Global Scientific JOURNALS

GSJ: Volume 8, Issue 1, January 2020, Online: ISSN 2320-9186

www.globalscientificjournal.com

Structure and Magnetic Properties of Copper, Gallium Substituted Manganese Ferrites.

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Abstract

Manganese Ferrites $MnFe_2O_4$and the system $Mn_{1-x}Cu_xFe_{1.7}Ga_{0.3}O_4$($x$ = 0, 0.25, 0.5, 0.75 and 1) were prepared by solid -state reaction method. Where, the effect of replacing Cu^{2+} for manganese as well as substituting iron with Ga^{3+}on the physical properties were investigated. X-ray diffraction measurements clearly showed the formation of single phase cubic spinel structure(fd_3m)for all the prepared samples.The compositions $Cu\ Fe_2\ O_4$and $CuFe_{1.7}Ga_{0.3}O_4$ were formed with a tetragonal structure with space group ($I4_1/amd$). The magnetic properties were studied using vibration sample magnetometer (VSM) and the magnetic parameters were determined. The saturation magnetization (M_S) was found to increase up to Cu content x = 0.5 and then gradually decrease with further increase of x content.

1. Introduction*:*

Spinel ferrite is one of the most important classes of magnetic ceramic materials owing to their interesting applications. In spinel structure, the magnetic ions are distributed among two different lattice sites, tetrahedral (A) and octahedral (B) sites. The magnetic as well as the electrical properties of these ferrites depend on the relative distribution of cations at these two sites as well as the preparation condition [1]. The magnetization of either site can be reduced relative to the other one, by substitution of non-magnetic ions in the corresponding sites.

It is well known that $MnFe_2O_4$ material has inverse spinel structure because 80% of Mn ions occupy tetrahedral site (A-site), which is surrounded by four O^{2-} ions, and the left 20% of Mn ions occupy octahedral site (B-site), which is surrounded by six O^{2-} ions [2,3].

2. Experimental:

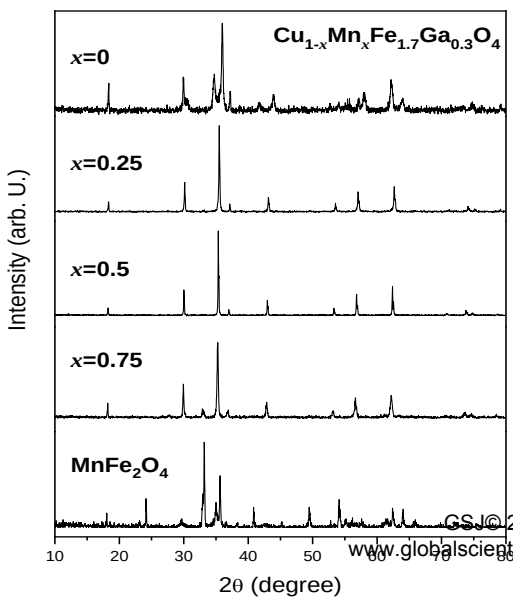
High purity samples of the chemical $MnFe_2O_4$ and the system $Mn_{1-x}Cu_xFe_{1.7}Ga_{0.3}O_4$($x$ = 0, 0.25, 0.5, 0.75 and 1) were synthesized through standard solid-state reaction method using Fe_2O_3, MnO, CuO, and Ga_2O_3 (purity 99.99%). The mixture of the oxide powders is preheated at (975^{o}C) for 24 h. Then the product is reground and heated again at the same conditions to improve homogeneity. The final powders are heated at (925–1470^{o}C)depending on the Mn content (x), and then slowly cooled to room temperature. X-ray diffraction (XRD) patterns were recorded using Philips diffractometer with CuKα radiation. Magnetic measurements on the prepared ferrites were carried out using vibrating sample magnetometerVSM (9600-1 LDJ, USA) with a maximum appliedfield of nearly 20 kG at room temperature.

