

Effect of zinc, magnesium, and copper substitutions on the initial permeability of nickel ferrite

SAREH SHAFIEE

Malek-Ashtar University of Technology

Ali Arab (✉ aa.arab@yahoo.com)

Malek-Ashtar University of Technology

NASTARAN RIAHI-NOURI

Niroo Research Institute

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Abstract

Zinc, magnesium, and copper substituted nickel spinel ferrite were synthesized in the form of $Ni_{1-x}(Zn_{0.6}Mg_{0.2}Cu_{0.2})_xFe_2O_4$ (where $x = 0.0, 0.3, 0.5$ and 0.7) via auto combustion method. The effect of the presence of these dopants on the average of crystallite, average particle size, the lattice constant, morphology, initial permeability, and magnetization of the synthesized ferrites was investigated. The structural properties, morphology, and magnetic properties were characterized via X-ray diffraction (XRD), field emission scanning electron microscopy (FE-SEM), energy dispersive spectroscopy (EDS), LCR meter and alternative gradient force magnetometer (AGFM). XRD study shows the formation of a single-phase cubic spinel structure. Also, the average crystallite size was found to increase from 6 nm to 11 nm with increasing the Zn^{2+} , Mg^{2+} , and Cu^{2+} doping ratio from 0.0 to 0.7. Moreover, FE-SEM results were indicated the presence of nanosized spherical shape of prepared particles with agglomeration. The lattice constant and the particle size were found to increase with the increase in Zinc, magnesium, and copper. The values of initial permeability and magnetization were increased to a maximum value of 76 H/M and 71.37 emu/g for $x = 0.7$ sample. Furthermore, coercivity was found to decrease with increasing Zinc, magnesium, and copper concentration, which is useful for power applications. The variations of initial permeability and magnetization as a function of average particle size were discussed and were compared with previous works. The results were indicated the increase in initial permeability and magnetization with the enhancement of average particle size. The constancy in permeability throughout the frequency range studied from 10 kHz to 1 MHz was indicated the compositional stability and quality of the samples. The results were indicated that the ferrites with high initial permeability can be an excellent choice as magnetic cores.

1. Introduction

ZnMgCu-substituted nickel ferrites have been indicated excellent magnetic properties in the high-frequency from several kHz to MHz [1]. They are applied in power supply transformers, choke coils, power transformers, and noise filters [1]. The lattice of oxygen ions in spinel ferrite determines the structure and morphology of ferrites. The radii of oxygen ions are larger than the metal ions [2, 3]. The tetrahedral (A) and octahedral (B) sites are surrounded by 4 and 6 oxygen ions, respectively. Electrical and magnetic properties of ferrites are dependent on cationic distribution, composition, and nature of impurity ions in the material [4]; the structural properties of ferrites are dependent on compositions and synthesis methods [5]. The growth of grains is influenced by grains' boundary mobility. Recrystallization and the growth of grains cause the movement of grain boundaries and the increase in initial permeability [6]. To improve the electronic and magnetic properties of ferrites numerous divalent cations, such as chromium, copper, manganese, and zinc, substitute on ferrites [4]. The auto combustion method in comparison with others is more acceptable for synthesizing ferrites due to the high sintering, homogenous particle size distribution, and lower time consumed [7]. This method is rather simple, safe, and economical [8]. Zn^{2+} ions have a strong preference to occupy the A sites [9], whereas the Ni^{2+} ions have a strong preference to occupy the B sites. Therefore, with increasing Ni^{2+} substitution on Zn ferrite, Fe^{3+} ions in B sites, due to the occupancy of B sites by Ni^{2+} ions, migrate to A sites [10]. Previous studies have reported the synthesise of CuMgZnNi-ferrites. Thorat et al. has investigated the initial permeability and magnetic properties of $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$; reported that the initial permeability reaches the maximum value $\mu = 2620$ at $x = 0.05$ [11]. Gangaswamy et al. investigated the magnetic behavior of $Ni_{0.65-x}Mg_xZn_{0.35}Fe_2O_4$; reported that the Mg substitution enhances the saturation magnetization [6]. Roy et al. has studied the electromagnetic properties of

$\text{Ni}_{0.25-x}\text{Mg}_x\text{Cu}_{0.2}\text{Zn}_{0.55}\text{Fe}_2\text{O}_4$ ferrite prepared by the auto combustion method; reported that the very high permeability of $\text{Ni}_{0.07}\text{Mg}_{0.18}\text{Cu}_{0.2}\text{Zn}_{0.55}\text{Fe}_2\text{O}_4$ due to better densification, lower magnetostriction constant and inner stresses [12].

In this work, $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ have prepared by the auto combustion method. The aim of this work is: (1) to find the regularities of the effect of zinc, magnesium, and copper substitutions on the initial permeability of NiFe_2O_4 spinel ferrite; (2) investigate the morphological, structural and magnetic features of ferrite samples; (3) discuss the application of ferrite samples in high-frequency and the variations initial permeability as a function of particle size. The samples prepared in this work displayed a considerable performance in soft magnetic characteristics and high-frequency permeability.

2. Experimental

2.1. Samples preparation

A series of ZnMgCu-substituted nickel ferrites with the formula of $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ ($x = 0.0, 0.3, 0.5$ and 0.7) were made by the auto combustion method. The preparation of nanopowders had been revealed in previous work [12]. A stoichiometric ratio of Merck materials $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$, $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ with purity $\geq 98\%$ were dissolved in water deionized using a magnetic stirrer. Then, the $\text{NH}_5\text{C}_2\text{O}_2$ solution was added to the mixtures slowly. The mixture was heated at a temperature of $80^\circ\text{C} - 200^\circ\text{C}$ for two hours with continuous stirring. The obtained gel was dried at 250°C for two hours. Then the prepared powders were calcined at 600°C for four hours.

The powders were granulated using polyvinyl alcohol (PVA) as a binder and were pressed at a pressure of 3 ton/cm^2 into toroidal shape (about 4 mm diameter and 3 mm thickness). The toroid samples were sintered at 900°C for 8 h in air and were wound by copper wire at room temperature.

