Structural and Magnetic Properties of Nano Ferrite for Magnetoelectric Applications.

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Abstract. A Series of ferrite samples Ni_{0.1} Cu_{0.2} Zn_{0.7-x} Mg_x Fe_2O_4 (where x = 0, 0.15, 0.25, 0.35, 0.45, 0.55 and 0.7) were synthesized using auto combustion flash method. The samples were annealed at 600 °C for 2 hours to eliminate the foreign phases of the constituent nitrate and internal stress. The purity of phase structure was confirmed by x-ray diffraction. Some structural and microstructural parameters like porosity, x-ray density, crystallite size and lattice constant were deduced from x-ray. Scanning electron microscope (SEM) analysis reveals that the grains are separated by pores in all samples and the average grain size decreases with increasing Mg content. Magnetic properties such as coercivity (H_c), saturation magnetization (M_s) and reentivity (M_r) were measured from M-H loops. The magnetization curves were characterized by low coercivity indicating that our samples are soft magnetic material. Both saturation magnetization and coercivity increase by increasing Mg ions where M_s reaches maximum value at x = 0.35 then decrease for higher Mg content.

1. Introduction

Nowadays magnetic nanoferrites are important due to their applications in many technological and versatile fields. The magnetic nanoferrites have been used in variety of applications such as microwave devices, magnetic refrigeration, magnetic storage devices, microwave absorbers, permanent magnets, electronic industry, magnetic ferrofluids, magnetic separation, switching, core, recording heads, magnetic shielding and high frequency devices. However, these applications are classified according to their soft and hard magnetic behavior. Generally, when the particle size is less than the critical diameter of the domain formation, the particle is said to be in a single domain state. However, the magnetic properties of the nanoferrites can be changed and improved accordingly. Super paramagnetic phenomenon in magnetic nanoferrites occurs due to their large surface to volume ratio. Therefore, these nanoferrites with superparamagnetic characteristics have moderate permeability, low coercivity, high saturation magnetization and low loss factor at wide range frequencies due to their better morphology, magneto crystalline anisotropy and small particle size [1].
Nanocrystalline spinel ferrite like Ni-Cu-Zn nanoferrites have been the most universal ferrite materials due to their low sintering temperature, high electrical resistivity, high permeability, hard mechanical properties and also high curie temperature. Doping with Mg may improve the electromagnetic properties of Ni-Cu-Zn ferrite samples. This is because of the fact that Mg containing composition may avoid the tendency of discontinuous grain growth [2].

A combination of two phases such as of ferrimagnetic and ferroelectric phases produce a new type of material with improved and unique properties. The magnetostriction deformation of the ferrite phase causes polarization of the ferroelectric phase in the composites and the electrical polarization of the ferroelectric phase causes changes in the magnetism of the ferrite phase due to the strong mechanical coupling between ferrite and ferroelectric phases. In the last years, the magnetoelectric (ME) materials showing simultaneous magnetic and ferroelectric phases became very important for their fundamental physics as well as for some particular applications [3].

The aim of this work is to synthesis a novel material suitable for magnetoelectric devices and as a probe for magnetic fields measurements, this aim will be achieved by synthesis a composite with ferroelectric material which consider a future work to this search.

2. Experimental Details.

2.1. Samples preparation.
A series of ferrite $\text{Ni}_{0.1}\text{Cu}_{0.2}\text{Mg}_x\text{Zn}_{0.7-x}\text{Fe}_2\text{O}_4$, ($x = 0.00, 0.15, 0.25, 0.35, 0.45, 0.55$ and $0.70$) system were prepared by the flash auto combustion method using nickel nitrate $(\text{Ni(NO}_3)_2\cdot6\text{H}_2\text{O})$, copper nitrate $(\text{Cu(NO}_3)_2\cdot3\text{H}_2\text{O})$, magnesium nitrate $(\text{Mg(NO}_3)_2\cdot6\text{H}_2\text{O})$, zinc nitrate $(\text{Zn(NO}_3)_2\cdot6\text{H}_2\text{O})$, ferric nitrate $(\text{Fe(NO}_3)_3\cdot9\text{H}_2\text{O})$ and urea $(\text{CO(NH}_2)_2$ as a fuel. The metal nitrates were mixed with the urea by using glass rod. The mixture was heated at 80°C with constant stirring using hot plate until become viscous and the internal ignition take place a tree, and finally forming brown ferrite powder. The powder was collected and annealed at 600 °c for 2 hours.

