ISSN 2278-7763

Synthesis and charcterization of Nanocrystalline NiCuZn Ferrite prepared by Sol-gel auto combution method.

Sopan M. Rathod*. Ashok B. Shinde

PG Department Of Physics, Abasaheb Garware College, Karawe Road, Pune-04, India.
Corresponding Auther: Dr.S.M.Rathod, Email: smragc@rediffmail.com

ABSTRACT

Promising future applications of ferrite nanoparticles in medicine, making many devices like permanent magnets, memory storage devices etc. Ferrite nanoparticles have been the emerging focus of the recent scientific research. Therefore nanostructured powders of ferrites having chemical compositions [$Ni_{0.8-x}Cu_{0.2}Zn_xfe_2O_4$], where x=0.3, 0.5, synthesised through nitrate citrate by sol-gel autocombustion method from stoichiometric mixture of their respective metal nitrate. The prepared powders were sintered at 400 ^{0}C and 600 ^{0}C for 4 hours. The structural, morphology, ferrite formation of powder were determined by X-ray powder diffractometry (XRD), Scanning Electron Microscope (SEM) photograph of the samples and Infrared (IR) spectroscopy technique. The X-ray revealed the formation of nano-sized ferrite particles with cubic spinel structure and the cubic phase in the ferrite matrix. The IR shows the characteristic ferrite bonds were confirmed. The average crystalline particles sized were calculated by Scherrer formula. The average crystalline size obtained from XRD was found between 40 and 44nm. The lattice parameters, X-ray density and bond length are different parameters are calculated from XRD patterns. The UV-Visible Spectroscopy of prepared sample shows that the band gap energy in the range of semiconductor materials. The Coercivity was found to change in proportionally and sintering temperature with the particle sizes of the investigated ferrites.

Keywords : NiCuZn Nanocrystalline ferrite,IR,XRD,SEM,VSM, UV.

1 INTRODUCTION

THE Recent interest in the study of several spinel types ferrites is in terms of the synthesis of their nanoparticles at low temperatures by different techniques, in view of the potential applications of these nanosized magnetic materials in different technological areas, as well as to study the intriguing magnetic properties of the nano-ferrite materials has been increased. The magnetic properties of the nanosized ferrites are entirely different from those of their bulk counterparts, such as the superparamagnetic behavior and associated properties. Nanosized ferrites with uniform particle size and narrow size distribution are desirable for a variety of applications like targeted drug delivery, ferrofluids, medical imaging and other biomedical applications, magnetic data storage, etc,[1-4].

Presently, NiCuZn ferrites have been the dominant materials for MLFCI (Multi Layer Ferrite Chip Inductors) due to its low sintering temperature (<950°C) and good electromagnetic properties in radio frequency range, good chemical stability. In addition, NiCuZn ferrites have better high frequency properties compared to that of MnZn ferrite and low densification temperatures than NiZn ferrites [5, 6]. If the dielectric properties can be effectively improved on the premise of low loss of the permeability, the ferrite has the potential application in the multi-layer electromagnetic interference filter (EMIF) as the material for both inductor layer and capacitor layer. [7].

The Ni–Zn ferrites are considered as the most versatile ferrites for their high resistivity and low eddy current losses. Electrical conductivity of nickel ferrite changes with Zn content and it is found to be minimum in the case of Ni0.7Zn0.3Fe2O4. The substitution of Cu brings about a structural phase transition accompanied by the reduction in the crystal symmetry [8].

