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Characteristics Exploration of NIiCuZn Nano-Composite coated Permanent Magnets

T. Narasimhulu, Mallikarjuna Rao Pasumarthi

Department of Electrical Engineering, Andhra University College of Engineering (A), Andhra University, Visakhapatnam, India

Abstract*—This paper presents the synthesis of*** $Ni_{0.5}\,Cu_{0.2}Zn_{0.3}\,Fe_2\,O_4$ ***compound using Citrate Precursor Sol- Gel Method and Ball millingfor grinding the compound. X-ray diffraction measurements (XRD) confirmed the formation of single-phase cubic spinel structure. The average crystallite size was calculated using XRD pattern and confirmed by Scanning Electron Microscope (SEM). The electromagnetic properties were investigated using Vector Network Analyzer (VNA) and molar magnetic susceptibility measurements. The magnetic measurements have proved that the entire preparation method has considerable effect in enhancing the magnetic properties of the system. And an application of PMBLDC machine design with ferrite coated permanent magnets having competitive power density and efficiency. The influence of temperature variation on the magnets on the electric machine performance is also observed.***

Keywords— ferrite coat; magnetic susceptibility; Citrate Precursor Sol- Gel Method; Ball Milling.

I. INTRODUCTION

At present surface-mounting devices have been developed for electronic applications, which are produced by coating ferrite and Ag electrode layers alternately and co-firing them. Low temperature sintered NiCuZn ferrites are the most universal ferrite materials to produce MLCIs because of their relatively low sintering temperature and high resistivity with good performances in the high frequency range [1–3].Typical NiCuZn ferrites with a regular particle size have sintering temperatures above 1000^0C. The usage of fine powder decreases the sintering temperature of ferrites. Fine powders can be prepared through various wet-chemical methods like co-precipitation [4],hydrothermal synthesis [5] and sol–gel processes [6]. Although theco-precipitation and sol–gel methods are the most popular, theyhave some disadvantages as most of them are highly pH sensitiveand require special attention for complex systems whereas the sol–gel technique requires expensive alkoxide precursor material and stringent process of gel product [7].Among the established synthetic methods, it is still critical tofind simple and cost-effective routes to synthesize nano-crystallineNiCuZn ferrites by using cheap, nontoxic and environmentally benign precursors. In addition to their high nutrition quality egg-white proteins are well known for their gelling, foaming and emulsifying characteristics [8-9]. NiCuZn ferrites of composition $Ni_{0.7-x}Cu_xZn_{0.3}Fe_2O_4$ (x = 0, 0.2, 0.4, 0.6) prepared by citrate precursor method, characteristics are investigated and reported[10]. X-ray diffraction(XRD) confirmed the formation of single-phase cubic spinel structure. The grain size, estimated by SEM micrograph, was found to increase with Cu content. The hysteresis data indicated that the maximum saturation magnetization was obtained for the composition with x = 0.2.Lima [11] synthesized $Ni_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ ferrite($0.2\leq x\leq 0.4$) nano-particles using the citrate precursor method.Vibrating sample magnetometer (VSM) showed that adding copperto NiZn ferrite decreases magnetization saturation and the calcining temperature. Ferrites with compositions of $Ni_{0.27}Zn_{0.64}Cu_xFe_{1.98O4}$ (x = 0.1,0.2) were prepared by conventional ceramic methods [12]. Theintergranular pores in the prepared ferrites were found to generate large demagnetizing fields, reduce the temperature dependence ofthe effective anisotropy field and thus decrease the temperature dependence of the relative initial permeability. Fine powders of $Ni_{0.6-x}Cu_xZn_{0.4}Fe_2O_4$, where $0\leq x\leq 0.4$ were prepared by the citrate precursor method [13]. XRD confirmed theformation of single-phase cubic spinel structure. The addition of copper was found to promote the grain growth, resulting in anincrease in the grain size. Curie temperature, however, was understandably lowered with the increase in Cu content. Ferrite with Cuconcentration of x = 0.4, showed the highest value of initial permeability.

