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# Low Temperature Hydrothermal Synthesis and Characterisation of Nano Size Ni-Zn Ferrites

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Abstract: Ni-Zn ferrites of composition NixZn1-xFe2O4 [x = 0.0, 0.5, 1.0] are synthesized at 140oC by hydrothermal method using EDTA as template . FTIR studies of NF1 before HT show the presence of moisture and after HT only two absorption bands are observed for all the ferrites, one around 420 cm-1 for the vibrations of octahedral metal – oxygen bond and another around 610 cm-1 for tetrahedral metal – oxygen bond vibrations. TGA-DSCanalysis indicate formation of spinel phase at 250oC. HoweverTGA-DSC analysis of NF1,NF2 and ZF1 after HT at 1400C show no significant loss of mass which confirms presence of stable single spinel phase. XRDstudiesshowtheformationof pure and

single spinel phase. The average crystalline size was determined from X-

ray diffraction line broadening using Scherrer equation. FESEMS tudies revealed that all the synthesized ferrites are having nearly octable dron crystals with an average particle size of 20 to 40 nanometer

Keywords: hydrothermal synthesis, Ni-Zn ferrites, TGA-DSC analysis, spinel ferrites

I.

## INTRODUCTION

Magnetic nano particles have received special attention over the last years due to their special propertiessuch as high surface area, easy separation from reaction media. Nano material ferrites have applications in making cores of audio frequency and high frequency transformers, coils(inductors), chokes, permanent magnets, magnetooptical displays, microwave absorbers, wave guides in the GHZ region, chlorine gas sensors [1], high density information storage[2], color imaging[3], bioprocess[4], medical diagnosis[5], electromagnetic wave absorption,[6] etc. Multi layer chip indicator(MLCI) has recently beendeveloped as one of the key surface mountingdevices[7,8]. 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 [9,10] because of their high resistivity. relatively high permeability and low eddy current loss [11,12]. These soft magnetic materials, crystallizes in the spinel structure of the type  $(Zn_{1-x}Fe_x)$  (Ni<sub>x</sub>Fe<sub>2-x</sub>)O4, where the metallic cations Fe<sup>3+/</sup>Zn<sup>2+</sup> occupy the tetrahedral A sites, and the metallic cations Fe3<sup>+/</sup>Ni<sup>2+</sup> occupy the octahedral B sites[13,14] In biomedical application, one can use nano magnetic materials as drug carriers inside body where the conventional drug may not work. For this purpose, the nanosize particles should be in the super paramagnetic form with a low blocking temperature. Ferrite nanomaterials are object of intense research because of their proper magnetic properties. It has been reported that when the size of particles reduced to small size or in range of nanomaterials, some of their fundamental properties are affected. It is known that magnetic properties of ferrites are sensitive to preparation technique and their microstructures [15]. 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 [16-18]. It is well known that zinc ions can be used to alter the saturation 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 [19]. The substitution of divalent ions in pure ferrites leads to the modification of the structural, electrical and magnetic properties [20]. Several researchers have reported the synthesis of Ni-Zn ferrites using different techniques like, refluxing process [21], ceramic [22], hydrothermal [23], combustion [24], co-precipitation [25], reverse micelle process [26], spark plasma sintering [27], micro emulsion [28] and ball milling, etc. In this work, we present the results of substitution of non-magnetic Zn on the magnetic Nickel ferrite synthesized by low temperature hydrothermal method.



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## II. EXPERIMENTAL

For the synthesis of  $Ni_xZn_{1-x}Fe_2O_4$  [x = 0.0, 0.5,1.0] appropriate mole ratios of Analargrade Cobalt chloride, Nickel chloride, ZincchlorideandFerricchloridewereweighedandeachofwhichwas dissolved in 50mlofasolution containing 0.5 gramofdisodiumsalt ofEthyleneDeammineTetraaceticAcid(EDTA)withconstantstirringbymagneticstirrer.4.4molarNaOHsolutionwasaddeddropbydropinto eachmixturetillthepHisincreasedto12.Theprecipitate producedwasstirredforanother3hoursatroomtemperature.Thentheprecipitate was transferred into an autoclave(23 ml capacity) and kept at a temperature of 140°C for 12 hours in an air oven. The precipitate obtained was centrifuged and washed withdistilledwaterandethanol. The synthesized materials were designated as follows:

