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DIELECTRIC PROPERTIES OF ALUMINIUM SUBSTITUTED MAGNESIUM-COPPER FERRITE BY SOL-GEL METHOD

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Short Profile

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ABSTRACT:

Aluminium substituted magnesium-copper ferrites; $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ ($0.1 \leq x \leq 0$. with step 0.1) has been synthesized by using the Sol-Gel auto-combustion method to study the effect of Al3+ doping on dielectric properties of ferrites. The samples were characterized by X-ray diffraction Electron Dispersive Spectroscopy (EDS) and scanning Electron Microscopy for structural properties, LCR meter for dielectric parameters and FTIR absorption techniques. The X-ray diffraction data shows that system exhibit a single phase with cubic spinel structure. Lattice constant was found to be decreases with Aluminium concentration due to larger ionic radius of Fe3+ ions replaced by small ionic radius of Aluminium ions. The dielectric constant, dielectric loss and dielectric loss tangent have been measured in the frequency of the range 100Hz -5MHz for samples sintered at 400^0C. Dielectric constant, dielectric loss and dielectric loss tangent all decreases with increase in frequency and also shows influence of the Aluminium substitution. The FTIR spectra have been analyzed in frequency range 400 to 4000 cm^{-1}. It shows two prominent bands ν_1 and ν_2 which were assigned to tetrahedral and octahedral complexes.

KEYWORDS

XRD, SEM,Ferrite, Dielectric constant, Dielectric loss, Dielectric loss tangent.

INTRODUCTION :

Ferro-spinels have attracted attention of scientist and technologist because of their interesting and controllable physical properties depend on the method of preppation , site preference, valence distribution, chemical composition, sintering temperature etc. Spinel ferrites with general chemical formula MFe_2O_4 (where M as Mg, Cu, Ni, Zn, Mn etc) are promising magnetic materials because of their applications of high density magnetic recording, microwave devices, drug delivery, magnetic resonance imaging, transformer core, memory chips, ferro-fluid etc [1, 2].The chemical methods has ability to produce the particles of nano size dimension opened new application for magnetic material like ferrite. Different techniques are used for synthesis nano-particles of several spinel ferrites relatively at low temperatures in view of their potential applications in several fields.

Magnetic particles have become a considerable interest in last few decades and many physical studies have been dependent on them. The structural, electrical and magnetic properties of nanosized ferrites are entirely different from those of bulk counter part [3]. The ferrites of uniform and narrow particle size distribution are excellent for variety of applications like magnetic data storage, magnetic drug delivery etc. A present study on ferrites is focused on production of nano size material at low temperature by Sol-gel method. It is assumed that the substitution of aluminium ions in magnesium-copper ferrite may alter their structural and dielectric properties and therefore $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ spinel ferrite system has been studied in the present paper because less work has been found on this ferrite system.

EXPERIMENTAL DETAIL-

In the present work, the spinel ferrites having general formula $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ ($x = 0.1 \leq x \leq 0.3$ with step of 0.1) has been prepared by Sol-gel method. Stoichiometric analytical reagent grade nitrates of magnesium, copper, aluminium, iron and citric acid were used as a raw material for the synthesis of $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ spinel ferrites. The as- prepared powder of $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$. This as-prepared samples were heat treated at 100^0C to remove water content. A small quantity of ploy vinyl alcohol was added to sample as binder. Resulting samples were pressed in the form of pellets of 13mm in diameter and 3mm in thickness by applying pressure of 3 tones for 3 minutes by PRO-VAK press machine.

The structural characterization of all samples was carried out by using X-ray diffractometer (BRUKER AXS MODEL-D8 advance).The wavelength of X- ray K α line 1.5406 A°was used.

The average particle size 'D' was determined form intense peak of reflecting plane (311) using Scherrer formula

$$D = \frac{0.9\lambda}{\beta \cos\theta}$$

Where β is angular line width at half maximum intensity and θ is the Bragg angle corresponding to maximum peak.

