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Dielectric Properties of Copper Substituted Nickel Nano-Ferrites

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ABSTRACT
Mixed $Ni_{1-X} Cu_X Fe_2 O_4$ ($0 \leq X \geq 1$) nano-ferrites have been synthesized by Citrate-gel auto combustion technique. The structural characterization was carried out by X-Ray Diffractometer (XRD) and the crystallite size was found to be in the range of 36- 58nm. The dielectric properties such as Dielectric constant (ε'), Dielectric loss tangent (tan δ) and AC conductivity (σ_{ac}) were studied at room temperature as function of frequency in the range 20Hz – 2MHz. The real part of dielectric constant (ε') and dielectric loss tangent (tan δ) showed a decreases with the increase in frequency where as the ac conductivity (σ_{ac}) increases with increase in frequency. The dielectric constant (ε') shows an increasing trend with increase of Cu substitution in Ni nano-ferrites.
Keywords: Nano - Ferrites; Citrate-Gel Technique; X-Ray Diffraction; Dielectric Properties

I. Introductions

In the recent years many physicists have studied the electrical and magnetic properties of nano crystalline spinel ferrites in view of their many applications [1, 2]. As a common feature, the nano crystalline spinel ferrites have very high dielectric constants at low frequencies, so they are very useful in designing microwave devices such as circulators and isolators [3]. These ferrites are very important group of magnetic materials due to their extensive use in a wide range of applications from low to high permeability devices including electronics, ferrofluid, magnetic drug delivery microwave devices and high density information storage devices [4-8].

The electrical conductivity and dielectric behavior in ferrites depends on many factors; preparation method, sintering temperature, amount and type of substitution. Several investigators have studied the frequency and temperature dependence of the dielectric constant [9-11]. The aim of the present work was to investigate the AC conductivity and dielectric constant as a function of frequency at room temperature for Ni nano-ferrite and the study includes effect of Cu^{2+} on the conduction mechanism.

II. Experimental details

The mixed Ni-Cu ferrite powders having the chemical formula $Ni_{1-x}Cu_xFe_2O_4$ (where x = 0, 0.2, 0.4, 0.5, 0.6, 0.8 and 1.0) were synthesized by citrate gel auto combustion technique. Analytical grade of Nickel Nitrate ($Ni(NO_3)_26H_2O$) Copper Nitrate ($Cu(NO_3)_23H_2O$) Ferric Nitrate ($Fe(NO_3)_29H_2O$) Citric Acid-Citrate ($C_6H_8O_7H_2O$) Ammonia (NH_3) raw materials were used to prepare the Ni-Cu nano ferrites. Metal nitrates and citric acid were dissolved in deionized water. Metal nitrate solutions were mixed with citric acid solution in 1:3 molar ratio of nitrate to citric acid. The pH of the solution was adjusted to 7 using ammonia. The solution was first heated at 80^oC to transform into gel and then ignited in a self-propagating combustion manner to form a fluffy loose powder. The as-burnt ferrite powders were grained by agate motor then calcined at 700^oC for 5hr. The calcined ferrite powders were again grained by agate motor.. The calcinations powder of each sample is prepared into pellets were sintered at 1100^oC for 12 hours.

The structural characterization was carried out using X-Ray Diffractomerter Bruker (Karlsruhe, Germany) D8 advanced system with a diffracted beam monochromatic Cu K_α radiation (λ = 1.5405 Å) radiation source between the Bragg Angles 20° to 80° in steps of 0.04°/Sec. The capacitance and tangent loss were recorded using a PRECISION LCR METER – E4980A in the frequency range from 20Hz to 2MHz at room temperature.

III. RESULTS AND DISCUSSION

The X-ray diffraction pattern (**Fig1.**) of crystalline phases were identified by PDF-4 reference data from the international centre for diffraction data (ICDD). All Bragg reflections have been indexed, which confirm the formation of cubic spinel structure in single phase without any impurity peak. The strongest reflection comes from the (311) plane, which denotes the spinel phase. The peaks indexed to (111), (220), (311), (400), (511) and (440) planes of a cubic unit cell, all planes are the allowed planes which indicates the formation of cubic spinel structure in single phase [12]. The values of the crystal size varied

from 36.7 nm to 58.91 nm. Though all the samples were prepared under identical condition, the crystallite size was not the same for all Cu concentrations. This was probably due to the preparation condition followed here which gave rise to different rate of ferrite formation for different concentrations of Cu, favoring the variation of crystallite size.

Compositional variation of dielectric constant at 2 MHz frequency is shown **Fig 2.** It was observed that dielectric constant increased with the increase of Cu concentration and the dielectric constant is maximum at X=1.0 and minimum at X= 0. It is due to the fact that resistivity decreases with the increase in Cu concentration which increases the probability of electrons reaching the grain boundary. This increases the polarization and hence the dielectric constant increases [13].