2.2. Characterization

The powders were characterized by XRD (X' Pert Pro MPD model), where the wavelength of Cu K_α radiation was 1.54 \AA . Their morphology was carried out by FESEM (Hitachi S- 4160 model). The compositions of the ferrites were measured through energy-dispersive x-ray spectroscopy (EDS). The initial permeability of toroid ferrites was measured by L.C.R meter (Hioki IM3536 model) in the frequency range from 10 kHz to 10 MHz. while the magnetic properties were characterized by AGFM in a maximum field of 9 kOe.

3. Results And Discussion

3.1. Analysis of structure and composition

Fig. 1 shows the X-ray diffraction (XRD) patterns of $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ ($x = 0.0, 0.3, 0.5$ and 0.7) samples. It can be seen that the prominent hkl planes (220), (311), (222), (400), (422), (511), and (440) were identified. The patterns were confirmed the formation cubic spinel structure of samples due to the well matching with the standard XRD patterns (JCPDS PDF cars No. 00-008-0234). Similar results have been revealed in previous work [13, 14]. The average crystallite size (D) was calculated by using the Scherrer-Debye equation [15]:

$$D = \frac{0.9\lambda}{\beta \cos \theta} \quad (1)$$

D is the average crystallite size, θ is the Bragg's diffraction angle, λ is incident beam wavelength, $\beta = (\beta_{\text{exp}}^2 - \beta_{\text{std}}^2)^{1/2}$, β_{exp} and β_{std} are FWHM peaks and the standard, respectively.

The lattice constant (a) was calculated by using Bragg's equation [16]:

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

Where "a" is the lattice constant, d is inter planer spacing, (hkl) are Miller indices of respective peaks.

From Table 1 which shows XRD parameters of ferrites can be seen that the average crystallite size raised from 6 nm to 11 nm with increasing the doping ratio of magnesium, copper, and zinc. The increase in the average crystallite size may be due to the liquid phase in the calcination process. With the addition of Zn^{2+} , Mg^{2+} , and Cu^{2+} ions, the contact area of solid reaction increases, and the growth of the crystallite size accelerates [18]. Also, the lattice constant found to be increased little from 8.331 Å to 8.372 Å with increasing the x content (0.0 to 0.7). The lattice constant is dependent on the difference between the substitution ions and host ions radii and the cationic distribution among the interstitial tetrahedral (A) and octahedral (B) sites [18]. The increase in lattice constant is due to larger ionic radii of Cu^{2+} (0.73 Å), Mg^{2+} (0.72 Å) and Zn^{2+} (0.82 Å) than nickel (0.69 Å) [19]. This agrees with other works observed by Where "a" is the lattice constant, d is inter planer spacing, (hkl) are Miller indices of respective peaks.

Roy et al. [12] and Thorat et al. [11]. The variations in lattice constant and crystallite size as x contents are shown in Fig. 2.

Fig. 3 indicates the FE-SEM micrographs for all the samples. The substitution of Cu, Mg, and Zn ions into nickel ferrites has influenced on their size and structure. The average particle size increases with the addition of Cu, Mg, and Zn ions from 21 nm to 26 nm, as listed in Table 1. The spherical nature of the particles and the agglomeration of grains is shown in the FE-SEM images. The variation in particle size as x concentration are shown in Fig. 4. The increase in lattice volume has an important role in grain boundary diffusion [20]. With increasing lattice volume, the diffusion path, and the rate of the cation interdiffusion increase. Furthermore, the growth of grain is related to the grain's boundary mobility [21]. Recrystallization and grains growth lead to the movement of grains boundaries and the variations in permeability, density, and resistivity [21]. So, the grain boundary diffusion was increased by increasing Cu^{2+} , Mg^{2+} , and Zn^{2+} substitutions.

Using EDS, the distribution of elements in selected samples with x= 0.3, 0.5 were analyzed. The atomic percentage of elements are given in Fig. 5. The spectra confirmed that the final compositions of ferrites were the same as those of compositions without any extra impurity elements.

3.2. Analysis of magnetic performance

Fig. 6 shows the initial permeability (μ_i) as a function of frequency for $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ ($x=0.0, 0.3, 0.5$ and 0.7) ferrites under the frequency range of 10 kHz-10 MHz. As depicted clearly, μ_i increases with the increasing zinc, magnesium, and copper and reaches the maximum value $\mu_i = 76$ at $x = 0.7$. The increase in initial permeability has been attributed to grain size. The variations of the initial Permeability as a function of x concentration and particle size in the frequency range of 10 kHz- 10 MHz are shown in Fig. 7 and Fig. 8. The observed variation in initial permeability can be due to the basis of changes that took place in grain size as evident from FE-SEM micrographs. The variations in initial permeability are parallel to the changes observed in the particle size. The initial permeability correlates by two different magnetizing mechanisms of ferrites; the movement of domain wall and the spin rotation [22]. The spin rotation is smaller than domain walls at the low-frequency region, the domain wall movement is reversible because of the presence of a weak magnetic field [21, 23].

As can see in Fig. 8, the increase in particle size causes an increase in domain wall contribution, and the increasing initial permeability [24]. It is a well-known fact that the increase in grain size reduces the number of grain boundaries in a sample and causes an increase in the initial permeability [25]. The higher μ_i obtained for the composition $x = 0.7$ at the frequency of 10 kHz is contributed by the domain wall oscillations in the bigger grains of this composition. Furthermore, the μ_i remained steady over a wide range of frequency 10 kHz -1 MHz, and then decreases rapidly greater than 1 MHz applied frequency, as indicated in Fig. 6. The stability of μ_i from 10 kHz to 1 MHz is due to the domain wall motion. The frequency of 1 MHz is named the zone of utility [26], which is equal to the external magnetic field frequency and a desirable characteristic for various applications of ferrites in high-frequency, such as broadband pulse transformers and wideband read-write heads for video recording [26]. After the resonance frequency (1 MHz) the μ_i decreases due to the absorption of magnetic energy by spin moments [24]. The required energy for the displacement of the domain wall is lower than that required for domain rotation. The constancy in permeability throughout the frequency range studied from 10 kHz to 1 MHz indicates the compositional stability and quality of the samples. The present investigation of Cu^{2+} , Mg^{2+} , and Zn^{2+} substituted Ni ferrites reveal that the ferrites with the high initial permeability are an excellent choice as magnetic cores.

