Structural and Dielectric Studies of Gadolinium Substituted Nickel Ferrite Nanoparticles

Rafeekali K, Muhammed Maheem, E.M. Mohammed

Research department of physics, Maharajas college, Eranakulam, Kerala

Abstract— Nanoparticles NiFe2-xGdxO4 (x=0, 0.1, 0.2, 0.3, 0.4) ferrite was prepared by solgel combution method. The samples were characterized with X-ray diffraction and SEM measurements. The effect of Gd3+ cations substitution on structure of prepared nanoparticles was investigated. From the analysis, the system was found to be inverse spinel cubic structure. The lattice parameter (a) increases with Gd doping content. . Room temperature DC electrical resistivity decreases .Dielectric properties have been studied in the frequency range of 1 kHz to 5 MHz. Permittivity and tangent loss (tan δ) decreases with the substitution of Gd3+ in parent crystal structure.

Keywords: Rare earth ions, XRD, Dielectric properties, Permittivity.

1 INTRODUCTION

In the recent years, so much attention has been paid to the nanomagnetic materials that show very interesting magnetic properties.

In this material, different properties and applications are appeared as compared to their bulk counterparts. The magnetic properties of nanomaterials are used in medical, electronic, and recording industries that depend on the size, shape, purity and magnetic stability of these materials. 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. nano ferrites are simultaneously good magnetic and dielectric materials. These properties of the nano ferrites are affected by the preparation conditions, chemical composition, sintering temperature and the method of preparation . Several chemical and physical methods such as spray pyrolysis, solgel, co-precipitation, combustion technique, high energy milling etc. have been used for the fabrication of stoichiometric and chemically pure nano ferrite materials. Among the available chemical methods, the sol-gel technique is an excellent method to synthesize rare earth substituted nanoparticles with maximum purity. In spite of the development of a variety of synthesis routes, the production of nickel ferrite nanoparticles with desirable size and magnetic properties is still a challenge. This would justify any effort to produce size tuned nickel ferrite nanoparticles with rare earth substitution. In the present paper, the structural and magnetic properties of gadolinium substituted nickel ferrite and XRD,SEM and DIelectrical properties were investigated.

2. Experimental

2.1. Synthesis

Nano particles of Gadolinium substituted nickel ferrite were synthesized by the sol-gel combustion method. A stoichiometric ratio of $NiFe_{2-x}Gd_xO_4$ (x=0, 0.1, 0.2, 0.3, 0.4) were dissolved in ethylene glycol using a magnetic stirrer. The five sample solutions was then heated at 60 °C for 2 hours until a wet gel of the metal nitrates was obtained. The gel was then dried at 120 °C. This resulted in the self ignition of the gel producing a highly voluminous and fluffy product. The combustion can be considered as a thermally induced redox reaction of the gel wherein ethylene glycol acts as the reducing agent and the nitrate ion acts as an oxidant. The nitrate ion provides an oxidizing environment for the decomposition of the organic component. The obtained powder of different samples NiFe_{2-x}Gd_xO₄ (x=0, 0.1, 0.2, 0.3, 0.4) was ground well collected in different packets for the measurements.

2.2. Characterization

The gadolinium doped nickel ferrite samples were characterized by an X-ray powder diffract meter (XRD, Bruker AXS D8 Advance) using radiation (*wavelength*= 1.5406 Ű) at 40 kV and 35 mA. Lattice parameter was calculated. The crystal structure, crystallite size and X-ray density were determined. The particle size was determined using scanning electron microscopy(TESCAN VEGA3SBH). Electrical studies conducted using RF material analyser(AGILENT E4991A)

3. Results and Discussion

3.1. Structural Analysis

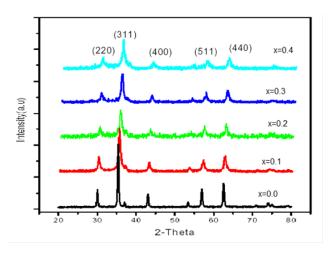


Fig.1. XRD patterns of NiFe_{2-x}Gd_xO₄ (x=0, 0.1, 0.2, 0.3, 0.4)

