STRUCTURE AND MAGNETIC PROPERTIES OF NANOCRYSTALLINE NiZnFe$_2$O$_4$ PREPARED VIA CONVENTIONAL CERAMIC METHOD

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Abstract

Nanocrystalline Nickel-Zinc ferrite was synthesized by conventional ceramic method. The metal oxide of Ni, Zn and Fe were used as precursors for NiZnFe$_2$O$_4$. The structural characterizations were made for the samples with a chemical formula Ni$_x$ Zn$_{1-x}$ Fe$_2$O$_4$ (x = 0.1, 0.3, 0.5 and 0.7mol). The obtained nanocrystalline Ni-Zn ferrite was analysed and discussed through structural, morphological and magnetic characterization. Formation of pure NiZnFe$_2$O$_4$ phase was confirmed by X-ray diffraction analysis (XRD). The determined material's nanocrystalline structure was additionally supported by scanning electron microscopy (SEM). The magnetic hysteresis loop was recorded by means of a Magnet-Physik(EP-3).

Keywords: nanocrystalline, nickel-zinc ferrite (NiZnFe$_2$O$_4$),conventional ceramic method, XRD,SEM.

Introduction

Among the different mixed ferrites, Ni-Zn ferrites have a good utility as a conducted noise suppressor material in various electromagnetic interfaces compared to other ferrites. Because of their high resistivity, relatively high permeability and low eddy current loss. These soft magnetic materials, crystallizes in the spinel structure of the type (Zn$_{1-x}$Fe$_x$) (Ni$_x$Fe$_{2-x}$) O$_4$, where the metallic cations Fe$_{3+}$/Zn$_{2+}$ occupy the tetrahedral A sites, and the metallic cations Fe$_{3+}$/Ni$_{2+}$ occupy the octahedral B sites. It is known that magnetic properties of ferrites are sensitive to preparation technique and their microstructures.

The electrical and magnetic properties of such ferrites depend strongly on distribution of cations at the tetrahedral (A) and octahedral (B) sites in the lattice. It is well known that zinc ions can be used to alter the saturation

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magnetization. It is believed that the addition of zinc ions also affects the lattice parameter and it would therefore be expected to change the Curie temperature of the material. The substitution of divalent ions in pure ferrites leads to the modification of the structural, electrical and magnetic properties. There are the synthesis of Ni-Zn ferrites using different techniques like, refluxing process , ceramic , hydrothermal , combustion, co-precipitation, reverse micelle process, spark plasma sintering, micro emulsion and ball milling, etc.

Among these methods, the conventional solid-state reaction route is widely used for the production of ferrite because of its low cost and suitability for large scale production. In this work, the systematic doping of Ni content on the magnetic properties of Ni-Zn ferrite was synthesized by solid state double sintering method.