FTIR analysis of $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1) of as-prepared samples was done at room temperature with wave number range from 400cm^{-1} to 4000cm^{-1}..

The dielectric measurement was carried using an LCR-Q meter (Model HIOKI 3532-50) at room

temperature in frequency range 100Hz to 5MHz.The dielectric loss tangent with respect to variation in frequency directly get from LCR meter. The dielectric constant (ε?) dielectric loss (ε?)and dielectric loss tangent tan δ were calculated using formulae as

$$\varepsilon' = \frac{Cd}{A\varepsilon_0}$$

$$\varepsilon'' = tan\delta\, \varepsilon'$$

where, C is capacitance in F, d is thickness of sample in meter, A is cross sectional area in m^2 and ε_0 is permittivity of free space.

From dielectric constant e?and dielectric loss e" , the a. c. conductivity s ac of the ferrites can be calculated by relation as

$$\sigma_{ac} = \omega\varepsilon_0\varepsilon' tan\delta$$

where ω is the angular frequency

RESULT AND DISCUSSION-

X-Ray diffraction analysis –

Fig.1(a) shows the X-ray diffraction pattern for $Mg_{0.5}Cu_{0,5}Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1) of as-prepared samples. The lattice constant 'a' calculated from the X-ray data for all samples. The lattice constant 'a' decreases with increase in aluminium as shown in

fig. 1.0(b). The decrease in lattice constant is due to replaced larger ionic radii (0.67 A°) of Fe^{3+} ions by smaller ionic radii (0.57 A°) of Al^{3+} ions. Hence lattice constant of present system decreases. Similar observations of lattice constant were observed in the literature [4].

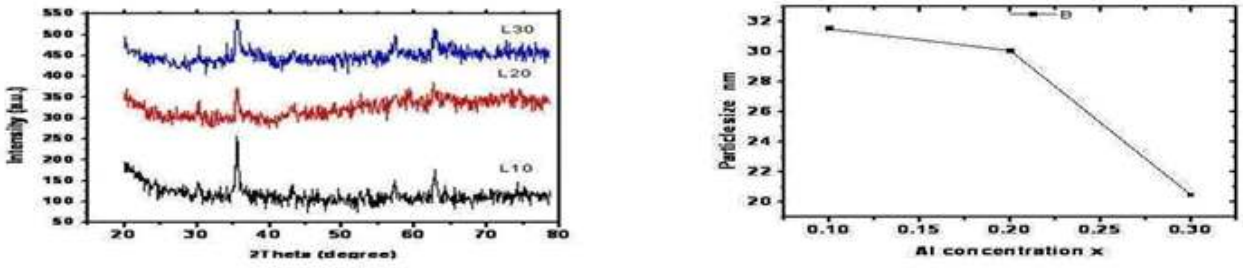

Fig.1-(a) XRD pattern of the $Mg_{0.5}Cu_{0,5}Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1)

(b) Variation of particle size with Al concentration for $Mg_{0.5}Cu_{0,5}Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1) at room temperature

Table 1: Lattice constant, Crystalline size and volume of unit cell of $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1)

Sample code	Content x	Crystal size(nm)	Lattice constant (Å)	Volume of Unit cell(Å)^3
L10	0.1	31.54	8.3728	586.9649
L20	0.2	30.06	8.3664	585.6199
L30	0.3	20.45	8.3532	582.8525

The XRD patterns shown in Fig.1(a) confirm that, single phase cubic spinel structure of all the samples. The crystallite sizes of all the samples were calculated from XRD data using Scherrer formula and were found that decrease in crystallite size with Al content as given in table 1. It is evident that samples were the spinel cubic ferrite. The observed value of lattice constant 'a' also decreases with Al concentration as listed in Table 1. and also as shown in figure.1(b) .

SEM Studies: The visualized micro structure of the samples taken by scanning electron micrograph are shown in Fig.2. SEM micrograph shows that particles were spherical in shape.