The characteristics of μ_i have been reported by Roy et al. [12] for Ni-Zn-Mg-Cu ferrites prepared by the auto combustion method and Thorat et al. [11] for Ni-Zn-Mg-Cu ferrites prepared by citrate assisted sol-gel method, which agrees with our reported values. The results are listed in Table 2.

The initial permeability is influenced by compositions, impurity contents, preparation methods, grain size, saturation magnetization, magnetostriction [17, 22]. The μ_i values reported by Roy et al. [12] and Thorat et al. [11] are higher than those obtained for the $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ in the present investigation. It may be due to the different preparation methods and the higher calcination and sintering temperature of samples than those ferrite samples in the present investigation. The density and grain size of the ferrites increase with increasing sintering temperature, and they would affect magnetic properties directly [27]. Higher crystallite and particle sizes, and higher temperature used for calcination and sintering samples cause to increase in μ_i . The magnetization mechanism of soft magnetic materials is domain wall motion, which generates high initial permeability (μ). Although pores and grain boundary would obstruct the movement of the domain wall, the fewer amounts of pores and grain boundary could be obtained at higher sintering temperature and lead to easy movement of domain wall

and high initial permeability [27]. Furthermore, the values of μ_{eff} reported by Roy et al. [12] indicated that the increase in permeability is attributed to the increase in particle size.

Fig. 9 illustrates the magnetization (M) as a function of field (H) curves for the $Ni_{1-x}(Zn_{0.6}Mg_{0.2}Cu_{0.2})_xFe_2O_4$ ($x=0.0, 0.3, 0.5$ and 0.7) ferrite at room temperature and maximum magnetic field of 9 kOe. The magnetization is not saturated until 9 kOe in all hysteresis curves. From the plotted M-H curves, the maximum magnetization (M), and coercivity (H_c) were measured as reported in Table 1. With increasing Cu^{2+} , Mg^{2+} , and Zn^{2+} ions in the nickel ferrite the magnetization increases. The increase in magnetization can be due to the variations of exchange interactions between A and B sites, an increase in the crystalline nature, and a narrow particle size distribution [26]. The magnetization is calculated about 57.94 emu/g, 68.41 emu/g, and 71.37 emu/g for samples with $x=0.3, 0.5$, and 0.7 respectively, which are higher than that of the unsubstituted nickel ferrite sample (32.5 emu/g). Moreover, the M value of ferrite with $x=0.7$ is higher than that of the substituted nickel ferrite samples. The magnetic properties are influenced by the composition and different cation distribution as described by the Neel model [24]. According to Neel's theory, the net magnetic moment can be defined as following [24]:

$$M = |M_B - M_A| \quad (3)$$

Where M_A and M_B are the magnetic moments of A and B sublattices [24].

The replacement of cations substituted in the spinel ferrites leads to weak or strong interaction among magnetic ions [13]. It is reported in previous research that the Zn^{2+} and Mg^{2+} ions have a strong preference for occupancy the A sites; the Ni^{2+} and Cu^{2+} ions have a strong preference towards B sites, while the Fe^{3+} ions distribute over both A and B sites [28, 29]. The magnetic moment of Zn^{2+} , Cu^{2+} , Ni^{2+} , Fe^{3+} and Mg^{2+} ions is 0, 1.3, 2.3, 5 and 0 μ_B , respectively [15, 19]. With the increasing of Cu^{2+} , Mg^{2+} , and Zn^{2+} ions on nickel ferrite, the copper ions occupy B sites, due to their high preference towards B sites, and a smaller concentration of copper may appear at A sites. The diamagnetic Zn^{2+} and Mg^{2+} ions occupy the A sites, where cause the movement of Fe^{3+} ions from A sites to B sites. Therefore the increase in magnetization is due to the higher magnetic moment of B than A sublattices [30]. The variations of magnetization and coercivity of ferrite samples as a function of x are shown in Fig. 10.

The magnetic characteristics are dependent on the particle size, anisotropy, density, cationic stoichiometry, random canting of particle spins, and surface effects [29, 31]. The increase of M can be explained with the particle size trend. As reported in the structural analysis of this series published elsewhere, the particle size has increased with an increase in the amount of doped copper, zinc, and magnesium. The variation of magnetization and coercivity dependence of average particle size is shown in Fig. 11. As can be seen the magnetization increases by the enhancement of particle size. As the particle size increases, the number of magnetic domains increases, and the movement of the domain wall facilitates, which leads to the enhancement of magnetization [32]. This behavior can also be justified according to the core-shell model [33]. In this model, each particle consists of a magnetically ordered core and a spin glass surface with a constant thickness without net magnetization [33]. The disordering of the surface layer spins may be due to the broken super-exchange bonds and unlike local symmetry for those atoms near the surface layer [33]. In smaller particles, the surface to volume ratio of particles is higher, which leads to a decrease in magnetization [33]. With an increase in the average particle size, due to the diminution in the surface to volume ratio, the magnetization increases.

Coercivity (H_c) gives the value of the applied field, at which the induced magnetization is zero. The values of H_c for ferrite samples are listed in Table 1. As can be seen from Fig. 10 the coercivity decreases with Cu^{2+} , Mg^{2+} , and Zn^{2+} substitution. The H_c is dependent on the particle size, grain boundaries, anisotropy, and precipitates [34]. The increase in particle size of ferrites decreases the surface area to volume ratio and surface anisotropy of the crystal. The coercivity property of the samples originates from a multi-domain structure. In the multi-domain region, the increase in particle size decreases the coercivity [35]. Referring to the variations of M and μ_{eff} as a function of average particle size, it is understood that the dependence of magnetization to the initial permeability is direct. Therefore, the increase in particle size leads to an increase in domain wall contribution and an increase in initial permeability [24].