In the present paper, synthesis of NiCuZn ferrites by a simple method uses Citrate Precursor Sol- Gel Method and Ball milling for grinding the compound. The synthesized nanocrystals have been characterized using thermal analysis techniques. Calcined nano-ferrite samples are characterized by X-ray diffraction (XRD) and Scanning Electron Microscopy (SEM). The magnetic properties of the ferrites

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were investigated using a Vector Network Analyzer (VNA) at room temperature and magnetic susceptibility measured at different magnetic fields and temperatures.

Permanent magnet technology is constantly developing. At present NdFeB magnets are being used in electrical machines. Due to their cost and low availability alternatives are being explored. An additional boost of the progress of permanent magnet synchronous machines (PMSMs) was got after establishing of the interior magnet rotor structure [14], [15] and with the development of tooth-coil winding approaches [16], [17]. [18] presents, At this power and speed areas conventional asynchronous machines should have similar performance characteristics as PMSMs with slightly smaller peak efficiency in the static efficiency map, less torque density and lower power factor [19]. After the rapid increase of the neodymium magnets' price in 2010, there appeared many companies and organizations searching for appropriate designs for so called "rare earth free" electric machines. The main purpose of the rare earth free electric machines is to reach almost the same torque density as in commercially available neodymium PMSMs, without efficiency deterioration. Major part of these attempts is done for hybrid electric vehicle applications [20-27]. Common measures in order to increase the power density of PMSMs are high angular speeds [28], increase the number of pole pairs and increasing the tangential stress [29].

This paper narrates the possibilities for improving the torque density with the use of ferrite coat on magnetic surface of PMBLDC machine. Permanent-magnet (PM) BLDC motor with rare-earth PMs is most popular,but rare-earth PMs have problems with high power low voltage applications, high cost and limited supply. Therefore, the electric motors with less or no rare-earth permanent magnets are available for numerous application.

II. SYNTHESIS OF NANO-COMPOUND

This section gives the details of synthesis of the nano compound $Ni_{0.5}\,Cu_{0.2}Zn_{0.3}\,Fe_2\,O_4$:

a. A magnetic spinel nano NiCuZn ferrite catalyst with composition $Ni_{0.5}\,Cu_{0.2}Zn_{0.3}\,Fe_2\,O_4$ was chosen for this study.
b. For the preparation of catalyst, aqueous solutions of stoichiometric amounts of Nickel nitrate, Copper nitrate and Zinc nitrate along with ferric citrate were reacted with citric acid in 1:1 molar ratio.
c. $p^H$of the solution was increased by the addition of ammonia to complete the reaction and ethane diol was added.
d. The solution was evaporated very slowly over a period of 24 hours to dryness. Viscosity and color were changed as the solution turned into puffy, porous dry gel. As soon as the solvent removal is completed, dried precursor goes under a self-ignition reaction to form a very fine powder known as synthesized powder.
e. The synthesized powder thus obtained was calcined in a muffle furnace at 600c for 2 hours to remove the residual carbon and furnace cooled. Then matter is subjected to Ball milling for 2 hours at speed of 450 rpm.

III. CHARACTERIZATION OF NANO COPPER FERRITE

a. X- RAY DIFFRACTION (XRD) ANALYSIS:

Fig1. shows typical XRD pattern for nano copper ferrite sample which was sintered at 600 degree celsius. The pattern shows all the characteristics peals of a spinal structure and confirms the phase formation indicating the absence of other impurity phases. The XRD parameters of various peals were compared with the standard data of the cubic copper ferrites and found to be in cubic phase. The particle size and other characteristics of the copper nano particles obtained from the XRD pattern using Scherer's formula was found to be 39nm and reported in table1. The peals can be indexed to (220),(311),(400),(422),(511) and (440) phases of a cubic unit cell are shown in fig 2.The X-ray diffraction pattern was studied in detail forthe determination of crystallite size by using the classical Scherrer equation [15]:

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

Where, D is the average crystallite size, k is a constant equal to 0.89, λ is the X-ray wave length (0.1542 nm), θis the angle of diffraction and β is the full width at half maximum (FWHM) of the peak.

