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ARTICLE

Structural and Magnetic Properties of $(Ni_{1-x}Mg_x)\ Fe_2O_4$ Ferrites

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Abstract: We have studied the structural and magnetic properties of $(Ni_{1-x}Mg_x)Fe_2O_4$ ferrite. X-ray diffraction and magnetization measurements were performed for all samples. X-ray results showed that a single phase ferrite was obtained for all samples. The lattice parameter, *a*, was estimated from the X-ray results and we found that the lattice parameter increases slightly at high Mg^{2+} contents ($x > 0.4$). Magnetic hysteresis measured for all samples showed that the coercivity (H_c) shows an appreciable change at low ($x = 0.2$) and high ($x = 1.0$) Mg^{2+} content. It was also found that the saturation magnetization (M_s) and the remanence (M_r) decreases with increasing Mg.

Keywords: Nickel ferrite; Lattice parameters; Magnetic hysteresis; Cationic distribution.

Introduction

Ferrite magnetic materials are among the most important materials used today in modern technology. They are used as an important part in many applications as in wave applications, radio electronics and sensors [1-4].

The physical properties of ferrites are dependent on several factors such as preparation method, sintering process, and the kind and amount of constituent elements or additives [5]. The effect of various cations substitution on the structural, electrical, and magnetic properties of ferrites was the subject of extensive research work [6-13].

Many researchers studied the spinel oxides MFe_2O_4 ferrites (where M is a divalent atom like Zn, Mg, Mn, Co, Ni etc.). These ferrites are usually denoted by AB_2O_4, where A and B refer to the tetrahedral and octahedral sites respectively in the oxygen lattice. Cations distribution in spinel ferrites depends on many factors, such as ionic radii and electronic configuration. This distribution affects the microstructure and the magnetic properties of these ferrites. The effect of cations distribution on spinel ferrites was studied widely and their structural and magnetic properties were also studied [7-14].

Ni ferrite ($NiFe_2O_4$) is considered one of the most widely studied spinel ferrites due to its high Curie temperature ($T_c = 585\ ^oC$), a fact which allows the use of this ferrite in room temperature applications. The aim of this work is to prepare a single phase $NiFe_2O_4$ ferrite, and then study the effect of Mg substitution on the structural and magnetic properties of the prepared samples.

Experimental Techniques

Sample Preparation

The system we prepared for this study was $(Mg_xNi_{1-x})(Fe_2O_4)$, where x = (0.0, 0.2, 0.4, 0.6, 0.8, 1.0). The conventional double sintering ceramic technique was used to prepare the samples. NiO, MgO and Fe_2O_3 powders with 99% purity or better were

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used. A digital analytical balance was used to measure the mass of the powder needed according to the required stoichiometry. The weighed powders were mixed using ceramic pestile and mortar**.** The samples were annealed at 1100˚C for 10 hours. The samples were cooled slowly while it is inside the furnace. The process was repeated once more.

X-Ray Diffraction (XRD)

X-ray diffraction system was used to check the structure of the prepared samples. Cu-K$_\alpha$ radiation with λ = 1.54 Å was used. The diffraction patterns were collected and from the peak positions, each corresponding to a set of (h, k, l) planes, we estimated the d spacing between planes using Bragg's law,

$$2d\sin\theta = n\lambda \qquad (1)$$

The lattice parameter a for the cubic ferrite structure can be estimated using the relation:

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

The X-ray density for all samples prepared was calculated using the equation:

$$d_x = \frac{8M}{N_A a^3} \qquad (3)$$

where M is the molar mass of the ferrite, a is the lattice parameter, and N_A is Avogadro's number.

Magnetic Measurements

The Vibrating Sample Magnetometer (VSM) with a maximum field of 1.5 T was used to study the magnetic behavior of the prepared samples. The initial magnetization curves and the hysteresis loops were measured for all samples.

Results and Discussion

Structure of $Ni_{1-x}Mg_xFe_2O_4$ System

The XRD patterns confirmed the formation of the single spinel phase for all samples. The measured XRD patterns, for all samples (x = 0, 0.2, 0.4, 0.6, 0.8, 1) are shown in Fig. 1 and Fig. 2.