4. Conclusion

Cu^{2+} , Mg^{2+} , and Zn^{2+} doped nickel ferrites have been successfully prepared by the auto combustion method. XRD results confirmed the formation of a cubic spinel structure. With increasing zinc, magnesium, and copper the crystallite size and lattice constant increased. FE-SEM analysis reported the spherical shape of ferrite nanoparticles along with the presence of high agglomeration due to magnetic interactions between ferrite particles. Also, the average particle size increased by addition zinc, magnesium, and copper on nickel ferrite. In the ferrite sample of $\text{Ni}_{0.3}\text{Zn}_{0.42}\text{Mg}_{0.14}\text{Cu}_{0.14}\text{Fe}_2\text{O}_4$ the average crystallite size, the lattice constant and particle size be larger than maybe understood due to larger ionic radii of Cu^{2+} , Mg^{2+} , and Zn^{2+} as compared to Ni^{2+} .

The initial permeability increased by increasing x content and average particle size at a frequency range of 10 kHz-10 MHz due to the grain growth. The constancy in initial permeability throughout the frequency range studied from 10 kHz to 1 MHz, due to the contribution of domain wall motion, indicated the compositional stability and quality of the samples. The reduction of initial permeability from 1 MHz to 10 MHz, illustrated the external magnetic field frequency. The investigation of Cu^{2+} , Mg^{2+} , and Zn^{2+} substituted Ni ferrites revealed that the ferrites with the high initial permeability are an excellent choice as magnetic cores. The μ_{eff} values of present work compared with the results of previous work. The μ_{eff} in other works were found to be higher than those obtained for the $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ ferrites in the present investigation. It can be due to the different preparation methods and the higher calcination and sintering temperature of samples than those ferrite samples in the present investigation. The density and grain size of the ferrites increase with increasing sintering temperature, and they would affect magnetic properties directly. Furthermore, hysteresis loops showed an increase in magnetization, due to the site's occupation of Zn, Mg, and Cu ions, with the enhancement of Cu^{2+} , Mg^{2+} , and Zn^{2+} ions. While the coercivity decreased due to a multi-domain structure of ferrites. The M and μ_{eff} influenced by the average particle size, due to the variation of the surface to volume ratio of particles. From the results of all characterization techniques in this paper, it is concluded that the sample of $\text{Ni}_{0.3}\text{Zn}_{0.42}\text{Mg}_{0.14}\text{Cu}_{0.14}\text{Fe}_2\text{O}_4$, due to the higher M and μ_{eff} , can be used as a magnetic core. Also, it can be applied in various technologies useful in many fields like medical, information and communication, etc. The obtained results were compared with previous works.

Declarations

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Tables

Table 1

The contents of D, a, d, μH , M, and H_c of $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ for different x values.

X	Samples	the average crystallite size (D) nm	Lattice constant (a) Å	Average particle size (d) nm	Initial permeability (μH) H/M 100 kHz-1 MHZ	Maximum magnetization (M) emu/g	Coercivity field (H_c) Oe
0.0	NiFe_2O_4	6	8.331	21	32	32.50	198.41
0.3	$\text{Ni}_{0.7}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_{0.3}\text{Fe}_2\text{O}_4$	8	8.357	23	58	57.94	131.95
0.5	$\text{Ni}_{0.5}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_{0.5}\text{Fe}_2\text{O}_4$	10	8.369	25	71	68.41	96.25
0.7	$\text{Ni}_{0.3}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_{0.7}\text{Fe}_2\text{O}_4$	11	8.372	26	76	71.37	92.12

Table 2

The contents of D, a, d, μH reported by Roy et al. [12] and Thorat et al. [11] for different ferrite samples.

X	Samples	the average crystallite size (D) nm	Lattice constant (a) Å	Average particle size (d) nm	Initial permeability (μH) H/M 100 kHz-1 MHZ	Method of synthesis
0.0	$\text{Ni}_{0.18}\text{Zn}_{0.55}\text{Mg}_{0.7}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$	124	8.3990	-	680	
0.3	$\text{Ni}_{0.12}\text{Zn}_{0.55}\text{Mg}_{0.13}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$	121.9	8.3991	-	940	Auto combustion
0.5	$\text{Ni}_{0.7}\text{Zn}_{0.55}\text{Mg}_{0.18}\text{Cu}_{0.2}\text{Fe}_2\text{O}_4$	127.6	8.4097	-	2420	
0.7	$\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Mg}_{0.2}\text{Cu}_{0.25}\text{Fe}_2\text{O}_4$	48	8.392	237	2620	
	$\text{Ni}_{0.1}\text{Zn}_{0.5}\text{Mg}_{0.15}\text{Cu}_{0.25}\text{Fe}_2\text{O}_4$	51	8.400	237	1930	citrate assisted sol-gel
	$\text{Ni}_{0.15}\text{Zn}_{0.5}\text{Mg}_{0.1}\text{Cu}_{0.25}\text{Fe}_2\text{O}_4$	47	8.405	338	1569	
	$\text{Ni}_{0.2}\text{Zn}_{0.5}\text{Mg}_{0.5}\text{Cu}_{0.25}\text{Fe}_2\text{O}_4$	49	8.407	180	522	

Figures

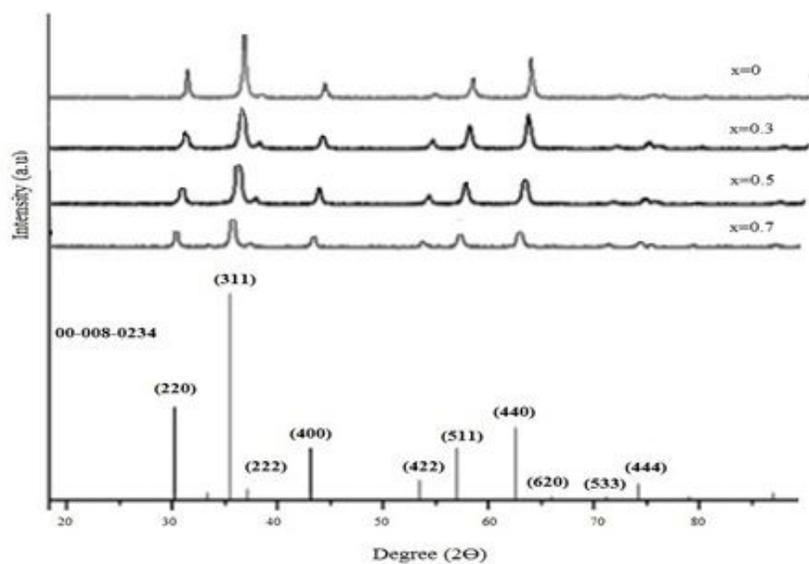


Figure 1

Indexed XRD patterns of $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ with x equal to: 0.0, 0.3, 0.5 and 0.7.