2.2 Characterizations.
The samples were examined by x-ray diffraction using a Philips model (PW-1729) diffractometer (Cu-Kα radiation source with $\lambda = 1.540598$ Å) with $2\theta$ from 4-80°. The microstructure of the samples was analyzed using a scanning electron microscope (SEM) (JEOL JSM-6460, Japan). The magnetic hysteresis loops were measured at room temperature using vibrating sample magnetometer (VSM) operating system v 1.6 control software Oxford OX8JTL England.

3. Results and Discussion.

3.1 Structural analysis.
The x-ray diffraction of $\text{Ni}_{0.1}\text{Cu}_{0.2}\text{Mg}_x\text{Zn}_{0.7-x}\text{Fe}_2\text{O}_4$, ($x = 0.00, 0.15, 0.25, 0.35, 0.45, 0.55$ and $0.70$) system were prepared by the flash auto combustion method using the nitrate salt as constituent are shown in figure 1. The pattern confirm the formation of single phase spinel structure as a major phase without any peaks belong to internal stress and a small minor phase belongs to hematite Fe$_2$O$_3$ at $2\theta = 32.17°$ and $2\theta = 39.07°$. These peaks are disappeared completely at high Mg content. The intensity of the 100% peak (311) decreases by increasing Mg content which indicates that the presence of Mg retard the crystal growth. The variation of the lattice parameter ‘a’ as a function of Mg content is shown in figure 3. The decrease was observed and the variation obeys Vegard’s law [4, 5]. The lattice constant which was calculated from XRD pattern was found to decrease with increasing Mg content may be due to the smaller of magnesium radius (0.72Å) compared with Zinc ion radius (0.74Å).

The attractive force of oxygen anions with Mg cations is smaller than that with Zn ions which mean that the bond length increase leading to the increase of lattice parameter, so the competition between the ionic
radii and attractive force is responsible for the decrease of lattice parameter, where the effect of ionic radii is the predominant factor controlling the lattice parameter.

The bulk density of the samples decreases at higher concentration of Mg beginning from x=0.35 which has lower atomic weight (24.305) than zinc (65.39) as shown in figure 4. The decrease of density at higher concentration could be attributed to the fact that Mg retards the grain growth and density. At low Mg content the effect of retardation process is negligible and so the density values still low than density at x = 0.35. The theoretical density was calculated from XRD pattern using the formula:

$$\rho_x = \frac{8M}{N_A a^3} \text{ g/cm}^3$$  \hspace{1cm} (1)

where M is molecular weight of the composition, N_A is Avogadro's number and (a) is the lattice parameter. The theoretical density decreases significantly with increasing Mg content and is higher than bulk density due to the presence of pores which may be formed during preparation process. The porosity of the samples was calculated using the following equation [6] and is shown in figure 5.

$$P = 1 - \frac{\rho_x}{\rho} \%$$  \hspace{1cm} (2)

The porosity increases at high Mg content due to the vacancies and pores during the preparation of the samples. The cation distribution at both tetrahedral and octahedral sites was expected on the bases of theoretical lattice parameter which is given by:

$$a_{ch} = \frac{8}{3\sqrt{3}} \left( [r_A + r_O] + \sqrt{3} [r_B + r_O] \right)$$  \hspace{1cm} (3)

where $r_O$ is the radius of Oxygen ion, $r_A$ and $r_B$ are the radii of tetrahedral and octahedral sites. The ionic radius of octahedral and tetrahedral site was calculated using the assumption:

![Figure 1. X-ray diffraction patterns of system ferrite Ni_{0.1} Cu_{0.2} Zn_{0.7-x} Mg_x Fe_2O_4 for different ratios of Mg content (x).](image)
Figure 2. The ionic radius of octahedral (r_B) and tetrahedral site (r_A) of "Ni-Cu-Zn" system for different Mg content (x).