Experimental

Nickel-Zinc ferrites were prepared by solid state sintering method. The starting materials were nickel oxide(NiO), and, zinc oxide (ZnO) and ferrite oxide(Fe_2O_3) all of analytical grade. The structural characterizations were made for the samples with a chemical formula $Ni_x Zn_{1-x} Fe_2O_4$ (x = 0.1, 0.3, 0.5 and 0.7) mol. The metal oxides will be mixed and grind in A-gate mortar. The mixture will be pre-sintered at 800°C in the furnace. Afterwards the powder was pressed into pellets of thickness 3 mm and a diameter of 10 mm with press by applying a pressure of 2 tons/in^2. The pellet will be sintered at 1000°C in the furnacefor one hour to obtain spinel phase. The final sintering was done at 1000°C. The structural characterizations of all samples were carried out by X-ray diffraction (XRD) and conforms the well defined single phase spinel structure. XRD data were taken at room temperature using CuKα radiation. Morphological, elemental composition characterizations of all prepared samples were performed by high resolution scanning electron microscopy. The Magnetic measurements were carried out by using Magnet-Physik (EP-3).
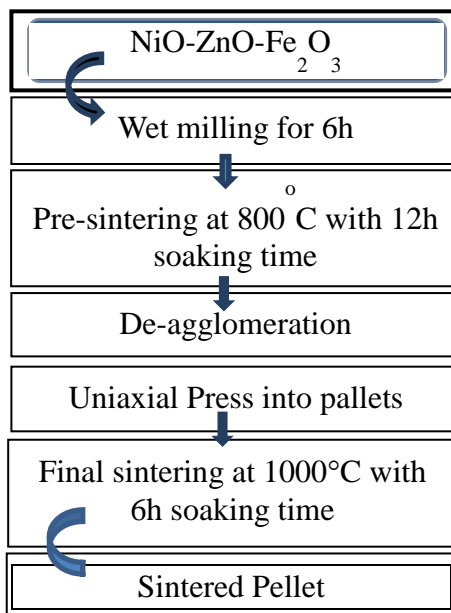
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┌─────────────────────────────────┐
│        NiO-ZnO-Fe₂O₃            │
└─────────────────────────────────┘
┌─────────────────────────────────┐
│        Wet milling for 6h       │
└─────────────────────────────────┘
┌─────────────────────────────────┐
│  Pre-sintering at 800°C with 12h│
│          soaking time           │
└─────────────────────────────────┘
┌─────────────────────────────────┐
│        De-agglomeration         │
└─────────────────────────────────┘
┌─────────────────────────────────┐
│    Uniaxial Press into pallets  │
└─────────────────────────────────┘
┌─────────────────────────────────┐
│   Final sintering at 1000°C with│
│         6h soaking time         │
└─────────────────────────────────┘
┌─────────────────────────────────┐
│        Sintered Pellet          │
└─────────────────────────────────┘
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Figure 1: Flow chart for coventional synthesis of $NiZnFe_2O_4$ fine powder

Results and Discussion

XRD Analysis

The measured results of metal ions are consistent well with the stoichiometric values of samples, indicating that the compositions of as-obtained samples are in agreement with those of anticipated stoichiometry. The X-ray diffraction patters of the samples are shown in Figure 2. The XRD pattern of metal ions presents a broad diffraction peak located at 22.04°. In Figure2, The main diffraction peaks appeared at $2\theta = 18.28°$, 30.17°, 35.52°, 43.17°, 53.53°, 57.06° and 62.76°correspond to (111), (220), (311), (400), (422),(511) and (440) crystal planes of the spinel structure with the characteristic reflections of the Fd3m cubic group (JCPDS CARD 01-1108).All the Nickle substituted zinc ferrites of the various compositions show the crystalline cubic spinel structure. The sharp peaks represents that all ferrites are crystalline nature of single phase.
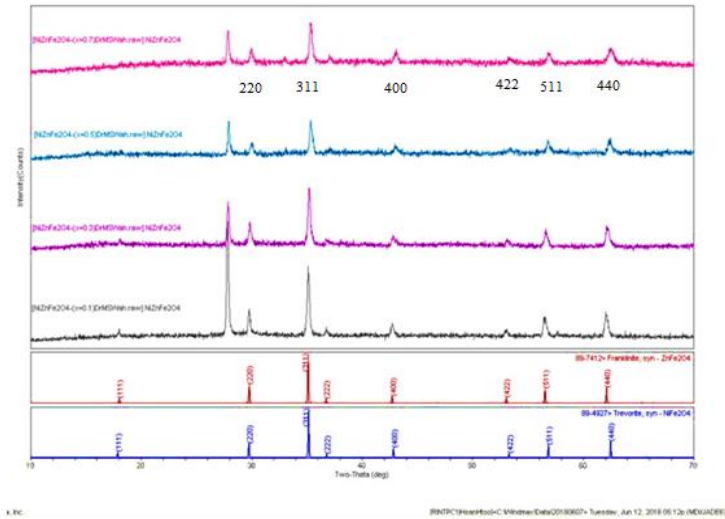
Figure 2: XRD spectrums of powder (NiZnFe$_2$O$_4$)

According to the Scherrer equation:

$$D = \frac{k\lambda}{\beta cos\theta} \qquad (1)$$

where D is the particle size (nm), b is the full width at half maximum diffraction peak, is the diffraction angle and k(0.15418 nm) is the wavelength of X-ray.