 $\label{eq:NF1-NiZnFe_2O_4} \begin{array}{l} [ \ X=0 ] \\ NF2-Ni_{0.5}Zn_{0.5}Fe_2O_4 [ \ X=0.5 \ ] \\ ZF1 \ - ZnFe_2O_4 [ \ X=1.0 \ ] \end{array}$ 

## III. CHARACTERISATION

PowderX-raydiffraction(XRD)patternsweremeasuredbyusingRICH-SIEFERT3000-TTdiffractometeremployingCuK\_radiation. The Fourier transform infrared (FTIR) was performed by the KBr tablet method in the range 4000–250 cm<sup>-1</sup>, in a BRUKER model IFS 66 V FTIR spectrometer. Themorphologiesofthesampleswereexamined with field emissions canning electron microscope (FESEM, FEI Nova-Nano SEM-600, The Netherlands). TGA- DSC were carried out using Mettler Toledo TGA/DSC1 in the temperature range from  $25^{\circ}$ C to  $1100^{\circ}$ C at heating rate of  $10^{\circ}$ C per minute to investigate thermal properties of prepared samples.

#### IV. RESULTS AND DISCUSSION

The FTIR spectra of the spinel ferrites after hydrothermal treatment are given in Fig.1. The vibrational spectra of absorption bands were observed only in the range between 420 to 610 cm<sup>-1</sup> which are characteristic to spinel structure of the synthesized ferrites. The absorption bands observed around 420 cm<sup>-1</sup> are due to the vibrations of octahedral metal – oxygen bond and around 610 cm<sup>-1</sup> are attributed to tetrahedral metal – oxygen bond vibrations. The difference in the frequency of the two vibrations might have resulted due to the longer bond length of oxygen – metal ions in the octahedral sites and shorter bond length of oxygen – metal ions in the tetrahedral sites .The absence of peaks at 1300–1650cm<sup>-1</sup> and 2000–3500 cm<sup>-1</sup> shows nonexistence of the O–H mode, C–O mode, and C=H stretching-mode of vibrations. However the FTIR spectra of NF1 before hydrothermaltreatment (Fig.2) shows additional peak around 3305 cm<sup>-1</sup> and 1583 cm<sup>-1</sup> which are attributed to O–H mode of vibrations. Very weak band at 591 cm<sup>-1</sup> and 404 cm<sup>-1</sup> are observed. These observations indicate the presence of moisture and no significant formation spinel phase before hydrothermal treatment.







Fig 2: FTIR Spectra of NF1 before HT

Fig 1: FTIR Spectra of (a)NF1, (b)NF2 and (c)ZF1 after HT

In order to findtemperature range for the growth of system, TGA/DSC in temperature range 25°C to 1100°C at a heating rate 10°C perminute were performed for NF1 before and after



hydrothermal treatment (HT). TGA curveanalysis of NF1 before HT(Fig.3) shows that there is mass loss with temperature. Around temperature  $120^{\circ}$ C , loss of mass is due tovaporization of surface water molecules and more loss ofmass up to  $160^{\circ}$ C , due to vaporization of trapped watermolecules. For temperature range  $200^{\circ}$ C to  $260^{\circ}$ C , mass loss of samplemay be due to loss of organic material. Ranges oftemperature corresponding to no significant loss suggestrecrystallization process. DSC Endothermic peak (Fig.3) at  $250^{\circ}$ Cshows that at this temperature , ferrite formation getscompleted and Enthalpy change is found to be  $325 \text{ Jg}^{-1}$  of synthesized samples as-prepared. Hence addition of EDTA as template significantly reduces the spinel phase formation compared to conventional co-precipitation methods without addition of any template. However in the present study zinc substituted ferrites are synthesized by hydrothermal methodwith EDTA as template at a temperature of  $140^{\circ}$ C. The TGA-DSC analysis of NF1 (Fig.4) after Hydrothermal treatment shows no significant weight loss indicating the presence of stable single phase spinel structured ferrites. The role of EDTA in controlling size and shape of ferrites with rapid the spinel phase formation is under investigation.





5. Which exhibit typical reflections of (220), (311), (222), (400), (422), (511)

and (440) planes that are indications of the presence of the cubic spinel structure. All of the diffraction peaks match well with the reported values (JCPDS file No: 10-0325 for Nickel Ferrite, and File No. 22-1012 for Zinc Ferrite).