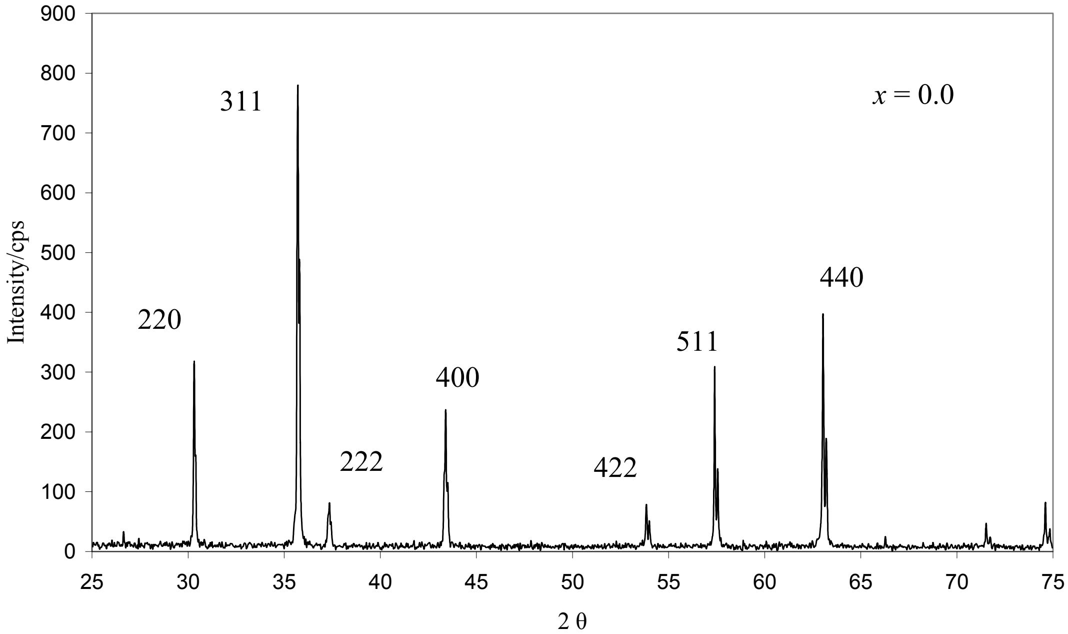

FIG.1. X-ray diffraction for $NiFe_2O_4$ (x = 0) sample

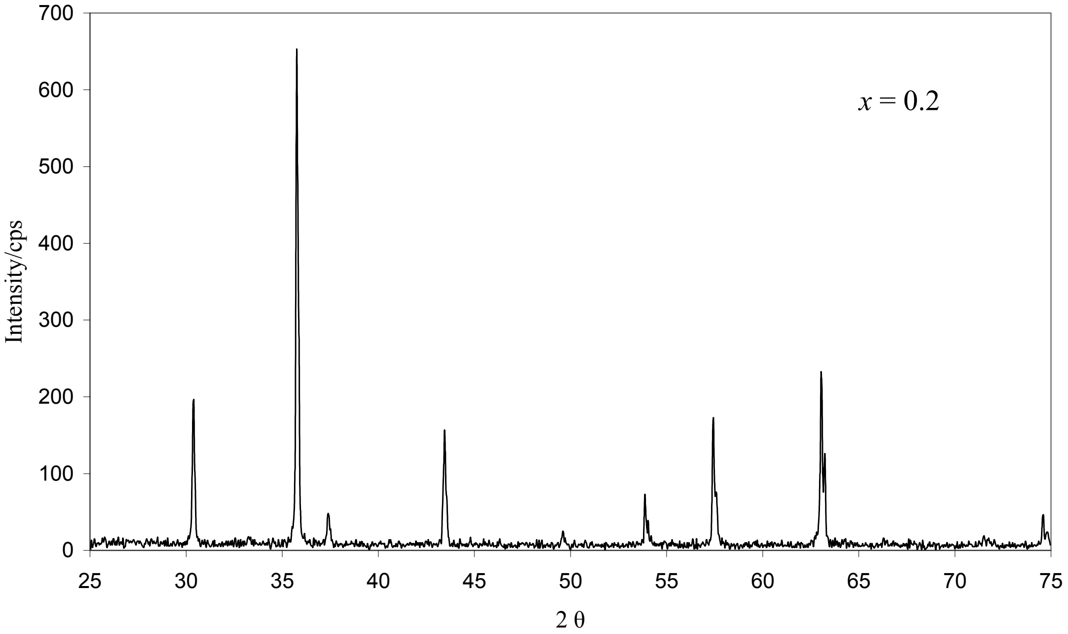

FIG.2(a). X-ray diffraction for $x = 0.2$ sample

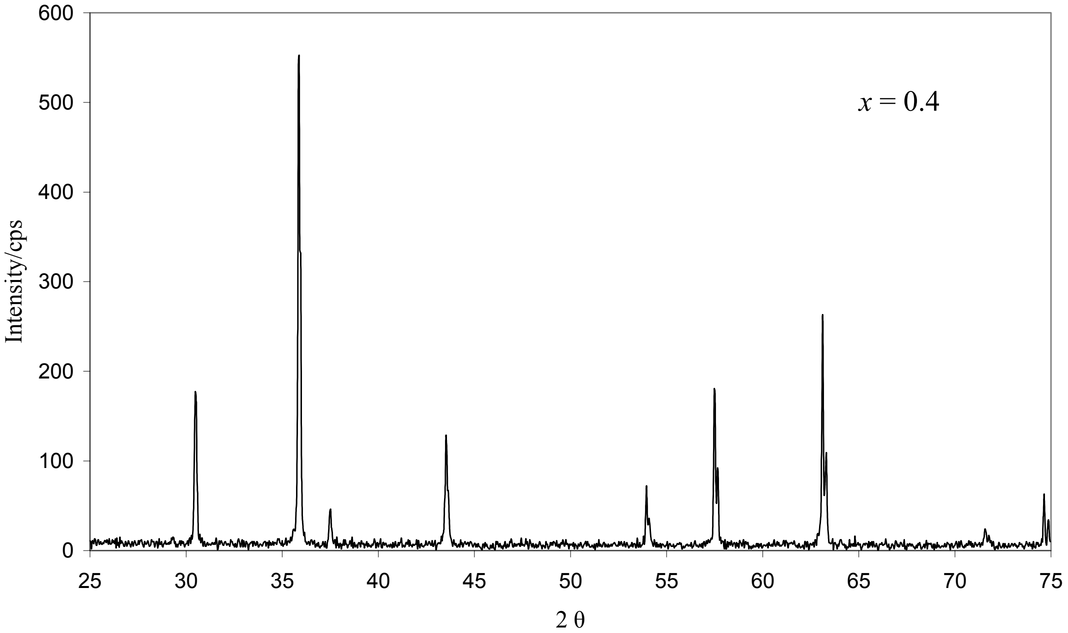

FIG.2(b). X-ray diffraction for $x = 0.4$ sample

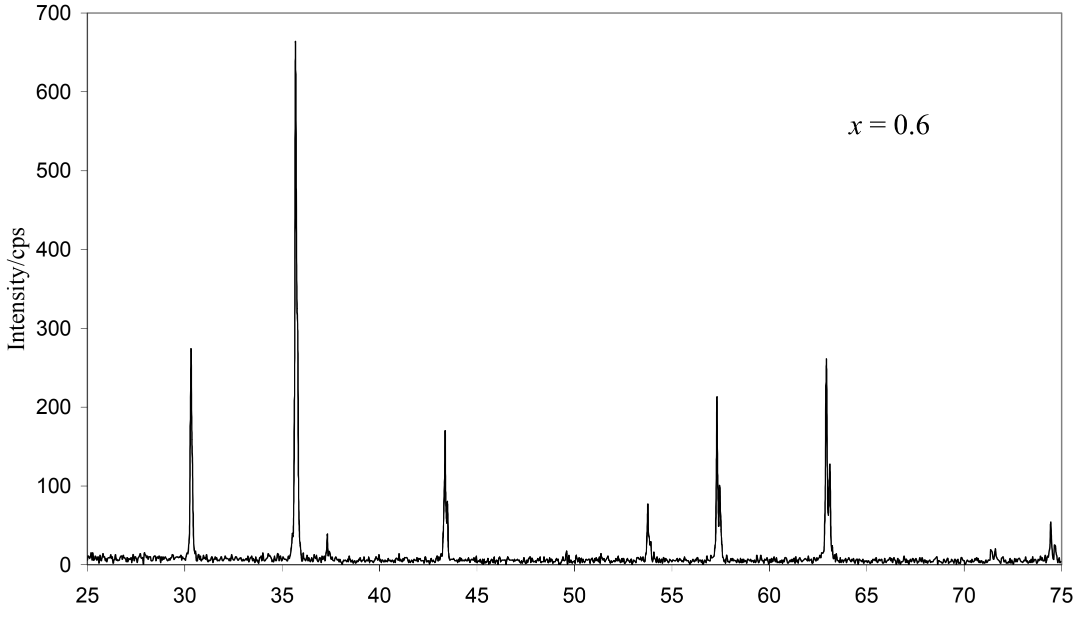

FIG.2(c). X-ray diffraction for x=0.6 sample

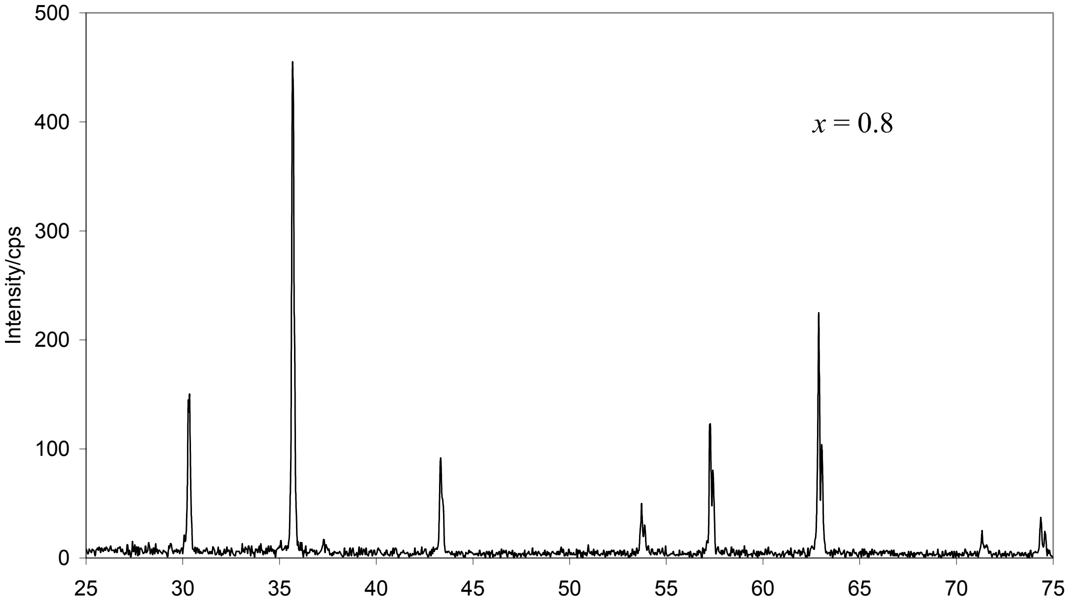

FIG.2(d). X-ray diffraction for x = 0.8 sample

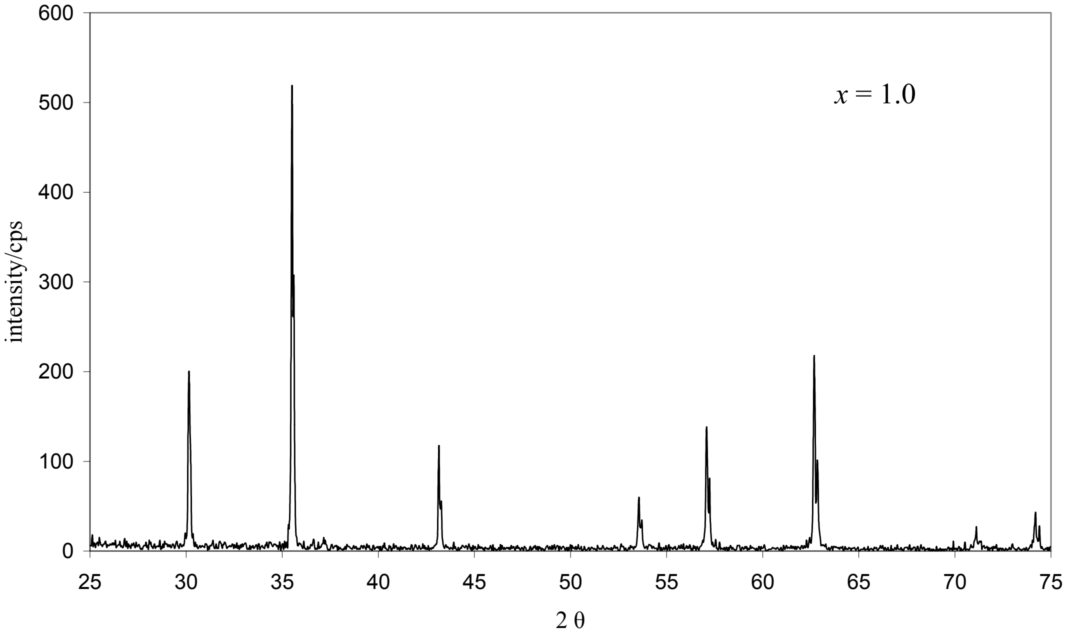

FIG.2(e). X-ray diffraction for $x = 1.0$ sample

The replacement of cations by others in the spinel lattice can change the lattice parameter, depending on their radii and whether they are entering the tetrahedral or octahedral sites. It is known that the replacement of cations by larger ones in the spinel lattice causes an increase in the lattice parameter, as is the case of $Cd_{1-x}Co_xFe_2O_4$ ferrites [14]. Fig. 3 shows the dependence of the lattice parameter on