The average crystallite sizes of the NiZnFe$_2$O$_4$(NZF) nanoparticles estimated from the equation (1) were showed in Table 1. The average size of NZF nanoparticle decreases with the increase of x. The lattice parameter of individual composition was calculated by using the formula:

$$a = d[h^2 + k^2 + l^2]^2 \qquad (2)$$

where, a = lattice constant; d = inter planar distance; and (h, k, l) are the Miller indices.

The calculated lattice constant "a" is seen to increase from 8.47Å to 8.38 Å with decrease in nickle content as reported in Table 1. The lattice parameter is found vary linearly with decreasing nickle concentration, there by indicating that the Ni-Zn ferrite system obeys Vegard's law . A similar behavior of lattice constant with dopant concentration was observed by

several investigators in various ferrite systems. The variation in lattice constant with nickle content can be explained on the basis of the ionic radii of Ni^{2+} (0.78 Å) ions is lower than that of Zn^{2+} (0.82 Å) .

Table 1: The variation of lattice parameter for intense reflection peaks $NiZnFe_2O_4$

Sample	Crystallite size (nm)	Lattice parameter (A^o)
$Ni_{0.1} Zn_{0.9} Fe_2O_4$	37.8	8.4702
$Ni_{0.3} Zn_{0.7} Fe_2O_4$	33.9	8.4490
$Ni_{0.5} Zn_{0.5} Fe_2O_4$	32	8.4160
$Ni_{0.7} Zn_{0.3} Fe_2O_4$	29	8.3860

Figure 2 showed the XRD spectrums of the powder heat treated at 1000°C using high temperature solid state sintering method. Theamount of crystalized size decreased with variable of molecular ratios. X-ray density (dx) was determined using the following relation

$$D_x = \frac{ZM}{Na^3}$$

where Z is the number of molecules per unit cell (Z = 8), "M" is the molecular weight and "N" is the Avogadro's number. The values of the bulk density, X-ray density and the percentage of porosity for Ni-Zn ferrites are given in Table 2.

Table 2: Bulk density-ray density and porosity data for mixed Ni-Zn ferrites

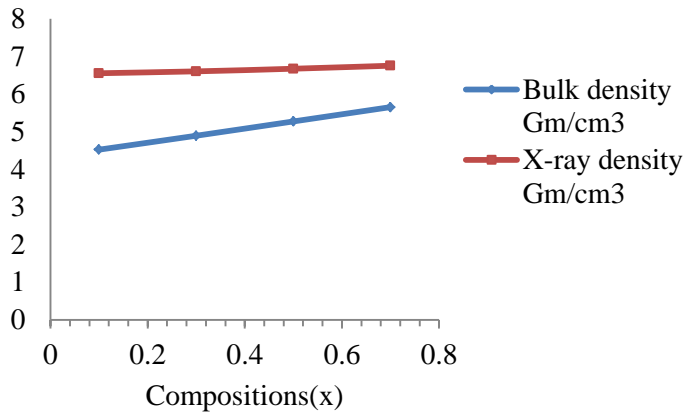| Sr. No.	Ferrite composition	Bulk density Gm/cm^3	X-ray density Gm/cm^3	Porosity
2	X = 0.1	4.52	6.55	30.99
3	X = 0.3	4.89	6.60	25.90
4	X = 0.5	5.27	6.67	20.98
5	X = 0.7	5.65	6.75	16.29

Figure 3: Variation of bulk density and X-ray density with composition

It can be seen from the table that the bulk density increases and the porosity decreases progressively with addition of nickel to zinc ferrite. Zinc ferrite having the least porosity, this conforms the observation that the addition of nickel to zinc ferrite results the densification of the material. The variation of bulk density with nickel content for mixed Ni-Zn ferrite is shown in the Figure 3. It may be seen from the figure that bulk density increases linearly with the increase of nickel content.

Scanning Electron Microscopy (SEM)

The scanning electron microscope (SEM) images of all prepared samples are given in Figure 3(a, b, c & d).The morphology and the size distribution of the $NiZnFe_2O_4$ nanoparticles were determined using SEM. Typical SEM images of $NiZnFe_2O_4$ synthesized particles are shown in Figure 4. SEM micrograph depicts that the samples contain micrometrical aggregation of tiny particles. The existence of high dense agglomeration indicates that pore free crystallites are present on the surface. The SEM images show the agglomerated form of $NiZnFe_2O_4$ nanoparticles. As the nanoparticles possess high surface energies, they tend to agglomerate and grow into larger assemblies.