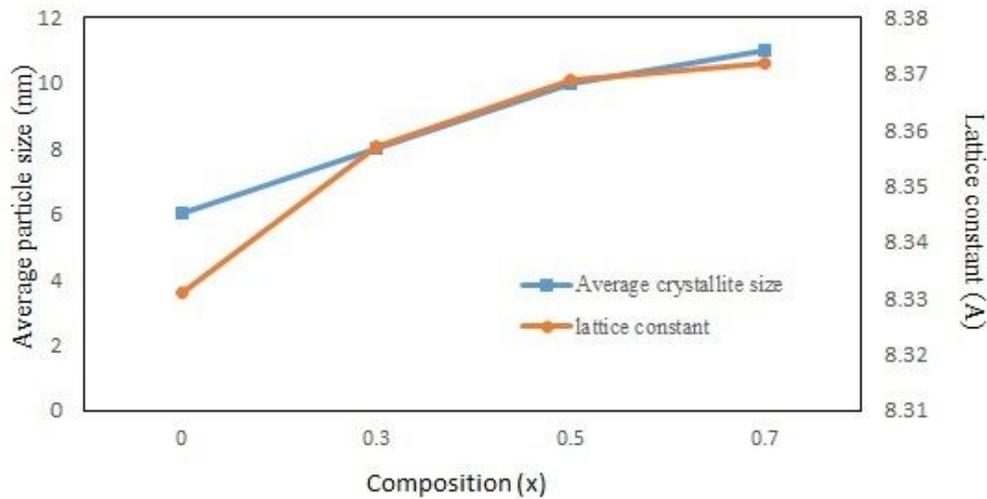


Figure 2

The variations of the lattice parameter and average crystallite size concerning x values for $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$. Morphology of $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ with x equal to: 0.0, 0.3, 0.5 and 0.7.

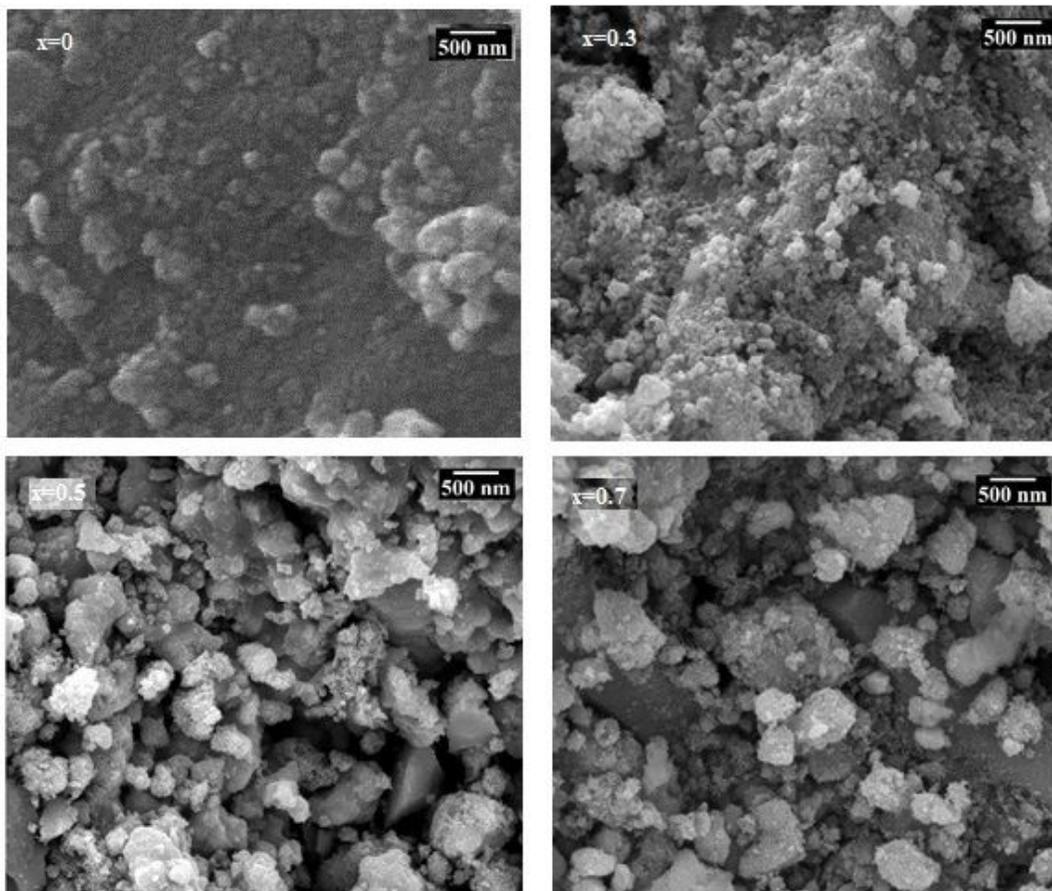


Figure 3

Morphology of $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ with x equal to: 0.0, 0.3, 0.5 and 0.7.