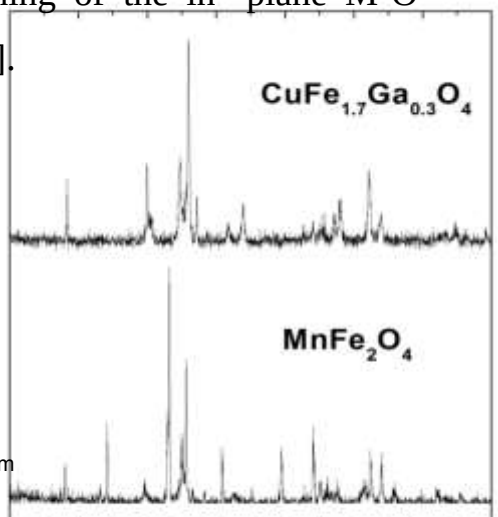
3. Results and discussion:

3.1. XRD measurements:

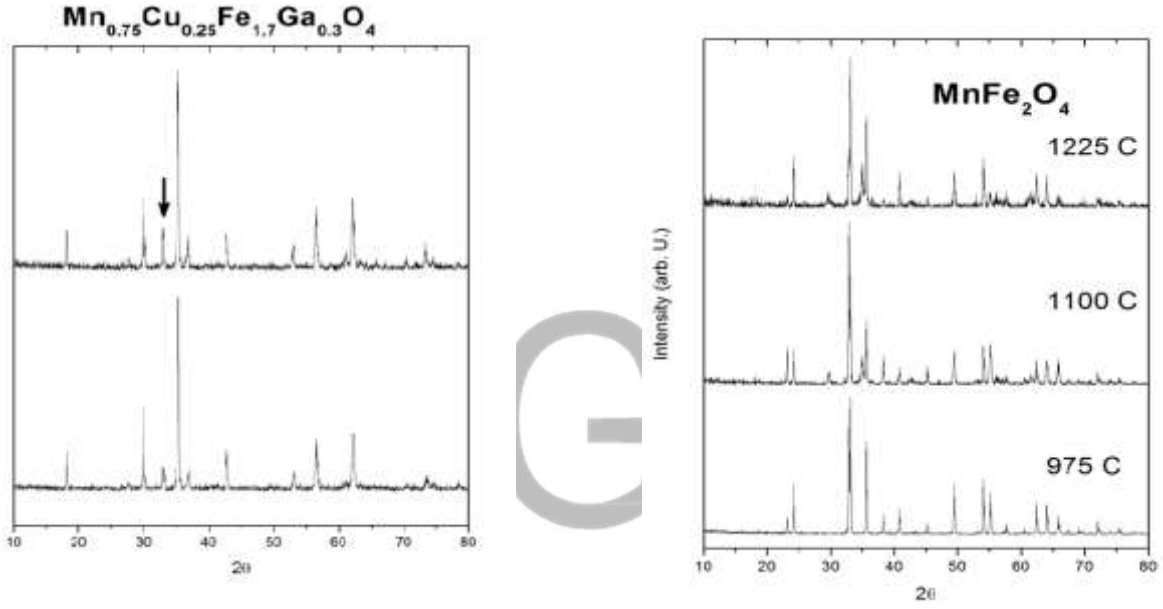
XRD patterns of the prepared samples, $Mn_{1-x}Cu_xFe_{1.7}Ga_{0.3}O_4$($x$ = 0, 0.25, 0.5, 0.75 and 1) are displayed in Fig. 1. XRD measurements have shown that, the nominal composition structures with different concentrations are single phase with no additional lines corresponding to any other phase. Analysis of thediffracted peaks by modified Gaussian functions, Fig.1 (a), (b)indicates that, allprepared samples are cubic structure with space group (fd_3m) except the compositions (Cu Fe_2 $O_4$and $CuFe_{1.7}Ga_{0.3}O_4$) where found a tetragonal structure with space group ($I4_1/amd$).Additional lines corresponding to other phaseare found in samples with high Mn concentrations shown in Fig.1 (c). These samples are annealed at different temperatures (925-1225 ^oC) to reach a single phase composition for these samples, as illustrated in Fig.1 (d).

