_c$ .

It is known that the initial permeability  $\mu_i$  is believed to arise as the result of reversible displacements of magnetic domain walls within the material. Rotation of spins within a domain, referred to as domain rotations, contribute little, on account of the relatively high magnetocrystalline anisotropy<sup>[18]</sup>. Figure 11 shows the relation between  $\mu_i$  and porosity. It is observed that the initial permeability decreases with increasing porosity. This behaviour may be related to samples whose porosity is too great (i.e. contain many pores) which hinder the movement of the domain wall causing the decrease in  $\mu_i$ . Also, the effect of grain diameter on the magnitude of the initial permeability is shown in TABLE 1. It is observed that the initial permeability increases with further increasing in grain diameter for  $D \geq 15 \mu m$ . This behaviour may be attributed to below 15 microns; the decrement in  $\mu_i$  may be related to the pores that occur in the crystals, and these, limit the permeability. Then for larger grains, the permeability increases due to domain wall displacements.

The magnitude of the initial permeability of a magnetic material as a result of rotations is proportional to

the square of the saturation magnetization and inversely proportional to the magnetic anisotropy energy according to the relation<sup>[25]</sup>:

$$\mu_i = \frac{M_s^2 \cdot D}{\sqrt{K_1}} \quad (4)$$

From this relation we calculate the value of the anisotropy constant  $K_1$  for each composition.

Guyot and Globus<sup>[26,27]</sup> have established a direct relation between the hysteresis losses and the domain wall energy. The domain wall energy is calculated according to the relation:

$$\sigma_w = 2\sqrt{AK_1} \quad (5)$$

where: A is the exchange constant, which is calculated from the relation<sup>[26]</sup>:

$$A(T) = \frac{k_B T_C}{a} \left( 1 - \frac{T}{T_C} \right)^{1/2} \quad (6)$$

The term (kb) is known the exchange energy stored in the transition layer resulting from the spin interaction<sup>[24]</sup>, and  $a$  is the lattice constant. The Right H.S. vs. T is represented in figure 12 for  $x = 0.0$  as a representative example. The extrapolated experimental value of the thermal variations of A(T) i.e. the values of A(0) are given in TABLE 1 and very coincident with the calculated values.

If  $\delta_w$  is thickness of the domain wall, the total wall energy per  $\text{cm}^2$  is given by:

$$\sigma_w = K_1 \delta_w \quad (7)$$

Form the two relations (5&6),  $\delta_w$  was calculated and tabulated in TABLE 1 for each composition. It is found that all the obtained results are in good agreement with the values in the literature<sup>[17,24]</sup>.

## CONCLUSION

The decrease in magnetization and Curie temperature with the addition of Ge content is suggested to be due to the decrease in A-B interaction according to Néel's molecular field theory.

The two parameters coercive field  $H_C$  and the relative magnetization  $m_r$  decrease with increasing temperature. This behaviour is explained through Brown's relation.

The thermal analysis of  $\mu_i$  show a secondary peak

(hump shape) below  $T_C$  for  $x=0.0$  up to 0.15. While the other samples of  $x=0.2, 0.25$  and  $0.3$  show a monotonic stabilization behaviour of  $\mu_i$  with T.

The domain wall energy and domain wall thickness were in the order of  $10^{-3} \text{ J/m}^3$  &  $10^{-8} \text{ m}$ , respectively and were dependent on Ge concentration.

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