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Influence of NI^{2+} ions on the electronic combustion of Mg-Cu-Zn Ferrite

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Abstract: Spinal ferrites of system $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ (x= 0, 0.05, 0.1, 0.15, 0.2 and 0.25) were synthesized using the citrate gel combustion method. The synthesized samples were studied electric properties such as a.c and d.c. resistivity as the function of temperature and Ni ions. The dielectric properties i.e dielectric constant, dielectric loss and dielectric loss tangent studied as function of frequency. The d.c. resistivity all samples like as semiconductor behavior the activation energy of samples in the range 0.20 to 0.44 eV and Curie temperature increases with increases in Ni^{2+} ions. The dielectric properties follow by Maxwell–Wagner interfacial polarization.

Keyword: Citrate gel Method, D.C. resistivity, dielectric properties, a.c. resistivity.

I. Introduction

Nano particles of ferrites are an important class of magnetic materials finding large number of applications in several fields including biomedical and consumer electronics.As an alternative to Ni-Cu-Zn ferrites, Mg-Cu-Zn ferrites have attracted the attention of number of researchers due to their inherent interesting properties. It is a pertinent magnetic material with high electric resistivity, relatively high Curie transition temperature, low Cost, high mechanical hardness and environment stability [1, 2]. Mg and Zn doped ferrites posses' high resistivity value that is 10^6-10^7 Ω/cm for high frequency. The resistivity ferrites material like Semiconductor behaviour with high resistivity variation over large range of temperature is an ideal characteristic essential for materials in sensor application [3]. The electrical conductivity in could be explained on the basis of transport of charge carriers through cation vacancies present on octahedral sites. These cation vacancies may be created in these ferrites during their preparation at higher temperature [4].The control of resistivity in ferrites are two general approaches (1) controlling sintering temperature and (2) substituted constituents to increase or decrease the conductivity [5]. Studied the role of Sm^{3+} substitution on electrical resistivity in Mg-Zn ferrites then Sm^{3+} increases the resistivity decreased [6]. The present paper is focused on the effect of Ni ion in electric properties of MgCuZn Ferrites synthesized by citrate gel.

II. Experimental

The Ni substituted Mg-Cu-Zn ferrites were synthesized using the citrate gel combustion method. All metal ions analytical reagent grade were used as starting precursors. The ratio of metal nitrates to fuel (citric acid) was kept at 3:5 moles continuous heating on the hot plate converted the precursor into the gel which gets auto ignited to produce voluminous powder. The as prepared powders were collected and annealed at 700ºC for 2 h to remove traces of undecomposition fuel, nitrates (if any), and their decomposition products to obtain the pure and well crystalline powder. These powders were then made into pellets using a hydraulic press machine. DC electrical resistivity measurements were carried out with two probe method. We had measured the resistance of the sample as function of temperature. The dielectric constant (ε′), dielectric loss (ε″) and dielectric loss tangent (tan δ) were measured as a function of temperature and frequency both using LCR-Q meter (Model Hioki 3532-50).

III. RESULTS AND DISCUSSION

D.C. resistivity.

Fig. 1 shows the variation of logarithm of DC electrical resistivity as a function of reciprocal of temperature for all samples. With increase in temperature the resistivity for all samples decreased. Change in the Conductivity mechanism with temperature was observed and the plots exhibited two different regions. The Conductivity mechanism at low temperatures is due to impurities, above the Curie temperature it is due to the change of magnetic ordering.