Tetragonal structure with space group (I41/amd) is obtained for $CuFe_2O_4$and$CuFe_{1.7}Ga_{0.3}O_4$, with c/*a*4> (~1.4). This can be explained as: the metal sites in most transition metal oxides (TMO) system have octahedral site symmetry (D_h), in which each TM ion is surrounded by six oxygen ions (MO_6).The point symmetry of the TM ion is reduced from cubic to tetragonal (D_{4h}) due to the lattice distortion with the out of phase M-O bonding length and the shortening of the in- plane M-O one [4].





(a) (b)



(c) (d)

Fig.1. (a)X-ray powder diffraction patterns of $Mn_{1-x}Cu_xFe_{1.7}Ga_{0.3}O_4$, and $MnFe_2O_4$.

(b) The samples $MnFe_2O_4$ has cubic phase(fd_3m) and $CuFe_{1.7}Ga_{0.3}O_4$has a tetragonal phase ($I4_1/amd$).

(c) The samples $Mn_{0.75}Cu_{0.25}Fe_{1.7}Ga_{0.3}O_4$ has impurity peak.

(d) The sample $MnFe_2O_4$ were fired at different temperature (975-1225 C^o).

The values of lattice parameters were obtained for all the samples using XRD data as follow:

- The lattice parameter values (a)was determined for cubic samples using the equation:

$$\frac{1}{d^2} = \frac{(h^2 + k^2 + l^2)}{a^2}$$

Where, d is the inter planner distance, and (h, k, l) are the Miller indices.- For tetragonal samples, to calculate a, c lattice constant, we use the equation:

$$\frac{1}{d^2} = \frac{(h^2 + k^2)}{a^2}\frac{l^2}{c^2}$$

It is observed that the lattice parameter decreases with increasing Mn content (x) as shown in Fig. (2). This behavior of lattice parameter with Mn content (x) is explained on the basis of difference in ionic radii of Mn^{2+}(0.70 Å) and Cu^{2+}(0.724 Å).

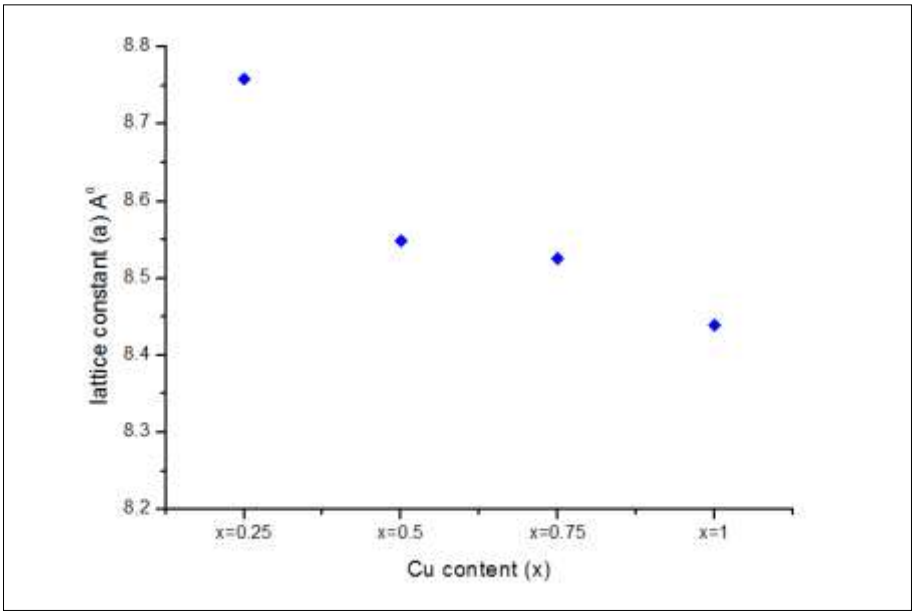


Fig. 2. The variation of lattice parameter (a) with Mn content (x).

	Lattice constant	Lattice constant	*c/a*

Sample	*a* (A°)	*c* (A°)	
$CuFe_2O_4$	5.8975	8.6974	1.4747
$CuFe_{1.7}Ga_{0.3}O_4$	6.1873	8.7652	1.4166
x=0.25	8.75837	----	----
x=0.5	8.54862	----	----
x=0.75	8.52482	----	----
x=1	8.43888	----	----

Table.1 the calculated lattice parameter a and c.