The average crystal size is determined by the Scherrer equation using the peak broadening (FWHM) of the most intense peak (311): t=0.9 $\lambda/\beta$ cos $\theta$  where  $\lambda$  is the wavelength of CuK $\alpha$ (1.54059°A),  $\theta$  is the angle of Bragg diffraction at full width half maximum (FWHM). The average crystallites ize obtained for all the spinel ferrites are given in table 1.



Fig 5: XRD Spectra of (a)NF1, (b)NF2 and (c) ZF1







Fig 6: FESEM images of (a)NF1, (b)NF2 and (c) ZF1



The morphology obtained by FESEM analysis(Fig6) shows incomplete octahedron structures for all the synthesized ferrites. The addition of EDTA, the complexing agent, controls the size as well as the shape of nanoparticles and prevents their agglomeration. The average crystal size observed from FESEM images are in good agreement with the calculated values by XRD. Lattice constant(a) and Volume of unit cell (a3) are also calculated for NF1, NF2 and ZF1 which are given in table 1.

## V. CONCLUSIONS

Hydrothermal treatment carried out at a lower temperature of  $140^{\circ}$ C using EDTA as template found to yield Ni –Zn ferrites of nano size and incomplete octahedron shape with spinel structure. FTIR investigation confirmed the presence of metal ions in the tetrahedral and octahedral holes. TGA – DSC analysis indicated spinel phase formation at  $250^{\circ}$ C. However hydrothermal treatment facilitated the spinel phase formation at much lower temperature of  $140^{\circ}$ C.XRD study showed presence of pure and single spinel phase in all HT treated samples. FESEM analysis clearly indicated the existence of isolated nanoparticle with nanosize and incomplete octahedran shape of ferrites.TheaveragecrystallitesizeobtainedbyXRDandFESEManalysisindicate significant size reduction inferriteparticles.

