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Synthesis, Characterization and Magnetic Properties of Ce³⁺ Substituted Cu Nanoferrite by Sol-gel Autocombustion Technique

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ABSTRACT: A simple and low cost effective method involving sol-gel auto combustion at low temperature is used to synthesize $CuCe_xFe_{2-x} O_4$. (Were x= 0.0, 0.04, 0.06, 0.08) Nanoferrite was synthesized. Metal nitrates are used analytical grade, such as Nickel nitrate, Cobalt nitrate and ferric nitrate, were used as the source materials. The present work is to study structural and magnetic properties of the synthesized samples. The role of Cr³⁺ (Cerium) substituted Cu (copper) shows the formation of crystal phase, which was identified by X-ray diffraction method. The magnetic properties of the synthesized samples were studied. The lattice constants increase with the increasing in Ce containt. We report the synthesis of nanoparticles with crystalline size is in the range of 48.79 to 30.71nm for 700^oc. The FTIR characterization shows the bond formation and synthesized material is ferrite. The Vibrating Sample Magnetometer was used to obtain the Hysteresis parameters. The magnetic property of the prepared samples shows remarkable changes with change of Ce percentage. UV-spectroscopy, to calculate the band gap energy is in the semiconductor range so that prepared sample is in the nature of semiconductor.

KEYWORDS: Sol-Gel, Cu-Ce nanoferrite, FTIR, XRD and VSM, UV.

I. INTRODUCTION

Ferrite nanoparticles are of great interest because of their scientific aspect and applications in permanent magnets, targeted drug delivery and high density information storage devices. From crystal structure point of view, ferrites are generally divided into two groups: cubic or spinel ferrites and hexagonal or hexaferrites, H. Gul, F. Amin et al [1,2]. Cobalt ferrite is a well known hard magnetic material with a high coercivity and a moderate magnetization. Copper ferrite (CuFe₂O₄) is one of the important spinel ferrites MFe₂O₄ because it exhibits phase transitions, changes semiconducting properties, shows electrical switching and tetragonality variation when treated under different conditions in addition to interesting magnetic and electrical properties with chemical and thermal stabilities, S.D. Sartale, C.D. Lokhande et al [3]. It is used in the wide range of applications in gas sensing et al [4], catalytic applications, K.-S. Kang, C.-H. Kim, S. Kameoka, T. Tanabe & N. Nasrallah, M. Kebir, Z. Koudri et al [5-7], Li ion batteries, R.K. Selvan, N. Kalaiselvi et al [8] high density magneto-optic recording devices, color imaging, bioprocessing, magnetic refrigeration and ferrofluids, S. Roy, J. Ghose, [9]. Moreover, CuFe₂O₄ assumes great significance because of its high electric conductivity, high thermal stability and high catalytic activity for O_2 evolution from alumina–cryolite system used for aluminum production, [10]. CuFe₂O₄ is known to exist in tetragonal and cubic structures. Under slow cooling Cu-ferrite crystallizes in a tetr R.K. Selvan, C.O. Augustin et al agonal structure with lattice parameter ratio c/a of about 1.06. Tetragonal phase of Cu-ferrite has inverse spinel structure with almost all Cu^{2+} ions occupying octahedral sublattice, whereas Fe3+ ions divide equally between the tetrahedral and octahedral sublattices, M. Sultan, R. Singh et al [11]. On other hand Cerium is a chemical element with symbol Ce and atomic number 58. It is a soft, silvery, ductile metal which easily oxidizes in air. Cerium was named after the dwarf planet Ceres (itself named for the Roman goddess of agriculture). Cerium is the most abundant of the rare earth elements. Cerium is the most abundant of the rare earth elements, making up about 0.0046% of the Earth's crust by