The average crystallite sizes of the powders were in the range 39 nm which indicates that the Cu substitution for Ni has no effect on the crystal size.

The lattice parameter (a) has been calculated from X-ray datausing the formula:

$$\frac{1}{d^2} = \frac{h^2 + k^2 + l^2}{a^2} \qquad (2)$$

Where, d is the lattice spacing and h, k and l are the miller indices ofthe plane.

The theoretical density or the X-ray density (D_x) was calculatedaccording to relation:

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$$D_X = \frac{ZM}{Na^3} \qquad (3)$$

Where, Z is the number of molecules per unit cell (Z = 8), M is themolecular weight, N is Avogadro's number and a3 is the volume ofunit cell.

The variation of the average crystallite size, lattice parameterand X-ray density, with copper content, are shown in Table 1.

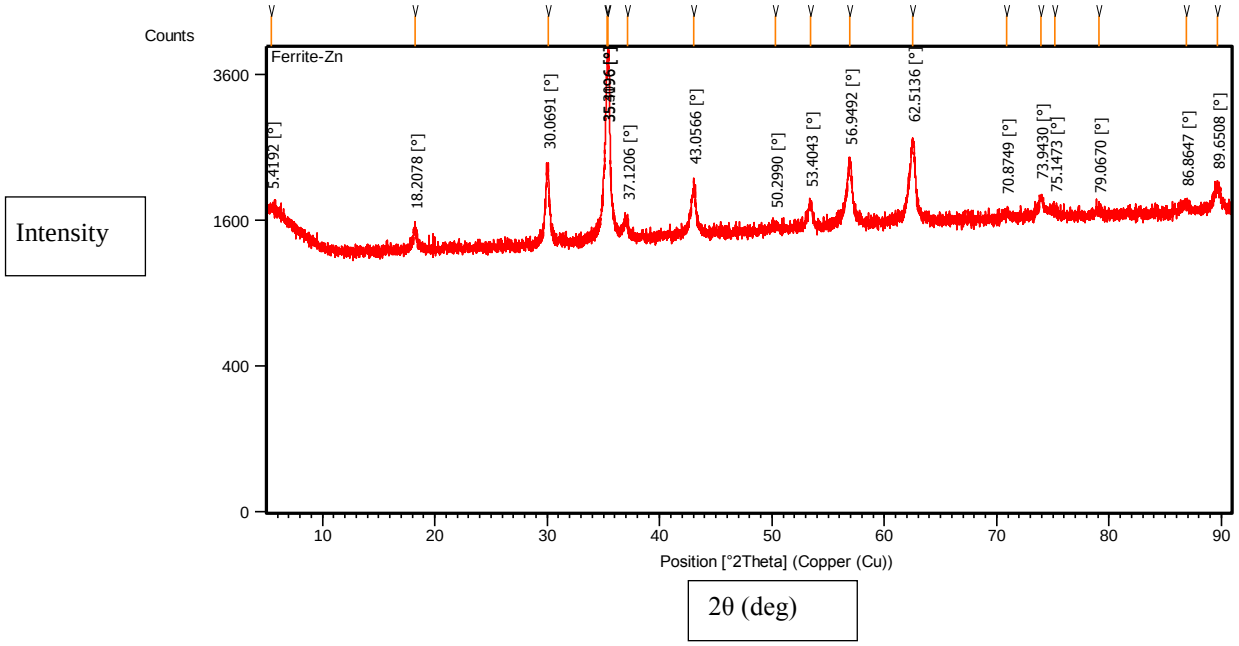

Fig.1: XRD Pattern of Nano-Composite $Ni_{0.5}\,Cu_{0.2}Zn_{0.3}\,Fe_2\,O_4$

b. Morphological and elemental analysis (SEM&EDS):

Fig3 shows the typical SEM image of the nano NiCuZn ferrite sintered at 600degree Celsius. The crystallite size calculated from XRD is in the range of below 30 nm which is in agreement with the SEM image. The structural composition and crystallinity of the NiCuZn ferrite nano particles was further examined by using SEM and TEM. The iron and copper ratio in the nano crystals as determined by EDX analysis was very much close to the atomic ratio in the formula NiCuZn ferrite.