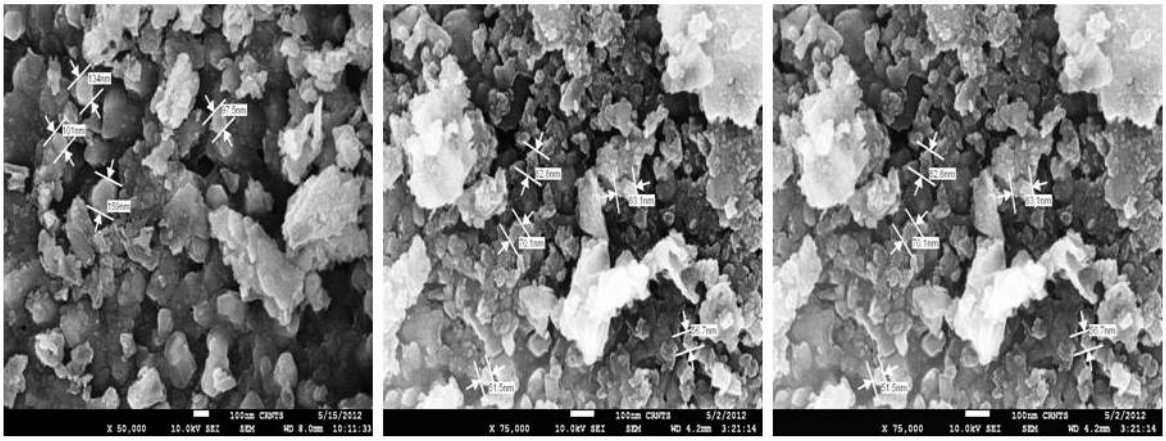

Fig.2: SEM micrographs $Mg_{0.5}Cu_{0.5}Al_xF_{2-x}O_4$ (x=0.1 to 0.3 with step of 0.1) of as-prepared samples.

Energy Dispersive Spectrograph (EDS) analysis -

The EDS spectra as shown in fig.3 indicates the presence of mainly Mg,Cu,Fe and O with small amount of Al content

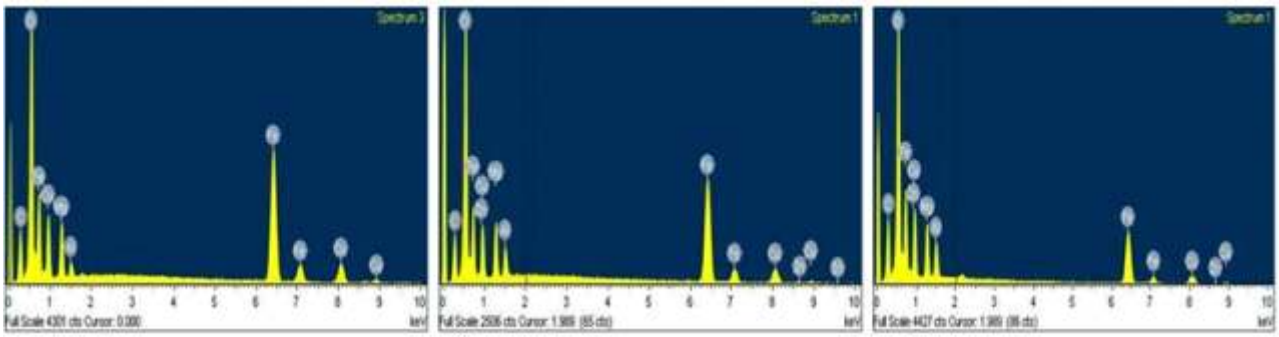

Fig.3 shows EDS patterns for $Mg_{0.5}Cu_{0,5}$ Al_xF_{2-x} O_4 (x=0.1 to 0.3 with step of 0.1) for as-prepared samples.