Figure 3. Experimental lattice parameters (a_{exp}) and theoretical lattice parameters (a_{th}) Vs. Mg content (x) for "Ni-Cu-Zn" system.
Figure 4. Bulk density ($\rho$) and X-ray density ($\rho_x$) Vs. Mg content (x) for "Ni-Cu-Zn" system.

Figure 5. Porosity (P) Vs. Mg content (x) for "Ni-Cu-Zn" system.
\[ r_A = (0.7 - x - t)r_{Zn}^{+2} + (0.3 + x + t)r_{Fe}^{+3} \]
\[ r_B = \left[ x r_{Mg}^{+2} + 0.1 r_{Ni}^{+2} + 0.2 r_{Cu}^{+2} + t r_{Zn}^{+2} + 2 - (0.3 + x + t)r_{Fe}^{+3} \right]/2 \]

where \( t \) is the traces of Zn present at octahedral site. The values of cation distribution parameter for which \( a_{th} \) and \( a_{exp} \) matches each other are taken as the expected cation distribution and are given in table (1). There is a cation preference for some ions as an example zinc ion has chemical affinity toward tetrahedral site and a fraction of Zn ion occupy octahedral site. The octahedral and tetrahedral radii are found to decrease with increasing Mg content as shown in figure 2. This is attributed to the decrease in lattice parameter (a) with increasing Mg content [7, 8]. Lavine [8] has suggested that there is an inverse relationship between the covalent character of the spinel structure and the bond length. Since the bond length \( R_A \) decrease with Mg content it can be concluded that there is an increase of ionic – covalent character of the spinel with Mg content. The decrease of zinc content at tetrahedral site leads to a noticeable decrease in \( r_A \) as shown in figure 2.

Table 1. X-ray cations distribution of "Ni-Cu-Zn" system for different Mg content (x).

| x  | A site          | B-site                                      |
|----|----------------|---------------------------------------------|
| 0  | Zn\(_{0.7}\) Fe\(_{0.3}\) | Ni\(_{0.1}\) Cu\(_{0.2}\) Fe\(_{1.7}\)       |
| 0.15 | Zn\(_{0.4}\) Fe\(_{0.6}\) | Ni\(_{0.1}\) Cu\(_{0.2}\) Zn\(_{0.15}\) Mg\(_{0.25}\) Fe\(_{1.4}\) |
| 0.25 | Zn\(_{0.3}\) Fe\(_{0.7}\) | Ni\(_{0.1}\) Cu\(_{0.2}\) Zn\(_{0.15}\) Mg\(_{0.25}\) Fe\(_{1.3}\) |
| 0.35 | Zn\(_{0.25}\) Fe\(_{0.75}\) | Ni\(_{0.1}\) Cu\(_{0.2}\) Zn\(_{0.15}\) Mg\(_{0.35}\) Fe\(_{1.25}\) |
| 0.45 | Zn\(_{0.15}\) Fe\(_{0.85}\) | Ni\(_{0.1}\) Cu\(_{0.2}\) Zn\(_{0.15}\) Mg\(_{0.45}\) Fe\(_{1.15}\) |
| 0.55 | Zn\(_{0.1}\) Fe\(_{0.9}\)  | Ni\(_{0.1}\) Cu\(_{0.2}\) Zn\(_{0.05}\) Mg\(_{0.55}\) Fe\(_{1.1}\) |
| 0.7  | Fe\(_{1}\)      | Ni\(_{0.1}\) Cu\(_{0.2}\) Mg\(_{0.7}\) Fe\(_{1.3}\) |

The net magnetic moment for NiCuZn ferrite system was calculated from x-ray cation distribution using Neal's theory and given in table (2):

\[ \mu_{th} = |\mu_B| - |\mu_A| \]

where \( \mu_B \) and \( \mu_A \) are the magnetic moments of octahedral site (B-site) and tetrahedral site (A-site). The magnetic moment of Fe\(^{+3}\) was taken (5\( \mu_B \)), (Cu\(^{+2}\)=1.73 \( \mu_B \)), (Ni\(^{+2}\)=2\( \mu_B \)), (Mg\(^{+2}\)= 0\( \mu_B \)) and (Zn\(^{+2}\)=0\( \mu_B \)). It is noticed that the x-ray magnetic moment decrease by increasing Mg content, this is due to the migration of Fe\(^{+3}\) ion from octahedral to tetrahedral site which lead to the decrease of net magnetization of the ferrite sample.
Table 2. Theoretical magnetic moment $\mu_{th}$ and $\mu_A/\mu_B$ of “Ni-Cu-Zn” system for different Mg content (x).