Fig.3 shows the variation of dielectric constant versus frequency at room temperature. It can be seen from the figure that the value of dielectric constant decreases initially with increase in frequency and reached a constant value at higher frequency. The decrease of dielectric constant with increase of frequency as observed in the case of Ni-Cu nano ferrites is normal dielectric behavior of spinel ferrites. The normal dielectric behavior can be explained on the basis of Maxwell-Wagner interfacial type polarization, which is in agreement with Koop's phenomenological theory [14, 15].A similar variation of the dielectric constant with frequency was observed in the case of $ZnFe_2O$, $MnFe_2O_4$ [16],$Zn_xCu_{1-x}Fe_2O_4$ and $Mn_xCu_{1-x}Fe_2O_4$ [17] ferrite systems. The dielectric polarization in ferrites is similar to the conduction hopping mechanism. Hopping between Fe^{2+} and Fe^{3+} results in the local displacement of electrons in the direction of the applied field and these electrons determine the polarization. The polarization decreased with increasing frequency and then reached a constant value due to the fact that beyond a certain external field frequency the electron exchange between $Fe^{2+} \Leftrightarrow Fe^{3+}$ cannot follow the alternating field. The large value of the dielectric constant at lower frequency is due to the predominance of species like Fe^{2+} ions, interfacial dislocations piled up, oxygen vacancies, and grain boundary defects, while the decrease in dielectric constant with frequency is natural because of the fact that any species contributing to polarizability is found to show lagging behind the applied field at some higher frequencies.

Fig 4. Shows the variation of dielectric loss tangent (tanδ) with frequency for $Ni_{1-x}Cu_xFe_2O_4$ nano-ferrites. By an increase in frequency, for X=0,0.2,0.6 the value of tanδ decreases linearly which is normal behavior of ferrites and for X=0.4,0.5,0.8 and 1.0 the value of tanδ increasing initially and exhibit the loss factor which is maximum between 3×10^5 Hz to 6×10^5Hz and further decrease by increasing the frequency. The loss factor depends upon various factors such as stoichiometry, density, grain size, Fe^{2+}

content and structural homogeneity, which in turn depend on the composition and processing temperature [18]. All the samples exhibit dispersion due to the Maxwell-Wagner interfacial type polarization in agreement with Koop's phenomenological theory [14, 15, 19-21].

Iwauchi [16] pointed out that there is a strong correlation between the dielectric behavior and the conduction mechanism in ferrites. In the low frequency region, which corresponds to high resistivity (due to grain boundaries), more energy is required for electron exchange between Fe^{2+} and Fe^{3+} ions and thus energy loss is high. In the high frequency range, which corresponds to low resistivity (due to the grains), a small energy is needed for electron transfer between Fe^{2+} and Fe^{3+} ions in grains and accompanied by a small eddy current and hence a decrease in the energy loss all the samples show dispersion in tan δ in lower frequencies.

Fig. 5 shows that ac conductivity rises with the rise in the frequency, which is in agreement with the reports [22, 23]. The conductivity of ferrites, in general, depends on the density, porosity, grain size. Grain boundary and chemical composition of samples [24, 25].

IV. CONCLUSION

The Citrate – Gel technique method is a conveniant and varsalite one for obtaining very homogeneous and reactive nanostructured Ni-Cu nano-ferrites. X-ray diffraction patterns confirmed the formation of single phased cubic spinel structure without any impurity peak. The Crystallite size of the nano-ferrite was in the range of 36-58 nm. Dielectric constant increases with the increase of Cu concentration and the dielectric constant is maximum at X=1.0 and minimum at X= 0. The dielectric constant (ε') and dielectric loss tangent (tan δ) decreases with increase of frequency. The ac conductivity (σ_{ac}) rises with the rise in the frequency.

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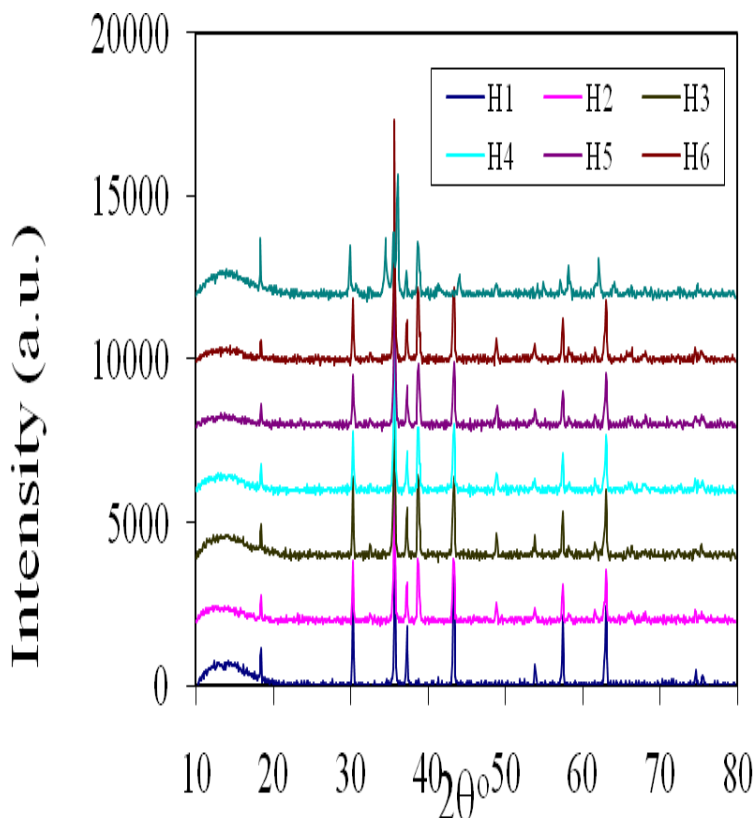[25]   Gul, I.H., Ahmed, W., Maqsood, A.: J. Magn. Magn. Mater. **320**, 270 (2008) 494 (2007)

