Effects of varying chemical composition with $x = 0.1 – 0.7$ on magnetic properties of soft ferrite $\text{Ni}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$

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Abstract. The preparation of NiZn ferrites with varying composition have been synthesized by mechanical milling method. The starting iron oxide ($\text{Fe}_2\text{O}_3$), nickel oxide ($\text{NiO}$), and zinc oxide ($\text{ZnO}$) were mixed with seven compositions of $\text{Ni}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ with $x = 0.1 – 0.7$. The effects of the composition on the structural and magnetic properties of the ferrite have been investigated. Structural properties were observed by X-ray diffraction (XRD) (Bruker’s). XRD patterns match well with the characteristic reflections of f d-3 cubic spinel structure, and it confirms that the all samples have single phase. Magnetic properties characterized by Permagraph Magnet Physik at room temperature. Hysteresis curve results show that varying composition influence the magnetic properties. Sample that have the highest $M_s$ and $M_r$ values is when $x = 0.3$ with magnetization of remanence ($M_r$) = 30.1538 emu/g, magnetization of saturation ($M_s$) = 39.1642 emu/g, coercivity ($H_cJ$) = 0.3040 kA/m, and density = 4.40 g/cm$^3$. The highest $H_cJ$ value when $x = 0.2$, coercivity ($H_cJ$) = 0.4050 k/m.

1. Introduction
NiZn ferrite is a soft ferrite magnetic material $\text{ABFe}_2\text{O}_4$ with the spinel structure base on a face- center cubic lattice of the oxygen ions. Each spinel unit cell contains eight formula units, were the metallic cation in occupy the 64 tetrahedral site (A) and metallic cations occupy the 32 octahedral site (B) [1]. The spinel structure of ferrites that indicating the tetrahedral and octahedral sites is shown in figure.1.

Figure 1. The crystal structure of spinel ferrite [2]
NiZn ferrite are of interest for researchers to sufficiently explore and study due to the ferrites that are widely used in many electronic applications such as; microwave, absorber, antenna, etc., [3-5]. The various forms of NiZn ferrite in electronic components are shown in figure 2. The magnetic and physical properties of ferrite are depend upon methodology adopted for the preparation [6]. Commonly ferrites produced by ceramic sintering involving high temperature solid state reactions between the constituent oxides or carbonates [7]. The other methods that use by researcher are wet chemical methods like sol gel [8], citrate-nitrate precursor method [9], microwave sintering [10], chemical coprecipitation method [11] etc. Magnetic properties are also influenced by composition a stoichiometric nickel-zinc ferrite, Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$ [12]. Le Zeong et.al. [13] have prepared Ni$_{0.5-x}$Zn$_{0.5}$Sr$_x$Fe$_2$O$_4$ with x = 0, 0.05, 0.10, 0.15, 0.20 were synthesized by the sol-gel auto-combustion method and added strontium in the materials. XRD results show that the increasing Sr substitution causes the lattice parameter increases and the average crystallite size decreases. Jun Hu and Mi Yan [14] reported that addition of CuO and V$_2$O$_5$ on preparation of NiZn ferrite can reduce the granulity of raw materials and magnetic properties of the ferrite obtained at low sintering temperature from 1200 °C to 930 °C.

![Figure 2. The various kinds of NiZn ferrite in electronic applications [5]](image)

The others previous studies have also reported effect of composition by added others elements such as Cu [15], Mn [9], Sr [1], etc. can effects the magnetic properties of NiZn ferrite. In the present work, the synthesis of NiZn ferrite were prepared by mechanical milling method with varying chemical composition of Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$ with x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7. The objective of this study is to confirm that various compositions can affect magnetic properties, so that it can provide information the best composition in preparing ferrite NiZn magnets with mechanical alloying methods. Characterized structural properties using X-Ray Diffraction (Bruker’s) and magnetic properties by Permagraph.