the x value (Mg content). The figure shows that the lattice parameter decreases firstly and then increases for $x \geq 0.6$ (see table (1)). Ni^{2+} ion radius is 0.69 Å, whereas the radius of an Mg^{2+} is 0.72 Å, therefore, the lattice parameter is expected to increase as the Ni^{2+} ions on the octahedral sites are replaced by the Mg^{2+} ions.

From table (1) we see that, the replacement of Ni^{2+} ion by Mg^{2+} on the octahedral sites causes a decrease in the lattice parameter for $x \leq 0.4$, and this is probably due to the distortion induced in the ferrite as a result of the vacancies on the O^{2-} sites, due to the larger radius of Mg^{2+} compared to that of Ni^{2+}.

The X-ray density was also calculated using equation (3), and it is shown in table (1). The decrease in X-ray density values with increasing Mg content shown in the table resulted from the replacement of the Ni^{2+} ions on the octahedral sites and/or the replacement of the Fe^{3+} ions on the tetrahedral sites in the conventional cubic cell by the lower-mass Mg^{2+} ions.

TABLE 1. The structural and magnetic parameters for $Mg_x\ Ni_{1-x}(Fe_2\ O_4)$ system

x	Lattice Parameter (Å)	Density (g/cm^3)	M_r (emu/g)	M_s (emu/g)	H_c (Oe)
0	8.331	5.38	4.4	35.7	151
0.2	8.317	5.29	4.3	34.1	193
0.4	8.290	5.23	2.9	30.2	152
0.6	8.335	5.04	3.1	27.1	145
0.8	8.335	4.92	2.8	21.2	154
1	8.372	4.75	2.8	21.3	115

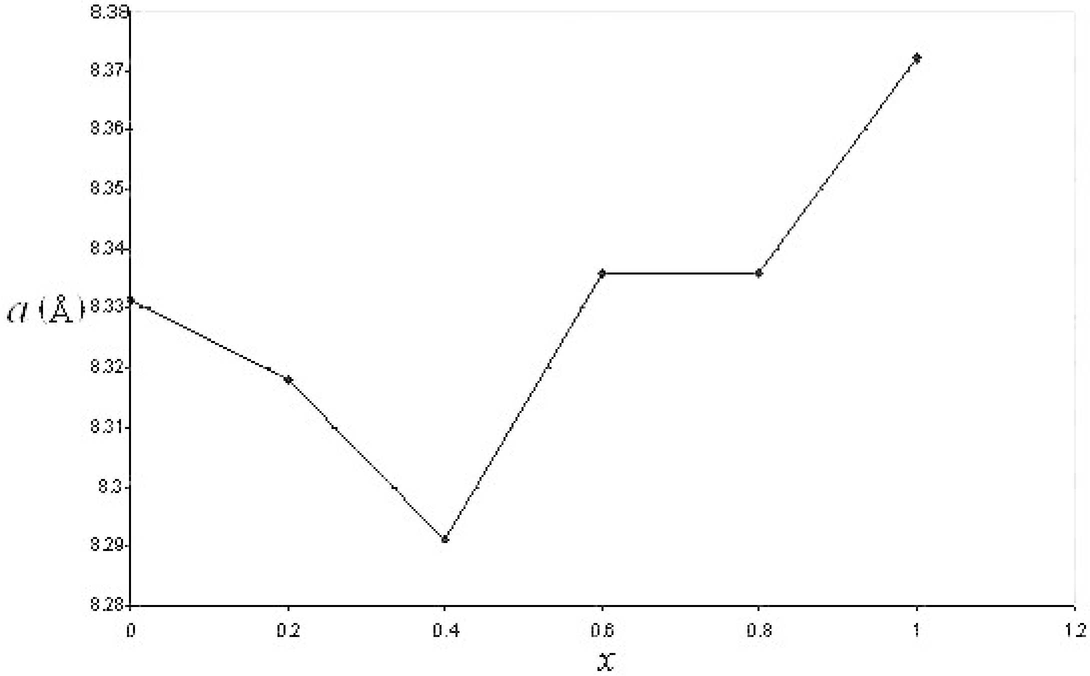

FIG.3. The lattice parameter (*a*) versus *x*

Magnetic Behavior of $Ni_{(1-x)}Mg_xFe_2O_4$ System

We measured the room temperature magnetic hysteresis for all the samples. The hysteresis loop for the base sample ($NiFe_2O_4$) is shown in Fig. 4. From the measured loop we obtained the magnetic parameters, M_s = 35.7 emu/g, M_r = 4.4 emu/g and H_c = 151 Oe. The hysteresis loops for the other samples x = (0.2, 0.4, 0.6, 0.8, 1.0) are shown in Fig. 5. The maximum applied magnetic field used in our study was (15 kOe).