FTIR studies: Fig.4 shows IR spectrograph of $Mg_{0.5}Cu_{0,5}$ $Al_{0.1}Fe_{1.9}$ O_4 sample. It provides information about position of ions in crystal lattice through vibrations. It shows two distinct peaks ,one around 1540.84 cm^{-1} (υ_1) and other 568 cm^{-1} ($?_2$).The high frequency ($?_1$) peak shows the metal oxygen vibration at tetrahedral sub-lattice (A-site) and the low frequency ($?_2$) peak shows to the intrinsic vibrations of metal oxygen bond at octahedral sub-lattice(B-site).These two absorption bands are expected for the spinel structure of present ferrites synthesized by Sol-gel auto ignition method..

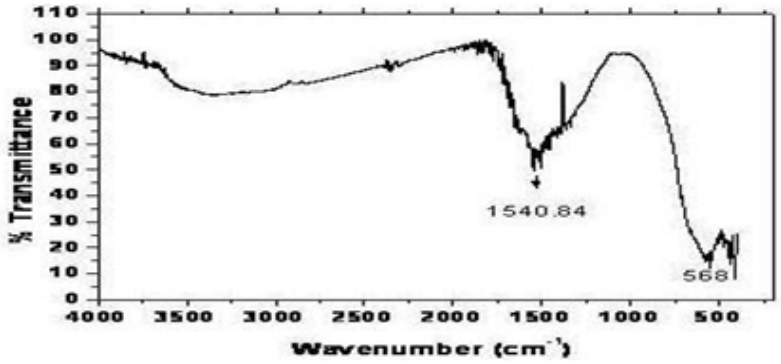

Fig.4 FTIR spectrograph of $Mg_{0.5}Cu_{0,5}$ $Al_{0.1}Fe_{1.9}$ O_4 sample.

DIELECTRIC ANALYSIS-

Dielectric constant

The dielectric properties of ferrite nano-particles are influenced by the synthesis method, grain size, cation distribution, doping ions concentration etc. Fig.3 shows the variation of dielectric constant ε?as a function of frequency ranges from 100 Hz to 5 MHz for as-prepared samples aluminium substituted for iron in magnesium–copper ferrites. The dielectric constant e?decreases with increase in frequency up to certain frequency of electric field and it takes low constant value independent of frequency at high frequency. All the samples show loss peak at low frequency. The peaking nature occurs when the jump

frequency of the electron between Fe^{3+} and Fe^{2+} is equal to the frequency of applied field. Thus there was small abnormal behaviour was observed for all samples at lower frequencies. According to Rezlescu model the relaxation peak may be due to the collective contribution of both p and n type carriers [6, 13].

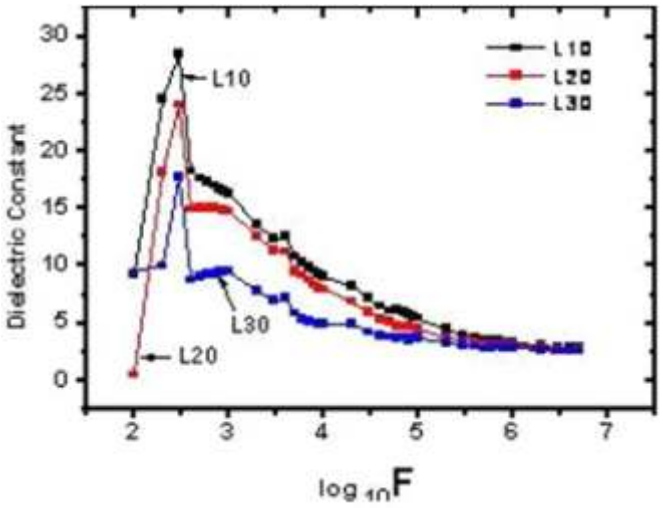

Fig.5: Variation of dielectric constant for $Mg_{0.5}Cu_{0.5}$ $Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1) w.r.t. frequency of electric field at room temperature