The XRD patterns of NiFe_{2-x}Gd_xO₄ (x=0, 0.1, 0.2, 0.3, 0.4) nanoparticles are depicted in Fig. 2 and are typical of spinel structure. Comparing the XRD pattern with the standard data, the sample with gadolinium concentration zero shows highest peak and concentration 0.4 shows lowest. The diffraction peaks are broad because of the nanometer size of the crystallite. The crystallite size 'D' of the samples has been estimated from the broadening of XRD peaks using the Scherrer equation.Lattice parameter 'a' for all the samples has been calculated by interplanar spacing (dhkl) and 2-theta values using the standard relation, Value of lattice constant for x=0.0 comes out to nbe 8.3865Å, well in agreement with reported value. Lattice constant has increased monotonically with increment in Gd³⁺ concentration. This increase can be easily explained due to substitution of large ionic radii of Gd³⁺ (0.94Å) in place of smaller Fe³⁺ (0.67 Å) ions. Also rare earth ions are usually present at grain boundaries that cause hindrance in the grain growth, therefore crystal size and unit cell parameters increases.

The crystallite size was observed to increase with the increase of gadolinium concentration. It has been reported that the doping process generally decreases lattice defects and strain, but this technique can cause the coalescence of smaller grains, resulting in an increased average grain size for the nanoparticles. Calculated values of lattice parameter of gadolinium substituted nickel ferrite samples were in close agreement with standard data.

3.2. SEM Analysis

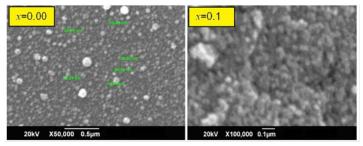


Fig.2. SEM Image of NiFe_{2-x}Gd_xO₄ at x = (0.00, 0.1)

The SEM system that has been used for morphology of sample was CAMSCAN MV2300 model with 15 KV applied voltage. Photo-

graph has been taken from the samples which were doped with gadolinium concentration 0.0 and 0.1. Particle size is obtained around 23.0 nm with mono disperesed nano particles as one can see from the photograph. It Shows the surface morphology and grain size for the prepared sample x=0.0 and x=0.1. It is clear from the image that uniformly distributed; less agglomerated and homogenous spherical particles have been formed in a controlled environment by sol-gel combustion technique. Micrographs also confirm the increase in grain size with Gd³⁺ doping in the parent crystal structure. The homogeneity of shape and grain size largely affects the electrical and magnetic properties of ferrites.

3.3. Dielectric study

Dielectric behavior of nano spinel ferrites mainly depends upon the nature and distribution of metal cations on A-sites and B-sites in the spinel lattice. Spinel nickel ferrites are considered good dielectric materials and the high frequency dielectric behavior is mainly dependent upon the particle size and method of synthesis of nano particles. Different studies have been provided relating the dielectricparameters of Gd3+ doped ferrites. Dielectric parameters (real and imaginary parts of relative permittivity, dielectric loss tangent) for the prepared series of NiGd_xFe_{2-x}O₄ (x=0.0 to x=0.4) have been studied in the frequency range 1 MHz to 1GHz at room temperature. Figs.3 shows the variation of relative permittivity with frequency at room temperature. It can be be be from the figure that relative permittivity for all the samples decreases with increase in frequency and ultimately becomes constant at higher frequencies. This decrease in permittivity is more rapid in the low frequency region and becomes sluggish as the applied frequency increases. This behaviour is subjected to dielectric polarization under the application of AC field.

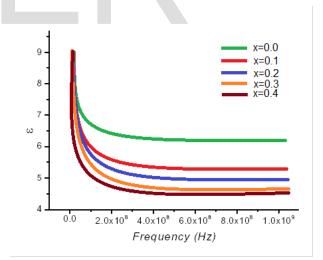
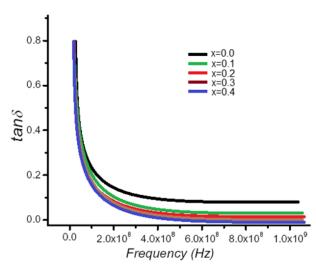
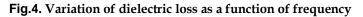


Fig.3. Variation of permittivity as a function of frequency

It can be seen that dielectric loss tangent has the same trend as permittivity losses. It decreases with increase in frequency and becomes constant up to 1GHz due to decreased polarization at high AC fields. At x=0.4 shows a low loss dielectric behaviour which allows its use in high frequency data reading/writing in electronic structures