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weight, W. Ponhan, S. Maensiri, et al [12]. The increase in the electrical conductivity at low temperature is attributed to the impurities, which reside at the grain boundaries [13]. In addition, their magnetic properties can be controlled and tailored to practical applications through the appropriate choice from a number of divalent cations in their structure , K. Muthuraman, [14]. Cu^{2+} ion concentration in Ni-Zn spinel ferrite by combustion method has been investigated single phase cubic structure and surface morphology. <u>V.V. Awati, S.M. Rathod</u> et al [15]. Permanent magnets, targeted drug delivery and high density information storage devices. From crystal structure point of view, ferrites are generally divided into two groups: cubic or spinel ferrites and hexagonal or hexaferrites, Y. Qi, Y. Yang, X. Zhao & .H. Gul, F. Amin, A.Z. Abbasi et al [16,17]. Magnetic nanoferrite particles have generated diverse technological interests because of their potential applications in magnetic fluids, high frequency magnets, magnetic bulk cores, microwave absorbers, and high-density data storage, I.H. Gul, F. Amin et al [18] Nano –sized ferrite with narrow size substitution and uniform particle size are of current research interest for important applications in technology field like drug delivery electric devices, W. C. Kim, S. J. Kim, A. W et al [19] Ni²⁺ substituted Mg-Zn spinel ferrite has been synthesized by sol-gel method and studied phase crystal structure and morphology, <u>D.H. Bobade, S.M. Rathod</u> et al [20]. The variation of Co²⁺ substitution has a significant influence on the grain size and magnetic properties. The mean crystalline size of the prepared ferrite was in the range of ~ 36 - ~45 nm, Patil Basavani K et al [21].

In this investigation, the effect of Ce^{3+} substitution in $Ce_x Cu Fe_{2-x} O_4$ is studied. The sol-gel method is used to synthesize the nanoparticles of $Ce_x Cu Fe_{2-x} O_4$. The structural, optical and magnetic properties of the synthesized samples have been discussed in the contents.

II. EXPERIMENTAL METHOD AND MATERIALS

Cu-Ce ferrite powders were synthesized by sol-gel auto combustion technique at low temperatures for different compositions Cu Ce_x Fe_{2-x}O₄ (where x=0.0, 0.02, 0.04, 0.06,) Raw materials are used in the experiments are AR grade nitrates i.e. Cu(NO₃)₂, Ce(NO₃)₂, Fe₂(NO₃)₂ and C₆H₈O₇ is used as a fuel in the ratio 1:3, all from Merck company of purity of 99 % using stoichiometric ratio and dissolved in distilled water. The mixture of the raw material was stirrered till to get the homogeneous solution then Maintaining pH 7 by adding ammonia. After at 80^oC on hot plate magneto-stirrer, it was continuously stirrered to obtain uniform gel. After 4-5 hours it converts from gel to ash form, which was sintered at 700^oC. The FTIR characterization shows the bond formation and synthesized material is ferrite. The structural and average grain size is studied by X-ray diffraction (XRD), it is in crystal nature and average particle size is 48.79 - 30.71 for 700^oc. Also lattice constant, (hkl) planes and grain size was calculated by Bragg's law and Scherer's formulae. From VSM the magnetic properties of the samples show remarkable changes with change of Ce percentage. Optical properties studied from UV-spectroscopy to calculate the band gap energy it is in the semiconductor range so that prepared sample confirms the nature of semiconductor material.

The general chemical reaction

 $CuCe(NO_3).6H_2O + 2Fe(NO_3)_3.9H_2O + 3C_6H_8O_7 - 100^{0}C \ For \ 5hrs \ CuCeFe_2O_4 + 4H_2O + 12CO_2 + 3N_2 + 4H_2O_3 + 100^{0}C \ For \ 5hrs \ CuCeFe_2O_4 + 4H_2O_3 + 12CO_2 + 3N_2 + 4H_2O_3 + 3N_2 + 3N_2 + 4H_2O_3 + 3N_2 + 3$

III. EXPERIMENTAL RESULTS AND DISCUSION

I. FTIR SPECTROSCOPY:-

The FTIR (Fourier Transform Infrared Spectrometer) characterization from figure 1 shows the bond formation and two main metals – oxygen bands at the range of 500-600 cm-1 so that it is conform that the synthesized material is ferrite.



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Figure 1.FTIR of $CuCe_xFe_{2-x}O_4$ (x= 0.06, for 700⁰C).

II. XRD CHARACTERIZATION:-



Figure 2 . XXR of CuCe_xFe_{2-x}O₄ for (x=0.00, 0.02, 0.04, 0.06 for 700⁰c)

Figure 2 shows the X-ray diffraction (XRD) patterns of typical samples of $CuCeFe_2O_4$ (where x=0.0, 0.02, 0.04, 0.06). The XRD patterns shows well developed diffraction line assigned to cubic spinel phase. The all measured XRD peaks match well with the standard patterns (JCPDS) of cubic spinel ferrite. Interplane distance and planes are calculated by Bragg"s diffraction law and index method using equation 1 and 2. The $CuCeFe_2O_4$ nanoparticles exhibit several diffraction peaks which can be indexed as cubic structure. The average crystalline size of the prepared $CuCeFe_2O_4$ nanoparticles was found in the range of 48.57-30.71 nm for 700^0c for the composition of (where x=0.0, 0.02, 0.04, 0.06,), by using Sherrer's formula. The intensity & width of the Bragg's peak Conforming good crystallinity & nano particle size it is also good agreement with research article.