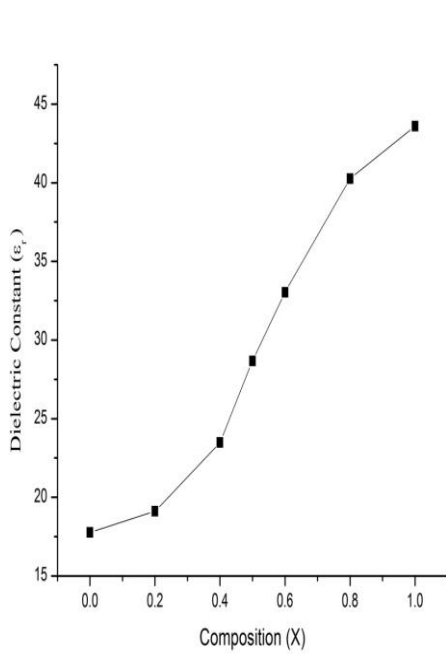**Fig 1. X-ray diffraction pattern of Ni-Cu nano-ferrites**

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Fig 2: Dielectric constant as a function of X (Cu) at 2M Hz in Ni-Cu nano- ferrites.
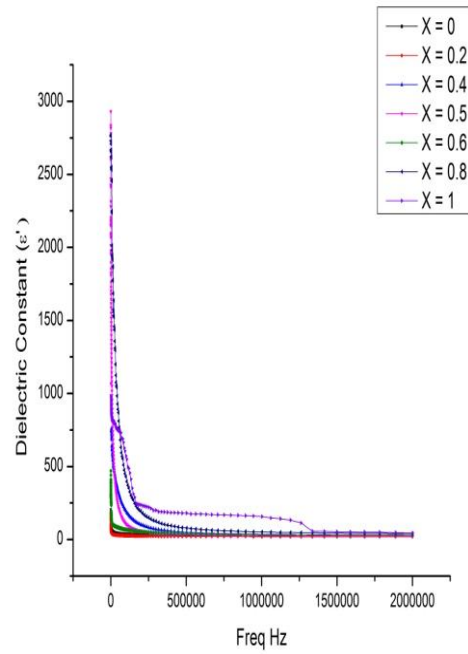

Fig 3: Frequency dependence of dielectric constant for Ni-Cu nano-ferrites.
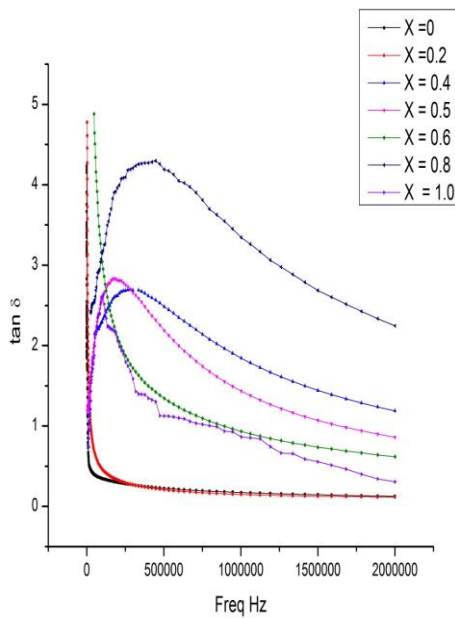

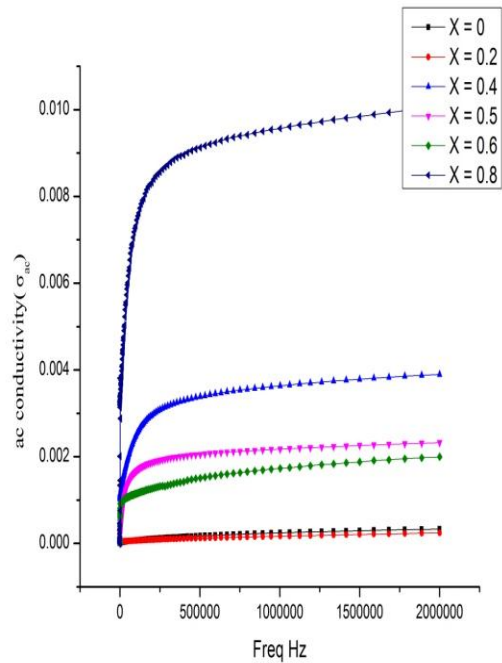**Fig 4: Frequency dependence of dielectric loss tangent for Ni-Cu nano - ferrites.**

Fig 5: Frequency dependence of ac conductivity for Ni-Cu nano- ferrites.