At small Mg^{2+} contents (x = 0.2), a relatively wider loop (H_c = 193 Oe) was observed compared with the ($NiFe_2O_4$) base sample, and this is probably due to the expected larger flux pinning when a less symmetrical structure resulted when a larger radius Mg^{2+} ions replaced the Ni^{2+} ions. At higher Mg^{2+} contents ($0.4 \leq x \leq 0.8$) no appreciable change in H_c was observed. For x = 1.0 (the sample that Mg totally replaced Ni) the coercivity decreases sharply compared with all other samples (Fig. 6).

The remanence and saturation magnetization for all the samples studied decrease with increasing Mg^{2+} as shown in Fig. 7 and Fig. 8, respectively. The saturation magnetization of the ferrite system can be estimated theoretically assuming total parallel and anti-parallel alignments for the ions from the moment in each ion, the distribution of the ions between the tetrahedral A site and the octahedral B sites, and by knowing that the exchange interaction between A and B sites is negative. The AB interaction is the strongest compared with the AA and BB interactions, so that all the A moments are parallel to one another and anti-parallel to the B moments.

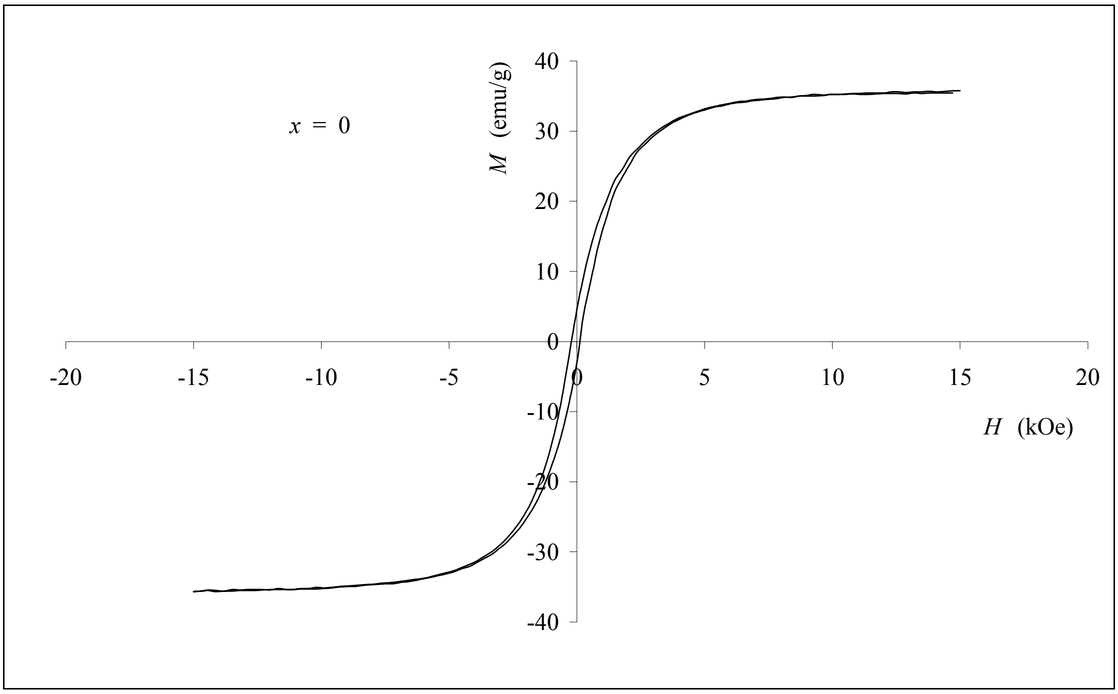

FIG.4. The hysteresis loop for $NiFe_2O_4$ ($x = 0$) sample

