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ORIGINAL ARTICLE

MAGNETIC PROPERTIES OF NANOCRYSTALLINE NICOZN FERRITE SYNTHESIZED BY CITRATE-NITRATE COMBUSTION METHOD

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Abstract:

Mixed Nano particles of $Ni_{0.57}Co_{0.03}Zn_{0.35}Fe_{2.05}O_4$, ferrite material were prepared by a chemical combustion route and powder was sintered at temperature 9500C. This method involves the addition of aqueous nickel nitrate, cobalt nitrate, zinc nitrate, ferric nitrate as oxidizing agent and fuel citric acid as a reducing agent to form a homogenous redox mixture. The structure and morphology of samples were studied using X-ray diffraction (XRD) and scanning electron microscopy (SEM). The crystallite size has been estimated by the Debye – Scherrer formula using the full width at half maximum of the line broadening of the (311) reflection and it is found to be around 66nm. The SEM was used to analyze the grain size distribution in ferrite. The SEM picture shows agglomerated grains with different shapes and sizes. The ferrite powder was pressed in the form of torroids and permeability spectra were analyzed. The initial permeability (μi) was found to increases and loss factor decreases as frequency increases. From thermal variation of loss factor, it is observed that because of low loss factor, the ferrite must be operated below Curie temperature.

KEYWORDS:

Ni(Co)Zn ferrite, Permeability, Combustion.

INTRODUCTION:

Ferrimagnetic cubic spinels ferrites are simultaneously good magnetic and dielectric materials. Spinel ferrites have been intensively studied for their fundamental understanding and applicability in a variety of areas such as, high density information storage system, ferrofluid technology, medical diagnostics, gas sensors etc. [1]. The interest in ferrite nanoparticles is due to their important physical and chemical properties and potential for various technological applications such as high density magnetic storage, electronic and microwave devices, sensors, magnetically guided drug delivery etc. [2]. Nanosized ferrites may have extraordinary electric and magnetic properties that are different from microstructured materials [3, 4].

A spinel ferrite nanopaticle belongs to the class of nanomaterials generating a lot of interest due to their technological applications. Their typical properties of having a high surface-to-volume ratio make them useful for potential applications in sensors, solar cells, photonics, and multiferroic materials [5], catalysis, biomedical separation, and microwave absorbers [6,7], amongst host of others. The advantage of nanosized ferrites are that it is possible to sinter them at relatively low temperatures for a short duration, which save time, cost, and other factors such as volatility of Zn occurring at higher temperatures. Nanosized ferrites are expected to give sintered density at relatively lower sintering temperatures, without

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considerable grain-growth [8].

Ni-Zn ferrite is one of the best soft magnetic materials suitable for high frequency applications, owing to its low magnetic coercivity, high electrical resistivity, low loss, fairly high mechanical hardness and chemical stability. Ni-Zn mixed ferrite nanoparticles are widely investigated and it is well known that their properties can be tuned by varying nickel or zinc content[2]. Different spinel ferrite compounds can be prepared by co-precipitation [3], sol-gel [9], microemulsion method [10], hydrothermal [11], spray pyrolysis [12], reverse micelle [13], precursor [14] etc. Above methods have various typical drawbacks such as wide particle size distribution, low crystallinity and high inhomogeneity. Also frequently there is a need to apply expensive reagents and complicated steps with high time consumption [15].

The aim of present work is to get nanoparticles of Ni0.57Co0.03Zn0.35Fe2.05O4 ferrite prepared using citrate-nitrate combustion route. The main advantages of these methods are that process is carried out in short time, don't require special equipment and huge costs, for process performing there is no need high energy consumption, and as reaction products is obtained as homogenous powders with high purity levels. Thus citrate-nitrate combustion route is chosen because it is a powerful tool for producing nanosized powders and nanocomposite materials. X-ray diffraction analysis (XRD), Scanning Electron Microscopy (SEM) for Ni0.57Co0.03 Zn0.35Fe2.05O4 was examined in this study. The frequency and thermal variation of initial permeability and loss factor were also studied by copper winding on the toroidal shape of sample sintered at 950oC.