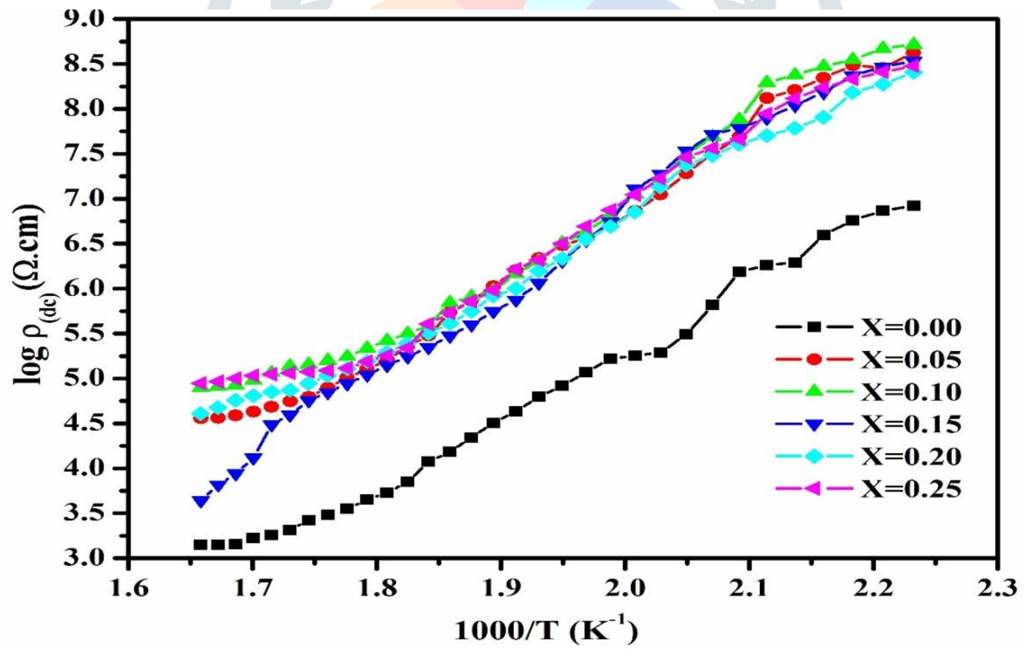

Fig.1: The logarithm of electrical resistivity as a function of reciprocal temperature.

The strong decrease in resistivity with temperature is because of thermally activated mobility of the charge carriers, but not due to a thermally activated creation of charge carriers. The change in slopes of regions separated by Curie point evidenced the influence of the ferrimagnetic ordering on the Conductivity process. The room temperature dc resistivity was found to increase with addition of Cobalt with maximum for sample with x=0.10 and however no systematic variation in resistivity with substitution was observed. The activation energy values in the range 0.21eV to 0.44 eV were strongly influenced by Ni addition and conductivity mechanism. The high values of activation energy for paramagnetic region than ferromagnetic region are in conformity with the theory developed [6].

Dielectric Properties.

Fig 2 depicted dielectric Constant with frequency for all samples the dielectric Constant decreased rapidly with an increase in frequency, and finally remains constant at higher frequencies. This is normal ferrimagnetic behavior and the samples in the present study exhibited low frequency dielectric dispersion which was previously reported by many researchers [6]. The incorporation of Ni into Mg-Cu-Zn ferrites has significantly affected the dielectric constant however no systematic variation with Ni Content was observed. At higher frequency dielectric Constant exhibited Composition independent behavior.

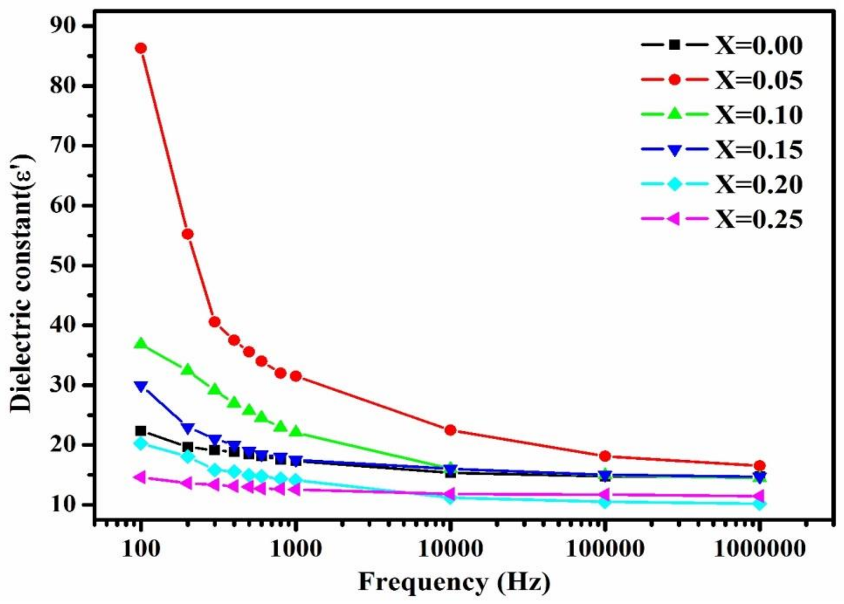

Fig.2: Variation of dielectric constant with frequency.

The dielectric polarization in ferrites is similar to the conduction hopping mechanism. Hopping between Fe^{2+} and Fe^{3+} leads to the local displacement of electrons in the direction of the applied field and these electrons determine the polarization. The polarization decreases with increasing frequency and then reaches a constant value due to the fact that beyond a certain external field frequency, electron exchange $Fe^{2+} \Leftrightarrow Fe^{3+}$ cannot follow the alternating field. The large value of dielectric constant at lower frequency is due to the predominance of Fe^{2+} ions, interfacial dislocation piles, oxygen vacancies, grain boundary defects, etc. [7] while the decrease in dielectric constant with frequency is due to the fact that all the above contributing factor to polarizability lag behind the applied field at higher frequencies.