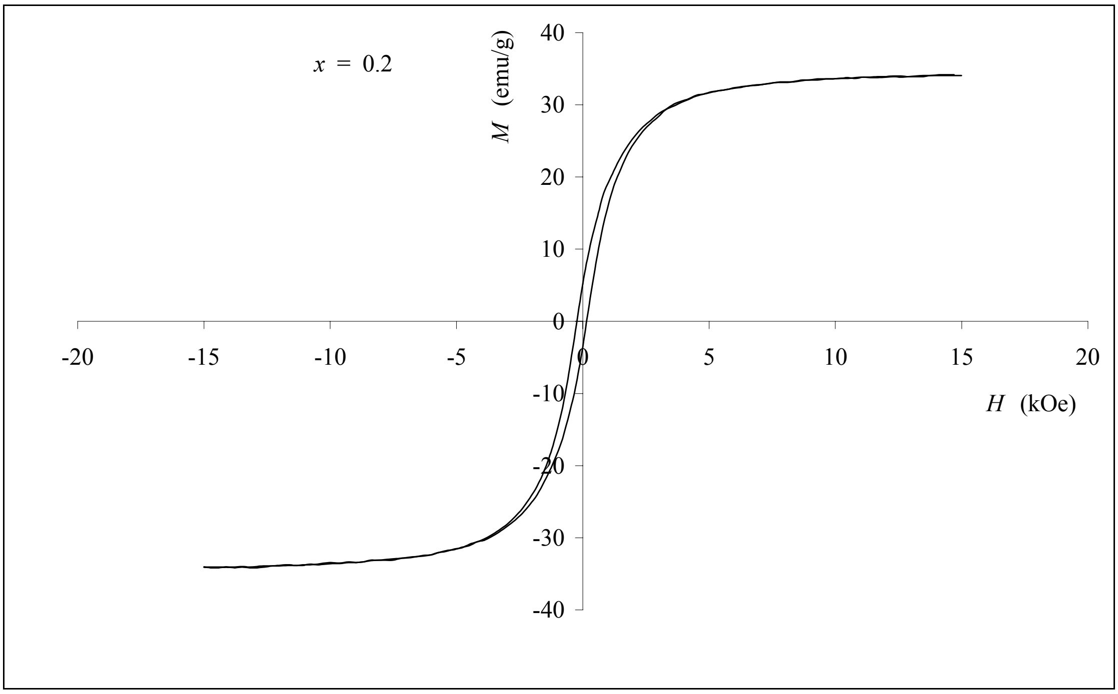

FIG.5(a). The hysteresis loop for the $x = 0.2$ sample

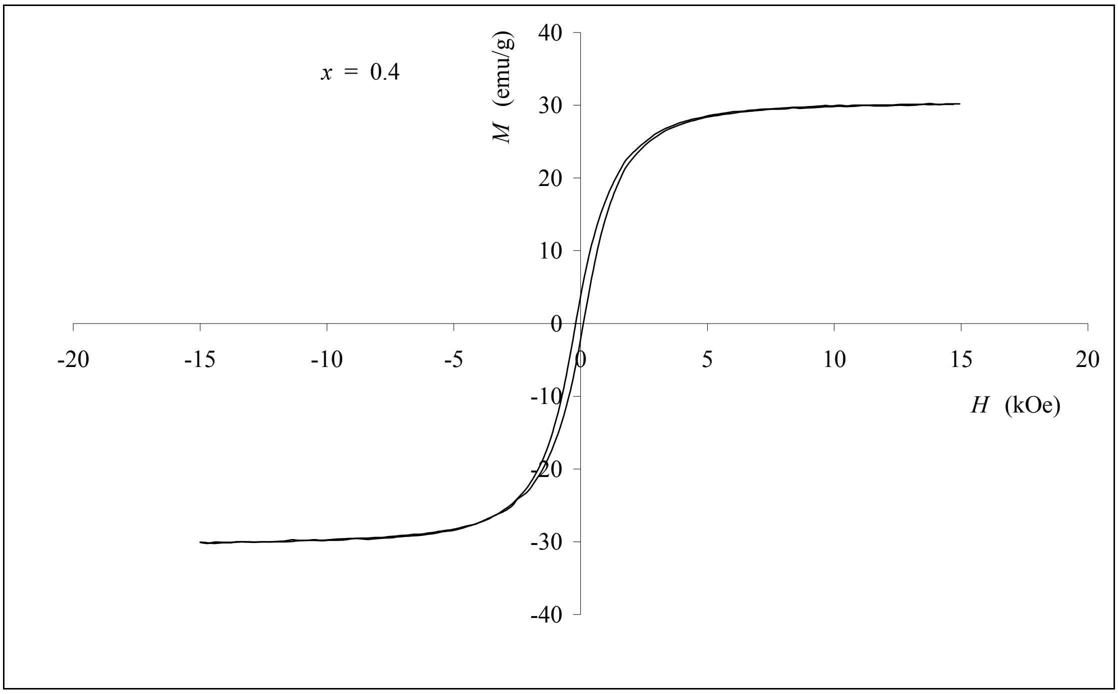

FIG.5(b). The hysteresis loop for the $x = 0.4$ sample

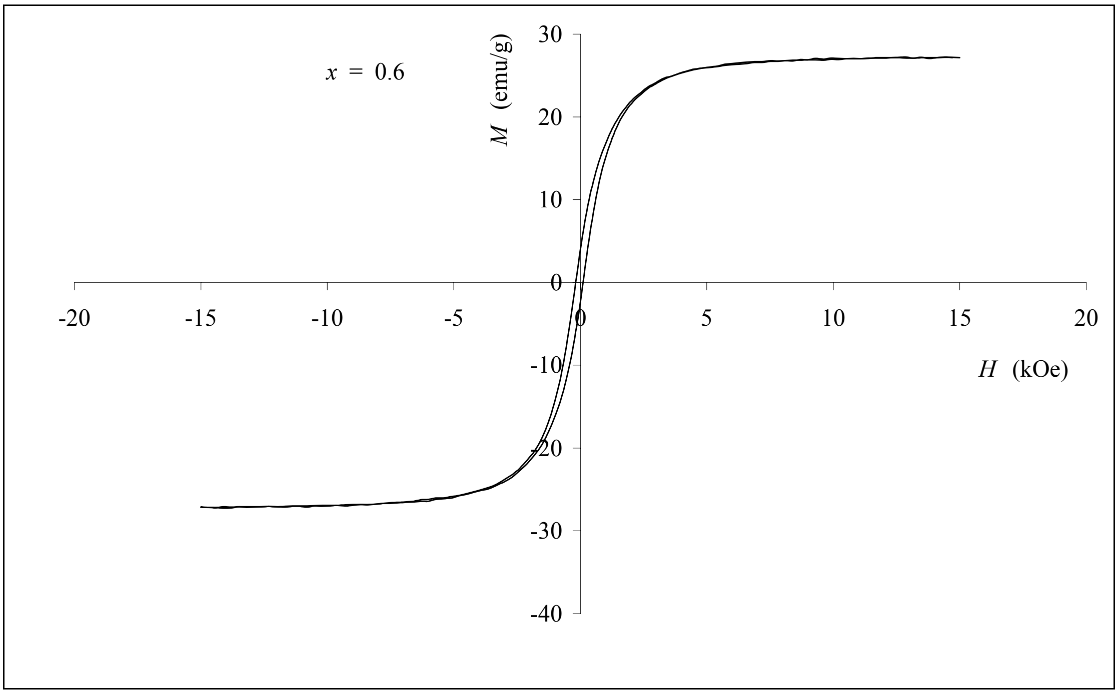

FIG.5(c). The hysteresis loop for the $x = 0.6$ sample

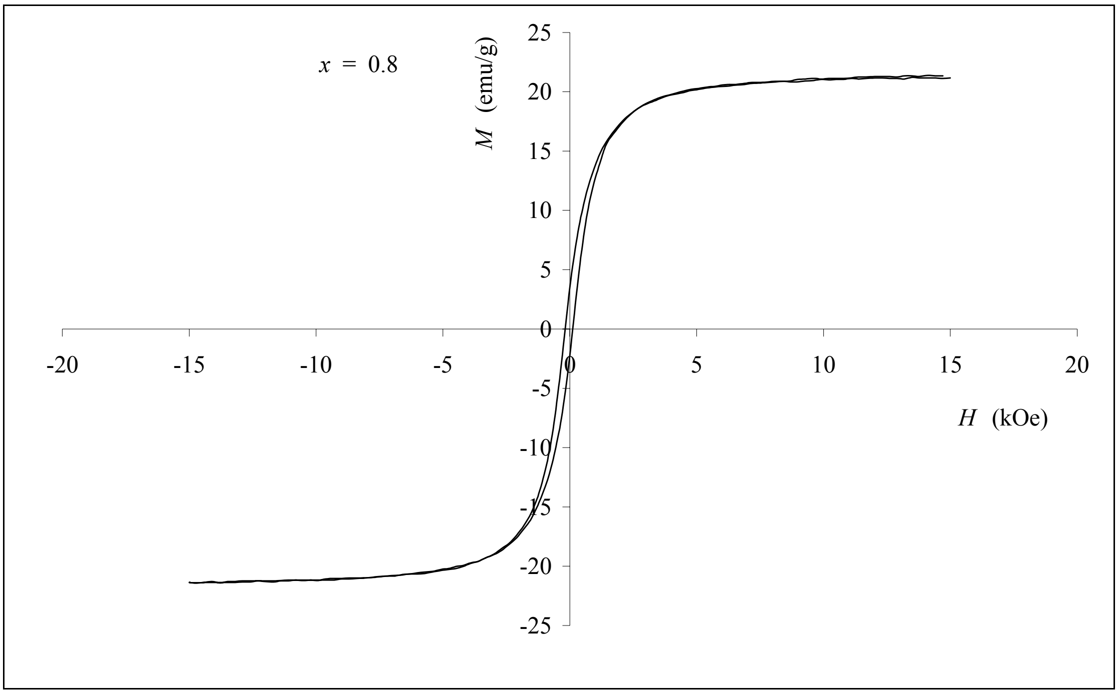

FIG.5(d). The hysteresis loop for the $x = 0.8$ sample

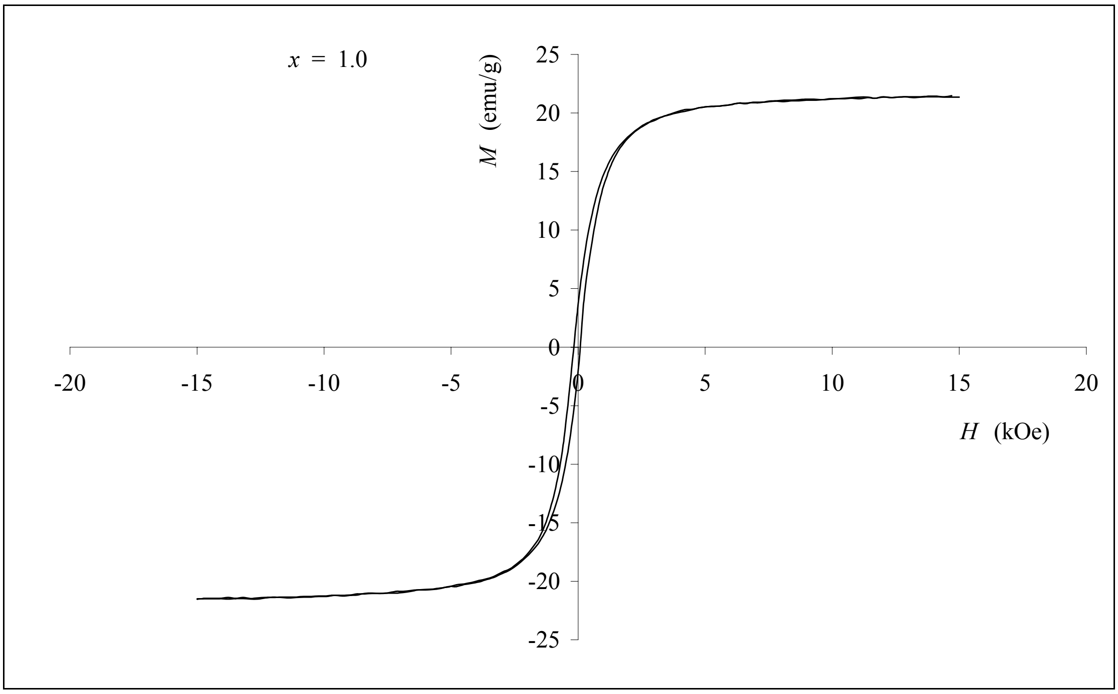

FIG.5(e). The hysteresis loop for the $x = 1.0$ sample

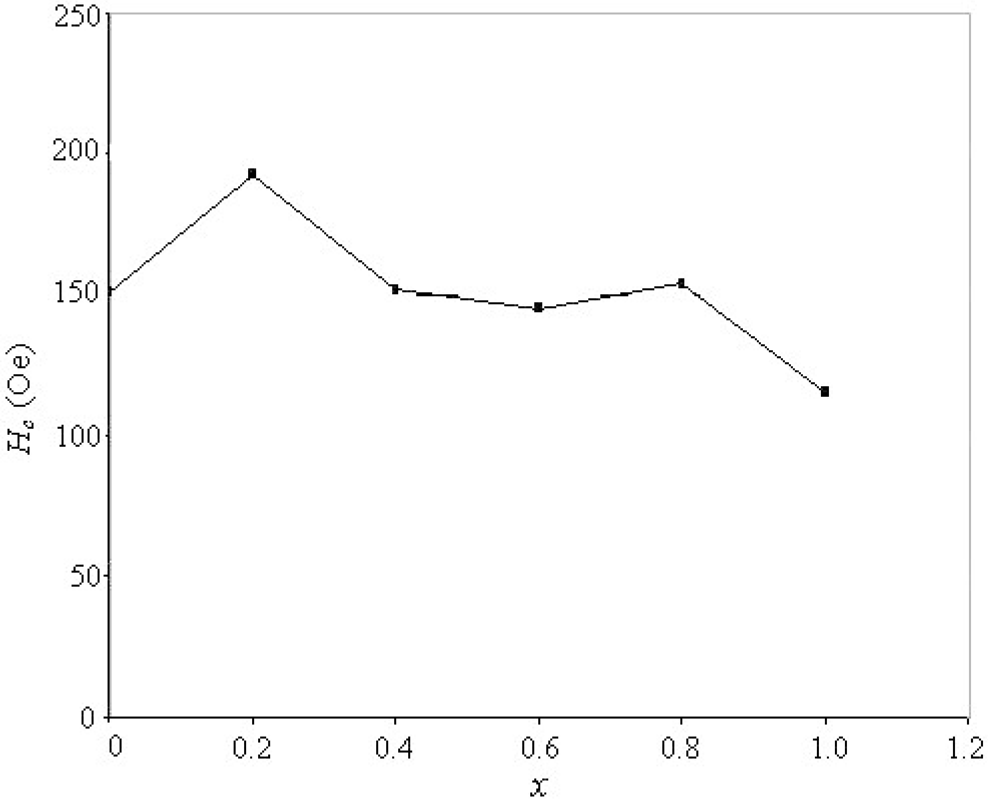

FIG.6. The coercivity versus *x* for all samples

We propose a moment distribution model, based on the magnetization results, to explain the magnetic behavior of the prepared ferrites and it is shown in table (2). For the base ($x = 0$) sample the moments structure is inverse, with all the Ni^{2+} ions in the B sites and the Fe^{3+} ions evenly distributed between A and B sites. The net moment is that of the Ni^{2+} ions (2 μ_B), since the moments of the Fe^{3+} ions cancel each other. For the rest of the samples, the Mg^{2+} ions are replacing Ni^{2+} ions which are in the octahedral sites (B), but the Mg^{2+} ions have the probability to occupy the tetrahedral site (A) or octahedral site (B), with high preference for the B sites, which means that the samples structures are