2. EXPERIMENTAL

2.1. Synthesis

Metal nitrates such as nickel nitrate [$Ni(NO_3)_2 \cdot 6H_2O$], Zinc nitrate [$Zn(NO_3)_2 \cdot 6H_2O$], Cobalt nitrate [$Co(NO_3)_2 \cdot 6H_2O$], iron nitrate [$Fe(NO_3)_2 \cdot 6H_2O$] and citric acid [$C_6H_8O_7 \cdot 6H_2O$] were used as raw materials. All metal nitrates used in fabrication of $Ni_{0.57}Co_{0.03}\ Zn_{0.35}Fe_{2.05}O_4$ ferrite were synthesized by chemical autocombustion route, in which the stoichiometric amount of corresponding metal nitrates acts as an oxidizing agent and fuel citric acid as a reducing agent for combustion reaction. Stoichiometry of the redox mixture for combustion is calculated based on the total oxidizing and reducing valencies of oxidizer (O) and fuel (F) [16]. The stoichiometric molar proportions of the reactants were mixed in a 300cm^3 pyrex dish and heated up to around 80^{0}C on magnetic stirrer, after evaporation of water content and other byproducts the stoichiometric amount of citric acid was added in to the melt slurry of nitrates and heated up to 500^{0}C for combustion. The as-burnt powder was mixed with small amount of polyvinyl alcohol and uni-axially pressed at 6 tones/inch to form pellet shaped sample with an internal and outer diameters 1cm and 2cm respectively and thickness 15mm. The samples were sintered at 950^{o}C for 1 hour.

2.2. Characterization

Powder acquired after combustion and sintering were characterized by X-ray powder diffraction (XRD) recorded for 2θ from 200 to 800 using an x-ray diffractometer with CuKα radiation. The microstructural aspects were studied with a scanning electron microscope (SEM: model JEOL-JSM 6360). The crystallite size was calculated from the X-ray diffraction (XRD) most intense peak (311) using the Debye Scherrer formula. The initial permeability, loss factor were calculated by using Ls and Q-factor values from Hioki (3532-50) LCR-Q meter. Also the thermal variation of initial permeability and loss factor were studied by using Ls and Q-factor values.

3. RESULTS AND DISCUSSION

3.1 Structural properties

Figure 1 shows a X-ray diffraction pattern of the $Ni_{0.57}Co_{0.03}\ Zn_{0.35}Fe_{2.05}O_4$ at 950^{o}C prepared by autocombustion route. All the peaks of the sintered samples can be clearly indexed to the eight major peaks of the spinel ferrite which are (220), (311), (222), (400), (422), (511), (440), (533) planes of cubic unit cell which correspond to cubic spinel structure. The value of lattice parameter (a) was 8.115A^o. The crystallite size of the material can be determined from X-ray diffraction using the well-known Debye Scherrer equation

$$D = \frac{0.9\lambda}{\beta Cos\theta} \qquad (1)$$

where, λ is the X-ray wavelength, θ the Bragg's angle and β is the full width of the diffraction line (311) at half the maximum intensity (FWHM) and was found to be 66.07 nm. In table (1) data on measured density (ρm) and X-ray density (ρ_x) are given. The measured density (ρ_m) and X-ray density (ρx) was evaluated using the relation

$$\rho_m = \frac{m}{\pi r^2 h} \qquad (2)$$

where, m is the mass, r is the radius and h is the height of the sample and

$$\rho_x = \frac{8M}{NV} \qquad (3)$$

where, M is the molecular weight of the samples, N is Avogadro's number, V is the volume of the unit cell and 8 stands for the number of formula units in cell.

The microstructural aspects were studied with a scanning electron microscope. Figure 2 display SEM micrograph of the fracture surfaces of the samples sintered at 950°C. The average measured grain size was calculated by using the line intercept method [17]. The micrograph shows agglomerated grains with different shapes and sizes, having an average grain size of 0.59μm.