| X   | $\mu_{th}$ (μB) | $\mu_A/\mu_B$ |
|-----|-----------------|----------------|
| 0   | 7.546           | 0.1658         |
| 0.15| 4.546           | 0.3976         |
| 0.25| 3.546           | 0.4967         |
| 0.35| 3.046           | 0.5518         |
| 0.45| 2.046           | 0.6750         |
| 0.55| 1.546           | 0.7443         |
| 0.7 | 0.546           | 0.9016         |

The variation of bond length $R_A$ and $R_B$ with Mg content are given in Table (3), where $R_A$ is considered the shortest distance between A-site and Oxygen ion O$^{2-}$ and $R_B$ is considered the shortest distance between B-site and Oxygen ion O$^{2-}$. The bond length was calculated using the equation:

$$R_A = a \sqrt{3} \left( \delta + \frac{1}{b} \right)$$

$$R_B = a \left( 3\delta^2 - \frac{\delta}{2} + \frac{1}{16} \right)^{(1/2)}$$

where $\delta = (u - 0.375)$ and $u$ is the Oxygen positional parameter. For ideal ferrite $u = 0.375$. The values of $R_A$ decrease by increasing Mg content which is attributed to the decrease of lattice parameter. The normality of spinel structure decrease by increasing Mg content which is directly proportional to the bond length $R_A$ and inversely proportional to $R_B$. Similar results have been reported in the case Li-Cu-Zn ferrite [5,9]. The oxygen positional parameter $u$ was calculated from the equation:

$$u = \frac{r_a r_o}{a} + \frac{1}{4}$$

and are given in table (3). The $u$ parameter is the distance between the cation ions and oxygen ions which varied by the accommodation of magnesium ions in the ferrite unit cell.

Table 3. Bond length $R_A$, $R_B$ and Oxygen positional parameter (u) for "Ni-Cu-Zn" system for different Mg content (x).

| X   | $R_A$(Å) | $R_B$(Å) | u(Å)  |
|-----|----------|----------|-------|
| 0   | 2.03     | 1.979    | 0.3898|
| 0.15| 2        | 1.991    | 0.3879|
| 0.25| 1.99     | 1.994    | 0.3873|
| 0.35| 1.985    | 1.996    | 0.3863|
| 0.45| 1.975    | 1.999    | 0.3864|
| 0.55| 1.97     | 2        | 0.3861|
| 0.7 | 1.96     | 2.003    | 0.3855|
The crystallite size of our ferrite samples were estimated from the broadening of x-ray peak (311) using Scherer's equation and shown in fig (6):

\[
D = \frac{k \lambda}{\beta \cos \theta}
\]  

(10)

where \( k = 0.89 \) is constant, \( \Theta \) is the peak location, \( \beta \) is the full width at half maximum of the peak, and \( \lambda \) is the wavelength of the X-ray for Cu-K\( \alpha \) radiation \( (\lambda = 1.540598 \ \text{Å}) \). The calculation shows that the crystallite size is ranged from 26.36 to 39.26 nm and decrease by increasing Mg content. The decrease trend of the crystallite size by increasing Mg content could be explained in the bases of crystal growth process which depends on many factors:

1) The molecular connection of the material at the surface of the crystal during growth process.
2) The site preference of the cations in the ferrite system.

The surface temperature affect the molecular concentration at the surface of the crystal and hence the crystal growth rate. In the second case the grain growth is retarded when the cation preference are not fully satisfied. Thus, we can say that as the Mg content increase, more heat may be liberated, leading to the decrease in molecular concentration at the crystal surface and hence obstructs the crystallite growth [2].