mostly mixed or partially inverse spinels; this was also reported earlier [15, 16]. The decrease in magnetization with increasing x observed in our samples could be related to the decrease in moment when Ni^{2+} is replaced by Mg^{2+} in our samples (see table (2)). Mg^{2+} ions (diamagnetic) have 6 electrons in the last orbit (2p), (3 up and 3 down) and therefore it has no net magnetic moment, while Ni^{2+} ions (ferromagnetic) have a magnetic moment of $2\mu_B$, so the gradual decrease in magnetization observed in our results comes from the gradual substitution of Mg^{2+} ions in place of Ni^{2+} ions, resulting in the decrease of the net magnetic moment per formula of the system (see table (2)).

One more thing to add concerning the nonlinearity of magnetic interaction and the randomly canted structure present in ferrite systems. Yafet and Kittel discussed this problem and proposed an interesting model that explains the deviations of the experimental results from the theoretical ones [17]. The change in the magnetic moments and the reduction of the dominant A-B interaction as a result of Mg^{2+} substitution in place of Ni^{2+} is behind the expected moment non co-linear arrangement on the octahedral B-sublattice which is reflected in the experimental results.

TABLE 2. Moment distribution per molecule of samples.

x	Cationic distribution		Net moment (μ_B/molecule)
	Site A	Site B	
0	(Fe^{3+}) 5 →	(Ni^{2+}) (Fe^{3+}) 2 5 ← ←	2
0.2	(Mg^{2+}) (Fe^{3+}) 0 4.85 →	$(Mg^{2+})_{0.2}(Ni\ ^{2+})_{0.8}Fe^{3+}$ 0 1.6 5.15 ← ←	1.9
0.4	(Mg^{2+}) (Fe^{3+}) 0 4.8 →	$(Mg^{2+})_{0.4}(Ni\ ^{2+})_{0.6}Fe^{3+}$ 0 1.2 5.2 ← ←	1.6
0.6	(Mg^{2+}) (Fe^{3+}) 0 4.7 →	$(Mg^{2+})_{0.6}(Ni\ ^{2+})_{0.4}Fe^{3+}$ 0 0.8 5.3 ← ←	1.4
0.8	(Mg^{2+}) (Fe^{3+}) 0 4.75 →	$(Mg^{2+})_{0.8}(Ni\ ^{2+})_{0.2}Fe^{3+}$ 0 0.4 5.25 ← ←	0.9
1	(Mg^{2+}) (Fe^{3+}) 0 4.5 →	(Mg^{2+}) (Fe^{3+}) 0 5.5 ←	1

Conclusions

This study showed that Mg^{2+} substitution in place of Ni^{2+} in the $Mg_xNi_{1-x}(Fe_2O_4)$ system decreases the lattice parameter (a) at low Mg^{2+} content ($x \le 0.4$), and increases the lattice parameter a at high Mg^{2+} contents ($x \ge 0.6$). Moreover, substitution of Mg^{2+} in place of Ni^{2+} decreases the X-ray density for all studied sample.

It was also found, that Mg^{2+} substitution in place of Ni^{2+} increases the coercivity at low Mg^{2+} content ($x = 0.2$), and decreases the coercivity at higher contents ($x \ge 0.4$), while this substitution decreases the saturation magnetization and the remanence for all the samples.

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