3.2. Magnetic properties measurements:

Magnetization measurements of $Mn_{1-x}Cu_xFe_{1.7}Ga_{0.3}O_4$ ($0 \leq x \leq 1$) samples were carried out using a vibrating sample magnetometer (VSM) at room temperature. The hysteresis loops were found to be well saturated with the available applied field. The obtained hysteresis loops are shown in Fig. 3.

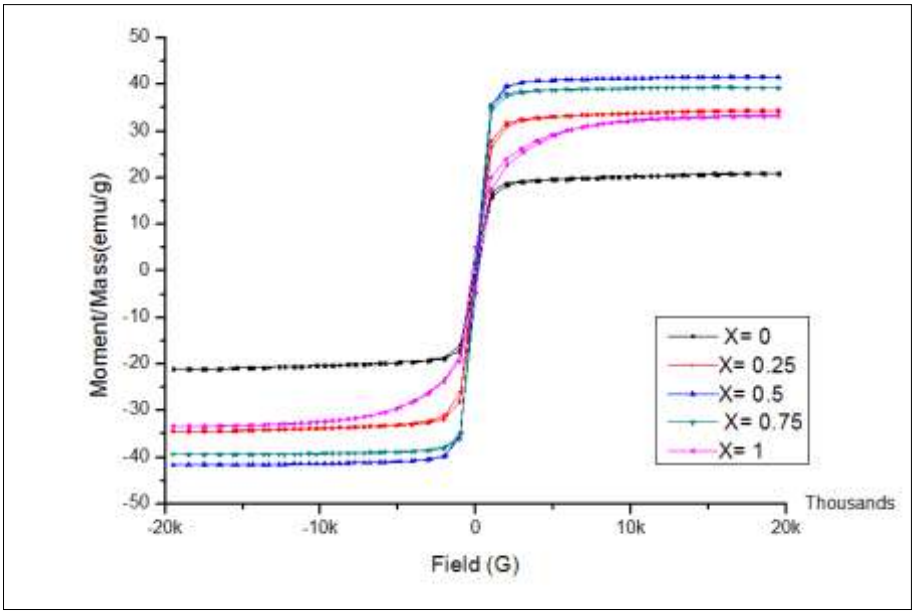


Fig. 3. Hysteresis loops of samples.

The hysteresis loops display the characteristics of soft-magnetic materials. The saturation magnetization (M_S) was found (from 14.071 to 41.672ηB). We found that, M_S increase with Mn content (x) up to x = 0.5 and then gradually decrease with further increasing of Mn content as shown in Fig. 4. This is due to Ga^{3+} doping and the change in the particle

size D. The experimental magnetic moment (ηB) is determined from the saturation magnetization data using the following relation [1]:

$$\eta_B = (M_W \times M_S)/5585 \qquad [1]$$

Where, M_W is the molecular weight of the sample and M_S is the saturation magnetization in emu/g.