![Figure 6. Crystallite size (D) for "Ni-Cu-Zn" system for different Mg content (x).](image)

3.2. Magnetic properties analysis

The magnetic hysteresis loop for the ferrite system \( \text{Ni}_{0.1} \text{Cu}_{0.2} \text{Mg}_x \text{Zn}_{0.7-x} \text{Fe}_2\text{O}_4 \), \( (x= 0.00, 0.15, 0.25, 0.35, 0.45, 0.55 \text{ and } 0.70) \) were recorded at room temperature and presented in figure 7 as M-H curves. The magnetization curve were characterized by low coercivity \( (H_c) \) indicating that our material are soft magnetic material and may be like superparamagnetic material. The unsaturated nature of magnetization even at high magnetic fields can be attributed to the core –shell structure of the magnetic nanoparticles at which the core possesses spinel ferromagnetic structure and the shell consist of variable co-ordination (spin- glass like structure at the surface) as shown in figure 8 [10].

Therefore it is difficult to align the magnetic particle along the applied field [11]. The saturation magnetization was calculated from the extrapolation of M to M axis. From Neal's theory of two sub-lattice model, the theoretical magnetization can be calculated using equation (6).
The experimental magnetic moment $\mu_{\text{exp}}$ of the studied ferrite system has been calculated from the following equation:

$$\mu_{\text{exp}} = \frac{M_w \times M_s}{5585}$$

where $M_w$ is the molecular weight of particular compositions, and $M_s$ is the saturation magnetization. The values of saturation magnetization ($M_s$), coercivity ($H_c$), remanent magnetization ($M_r$), experimental ($\mu_{\text{exp}}$) and theoretical ($\mu_{\text{th}}$) magnetic moment and Yafet- kettle angle ($Y_\alpha$) are given in table (4). It can be seen that both $M_s$ and $H_c$ increase by increasing Mg content and $M_s$ reach maximum value at $x = 0.35$ Mg content. The magnetic
behavior of ferrite nanoparticles depends on many aspects like the method of synthesis, cation distribution between tetrahedral and octahedral site and crystallite size [12]. It is well known that the nickel ferrite is inverse spinel structure, whereas zinc ferrite has normal spinel structure. Our cation distribution indicates that the substitution of magnesium at octahedral site cause the Fe$^{3+}$ to migrate at tetrahedral site resulting in the strengthening the A-B super exchange interaction and resulting in the increase of magnetic moment and saturation magnetization. However as magnesium concentration is being increased, the saturation magnetization begins to decrease at higher value $x \geq 0.35$. Similar trend of the variation in saturation magnetization was observed in NiCuZn ferrites [13-15]. To understand the possible reason for this behavior, the magnetic moment per molecule of the studied samples from the cation distribution were calculated as shown in Fig(9). The behavior of magnetic moment is the same as saturation magnetization so we can say that the cation distribution can explain the magnetization behavior of the present samples. There are other reasons that affect the behavior of our magnetic samples like core-shell interaction, the magneto crystalline anisotropy and spin canting.

### Table 4

| $x$  | $M_s$ (emu/g) | $H_c$ (G)  | $M_r$ (emu/g) | $\mu_{exp}$ ($\mu_B$) | $\mu_{th}$ ($\mu_B$) | $Y$ (°) |
|------|--------------|-----------|--------------|-----------------------|----------------------|---------|
| 0    | 15.591       | 47.520    | 0.29061      | 0.6701                | 7.546                | 76.1196 |
| 0.15 | 23.340       | 49.479    | 0.82521      | 0.9774                | 4.546                | 58.1947 |
| 0.25 | 25.252       | 58.399    | 1.1598       | 1.0389                | 3.546                | 49.896  |
| 0.35 | 28.487       | 50.109    | 1.1426       | 1.1510                | 3.046                | 43.8497 |
| 0.45 | 28.014       | 87.936    | 1.9664       | 1.1113                | 2.046                | 31.5934 |
| 0.55 | 27.128       | 106.13    | 2.2534       | 1.0562                | 1.546                | 23.2219 |
| 0.7  | 16.229       | 165.37    | 1.9476       | 0.6139                | 0.546                | -       |

The coercivity for multi-domain particles increase as the particle size decrease and at critical size the particle become single domain in nature and coercivity decrease. As particle size if further decrease, the coercivity increase again as multi-domain particle. So at the sample $x=0.35$, the particle transform from single domain to multi-domain particle. For single domain particles the coercivity decreases with the increase of particle size as shown in figure10.
The $H_c$ has maximum peak value at $x = 0.35$ whereas the porosity has minimum value at the same ratio. At higher value the two factors have direct proportionality below $x = 0.35$ the $H_c$ has inverse proportionality. The variation in $H_c$ values can be explained, as shown in figure 11 due to the relation:

$$H_c = \frac{2K}{\mu_0 M_s}$$

(12)

where $\mu_0$ is universal constant permeability at free space, $K$ is magnetic crystalline anisotropy and $M_s$ is saturation magnetization.