2 EXPERIMENTALMETHODOLOGY

2.1 Review Stage

Analytical grade Nickel nitrate, copper nitrate, zinc nitrate, iron nitrate and citric acid were used stoichiometrically to prepare the ferrite compositions ($Ni_{0.3}Cu_{0.2}Zn_{0.5}Fe_2O_4$) and ($Ni_{0.5}Cu_{0.2}Zn_{0.3}Fe_2O_4$.) The specified amount of metal nitrate and citric acid was first dissolved in to deionized water to form the sol. Ammonia was also slowly added to the sol to adjust the pH value at about 7. During this procedure, the sol was continuously stirred by a magnetic agitator. Then the sol was heated at 100 ^{0}C with continuous stirring till it transforms into a xerogel. At a proper temperature ignition started and the dried gel burnt in a self-propagating combustion manner until all the gel was burnt out completely to form a fluffy loose powder. The entire combustion process was done in a few minutes. Finally, the as-burnt powders were sintered in the muffle furnace at 400–600 ^{0}C for 4 hours with a heating rate of 10 ^{0}C /min to obtain the single phase ferrite.

An infrared spectrum (IR) of the as-burnt powder sintered at 600 ^{0}C was recorded on a spectrophotometer from 400 to 4000 cm^{-1} by the KBr pellet method.

The phase identification of the calcined powders was performed by X-ray diffraction (XRD) on a X-ray diffractometer using CuK_α radiation (K= 1.5405 A°). Powder X-ray diffraction studies (XRD) have been carried out on the sintered samples at 400^{0}C and 600^{0}C for $Ni_{0.3}\ Cu_{0.2}\ Zn_{0.5}\ Fe_2\ O_4$ as well as $Ni_{0.5}\ Cu_{0.2}$

ISSN 2278-7763

$Zn_{0.3}$ Fe_2 O_4 . The dried gel powder is amorphous in nature. The crystallite size of nano particles was calculated by Scherrer formula given by,

$$t = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

β=full width at half maxima, θ=bragg angle for the actual peak.
The X-ray density of prepared sample was calculated using this formula.[9].

$$D_x = \frac{ZM}{N_a a^3} \quad (2)$$

Vibrating sample magnetometer (VSM) (model 7307, Lake Shore with a maximum magnetic field of 10 kOe was carried out at room temperature to evaluate the magnetic properties like saturation magnetization (M_s), remenant magnetization (M_r) and coercivity (H_c) of the sintered powders. Scanning electron microscopy (SEM) was used to determine the microstructure of the sintered specimens.

2 Phase Analysis

The average crystallite size of the sintered samples was found to be in between 40-44nm. This shows that synthesized powder has nano-sized crystallites. All the Bragg's angles of corresponding peaks in XRD pattern matched exactly with the characteristic of reflection peaks of NiCuZn Ferrite reported in JCPDS by Barakat, m et al., J Therm. Anal 37.241(1991).

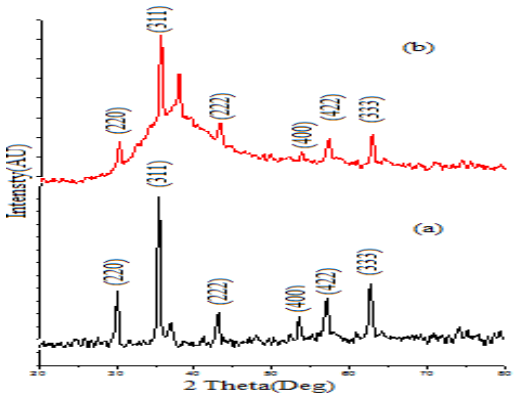


Fig1.XRD of $Ni_{0.3}Cu_{0.2}Zn_{0.5}Fe_2O_4$.at(a)400^{0}C,(b)600^{0}C

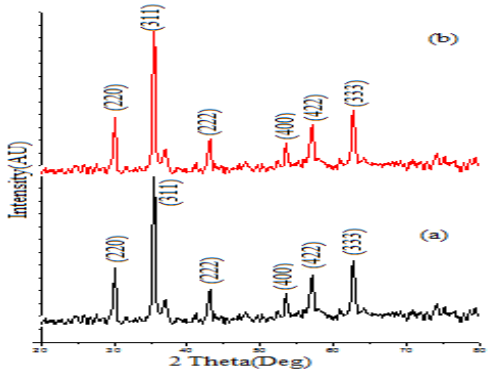


Fig2.XRD of $Ni_{0.5}Cu_{0.2}Zn_{0.3}Fe_2O_4$ (a)400^{0}C(b)600^{0}C

Table1.XRD Calculation

Sintring Temp(^{o}C)	Composition (x)	Particle size(nm)	Lattic Constant(^{o}A)	X-Ray Density(gmcm^{-3})
400	0.3	44	8.3921	5.321
	0.5	41	8.3961	5.336
600	0.3	45	8.4588	5.298
	0.5	40	8.5588	5.037