2. Experiment
The preparation of NiZn ferrite with compositions of Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$ (where x= 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7) were synthesized by mechanical milling method. This composition was chosen, because there were no reports of articles as same as on the composition done in this research. A. Silva [16] used the composition x = 0.5 at temperature 1000 °C, whereas M. Sorescu [17] prepared NiZn ferrites with composition x = 0.1 at temperature 1275 °C. The following chemical analytical grade were used as raw materials Fe$_2$O$_3$ (>99% E-Merck), NiO (>99% E-Merck), and ZnO (>99% E-Merck). The schematic procedure of the process can be described as follows on figure 3. Stoichiometric quantities of raw materials that have been calculated with mol percentages were first mixed by ball milling machine with ball steels in alcohol medium for 8 hours. After all raw materials completely mixed, followed by calcination process at temperature 900 °C for 2 hours. The calcined powder was crushed by ball milling machine for 8 hours to get fine powder up to 400 mesh. The finely powder then compacted in ring shape
at pressure 110 kg.cm\(^{-2}\). Finally the samples were sintering at temperature 1260 °C for 3 hours. H. Jun [18] also prepared NiZn ferrite with same raw materials with the composition (Ni\(_{0.17}\)Zn\(_{0.63}\)Cu\(_{0.20}\))Fe\(_{1.915}\)O\(_4\) by mechanical milling method, was sintered at 930°C in air for 5 h.

Figure 3. The schematic preparation of soft ferrite Ni\(_{1-x}\)Zn\(_x\)Fe\(_2\)O\(_4\)

In order to investigation the influence chemical composition of Ni\(_{1-x}\)Zn\(_x\)Fe\(_2\)O\(_4\) with x = 0.1-0.7, the magnetic properties were characterized by Permagraph Magnet Physik and the formation of NiZn ferrite compounds by XRD Bruker with Cu Kα radiation.

3. Result and Discussion

3.1. Magnetic Properties

Figure 4 shows the hysteresis loops of Ni\(_{1-x}\)Zn\(_x\)Fe\(_2\)O\(_4\) ferrite as a function of varying composition with x = 0.1-0.7. The hysteresis curve shows that the variation of the composition greatly influences the magnetic properties, every composition has a different hysteresis curve. The curves show the change of saturation magnetization (\(M_s\)), magnetization remanence (\(M_r\)), and coercivity (Hc). The \(M_s\) and \(M_r\) of sample when x = 0.1 is low, furthermore both magnetic properties increase for x = 0.2-0.3, and begin to decrease when x = 0.4. Whereas the coercivity value (Hc) initially decreases with x more than 0.2. The
difference in the hysteresis curves are due to the sensitive magnetic properties of ferrites dependent heavily on chemical composition and structure, but defects, crystallite size, internal strain and cation distribution will also have an effect.

![Hysteresis loops of Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$ ferrites](image)

**Figure 4.** Hysteresis loops of Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$ ferrites

A summary of the magnetic properties that obtained in this research along with the density, can be seen in table 1. Sample that have the highest $M_s$ and $M_r$ values is when $x = 0.3$, whereas the highest $H_c$ value when $x = 0.2$.

| Sample | $M_s$ (emu/g) | $M_r$ (emu/g) | $R = M_r/M_s$ | $H_c$ (kA/m) | Density $p$ (g/cm$^3$) |
|--------|---------------|---------------|--------------|---------------|---------------------|
| x1     | 8.4698        | 4.3322        | 0.51         | 0.3600        | 4.20                |
| x2     | 24.8520       | 17.6373       | 0.71         | 0.4050        | 4.30                |
| x3     | 39.1642       | 30.1538       | 0.77         | 0.3040        | 4.40                |
| x4     | 38.3544       | 27.8003       | 0.72         | 0.2440        | 4.29                |
| x5     | 37.9974       | 25.9064       | 0.68         | 0.1510        | 4.38                |
| x6     | 32.7354       | 17.2575       | 0.53         | 0.0950        | 4.60                |
| x7     | 22.7854       | 9.7287        | 0.43         | 0.0387        | 4.40                |

Figure 5 shows dependence of saturation and remanence of Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$. This figure indicates that $M_s$ and $M_r$ increased as the $x = 0.1$, 0.2, and 0.3 and decrease when $x = 0.4$, 0.5, 0.6, and 0.7. The remanence ratio, $R = M_r/M_s$ is a characteristic parameter of the material that indicates the isotropic properties of the material [19]. The lower the R value the more isotropic, the higher the R value the more anisotropic. The value of the R ratio shows the ease with the magnetization direction of reorientation to the nearest easy axis magnetization after the magnetic field is removed.
Figure 5. Dependence of magnetization saturation and magnetization remanence of Ni1–xZnxFe2O4 ferrites (x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, and 0.7)