(a)(x=0.1)   (b)(x=0.3)

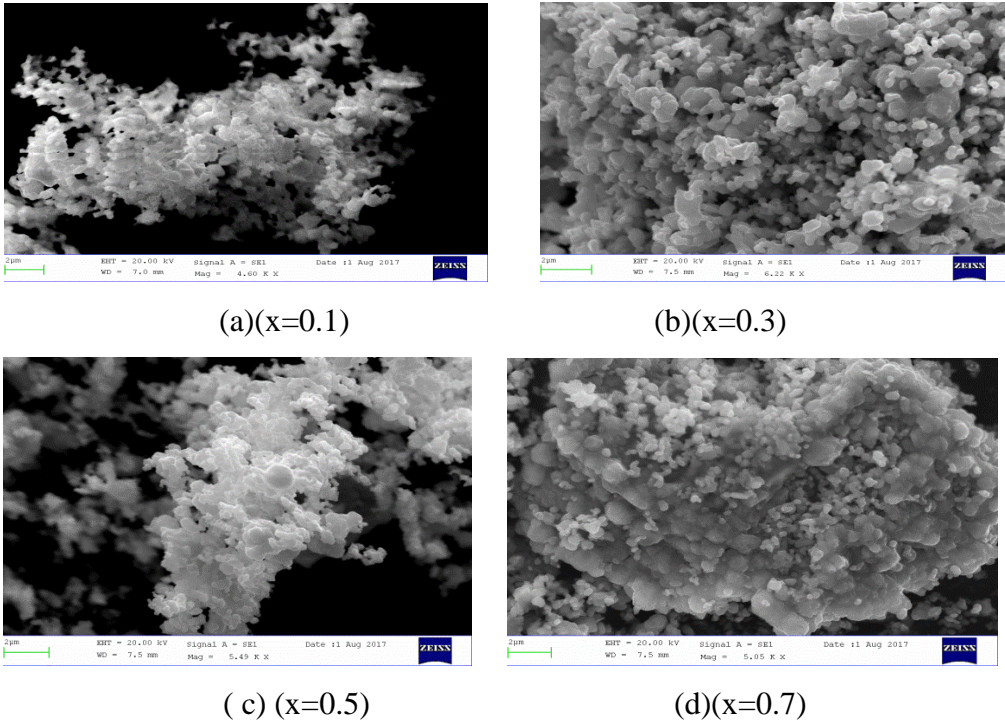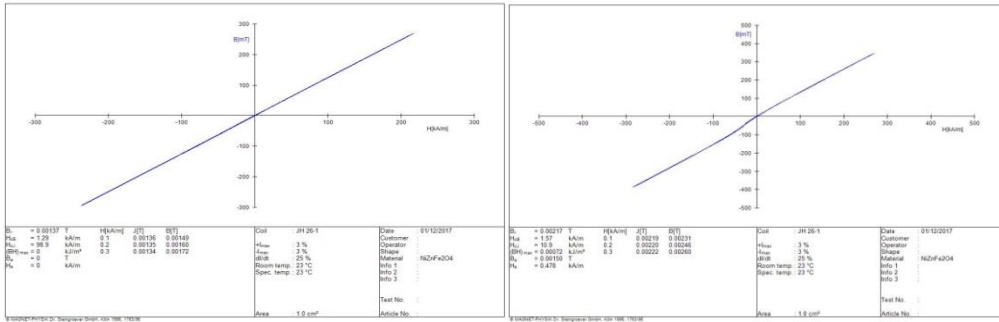(c) (x=0.5)   (d)(x=0.7)