Fig. 3 shows IR spectra of calcined powder at 600 ^{0}C in the range 400– 4,000 cm^{-1}. The appearance of the characteristic bands of NO_3 indicates that the NO_3 exists as a group in the structure of citrate gel during the gelation of mixed solution formed from nitrates and citric acid. In the IR spectrum curve of the powder after combustion the absorption band which corresponds to NO_3 ions, disappears and the bands that correspond to the carboxyl group and hydroxyl group decrease significantly. On the other hand, a significant spectroscopic band at 566 cm^{-1} appears which seems to be the characteristic absorption band of ferrite. This decrease of the characteristic bands of carboxyl group and NO_3 ions suggests that carboxyl group and NO_3 ions take part in a reaction during combustion. Therefore, the combustion is an exothermic reaction in the dried gel where nitrate ions act as oxidant and carboxyl group is reductant as observed in the other systems [6].

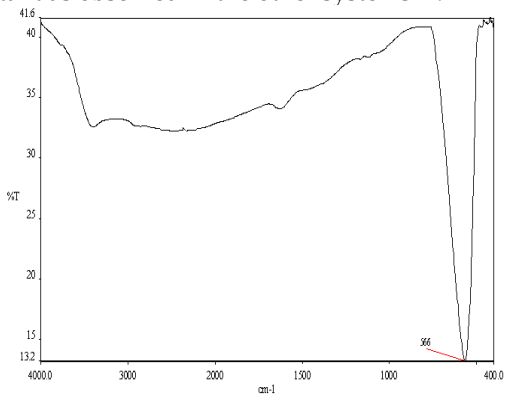


Fig3.IR Of $Ni_{0.3}Cu_{0.2}Zn_{0.5}Fe_2O_4$ at 600^{0}C

Saturation Magnetisation has been determined from the force experienced by the ferrite specimen in the field gradient. It was measured using Vibrating Sample Magnetometer (VSM) (model 7307, Lake Shore). The principle of VSM is the measurement of electromotive force induced by magnetic sample when it is vibrated at constant frequency in the presence of a static and uniform magnetic field. A small part of sintered powder (0.0125 g) was weighted and made to avoid movements inside the sample holder. The VSM was operated to 1 Tesla. Fig. 4 shows the magnetization curve of the $Ni_{0.3}$ $Cu_{0.2}$ $Zn_{0.5}$ Fe_2 O_4 sintered at 600 ^{0}C. Fig. 5 (c) indicates well densified microstructure of NiCuZn Ferrite sintered at 600 ^{0}C. The densification temperature of the as synthesized NiCuZn Ferrite is much lower than that of powders prepared by conventional method. In general, the sintering temperatures of pure NiCuZn Ferrite powders synthesized via the conventional mixed oxides method are higher than 1050 ^{0}C. As a magnetic material for MLCIs small grain size is preferable to high performance MLCIs with high electrical resistivity and high reliability [6].