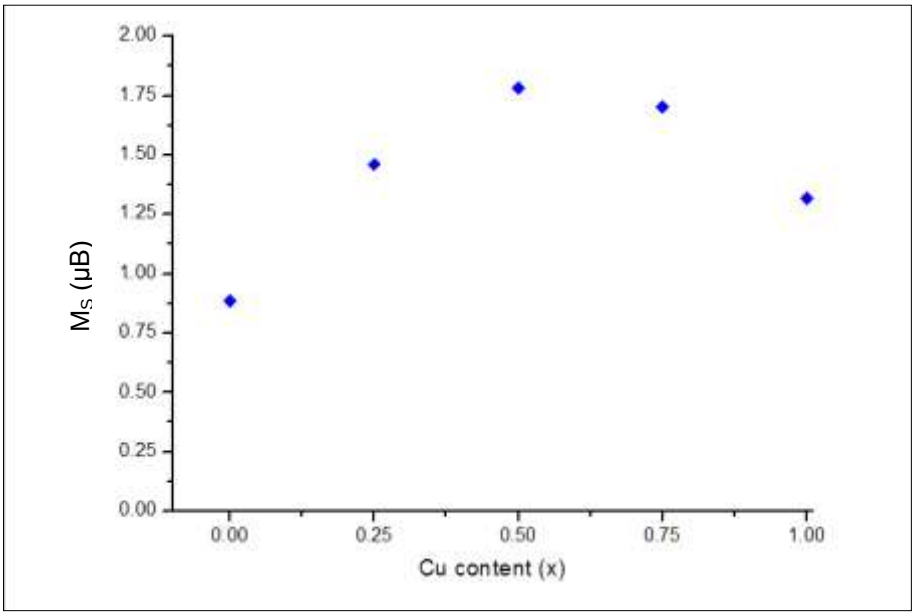


Fig. (4) Shows the variation of Saturation magnetization (μB) with Cu content x.

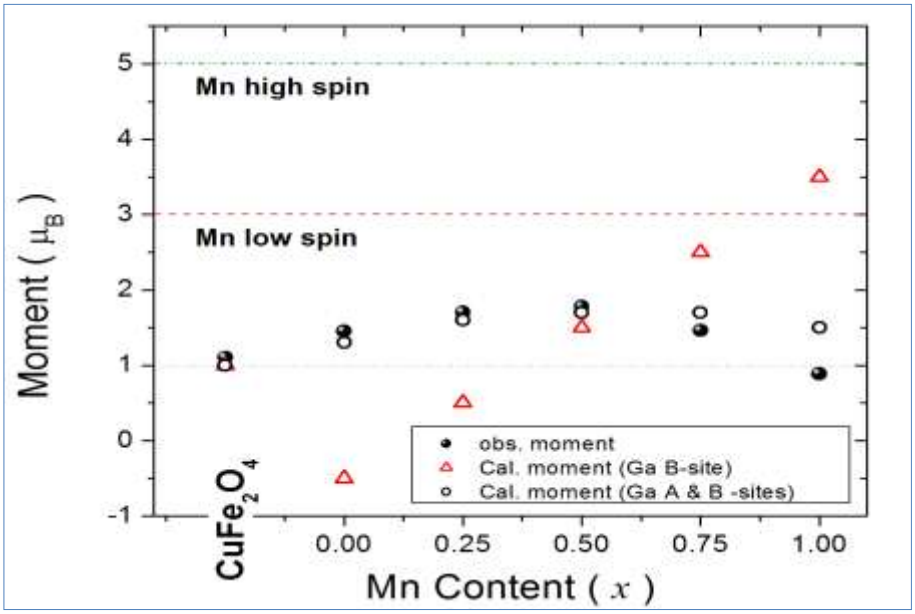


Fig. 5. The theoretically calculated moments according to Neel model and depending on different possible cation distribution.

Also the observed moments were compared to the theoretically calculated moments according to Néelmodel and depending on different possible cation distribution. The results clearly suggest that Mn^{2+} ions are in the low spin state as shown in fig. 5. The cation distribution calculated theoretically using η_B values of the composition, and we found that Ga^{3+}cation distributed in both A- Site and B- Site and this was in agreement with the observed data at fig. 5.