According to Maxwell-Wagner two layer model [7, 8], space charge polarization is due to inhomogeneous dielectric structure of the material. It is formed by large number of well conducting grains separated by thin poorly conducting intermediate grain boundaries. This variation of dielectric constant with frequency can be explained on the basis of dispersion is due to Maxwell-Wagner interfacial polarization with Koop's phenomenological theory [12]. According to Koops (1951) the effect of grain boundaries is predominant at lower frequencies. For thinner the grain boundary higher is the dielectric constant e?value. The decrease of dielectric constant with increasing frequency up to certain frequency of electric field, beyond that frequency the electron exchange between Fe^{2+} and Fe^{3+} ions cannot follow the electric field. Hence value of dielectric constant is low and constant at high frequency i.e. it is independent of frequency of the field.

Some other researchers [8] have also explained the compositional dependence of dielectric constant e?decrease with increasing frequency for Al ions substituted ferrites can be explained on the basis of the mechanism of polarization process which is similar to that in the conduction process. Fe ions exist in 2+ as well as 3+states occupy both tetrahedral (A-) sites and octahedral (B-) sites. When Al is added in place of Fe, some of Fe3+ converted to Fe^{2+} ions for charge neutrality. i.e. The electronic exchange between $Fe^{2+} \Leftrightarrow Fe^{3+}$. The decrease of dielectric constant with increase in frequency due to decrease in polarization. The complete polarization in ferrites is contributed by space charge polarization governed by number of space charge carriers and conductivity in materials [10] and hopping exchange of charges between two localized states governed by density of the localized state and resultant displacement of charges with respect to external electric field. The doping of Al ions reduces the Fe^{3+} ions on octahedral B-sites and Al^{3+}ions takes place in octahedral sites which is responsible for both space charge polarization and hopping exchange between localized states. Therefore increase of Al ions concentration causes a decrease

in a polarization which produces a decrease in dielectric constant ε?of the composition. Similar results were found in Mn substituted Ni-Zn ferrites synthesized by citrate method and Cd, Cr substituted Co ferrite [18] etc. Some researchers have also detected that similar kind of behavior [12, 13].The variation in dielectric constant may be explicated on the basis of space charge distribution.

Dielectric constant large value at lower frequencies due to predominance of species like Fe^{2+} ions, oxygen vacancies, grain boundary defects, interfacial dislocation pile-ups, voids etc[14, 15]. The decreasing trend in dielectric constant with the increasing frequency in natural due to the fact that any species contributing to polarizability is found to indicate lagging behind the applied field at higher frequencies [16].

Dielectric loss –

Fig.6 shows the variation of dielectric loss ε2s function frequency for $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1) at room temperature.

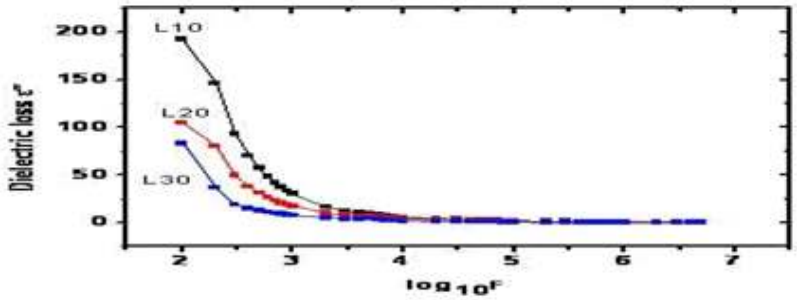

Fig.6: Variation of dielectric loss for $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1) w.r.t. frequency of electric field at room temperature

The dielectric loss ε" decreases with increase in frequency from 100Hz to 5MHz as shown in Fig.6. The dielectric loss is most important part of the total core loss in ferrites. The dielectric loss plot is seen to be similar to that of dielectric constant. The dielectric loss factor e" depend on number of factors such as structural homogeneity, grain boundary defects, interfacial dislocations, stoichiometry, Fe^{2+}content which in turn depend on the composition and sintering temperature of samples[17]. The initial decrease in dielectric loss with an increase in frequency may explain on the basis of Koop's phenomenological theory [12].None of samples show loss peak. The peaking nature occurs when the jump frequency of the electron between Fe^{3+} and Fe^{2+} is equal to the frequency of applied field. All the samples show dispersion in dielectric loss at lower frequencies. It is reasoned out that the frequency at which maximum in dielectric loss in these samples is outside the frequency range studied [16].