ISSN 2278-7763

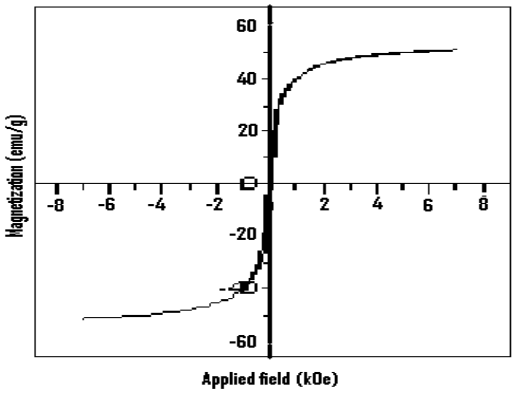
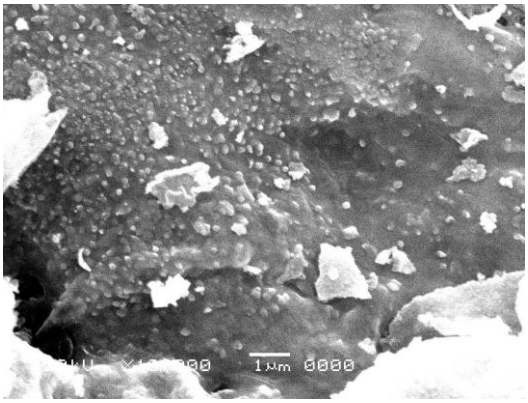


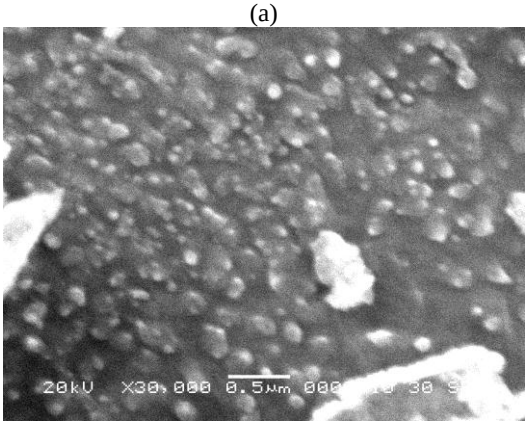
Fig4.Magnetic Hysteresisloop for the $Ni_{0.3}Cu_{0.2}Zn_{0.5}$ sintred at 600^0C

Table2.Magnetic Properties of $Ni_{0.3}Cu_{0.2}Zn_{0.5}$ sintred at 600^0C

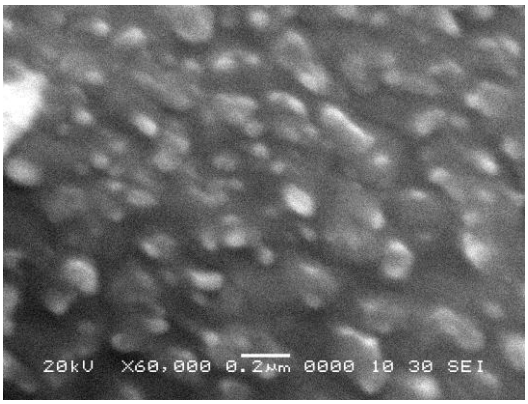
Saturation Magnetization() (emu/g)	Remenence (Mr) (emu/g)	Coercivity (Hc) (Oe)
51.366	7.5680	63.96



(a)



(b)



(c)

Fig5.Micrograph of $Ni_{0.3}Cu_{0.2}Zn_{0.5}Fe_2O_4$ sintred at 600^0C(a)1 μ m.(b)0.5 μ m (c)0.2 μ m

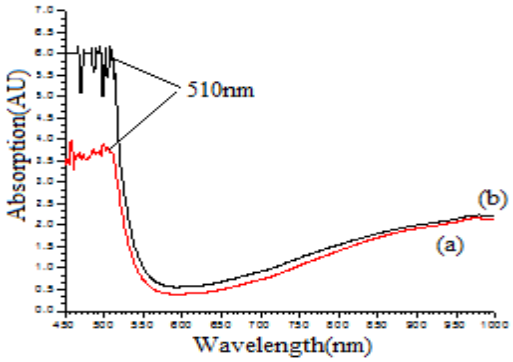


Fig6.UV spectrograph of $Ni_{0.8-x}Cu_{0.2}Zn_xFe_2O_4$ where(a) x=0.3(b)x=0.5.