**Figure 4:** SEM images of Ni-Zn ferrite (x=0.1,0.3,0.5 and 0.7)

Magnetic Properties

Figure 5 shows the magnetic hysteresis loop of four NZF samples in applied magnetic field in the room temperatureby using Magnet-Physik (EP-3). The corresponding magnetic parameters such as the remanence (Br), intrinsic coercivity (H_{CJ}), normal coercivity (H_{CB}) and maximum energy product $(BH)_{max}$ of ferrites were described in table 3. The Hc is the physical quantity used to measure the magnetic field intensity. In general, the Hc is influenced by many factors, including the anisotropic constant, the saturation magnetization, the comprehensive factors such as grain size, lattice stress and the defects of crystal surface and internal.
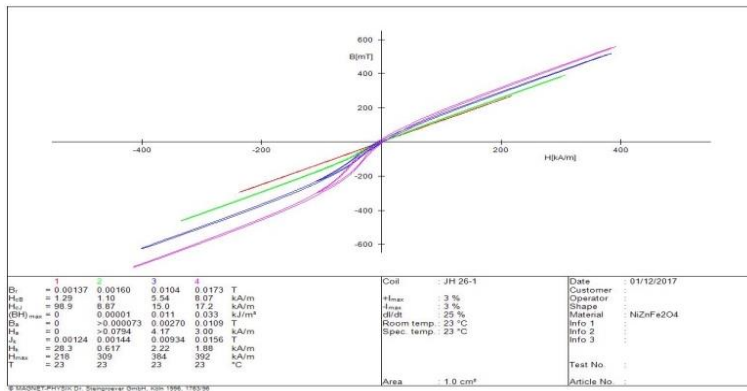
(a) (x=0.1)   (b)(x=0.3)
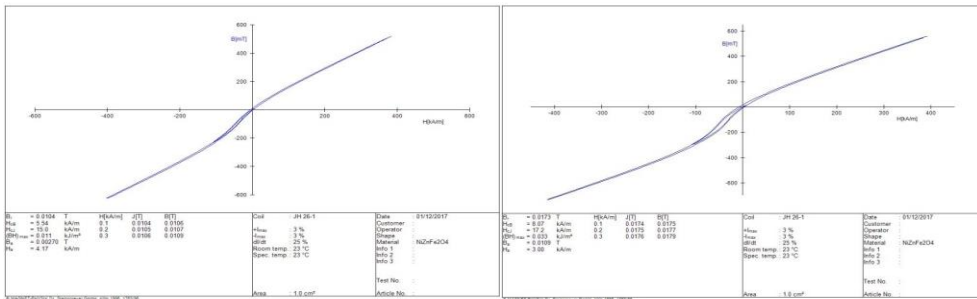
(c) (x=0.5)   (d) (x=0.7)

(e)

Figure 5: B-H graph of $NiZnFe_2O_4$ (a) x = 0.1, (b) 0.3, (c) 0.5 (d) 0.7 mol (e) comparison of all compositions

Table 3: The characteristic quantities magnetic field in ferrite samples.

Samples	Remanence (B_r) (T)	Normal coericivity (H_{CB}) (kA/m)	Relative permeability(μ_r)
$Ni_{0.1} Zn_{0.9} Fe_2O_4$	0.00137	1.29	8.45E-01
$Ni_{0.3} Zn_{0.7} Fe_2O_4$	0.00160	1.10	1.16E+00
$Ni_{0.5} Zn_{0.5} Fe_2O_4$	0.0104	5.54	1.49E+00
$Ni_{0.7} Zn_{0.3} Fe_2O_4$	0.0173	8.07	1.71E+00

The magnetic properties of ferrite materials showed that the values of the relative permeablility greater than one as a paramagnet type. Only one ratio showed that less than one as a diamagnetic type in $Ni_{0.1} Zn_{0.9} Fe_2O_4$. It will be focusing on optimizing the material properties and implementing the material in various electronics and magnetic application.

Conclusion

The mixed Ni-Zn ferrite samples are prepared and observed the following conclusions. The fact that the $NiFe_2O_4$ nanoparticles belonged to the cubic spinal structure was established by XRD. That the nanoparticles agglomerated to form spherical-shaped particles was also confirmed and made clear by the SEM analysis. The lattice parameter decrease with increase of nickel content, the bulk density increases linearly with nickle content, SEM pictures shows that the morphology of the particles is very similar. The values of bulk density, x-ray density increase gradually reaches the maximum value as Ni^{2+}composition is increased.

Acknowledgements

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