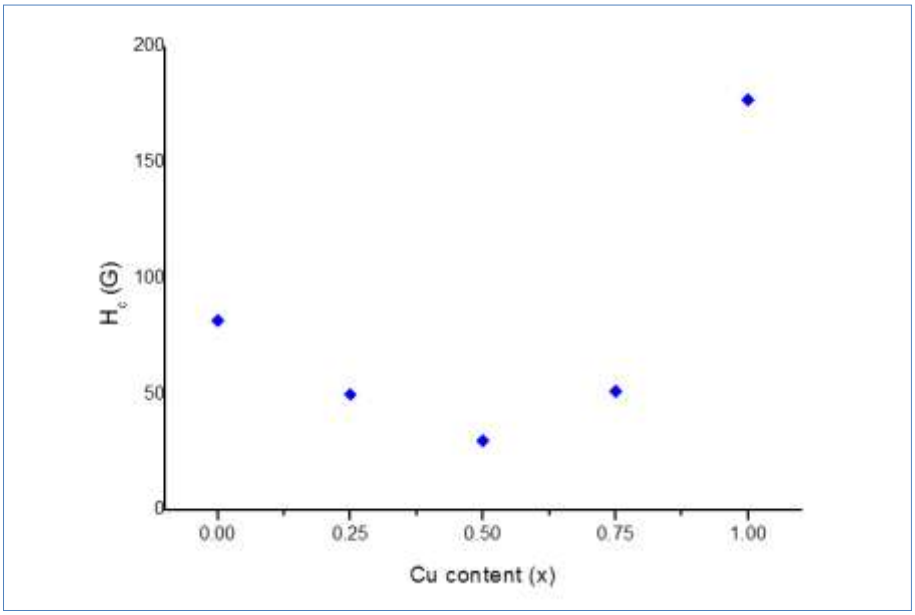


Fig. 6. TheCoercivity (H_c) of all samples.A strong dependence on *x* was observed. The maximum M_s is in coincidence with the minimum H_c at *x*=0.5.

Fig. 6.Indicatedthe coercivity (Hc),which found in the range (from30.017 to 177.23 G). Hcdecrease with Mn content (x) up to x= 0.5, then increase with further increase of Mn content.

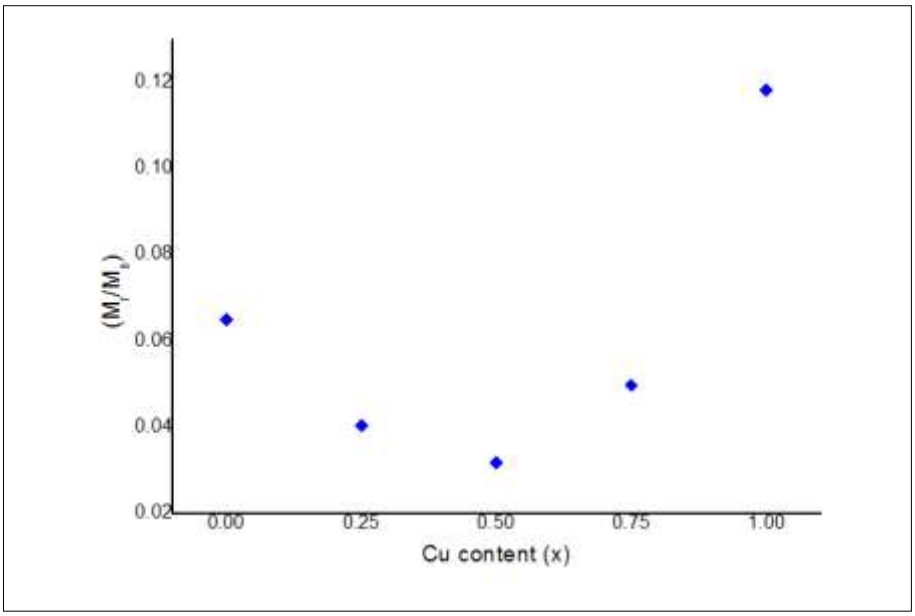


Fig.7. The (M_r/M_S) for all samples.

The retentivity magnetization (M_r) was found (from 0.02082 emu/g to 0.1183emu/g) for the studied samples. Fig.7.Shows that (M_r/M_s) decrease with Mn content up to x=0.5, then increase as x increase.

Conclusion:

From XRD analysis a single phase spinel ferrite structure in all the prepared ferrite compositions in cubic phase with space group (fd_3m) except two samples ($Cu\ Fe_2\ O_4$, $CuFe_{1.7}Ga_{0.3}O_4$) which are tetragonal structure with space group ($I4_1/amd$). Thecalculated lattice parameter was found to decrease with increasing Mn content (x). The saturation magnetization (M_s) was found to increase as Mn content (x) increase up to x=0.5 and then gradually decrease, in contrast the Coercivity (H_c) were found to decease with x content increasing up to 0.5, then decease.The calculated cation distribution using η_B values of the composition showed that, Ga^{3+} cation distributed in both A- Site and B- Site

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