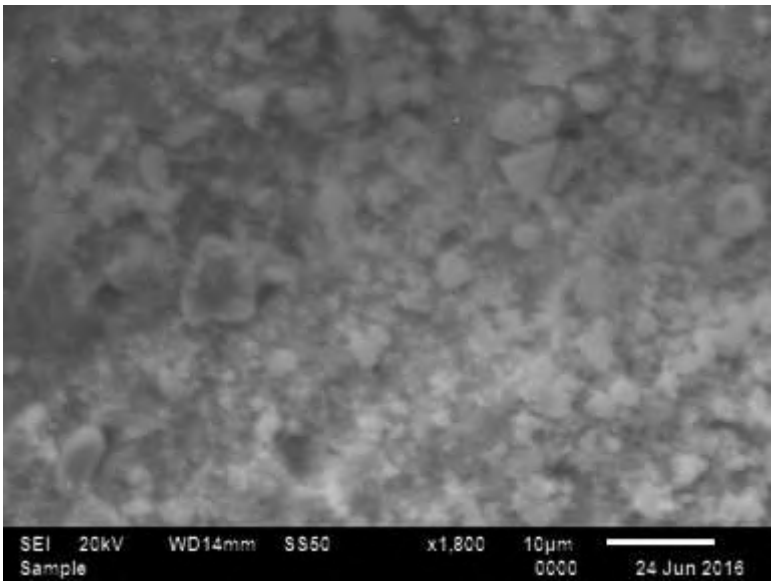

Fig.3: Obtained SEM image of NiCuZn ferrite

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Table.1: particle size and other characteristics of the nano $Ni_{0.5}Cu_{0.2}Zn_{0.3}Fe_2O_4$ *ferrite obtained from the XRD analysis.*

S. NO	Parameters	Values
1	Lattice parameter (a)	8. 379 A
2	Density (%)	94.2
3	X-ray density (D_x)	5.35g/m
4	FWHM	0.284
5	Grain size	1.42
6	Average crystallite size (D)	39 nm
7	Saturation magnetization (M_s)	47. 4 emu/g
8	Magnetic moment (η_B)	2.03 µB
9	Remnant magnetization, M_r	7.15 emu/g
10	Corecivity, H_c	67.8 Oe
11	Curie Temperature, T_c	690 ^{0}C
12	Effective magnetic moment (μ_{eff})	4. 65 µB

IV. RESULTS AND DISCUSSION

Electromagnetic properties measurement of $Ni_{0.5}Cu_{0.2}Zn_{0.3}Fe_2O_4$ system is using a Vector Network Analyzer(VNA). Fig. 3 **S**EM image of the sample with a field scan up to ±5.0 kOe at room temperature. The hysteresisloop of Ni0.5Cu0.2Zn0.3Fe2O4 is shown in fig.4. The sampleshows a ferromagnetic nature with &curves typical for soft-magneticmaterials.The values of saturation magnetization (M_S), remnant magnetization(Mr) and coercivity (H_C) areshown in Table 1. In ferrites,the magnetic moment arises mainly from the parallel uncompensatedelectron spin of individual ion. The intensity of magnetizationcan thus be explained by considering the metal ion distribution andantiparallel spin alignment of the two sub lattice sites as given byNeel's Model [19]. According to Neel's model, three types of interactionsAA, AB and BB are present with the intersub-lattice ABsuperexchange interaction is the strongest one of them. Since Zn^{2+}ions are non-magnetic, the contribution to the magnetization ismostly due to Ni^{2+}, $Cu^{2+}$and Fe^{3+} ions having magnetic momentsof 2.3, 1.3 and 5 µB respectively.

