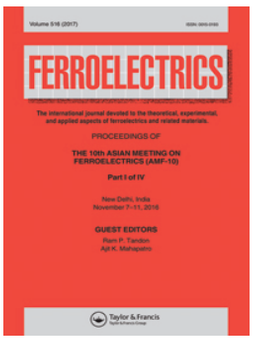

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Structural properties of $Cu_xNi_{1-x}Fe_2O_4$ nano ferrites prepared by urea-gel microwave auto combustion method

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ABSTRACT

Copper-substituted nickel ferrite nanoparticles were prepared via a sol-gel route using urea as a chelating agent. The influence of copper concentration on the microstructure, crystal structure of copper-substituted nickel ferrite nanoparticles has been studied. The results indicate that the substitution of copper influences strongly the microstructure, crystal structure and particle diameter. Powder XRD confirmed single phase cubic structure. Morphology of the particles were studied and particle size was confirmed using SEM. Magnetic properties were studied using VSM method. The dielectric constant and the dielectric loss were increased as the temperature increased at a constant frequency. Dielectric constant (έ) is determined by measuring capacitance (C). Variations of dielectric constant (έ) with frequency as well as temperature have been studied.

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Auto combustion; SEM; XRD; spinel ferrites; nickel ferrites substitution; dielectric; magnetic properties

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1. Introduction

Nanotechnology has been termed as the technology of the century. It includes the key understanding of the connexion between the different physical and chemical properties and material dimensions. Nanotechnology has an extensive range of application from nano-scale, optics to nano-biological system and nano-medicine.

Spinel ferrites are important class of ferrimagnetic material. They exhibit room temperature ferrimagnetism in dielectrics. These are one of the category of ferrites which have been widely studied and there emerged many excellent synthesis methods such as combustion, micro-emulsion, co-precipitation, sol–gel, hydro-thermal, mechanical milling for preparing it at low temperature [1–4]. Microwave assisted sol-gel auto-combustion method offers advantages of short reaction time, high purity, simple, energy efficient and results in ultrafine nanoparticles [1–4].

Nickel ferrite is an inverse spinel ferrite with all nickel ions located in the B-sites and ferric ions occupying both A-sites and B-sites [5]. The substitution of copper in nickel ferrite enhances the properties of nickel ferrite which are useful in many device applications. Nickel-copper ferrites play significant role among magnetic materials due to their high

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electrical resistivity, high saturation magnetization, high magnetic permeability [6]. Copper nickel ferrites have emerged as an important materials in recent years owing to their potential applications in power transformers in electronics, antenna rods, loading coils, microwave devices and telecommunication. Thus, studying the effect of the spinel ferrites composition is required for the understanding of the connectivity between structural phase heterogeneity and physical properties [7–9].

2. Experimental method

The samples of cubic spinel ferrite with formula $Cu_xNi_{x-1}Fe_2O_4$ were synthesized by microwave assisted sol-gel auto-combustion technique. The synthesis technique involved the combustion of redox mixtures, in which metal nitrates acted as an oxidizing reactant and urea as a reducing reactant. The initial composition of solution containing metal nitrates and urea was based on the total oxidizing and reducing valences of the oxidizer and the fuel using the concept of propellant chemistry. The stoichiometric amounts of AR grade $Ni(NO_3)_2{\cdot}6H_2O$, $Cu(NO_3)_3{\cdot}9H_2O$, $Fe(NO_3)_3{\cdot}9H_2O$, and $CO(NH_2)_2$ dissolved in an unionized distilled water at the temperature of 50°C in a beaker. The solution was continuously stirred for 15 minutes and then heated at 70°C for about 6 hours to get viscous gel. The beaker containing the gel was introduced into a microwave oven.

Initially the solution boils and undergoes dehydration followed by decomposition with the evolution of a large volume of gases (N_2, NH_3, and HNCO). After the solution reaches the spontaneous combustion, it begins burning and releases lots of heat, vaporizes all the solution instantly and becomes a solid, burning at temperatures above 1000°C. The entire combustion process, which produces nickel copper ferrite powders in microwave oven, takes about 7 min. This ferrite is ground in an agate mortar for 4 hours to get fine powder to reduce the particle size and to promote the mixing of any unreacted oxides. These ferrites in their respective crucible are sintered at 800°C for 4 hours by slowly raising the temperature of the furnace at the rate of 100°C per hour. After sintering the furnace was cooled in decreasing order of 100°C per hour [10–18].