The dielectric loss (ε″) as a function of frequency for all samples is shown in Fig. 3. It can be seen that all the samples show similar behavior, i.e. dielectric loss decreases with increase in frequency and reaches a constant value at higher frequency. After certain increase in frequency all the samples exhibit frequency independent behavior which can be explained

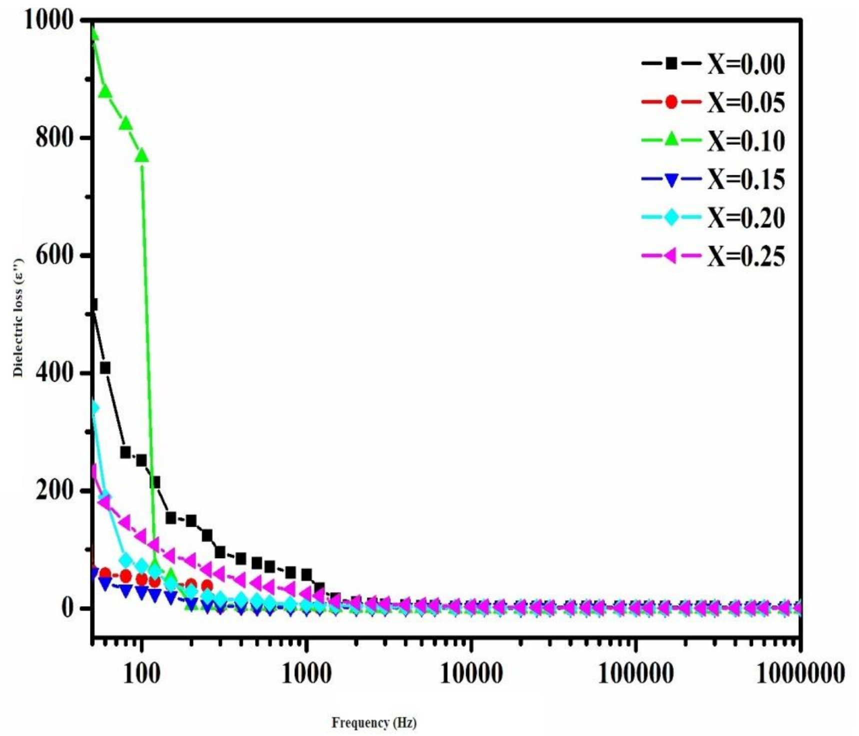

Fig.3: Variation of dielectric loss with frequency.

On the basis of Maxwell–Wagner interfacial type polarization, this is in agreement with Koop's phenomenological theory [8]. The large values of dielectric loss at low frequencies are observed in case of ferrites are due to the predominance of the species like Fe^{2+}ions, interfacial dislocation piles up, heterogeneity, oxygen vacancies, grain boundary defects etc [9].

Fig. 4 shows the dielectric loss tangent as a function of frequency of all samples at room temperature within the frequency range of 100 Hz–1MHz. With increase in frequency the dielectric loss tangent exhibited a decreasing trend up to a particular frequency then it remains constant at higher frequencies. It is known that, there is a strong correlation between the conduction mechanism and dielectric constant behavior in ferrites, for these two considerations it can be observed that the behavior of loss tangent is showing the expected decrease with frequency. The dielectric losses in ferrites are generally reflected in the resistivity i.e., materials with low resistivity exhibit high dielectric loss and vice versa. Similar type of variation is observed in the present study. However no systematic effect of Ni content on dielectric loss tangent was observed.