3.2 Magnetic properties

3.2.1 Frequency dependence of initial permeability (μi)

To study the initial permeability (μi), the powder was compacted at 1.07×10^9 dyne cm^{-2} for 1 min in to toroid form. The toroid were sintered at 9500C for 1 hour. The rate of heating was 1000C h- 1 and the rate of cooling was 60^0C h^{-1}. The initial permeability measurements of toroid samples were carried out using HIOKI (3532-50) LCR meter, from low field inductance measurements of coils with toroidal cores using the formula

$$\mu_i = \frac{L}{\left(0.0046N^2 h \log\frac{d_2}{d_1}\right)} \qquad (4)$$

where L is the inductance in μH; N is the number of turns; d_2 is the outer diameter; d1 is the inner diameter; h is the height of the core in cm; μi is the initial permeability.

The real part (μ') and imaginary part (μ") of initial permeability was calculated using the formula

$$\mu' = \frac{\mu_i}{\sqrt{1+\tan^2\delta}}, \qquad \mu''=\mu'\tan\delta, \qquad \tan\delta=\frac{1}{Q} \qquad (5)$$

Figures 3 and 4 show the variation of real (μ') and imaginary (μ") part of initial permeability with frequency in the range 40Hz–2MHz for the composition $Ni_{0.57}Co_{0.03}$ $Zn_{0.35}Fe_{2.05}O_4$. The value of initial permeability is greatly influenced by the method of preparation, firing temperature and porosity. The real part of permeability of the $Ni_{0.57}Co_{0.03}Fe_{2.05}$ $Zn_{0.35}O_4$ ferrite were slightly increases with frequency from 10 kHz to 1MHz. Samaila etal [18] observed that μ' characteristics depend not only on chemical composition

but also on the microstructure of the sintered body. However, for a given material, the permeability is determined by in the domain wall motion and spin rotation magnetizing mechanism.

Rado and others [19] observed high frequency dispersion and absorption in i and attributed it to rotational resonance in the combined anisotropy and demagnetizing fields while the low frequency dispersion was attributed to domain wall displacement. Figure 3 and 4 clearly indicate the low frequency dispersion which may be attributed to the domain wall movement.

3.2.2 Loss factor (L. F)

The ratio of the imaginary part of the permeability representing the losses in the material to the real part of the permeability is a measure of the inefficiency of the magnetic system. It is called the loss tangent.

$$\text{Loss tangent} = \tan\delta = \frac{\mu''}{\mu'}, \qquad \text{Loss factor} = \frac{\tan\delta}{\mu_i} \tag{6}$$

This loss factor (L. F.) Parameter should be as low as possible. The loss factor of the composition $Ni_{0.57}Co_{0.03}$ $Zn_{0.35}Fe_{2.05}O_4$ decreases with frequency increases are shown in figure 5. It is observed that with increase of frequency from 10kHz to 300kHz, the value of loss factor decreases and it is almost constant in the frequency range 300KHz to 2MHz. P. K. Roy etal [20] observed that all ferrites have higher loss factor at lower frequencies (kHz) range, which may be due to higher hysteresis loss arising from their porous structures.

3.2.3 Thermal variation of initial permeability (I)

Figure 6 shows how the initial permeability (i), its real part ('), and imaginary part (") vary with temperature for the compositions $Ni_{0.57}Co_{0.03}Zn_{0.35}Fe_{2.05}O_4$ in the range from room temperature to the Curie temperature (Tc). Near Tc, both i and ' drops to zero sharply. A sharp decrease in i and ' suggests single-phase formation of the ferrite material. This observation supports the conclusion drawn from the XRD analysis that the compositions are single phase. In most of the magnetic materials, i increase with temperature up to the Curie temperature Tc. This is because of the anisotropy field usually decreases faster with temperature than saturation magnetization[18]. From the thermal variation of ", it is seen that with the increase of temperature " increases, reaches a maximum near Tc and then falls sharply near to Tc. The loss becomes large near Tc, which may be due to damping effect to the domain walls which may be very small. From thermal variation of the initial permeability for the compositions $Ni_{0.57}Co_{0.03}$ $Zn_{0.35}Fe_{2.05}O_4$, peaking behavior is observed at Tc.