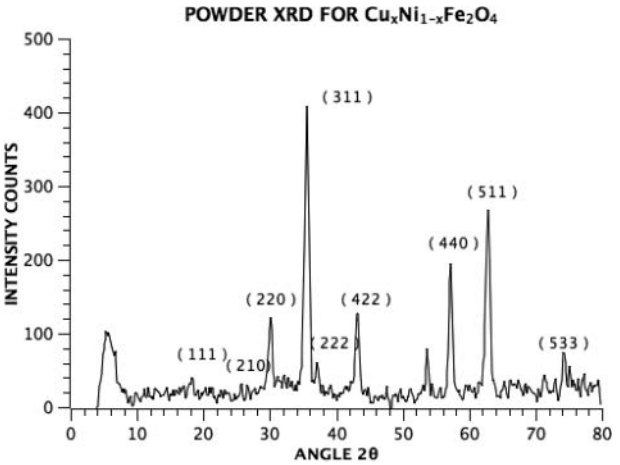

Figure 1. Powder XRD of $Cu_xNi_{1-x}Fe_2O_4$ spinel ferrite.

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Table 1. The lattice parameters of the samples.

Conc.(x)	a(Å)	V (Å ^{3})	M (gm)	X-ray Density g/cm^3	Bulk Density g/cm^3	Porosity (%)	Particle Size (nm)
x = 0	8.3681	585.977	234.344	5.3128	1.68344	68.313	33.16
x = 0.5	8.3030	572.4072	230.5291	5.3022	1.7308	67.65	24.85

3. Results and discussion

3.1 XRD analysis

The XRD pattern has been used to calculate the grain size of the prepared compound. Grain size of all the compound have been calculated from most intense peak (311) using Scherrer's formula $D = 0.9\lambda/\beta\cos\Theta$ where λ is the wavelength of X-ray beam used, β = The full width at half maxima (FWHM), Θ = The corresponding position at particular angle. Powder XRD of doped spinel ferrite samples are as shown in figure 1. The investigated samples of $Cu_xNi_{1-x}Fe_2O_4$ reveals a cubic spinel structure with space group Fd3m without any impurity phase. The miller planes indices are indexed by using Powder-X software. The lattice parameter, X-ray density, bulk density and porosity calculated from therein of samples is summarized in table format [18–20].

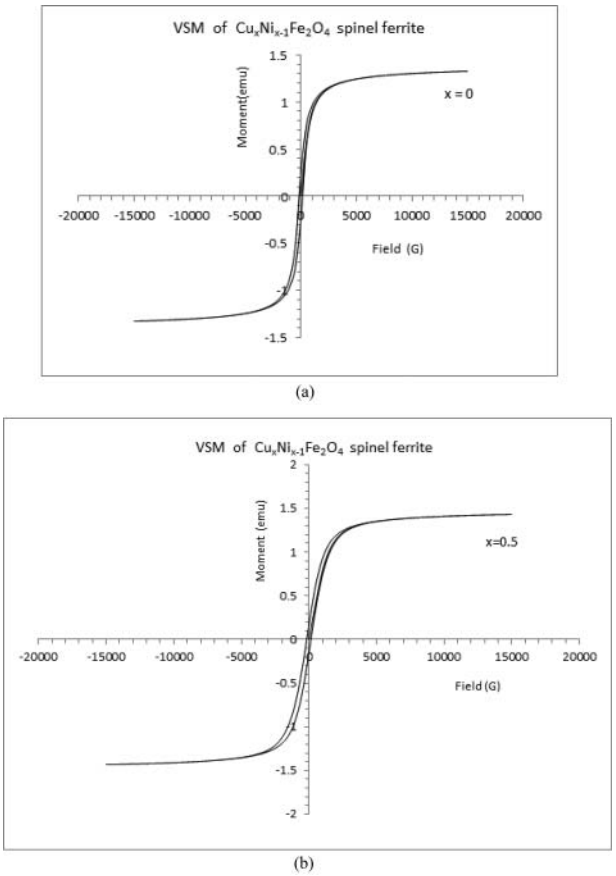

Figure 2. VSM of $Cu_xNi_{x-1}Fe_2O_4$ spinel ferrite.

Table 2. Magnetic properties of the samples.

Compound	Ms	Mr	Hc
$Cu_xNi_{1-x}Fe_2O_4$	1.437 emu/g	0.16262 emu/g	160.64 G
$NiFe_2O_4$	1.0411emu/g	0.19440 emu/g	219.49 G

3.2 VSM of samples

The magnetic properties of the samples were investigated by vibrating sample magnetometer (VSM) at room temperature. Table shows the magnetic hysteresis (M-H) loops of measurements of spinel $Cu_xNi_{1-x}Fe_2O_4$ samples and the observed values such as saturation magnetization (Ms), remnant magnetization (Mr) and coercivity (Hc) have been reported in table 1 and figure 2 (a) and (b) [21–22].