Composition(x)	Band Gap Energy(eV)	Absorption (AU)
$Ni_{0.3}Cu_{0.2}Zn_{0.5}Fe_2O_4$	3.8	6.0
$Ni_{0.5}Cu_{0.2}Zn_{0.3}Fe_2O_4$	3.8	3.5

The Fig.6 shows UV-Visible spectroscopy absorption and reflection in the UV region. In the absorption molecules of Π-electron or non –bonding (n-electron) can absorb the energy in the form of ultraviolet or visible light to excite this electron to higher or anti-bonding molecular orbit [10].The band gap energy was calculated by following formula.

$$E = \frac{hc}{\lambda} \qquad (3)$$

3. Results and Discussion

In the present study $Ni_{0.3}Cu_{0.2}Zn_{0.5}Fe_2O_4$ and $Ni_{0.5}Cu_{0.2}Zn_{0.3}Fe_2O_4$ have been synthesized using low temperature Sol-Gel Auto-Combustion method. The two compositions of NiCuZn ferrite was taken by keeping composition of Copper constant. The synthesis process is carried out using analytical grade compounds of Nickel Nitrate, Copper Nitrate, Zinc Nitrate and Ferric Nitrate. Ferric Nitrate was used as a precursor and citric acid was used as reducing agent in the reaction In the IR curve of the sintered powder there is only one significant band at about 560 cm^{-1}. This is the characteristic band of $Ni_{0.3}Cu_{0.2}Zn_{0.5}Fe_2O_4$. The average particle size (Grain size) for

ISSN 2278-7763

two different compositions was found in between 40-44 nm. The average grain size remains almost same if the parcentage of composition change, sintering temperature at 400^0C and600^0C but lattice constant increases respectively. The X-Ray density increases with increasing zinc percentage at 400^0C sintering temperature but it decreases at600^0C sintering temperature. The coercivity of the material is low therefore the material formed is soft ferrite. From UV-Visible spectroscopy band gap energy was detrermined it found to be prepared material is semiconductor in nature.

4 AKNOWLEDGEMENT

We thank the Department of Physics and Chemistry, Abasaheb Garware College and Department of Physics, University of Pune for their kind cooperation and providing their infrastructure for the characterization purpose.

5. REFERENCES

[1] M. R.Barati on *J Sol-Gel Sci Technol* (2009) 52; 171-178.
[2] Dyal A, Loos K, Noto M et al (2003*) J Am Chem Soc* 125:1684.
[3] Choi EJ, Ahn Y, Kim S et al (2003) *J Magn Magn Mater*
[4] 262:L198.
[5] Sinha A, Nayar S, Murthy GVS et al (2003) *J Mater Res* 18:1309.
[6] Pradip Kumar Roy, Ph.D. Thesis Influence of Substitutions and Sintering aids on structural and Electromagnetic Properties of NiCuZn Ferrite Zhenxing Yue, Ji Zhou, Longtu Li, hongguo Zhang, Zhilun Gui *Journal of Magnetism and Magnetic Materials* 208 (2000) 55-60.
[7] Weiwei Ling , Huaiwu Zhang Ying He,Yang Wu, Kai Yang, Yuanxun Li, Sheng Li, *Journal of Magnetism and Magnetic Materials* 322 (2010) 819–823
[8] Ke Sun, Zhongwen Lan, Zhong Yu,Xiaona Jiang, Jiaomin Huang, *Journal of Magnetism and Magnetic Materials* 323 (2011) 927–932..
[9] Ferea AMM, Kumar S, Batoo KM, Yousef A, Lee CG, Alimuddin :Structure and electrical properties of $Co_{0.5}Cd_xFe_{2.5-x}O_4$ Ferrites. *J Alloy Compd 2008,464:361-369*
[10] Suryawanshi SS, Deshpand V,Sawant SR: XRD analysis and magnetic properties of Al^{3+} substituted Cu-Cd ferrites. *J Mater Chem Phy 1999,59:199-203.*

Corresponding Auther:
Dr.S.M.Rathod
PG & Reasearch Depaertment Of Physics ,
Abasaheb Garware College ,Karve Road ,
Pune-411004.
Email: smragc@rediffmail.com
Mobile No: +919850501291