4. Conclusions

Nano spinel NiFe_{2-x}Gd_xO₄ with x in step increment has been synthesized by sol-gel Combustion method. All the studied samples are pure cubic spinel phase ferrites without any impurity metal oxides. Lattice constant and crystallite size increases with increase in Gd³⁺ concentration, due to increase ionic radii and atomic weight of gado-linium as compared to Fe³⁺. Substitution of Gd³⁺ ion in parent crystal causes a lattice distortion that can be observed by increased lattice strain in W-H plots. Dielectric constant and loss tangent decreases to 4.92 and 0.016 respectively with increase in the dopant concentration showing that the material with x=0.4 is a low loss dielectric.

5. Acknowledgements

RK and MM acknowledges the Maharajas College under Mahathma Gandhi Universty for providing LAB facilities. EMM thanks DST and UGC for the financial support. Authors thank SAIF,CUSAT Kochi, SAIF, IIT Madras and SAIF IIT Mumbai for providing XRD, FTIR, VSM and TEM measurement facilities.

Reference

[1] Peng J, Hojamberdiev M, Xu Y, Cao B, Wang J, Wu H 2011 Hydrothermal synthesis and magnetic properties of gadolinium doped CoFe2O4 nanoparticles J. Magn. Magn. Mater. 323 133-138. [2] Pillai V, Shah D O 1996 Synthesis of high-coercivity cobalt ferrite particles using water-in-oil microemulsions J. Magn. Magn. Mater. 163 243-248. [3] Cai W, Wan J Q 2007 Facile synthesis of superparamagnetic magnetite nanoparticles in liquid polyols J. Colloid. Interface. Sci, 305 366-370. [4] Sileo E E, Jacobo S E 2004 Gadolinium-Nickel ferrites prepared from metal citrates precursors Physica. B. 354 241-245 [5] Rashad M M, Mohamed R M, El-Shall H 2008 Magnetic properties of nanocrystalline Sm substituted CoFe2O4 synthesized by citrate precursor method J. Mater.

Proc. Technol. 198

139–146.

[6] Rao K S, Kumara A M, Varmaa M C, Choudary R K, Rao K H 2009 Cation dirtibution of Ti

doped cobalt ferrites J. Alloys. Compd. 488 6-9

[7] Yue Zhang, Zhi Yang, Di Yin, Yong Liu, ChunLong Fei Composition and magnetic properties of cobalt

ferrite nanoparticles prepared by the co-precipitation method. J.Magn.Magn.Mater, 322, P. 3470–3475

(2010).

[8] Raming T.P., Winnubust A.J.A., Van Kats C.M., Philipse P., The synthesis and Magnetic Properties

of nanosized Hematite particles. J. Colloid Interface Sci. 249, P. 346 (2002).

[9] Veena Gopalan E., I.A Al-Omari, Sakthi Kumar D., Yasuhiko Yoshida, Joy P.A. et.al. Inverse magnetocaloric

effect in sol-gel derived nanosized cobalt ferrite. Appl. Phys. A: Materials Science &

Processing, 99(2), P. 497-503 (2010).

[10] Gul I.H., Masqood A., Structural, magnetic and electrical properties of cobalt ferrites prepared by the

sol-gel route. J. Alloys compd, 465, P. 227-231 (2008).

[11] Pawan Kumar, Sharma SK., Knobel M., Singh M., Effect of La3+ doping on electric, dielectric and

magnetic properties of cobalt ferrite processed by co-precipitation technique. J. Alloys compd, 508,

P. 115–118 (2010).

[12] Binu P. Jacob, Smitha Thankachan, Sheena Xavier, E M Mohammed, Effect of Gd3+ doping on

the Structural and Magnetic Properties of nanocrystalline Ni-Cd mixed ferrite. Physica Scripta, 84,

P. 045702-045708 (2011).

[13] C. N. Chinnasamy, M. Senoue, B. Jeyadevan, Oscar perales-Perez, K.Shinoda and K.Tohiji, Syntheis

of size controlled cobalt ferrite particles with high coercivity and squareness ratio. Journal of colloids

and Interface Science 263, P. 80-83 (2003).

[14] Maldron R.D. Phys. Rev. 99, P. 1727–35 (1955).