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#### REFERENCES

- [1] Gopalreddy C V, Manorama S V, and Rao V J., semiconducting gas sensor for chlorine based oninverse spinel Nickel ferrite., Sensors and Actuators B, 1999,55(1), 90-95.
- [2] R.F. Zolio, US patent 4, 474 (1984) 866
- [3] Nixon L., Koval C. A., Noble R.D., SlaffG.S., Preparation and characterization of novel magnetic coated ion exchange particles, Chem. Mater., 1992, 4, 117.
- [4] MacMichael R.D., Shull R.D., SwartzendruberL.J, Watson R.E, Magneto caloric effect in superparamagnets., J.Magn. Magn. Mater, 1992,111, 29-33.
- [5] Bulte J.W.M., Magneto ferritin :Charcterization of novel super paramagnetic M R contrast agent J. Magn.Reson. Imaging., 1994, 4, 497-505.
- [6] Wei F., Baoshum L.,,Jizhong Y., Xi L., Muyu Z., J. Magn. Soc. Jpn. ,1998,22, 366.
- [7] R J. Charales, A.RAchuta, U.S Patent No.4966625, 1990.
- [8] H. Watannabe, Y. Kanagawa, T. suzuki, T. Nomura, U.S Patent No. 4956114,1990.
- [9] N. Matsushita, K. Kondo, S. Yoshida, M. Tada, M. Yo-shimura and M. Abe, "Ni-Zn Ferrite Films Synthesized from Aqueous Solution Usable for Sheet-Type Conducted Noise Suppressors in GHz Range," Journal of Electroce- ramics, Vol. 16, No. 4, 2006, pp. 557-560.
- [10] K. Kondo, T. Chiba, H. Ono, S. Yoshida, Y. Shimada, N. Matsushita and M. Abe, "Conducted Noise Suppression Effect up to 3 GHz by Ni-Zn Ferrite Film Plated at 90°C Directly onto Printed Circuit Board," Journal of Applied Physics, Vol. 93, No. 10, 2003, pp. 7130-7132.
- [11] V. L. O. de Brito, A. C. da C. Migliano, L. V. Lemos and F. C. L. Melo, "Ceramic Processing Route and Characteri-zation of a Ni-Zn Ferrite in Application in a Pulsed-Current Monitor," Progress in Electromagnetics Research, Vol. 91, 2009, pp. 303-318
- [12] T. Jahanbin and M. Hashim, "Magnetic and Morphologi-cal Characterization of Ni-Zn Ferrite Prepared from Nano Size Starting Powder via Coprecipitation Technique," Solid State Science and Technology, Vol. 17, No. 2, 2009, pp. 243-249.
- [13] S. Albuquerque, J. D. Ardisson, W. A. A. Macedo and M. C. M. Alves, "Nanosized Powders of Ni-Zn Ferrite: Syn-thesis, Structure, and Magnetism," Journal of Applied Physics, Vol. 87, No. 9, 2000, pp. 4352-4358.
- [14] D. Cullity, "Elements of X-Ray Diffraction," Addi-son-Wesley Publishing Company, Inc., Boston, 1956.
- [15] H. Su, H. W. Zhang, X. L. Tang, Y. L. Jing and Y. L. Liu, "Effects of Composition and Sintering Temperature on Properties of NiZn and NiCuZn Ferrites," Journal of Magnetism and Magnetic Materials, Vol. 310, No. 1, 2007, pp. 17-21
- [16] . L. Dormann and M. Nogues, "Magnetic Structures in Substituted Ferrites," Journal of Physics: Condensed Mat- ter, Vol. 2, No. 5, 1990, pp. 1223-1237.
- [17] N. Rezlescu, E. Rezlescu, C. Pasnicu and M. L. Craus, "Effects of the Rare-Earth Ions on Some Properties of a Nickel-Zinc Ferrite," Journal of Physics: Condensed Mat- ter, Vol. 6, No. 29, 1994, pp. 5707-5716.
- [18] A. E. Virden and K. O'Grady, "Structure and Magnetic Properties of NiZn Ferrite Nanoparticles," Journal of Magnetism and Magnetic Materials, Vol. 290-291, Part 2, 2005, pp. 868-870. doi:10.1016/j.jmmm.2004.11.398
- [19] A. M. El-Sayed, "Influence of Zinc Content on Some Properties of Ni-Zn Ferrites," Ceramics International, Vol. 28, No. 4, 2002, pp. 363-367. \
- [20] . Zhong, Q. Li, Y. L. Zhang, H. S. Zhong, M. Cheng and Y. Zhang, "Synthesis of Nanocrystalline Ni-Zn Ferrite Powders by Refluxing Method," Powder Technology, Vol. 155, No. 3, 2005, pp. 193-195.
- [21] A. Dias and R. L. Moreira, "Chemical, Mechanical and Dielectric Properties after Sintering of Hydrothermal Ni- ckel-Zinc Ferrites," Materials Letters, Vol. 39, No. 1, 1999, pp. 69-76.
- [22] S. E. Jacobo, S. Duhalde and H. R. Bertorello, "Rare Earth Influence on the Structural and Magnetic Properties of NiZn Ferrites," Journal of Magnetism and Magnetic Materials, Vol. 272-276, No. 3, 2004, pp. 2253-2254.
- [23] S. D. Shenoy, P. A. Joy and M. R. Anantharaman, "Effect of Mechanical Milling on the Structural, Magnetic and Dielectric Properties of Coprecipitated Ultrafine Zinc Fer- rite," Journal of Magnetism and Magnetic Materials, Vol. 269, No. 2, 2004, pp. 217-226.
- [24] S. A. Morrison, C. L. Cahill, E. E. Carpenter, S. Calvin, R. Swaminathan, M. E. McHenry and V. G. Harris, "Mag-netic and Structural Properties of Nickel Zinc Ferrite Na- noparticles Synthesized at Room Temperature," Journal of Applied Physics, Vol. 95, 2004, pp. 6392-6395.



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- [25] J. Sun, J. B. Li, G. L. Sun and W. G. Qu, "Synthesis of Dense NiZn Ferrites by Spark Plasma sintering," Ceramics International, Vol. 28, No. 8, 2002, pp. 855-858.
- [26] Verma, T. C. Goel, R. G. Mendiratta and M. I. Alam, "Dielectric Properties of NiZn Ferrites Prepared by the Citrate Precursor Method," Materials Science and Engin- eering: B, Vol. 60, No. 2, 1999, pp. 156-162.
- [27] G. P. Lópeza, S. P. Silvetti, S. E. Urreta and E. D. Cabanil-las, "Magnetic Interactions in High-Energy Ball-Milled NiZnFe2O4/SiO2 Composites," Physica B: Condensed Mat- ter, Vol. 398, No. 2, 2007, pp. 241-244.
- [28] C. Upadhyay, D. Mishra, H. C. Verma, S. Anand and R. P. Das, "Effect of Preparation Conditions on Formation of Nanophase Ni-Zn Ferrites through Hydrothermal Te- chnique," Journal of Magnetism and Magnetic Materials, Vol. 260, No. 1-2, 2003, pp. 188-194.











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