3.2.4 Thermal variation of loss factor (L.F)

Figure 7 shows thermal variation of L. F. for the compositions $Ni_{0.57}Co_{0.03}$ $Zn_{0.35}Fe_{2.05}$ O_4 sintered at temperatures 950^0C are shown. In the temperature range 25^0C to near Tc, it is found that L.F. is almost constant. While above Tc, the loss factor increases exponentially. The thermal variation of tan seems to be responsible for increase in loss factor. In order to have low loss factor the ferrite must be operated below Curie temperature.

4. CONCLUSION

Mixed Nano particles of $Ni_{0.57}Co_{0.03}$ $Zn_{0.35}Fe_{2.05}O_4$, ferrite material were successfully synthesized by a chemical combustion route. Structural and magnetic properties of the prepared samples were studied. The X-ray diffraction confirmed formation of single phase ferrite in nano size. The average crystalline size around 66nm which was evaluated by X-ray diffraction technique using the Debye – Scherrer formula. The scanning electron micrograph was used to analyze the grain size distribution of ferrite. Nanocrystalline $Ni_{0.57}Co_{0.03}Zn_{0.35}Fe_{2.05}O_4$ ferrite material were achieved with lower temperature by a chemical combustion method as compared to convential ceramic method. The $Ni_{0.57}Co_{0.03}$ $Zn_{0.35}Fe_{2.05}O_4$ powder prepared at temperature 9500C exhibit good crystal structure, fine grain size and good magnetic properties. The initial permeability was found to increase and loss factor decreased at high frequency. From thermal variation of

loss factor, it is observed that for low loss factor the ferrite must be operated below Curie temperature.

REFERENCES

[1] H. Sozeri, Z. Durmus, A.Baykal, Materials Research Bulletin, 47 (2012) 2442-2448.

[2] Binu P. Jacob, Smitha Thankachan, Sheena Xavier, E.M. Mohammed, J. Of Alloys and Compounds, 541, (2012), 29-35.

[3] Gul I H, Ahmed W and Maqsood A, J. Magn. Magn. Mater. 320 (2008) 270

[4] Sutka A,Gross K A, Mezinkis G, Bebris G, and Knite M Phys. Scr. 83 (2011) 025601 (6pp)

[5] Xie, S H, Li J Y etal, Applied Physics Letters 92(6) (2008) 062901-3

[6] Hwang, Y, Materials Letters, 60(27) (2006) 3277-3280

[7] Nam, J H, Joo Y H, J. Magnetism Magnetic Materials 321(10) (2009) 1389-1392

[8] Kambale R C, Adhate N R, Chougule B K, Kolekar Y D, Journal of Alloys and Compounds 491(2010) 372-377

[9] Zahi S, Hashim M, and Daud A R, J. Magnetism Magnetic Materials 308 (2007) 177

[10] Pillai V, Kumar P, Multani M S Shah D O, Colloid. Surface A80 (1993) 69

[11] Liao X, Chen D, and Hu Y , Mater. Res. Bull. 37 (2002) 1583

[12] Takayama A, Okuya M and Koneko S, Solid State Ionics 172 (2004) 257

[13] Thakur S, Katyal S C and Singh M, J. Magnetism Magnetic Materials 321 (2009) 1

[14] Sarangi P P , Vadera S R, Patra M K and Ghosh N N, Powder Tech. 203 (2010) 348

[15] Sutka A, Lagzdina S, Mezinskis G, Pludons A, Vitina A, Timma L, 5th Baltic Conference on Silicate Materials, IOP Conf. Series: Materials Science and Engineering 25 (2011) 012019

[16] Jain S R, Adiga K C, Pai V R, Vernecker Combust. Flame 40 (1981) 71

[17] Pjostupolski, Prace Inst. Telei Radio. Zesz 107/87 (1988) 1-50

[18] Samalia Bawa Waje and Mansor Hashim, W. Daud W. Yusoff and Zulkifly Abbas, Australian Journal of Basic and Applied Sciences, 3(3):2716-2723,2009

[19] Ghodake J S, Kambale R C, Kulkarni S D, Sawant S R, and Suryavanshi S S Smart Mater. Struct. 18 (2009)

[20] K. Roy, and J. Bera, J. Mat. Proc. Tech., vol. 197, pp. 279-283, Feb. 2008.

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