As a comparison, A.S. Albuquerque [16] prepared NiZn-ferrite by sol-gel processing. The magnetic properties of samples were obtained with saturation magnetization between 1.3 and 68 emu/g and coercivity ranging from 0 to 123 Oe. J. Azadmanjiri [20] reported the study of Ni–Zn ferrite with composition of Ni1–xZnxFe2O4 (where x = 0, 0.1, 0.2, 0.3, 0.4) were prepared by the chemical sol–gel method. The VSM measurement results of Ni–Zn ferrite sintered at 800 and 950°C are shown that the saturation magnetization ferrite sintered at 800°C is 62 emu/g and increased to 73 emu/g at 950°C.

Besides chemical composition, magnetic remanence and saturation is an extrinsic property that also depends on phases, microstructure, grain size, grain boundary, porosity, and structural defects, are important factors for obtaining good magnetic material performance [11].

3.2. Structure properties

X-ray diffraction patterns Ni1–xZnxFe2O4 are shown in figure 6. The all patterns match well with the characteristic reflections of f d-3 cubic spinel structure, and it confirms that the all samples have single phase referred to reference code ICSD 98-006-8765 with compound name NiZn Ferrite.

Figure 6. XRD patterns of Ni1–xZnxFe2O4. The h k l index is based on the cubic cell.
All the XRD peaks indicate the same crystal structure of the NiZn ferrite on database of XRD bruker’s raw data. The lower peaks show the composition when $x = 0.1$ and 0.4, the others peak show the same pattern. Changes in composition do not much affect structure properties of the NiZn ferrite NiZn, but can affect the lattice parameters. K. Rama Krishna [21] have been studies x-ray diffraction pattern of $\text{Ni}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ with $x = 0.0, 0.2, 0.6, 0.8,$ and 0.9 were prepared by citrate gel method. The X-Ray diffractogram show that all a single phase cubic spinel structure and the lattice parameter increases with Zn$^{2+}$ composition. Change in composition increases the lattice parameters of NiZn ferrite, this is in accordance with vegard’s law [22]. The variation of Zn$^{2+}$ composition with lattice parameter is shown in the figure 7.

![Figure 7](image.png)

**Figure 7.** Variation of lattice parameter as a function of composition $\text{Ni}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ with $x = 0.0, 0.2, 0.6, 0.8,$ and 0.9 [19].

In a previous study by Ravender [23] has been reported similar things in case of Cu-Zn ferrites that the variation of lattice parameter can be clarified on the basis of ionic radius of Zn$^{2+}$ (0.82 Å) is greater than Ni$^{2+}$ (0.78 Å). Alexandre R has reported that addition of Mn$^{2+}$ in composition $\text{Ni}_{0.50-x}\text{Zn}_{0.50-x}\text{Mn}_{2x}\text{Fe}_2\text{O}_4$ composition (where $x = 0.00, 0.05, 0.10$ and 0.15). The addition of Mn increases the lattice parameter of the crystal because the ionic radius of Mn$^{2+}$ (0.091nm) is larger than that of Ni$^{2+}$ (0.078 nm) and Zn$^{2+}$ (0.082 nm), this case explains the increase in the crystalline lattice parameters [9]. The X-RD observation can inform that the composition can affect the phase, structure, and parameters of the lattice parameter of crystal.

### 4. Summary

We have investigated the influence of chemical composition on a series of $\text{Ni}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ ferrites by varying the composition with $x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6,$ and 0.7. The magnetic properties value $M_s$, $M_r$, $R$, and $H_c$ are influenced by the composition. The maximum magnetic properties were obtained when $x = 0.3$. $M_s = 39.1642 \text{ emu/g}$, $M_r = 30.1538 \text{ emu/g}$, $R = 0.77$, and $H_c = 0.3040 \text{ kA/m}$. Dependence of magnetization saturation and magnetization remanence increase at $x = 0.1, 0.2,$ and 0.3 and than decrease at $x = 0.4, 0.5,$ and 0.6. The XRD results indicate that all samples are single phase with f d-3 cubic spinel structure, but peak of XRD pattern quite different. The lower peak obtained when $x = 0.1$ and 0.4 otherwise the others composition have higher peak.

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