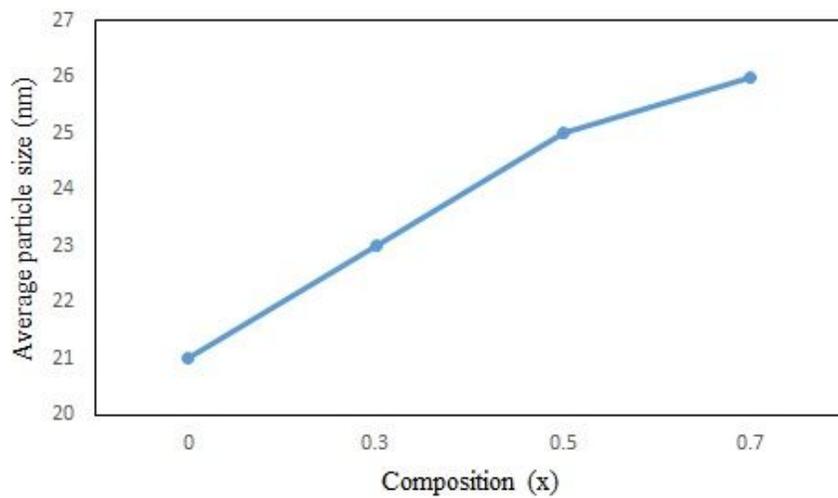


Figure 4

The variations of the average particle size concerning x values for $\text{Ni}_{1-x} (\text{Zn}_{0.6} \text{Mg}_{0.2} \text{Cu}_{0.2})_x \text{Fe}_2\text{O}_4$.

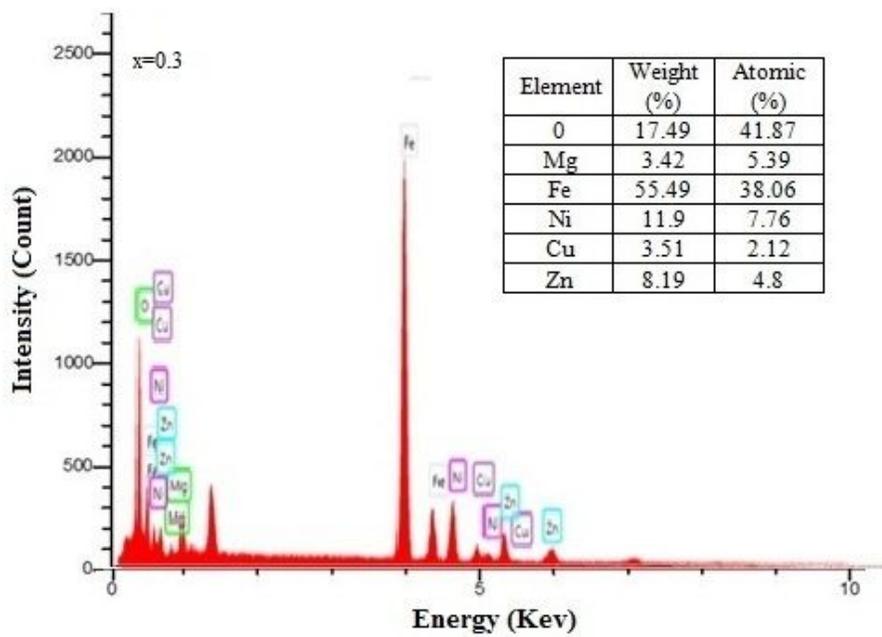
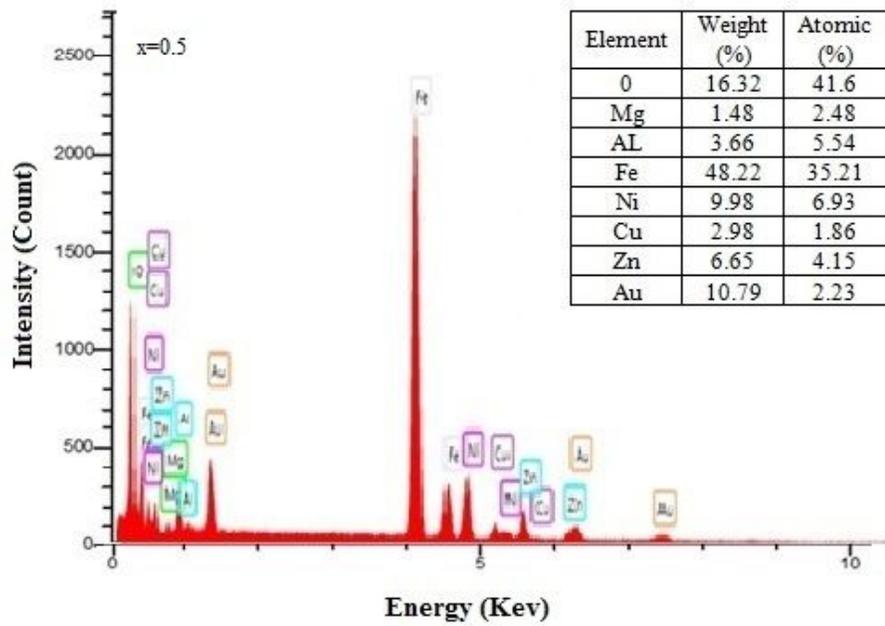


Figure 5

EDX spectrum of $Ni_{1-x} (Zn_{0.6} Mg_{0.2} Cu_{0.2})_x Fe_2O_4$ with x equals to 0.3 and 0.5