The experimental magnetic moment (η_B) is determined fromthe saturation magnetization data using the following formula [14]:

$$\eta_B = \frac{MW_X M_S}{5585} \qquad (4)$$

Where MW is the molecular weight of the sample

M_S is thesaturation magnetization in emu/g

The calculated values of the experimental magnetic moment (η_B) is presented in Table 1. The gradual decrease in the values of saturation magnetization and experimental magnetic moments with increasing copper content is accounted for the weakening of the AB interaction, which holds well with the decrease in theoretical values of magnetic moment. The decrease in coercivity (H_c) with increasing copper concentration may be attributed to lower magneto-crystalline anisotropy of Cu^{+2} ions as compared to Ni^{+2} that leads to lower coercivity according to the Stoner–Wolfforth model for coercivityof nano-particles [21].

Saturation magnetization value is obtained at room temperature is tabulated in Table 1 are relatively high especially at higher concentrations of copper content as compared with the results of Jadhav et al. [10]. On the other hand, the obtained coercivities show lower values as compared with the same results. This suggests that, the present method of synthesis used is Citrate Precursor Sol-Gel Method and Ball milling for grinding the compound, has an impact on improving the magnetic properties of the system. The temperature dependence of the molar magnetic susceptibility (χ), as a function of the magnetic field intensity is investigated for sample. The Curie temperatures and the effective magnetic moments are reported in Table 1.

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Table.2: Comparison of Permanent Magnet Materials:

Material	Remanence B_r(T)	Coercivity H_c (kA/m)	Curie temperature (^{0}C)	Comparisons
$Ni_{0.5}\,Cu_{0.2}Zn_{0.3}\,Fe_2\,O_4$	0.5….1.35	33.1…76.8	550….850	+ low Cost material +High magnetic properties +linear +Availability
SmCo	0.9…1.1	700…2400	500…850	+ High magnetic properties + linear + very High Cost
NdFeB	1.0…1.4	900…3200	310	+ High magnetic properties + linear -High temperature Coefficients -prone to corrosion

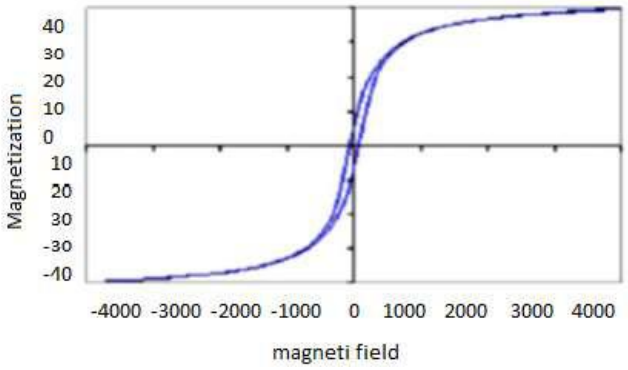

Fig.4: Magnetic hysteresis loops for $Ni_{0.5}\,Cu_{0.2}Zn_{0.3}\,Fe_2\,O_4$

For all the samples, the absence of any thermal stability of $\eta_M$with increasing temperature indicates that the thermal energy isquite sufficient to disturb the ordered spins even at lower temperatures.The measured Curie temperature decreases with increasing Cucontent (Table 1). This observed variation can be explained in termsof the magnetic super exchange interaction which has a direct relationwith Curie temperature [13,22]. Further, the strength of A–Binteraction, which is the interaction existing between the antiparallel uncompensated electron spin of A and B sub-lattices is the mostdominant [23]. This interaction is observed to decrease with Cusubstitution as indicated by the magnetization measurements thusaccounting for the fall in Curie temperature.

V. CONCLUSION

Nano-crystalline $Ni_{0.5}Cu_{0.2}Zn_{0.3}Fe_2O_4$wassuccessfully synthesized and prepared using Citrate Precursor Sol- Gel Method and Ball milling for grinding the compound. The obtained powders were characterized using TG,XRD, FT-IR and TEM techniques. The results indicate that, single phase cubic ferrites were obtained after calcining the precursors at 600^{0}C for 2hours. On investigation of characteristics and properties it is observed that, instead of the copper substitution has weak effect on the structural properties of the system, but it greatly affects the magnetic properties.PMBLDC machine with ferrite coated magnets is simulated and its performances have been evaluated.

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