The interplane distance were calculated by using bragg's equation from the relation

$$n\lambda = 2d\sin\theta$$
[1]

The lattice parameter 'a' was calculated using following relation

$$a = d\sqrt{h^2 + k^2 + l^2}$$
 [2]



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Where (hkl) is the miller indices, λ is the wavelength of X-ray radiation and θ is the Bragg angle. The results of interplane distance and lattice parameter are shown in table 1.

The particle size were calculated using Scherrer"s formula

$$t = \frac{0.9\lambda}{\beta\cos\theta} \qquad \qquad [3]$$

Where, "t" is the particle size, and β is the FWHM (full width half maxima) of the peak θ .

Composition of Cu Ce _x Fe _{2-x} O ₄	particle size't' nm	Lattice constant'a'	Inteplane distance'd'nm (311)
X= 0.0	48.7949	8.3073	0.2534
X=0.02	40.1053	8.4063	0.2508
X=0.04	35.5395	8.4257	0.2523
X=0.06	30.7125	8.4400	

TABLE I. : LATTICE PARAMETER AND PARTICLE SIZE

III. VSM CHARECTERIZATION (HYSTREESSIS LOOP)

Figure 3 shows the magnetic properties of the synthesis samples, it shows that the compositions of Ce^{3+} containt increase the Hc (Coercievity), Mr (Magnetic remenance) and Ms (Magnetic saturation) all are decreasing. The results are illustrated in table2. It shows that the Ce^{3+} containt increases the magnetic properties are increases.







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TABLE 2: VSM (HYSTERESIS LOOP) MAGNETIC PROPERTIES

Composition	VSM of Co _x -Ni _{1-x} Fe ₂ O ₄		
Cu Ce _x Fe _{2-x} O ₄ of x	Hc (Oe)	Mr emu/gm	Ms emu/gm
0.00	107814.9371	16.2893	25.8184
0.02	121620.9853	17.2786	26.9578
0.04	128930.8176	17.1776	28.8564
0.06	140985.3245	18.6579	29.2778

IV. OPTICAL (UV SPECTROSCOPY)PROPERTIES

Figure 4 shows Optical properties were studied from UV-spectroscopy to calculate the band gap energy, Band gap of given figure 4 is 2.5306 eV & wavelength absorb by 490 nm. It is in the semiconductor range so that prepared sample is in the nature of semiconductor material.



Figure 4 .UV of $CuCe_{x}Fe_{2-x}O_{4}$ (x=0.06, for 700[°]c)

V. CONCLUSION

The CuCe_xFe_{2-x}O₄ nanoferrites were synthesized using sol-gel technique. The increase in the Ce³⁺ concentration gives the significant changes in the particle size and magnetic properties of the composition Ce_xFe_{2-x}O₄ (where x=0.0, 0.02,0.04 0.06,). The FT-IR spectroscopy study shows two main metal oxygen bands at in the range of 480-490 cm-1 confirming the formation of cubic spinel phase structure of Ce substitute in Cu ferrite. The synthesis of nanoparticles with crystalline size decreases and lattice constant increases as the concentration increases and is in the range of 48.79 -30.71nm for 700^oc. The UV study shows band gap energy Band gap of given wavelength absorb by 480 nm. Band gap of given figure 4 is 2.5306 eV & wavelength absorb by 490 nm. It is in the range of semiconductor materials. So that synthesized samples is in the nature of semiconductor materials. From figure 3 shows that the hysteresis loop, it is clear that the synthesized sample is hard magnet. The magnetic properties are increases with increasing contain of Ce³⁺ in Cu. We are thanking to IIT Mumbai and TIFR Mumbai for providing the characterizations facility. I am thankful to, Department of Physics, Abasaheb Garware College, for their kind cooperation and providing their infrastructure for the characterization purpose. I, also thank to all those giving all type of valuable help in doing the research work.

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