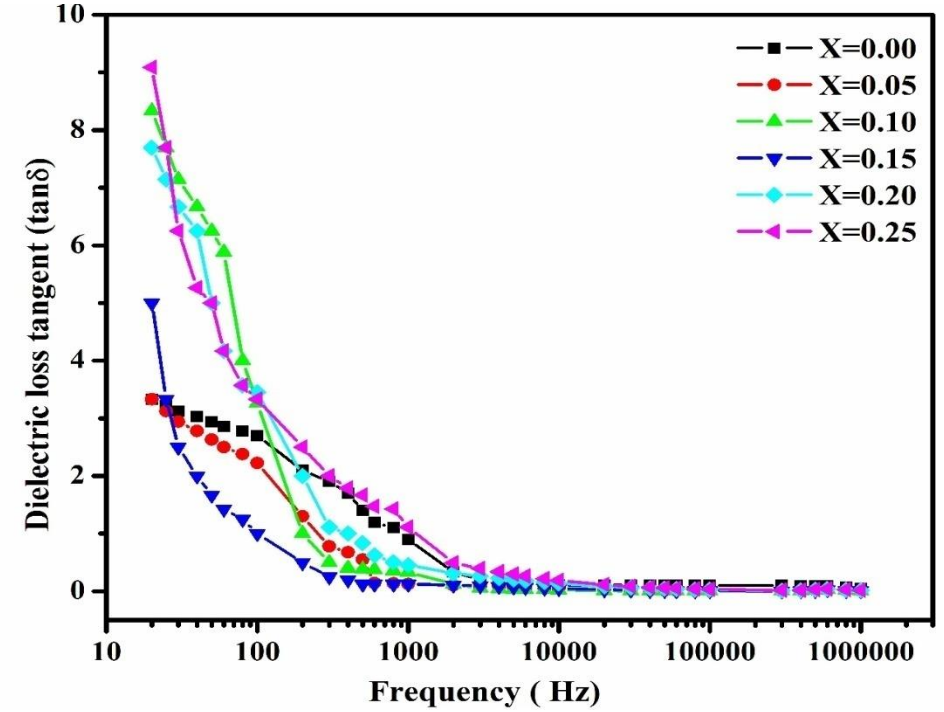

Fig.4: Variation of dielectric loss tangent with frequency

The AC resistivity of ferrites gives primary information related to the localization of charge carriers at grain and grain boundaries, inter-granular tunneling of charge carriers across the grain boundary and dielectric polarization of magnetic ions [10].The variation of ac resistivity for all samples is shown in Fig. 5. Decrease in ac resistivity with increase in frequency was observed for all samples. No prominent influence of Ni addition on room temperature ac resistivity was observed. The ac resistivity behavior in the present study is useful for high frequency applications. At lower frequency, the grain boundaries are more active, hence the hopping frequency of electrons between Fe^{3+} and Fe^{2+} ions is less.

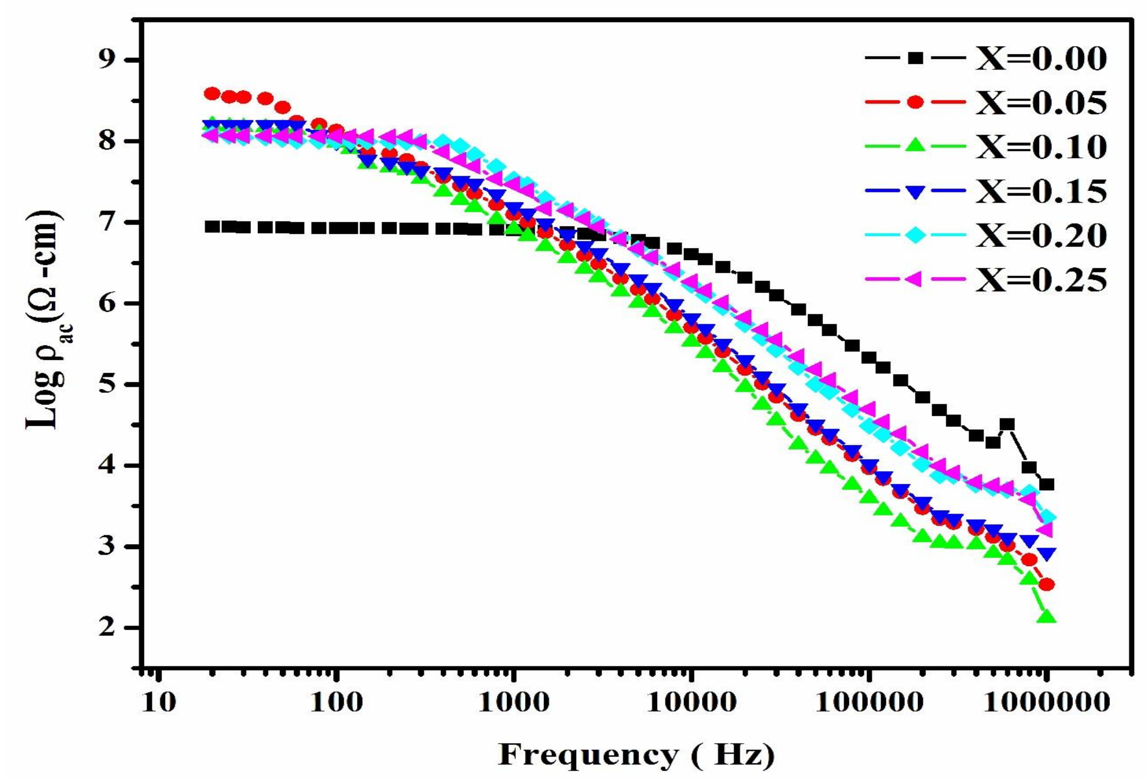

Fig .5: Variation of ac resistivity for all samples.

At higher frequencies, the conductive grains become more active by promoting the hopping of electrons between Fe^{3+} and Fe^{2+} ions therefore increasing the hopping frequency [11]. So we observe the decrease in resistivity with the increase in frequency. The linearity of the plots is attributed to small polaron type conduction [12].

Conclusions:

$Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_{4}$ ferrites were synthesis by citrate gel combustion method. The temperature dependent D.C. electrical resistivity decreases with increases in temperature nature of all samples semiconductor behavior. Curie temperature calculates from d.c. resistivity plots the temperature increases with increases in Ni content x.

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