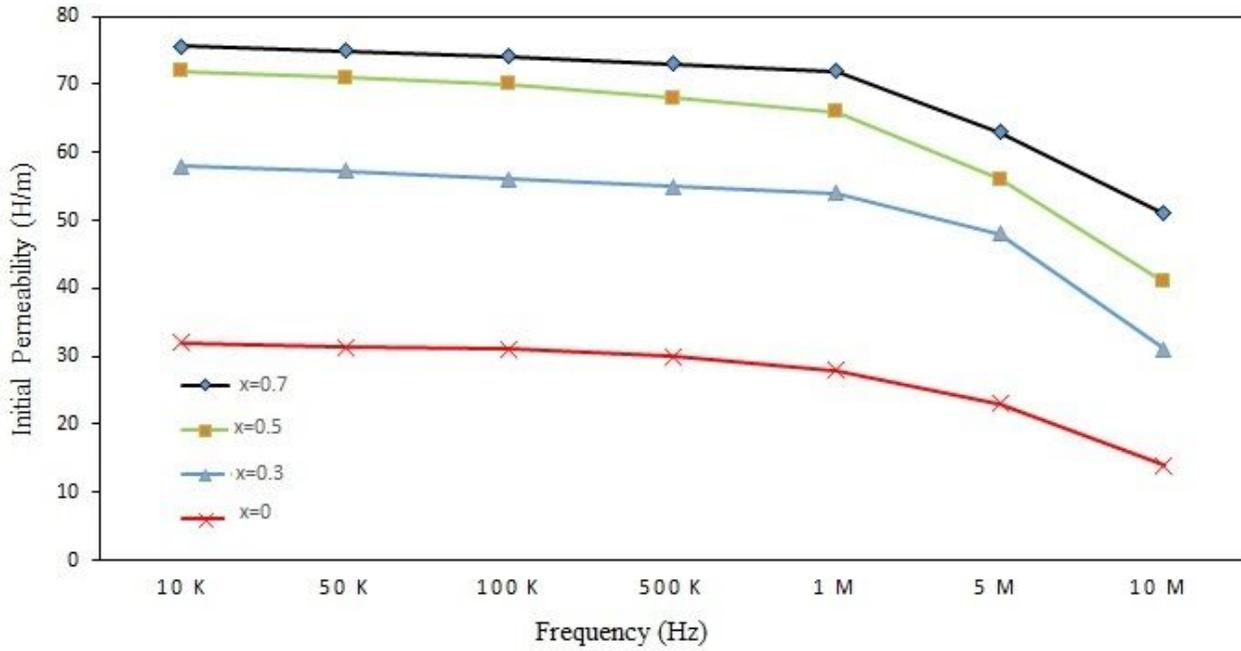


Figure 6

The variations of initial permeability concerning frequency for ferrite samples with x equal to 0.0, 0.3, 0.5, and 0.7.

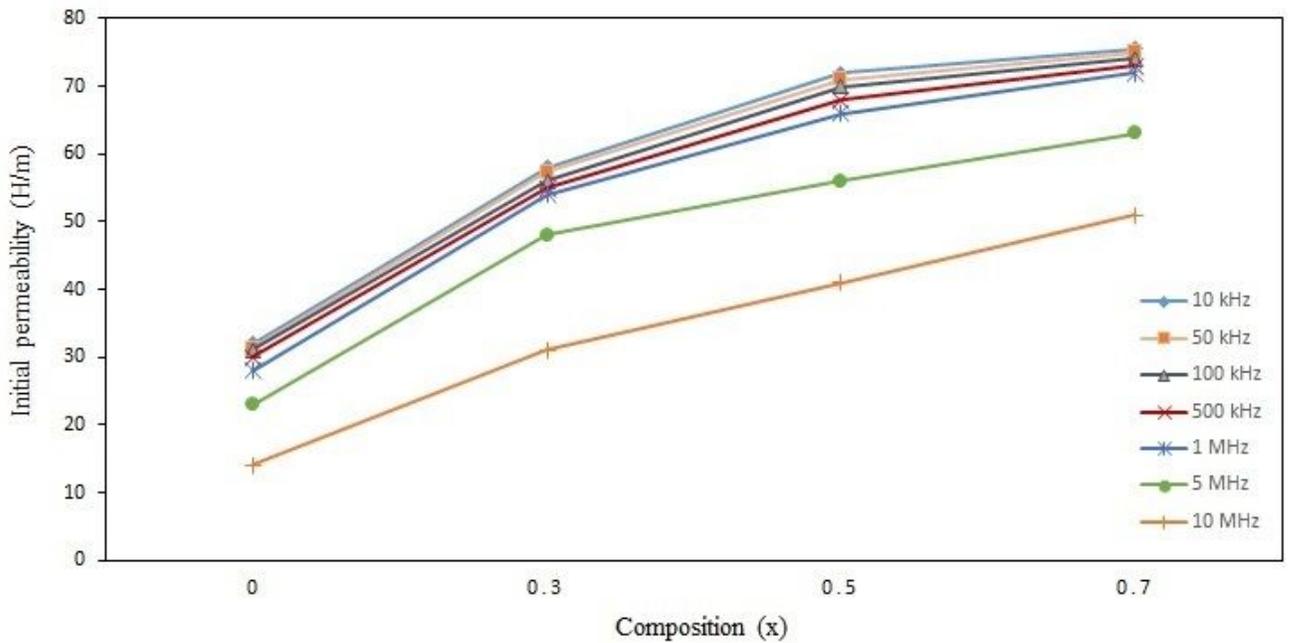


Figure 7

The variations of initial permeability concerning x values for Ni_{1-x} (Zn_{0.6} Mg_{0.2}Cu_{0.2}) x Fe₂O₄.

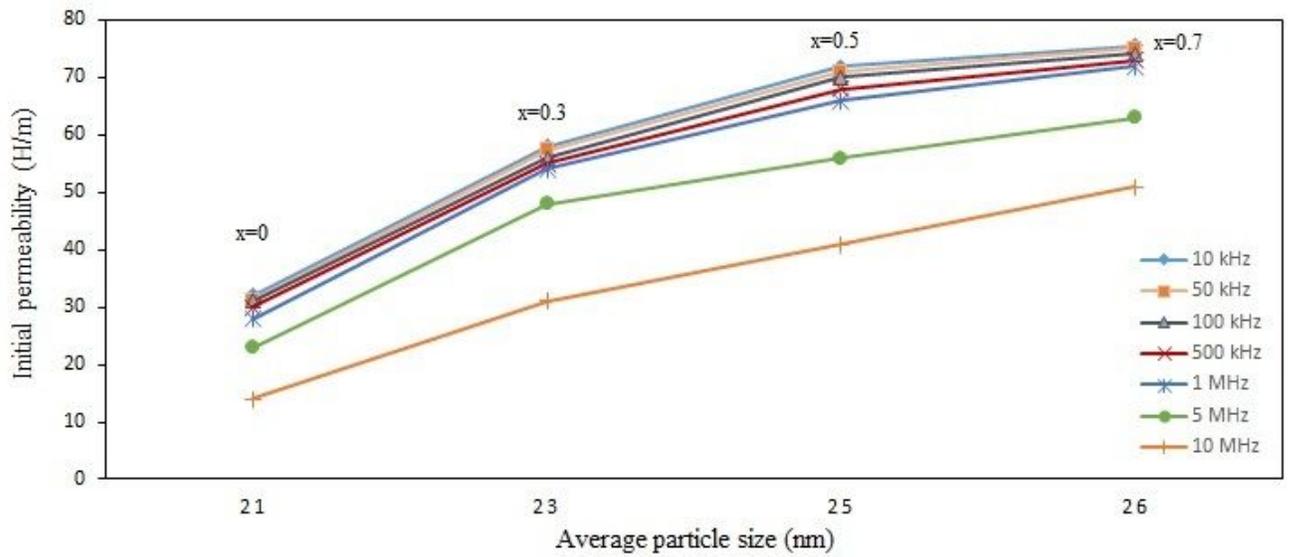


Figure 8

The variations of initial permeability as a function of mean particle size for $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$.