Both dielectric constant and dielectric loss obtained for present nano-ferrites synthesized by Sol-gel method possess lower value than that prepared by conventional ceramic method for the same composition. The low dielectric values make these ferrites useful in the high frequency applications.

Dielectric loss tangent

Fig.4 shows the variation of dielectric loss tangent tand as function frequency for all samples.

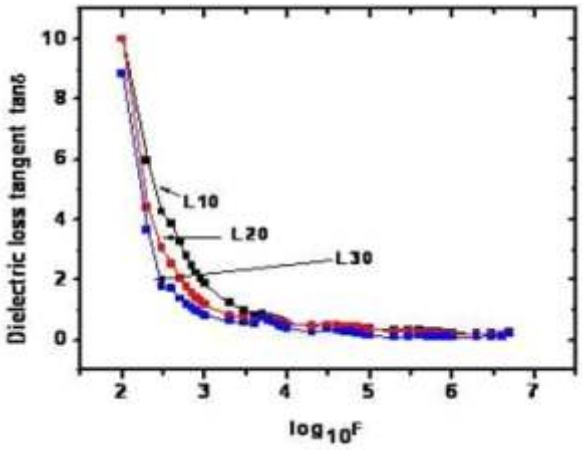

Fig.7: Variation of dielectric loss tangent, tanδ for $Mg_{0.5}Cu_{0.5}$ $Al_xFe_{2-x}O_4$ system (x=0.1 to 0.3 with step of 0.1) w.r.t. frequency of electric field at room temperature

Fig.7shows the dielectric loss factor tand decreases exponentially with increase in frequency. The decrease in tand with increase in frequency is due to the hopping frequency of charge carrier can't follow changes of the polarity of external field beyond certain frequency [6]. According to the Iwauchi [5] there is strong co-relation between the conduction mechanism and the dielectric behaviour of the ferrites.

The maximum value of tand observed when hopping frequency is approximately equal to that of applied electric field. The dielectric dispersion curve can be explained on the basis of two layer model and Maxwell–Wagner polarization theory. To understand the frequency dependent dielectric constant in ferrite materials, Koop's theory [12] proposed a theory in which relatively good conducting grains and insulating grain boundary layers of ferrite material represent with behavior of an inhomogeneous dielectric structure as discussed by Maxell [7]and Wagner [14]. Since an assembly of space charge carrier in the inhomogeneous dielectric structure described required finite time to line up their axes parallel to an applied alternating field ,the dielectric constant decreases as if the frequency of reverse field increases. Furthermore the jumping frequencies of localized charge carriers are almost equal to that of the applied a.c. electric field. The low dielectric parameters obtained for the present samples attributed to be more homogeneous and structurally perfect ferrites prepared by sol–gel method. The decrease in dielectric loss tangent with change in the composition and frequency is in accordance with Koops phenomenological model [12].

CONCLUSIONS-

The $Mg_{0.5}Cu_{0.5}Al_xFe_{2-x}O_4$ spinel ferrites have been synthesized by sol-gel method. Nano- size particles have been obtained using Sol-gel method. The dielectric constant, dielectric loss and dielectric loss tangent all three parameters decrease with increase in frequency. Thus the sol-gel method is found to be

one of easiest, simple and effective chemical technique for preparation of nano-size particles. Hence lower values of dielectric constant of ferrites are used in high frequencies applications as microwave absorbers.

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