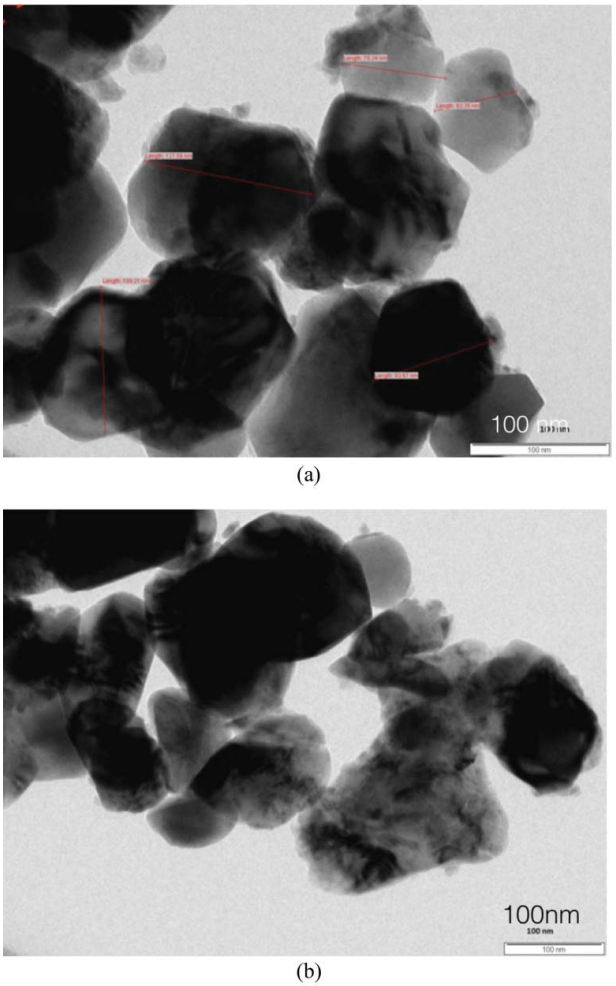

Figure 3. SEM of $Cu_xNi_{x-1}Fe_2O_4$ spinel ferrite.

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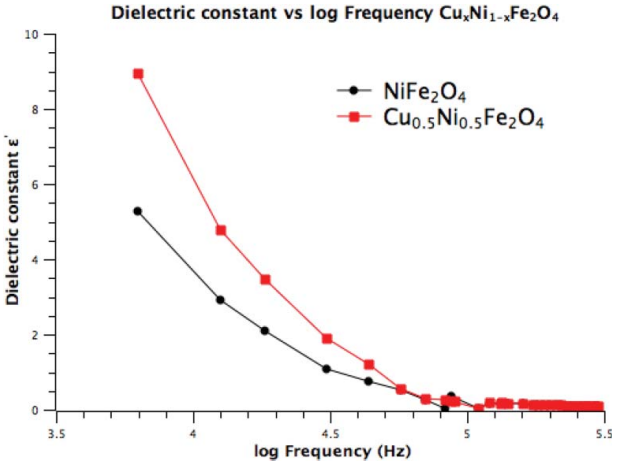

Figure 4. Variation of dielectric constant with frequency.

3.3 SEM results

Scanning electron microscopy results of the prepared samples are shown in the figure 3 (a) and (b). The particle size from SEM pictures is well consistent to the grain size (crystallite size) calculated from the XRD.

3.4 Dielectric properties

It is well known that in the case of low mobility semiconductors, such as ferrites, the activation energy is often associated with the mobility of charge carriers rather than their concentration. The temperature dependence of DC electrical conductivity is shown in figure 4. The DC electrical conductivity reaches a maximum value at a particular temperature known as the transition temperature for $Cu_xNi_{x-1}Fe_2O_4$ ferrite Cu substituted nickel ferrites. The initial increase in conductivity with temperature indicates metallic character and the subsequent increase represents a semiconductor. The activation energy of synthesized samples is calculated from graph of conductivity with temperature. Graph was plotted of log of frequency versus dielectric permittivity for ferrite sample. Dielectric loss decreases with increase in log frequency [23–26].

5. Conclusion

In summary, spinels $Cu_xNi_{1-x}Fe_2O_4$ nanoparticles were successfully synthesized by a facile microwave irradiation sol-gel method using urea as a fuel. Powder XRD, Rietveld refinement XRD, SEM analysis confirmed the formation of spinel nanoferrite of $Cu_xNi_{x-1}Fe_2O_4$ powders with well crystallized nanoparticles without any other secondary phase formation. The average crystallite sizes of the samples are in the range of 24 to 120 nm in size, which was calculated by Debye-Scherrer's formula. HR-SEM images showed the morphology of the samples with slight agglomeration. VSM technique showed that the Ms values of the undoped sample is 1.0411 emu/g, and it is decreased with increasing Ni content, and the M-H loops

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confirmed the ferromagnetic behaviour. The incorporation of Cu in the lattices of $Ni_{x-1}Fe_2O_4$ has shown a remarkable increase of dielectric permittivity in the composition. We have also noted a systematic modification of electrical conductivity with the increase of Cu content. Dielectric properties of doped samples are controlled by both grain and grain boundary kinetics. Dielectric properties are assumed to be affected by the exchange of cations between tetrahedral (A) and octahedral (B) sites and hopping of holes ($Cu^{3+} \rightarrow Cu^{2+}$) and electrons ($Fe^{2+} \rightarrow Fe^{3+}$) among B site cations.

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