The $M_s$ and porosity have opposite behavior with magnesium content. According to the following equation:

$$M_s = \frac{2K}{\mu_0 H_c}$$

(13)

As the porosity decrease the magnetic anisotropy increase and so the saturation magnetization increase up to ratio $x = 0.35$ above this ratio the porosity increase leading to the increase of magnetic anisotropy and hence the saturation magnetization decrease as shown in figure 12. The increase of magnesium content at B-site dilutes the magnetization of B-site leading to the decrease of the magnetic moment. The canting angle between the magnetic moment at B-site was calculated and this angle considered the reason of the difference between theoretical and experimental magnetic moment. This angle determines the strength of B-B interaction and given by the following expression:

$$\mu_{\text{exp}} = \mu_B \cos Y - \mu_A$$

(14)

At high magnesium value the result of $\mu_{\text{th}}$ are very near to the experimental which mean that the Yafet angle is very small whereas at low magnesium value the value of Y- kattle angle is high.

![Figure 10](image_url)  
**Figure 10.** a) Coercivity, b) particle size for "Ni-Cu-Zn" system for different Mg content ($x$).
3.3 Microstructural analysis.

Figure 13 shows scanning electron microscope photography of the samples Ni$_{0.1}$ Cu$_{0.2}$ Mg$_x$ Zn$_{0.7-x}$ Fe$_2$O$_4$ synthesis by auto combustion flash method which annealed at 600 °C. It was shown that the average grain size decreases with increasing Mg content due to the presence of magnesium oxide which acts as microstructural stabilizer. Nevertheless, magnesium oxide is stable oxide which prevents the tendency of discontinuous grain growth [2]. The SEM micrographs of all samples show non-uniform grain growth. The particles of varying sizes and different shapes have been observed. The magnetic dipole moment of the studied samples increases by increasing Mg content which leads to a grain agglomeration due to the attractive force [16]. The average grain size was measured using the intercept method followed by the equation [1].

\[
\text{Grain size} = \frac{1.5L}{MN}
\]  

(15)
where $L$ is the total length of the test line, $M$ is the magnification and $N$ is the total number of intercepts in the SEM micrographs. The average grain size for all samples are given in table (5). The micrographs reveal that the grains are separated by pores in all samples. The segregation of the impurity phase was not observed, this indicates that the complete solid solubility was obtained with the magnesium substitution.

**Table 5. Grain size for "Ni-Cu-Zn" system for different Mg content (x).**

| x     | Grain size (μm) |
|-------|-----------------|
| 0     | 0.57            |
| 0.15  | 0.46            |
| 0.25  | 0.42            |
| 0.35  | 0.30            |
| 0.45  | 0.16            |
| 0.55  | 0.15            |
| 0.7   | 0.135           |

a) x = 0   b) x = 0.15   c) x = 0.25   d) x = 0.35
e) $x = 0.45$

f) $x = 0.55$

g) $x = 0.7$

Figure 13. Micrographs (a-g) for "Ni-Cu-Zn" system for different Mg content (x).

4. Conclusion

$\text{Ni}_{0.1} \text{Cu}_{0.2} \text{Zn}_{0.7-x} \text{Mg}_x \text{Fe}_2\text{O}_4$ (where $x = 0, 0.15, 0.25, 0.35, 0.45, 0.55$ and $0.7$) were prepared by flash method. The x-ray diffraction patterns for $\text{Ni}_{0.1} \text{Cu}_{0.2} \text{Zn}_{0.7-x} \text{Mg}_x \text{Fe}_2\text{O}_4$ showed the formation of single phase spinel structure. The crystallite size is ranged from 26.26 to 39.36 nm. The x-ray magnetic moment decreases by increasing Mg content. At high Mg value the result of experimental magnetic moment are very near to the theoretical. The micrographs reveal that the grains are separated by pores in all samples.

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