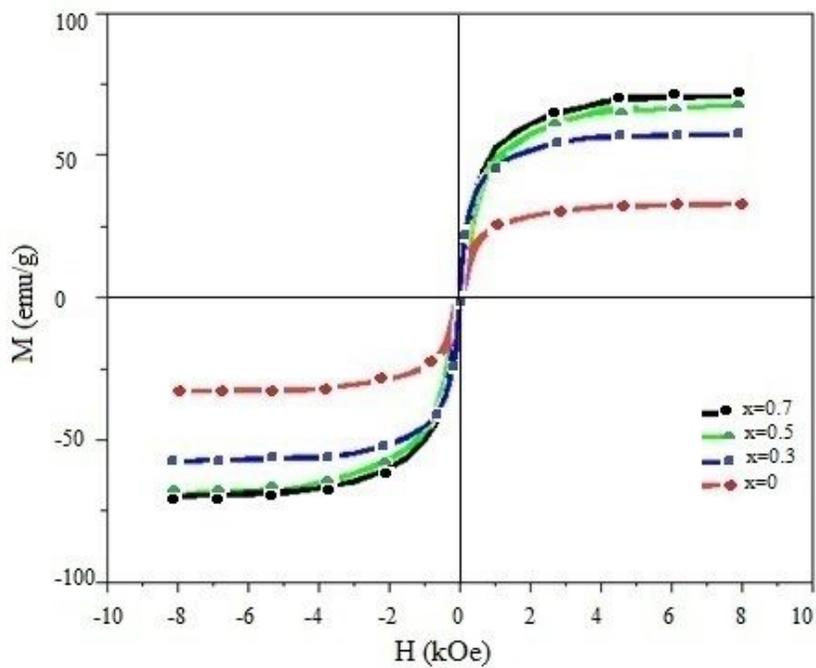


Figure 9

Room temperature hysteresis loops for $\text{Ni}_{1-x}(\text{Zn}_{0.6}\text{Mg}_{0.2}\text{Cu}_{0.2})_x\text{Fe}_2\text{O}_4$ with x equal to: 0.0, 0.3, 0.5 and 0.7.

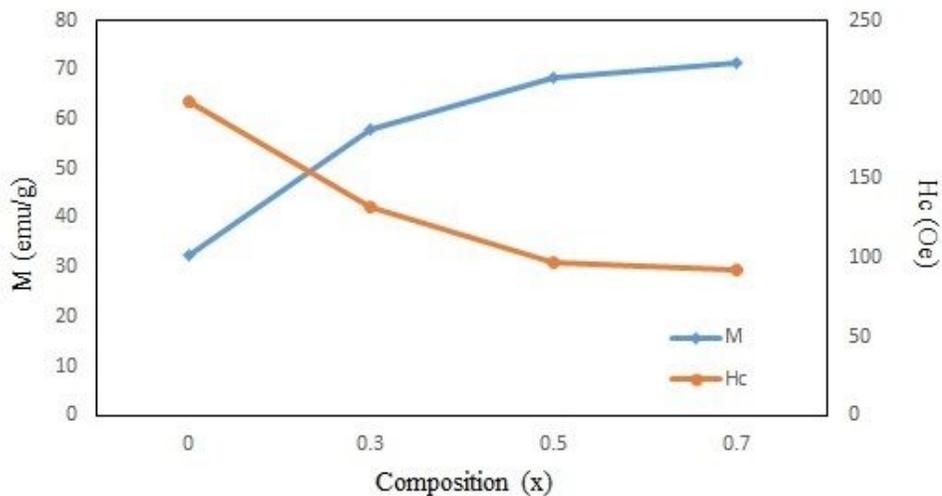


Figure 10

The variations of magnetization and coercivity of $Ni_{1-x}(Zn_{0.6}Mg_{0.2}Cu_{0.2})_xFe_2O_4$ as a function of x.

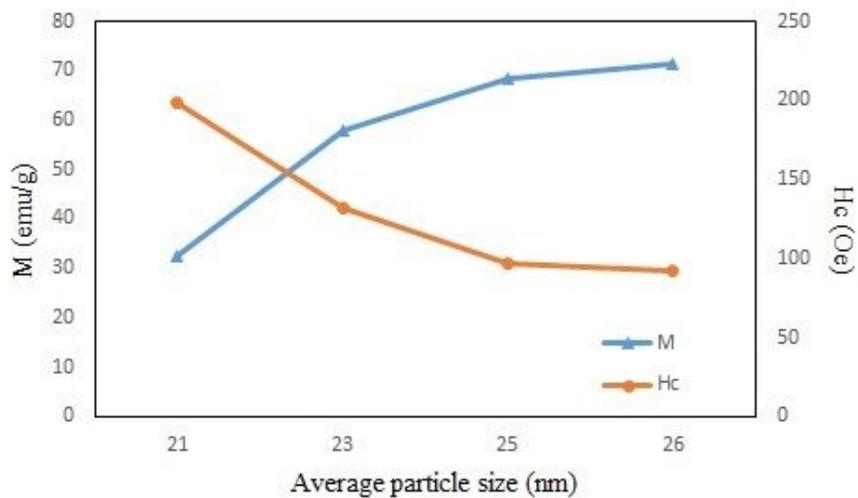


Figure 11

The variations of magnetization and coercivity of $Ni_{1-x}(Zn_{0.6}Mg_{0.2}Cu_{0.2})_xFe_2O_4$ as a function of average particle size.