

# STUDIES ON ELECTRICAL PROPERTIES OF TIN SUBSTITUTED COPPER FERRITE

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## Abstract:

A series of polycrystalline spinel ferrites with composition  $\text{Cu}_{1-x}\text{Sn}_x\text{Fe}_{2-2x}\text{O}_4$  where  $x = 0.0, .5, 1.0, 1.5, 2.0$  and  $3.0$  were prepared by standard ceramic method. The single phase cubic spinel structure of all samples has been confirmed by X ray diffraction analysis. The electrical transport properties like a c conductivity, thermoelectric power were studied. The measurements were carried out from room temperature 300K to well beyond Curie temperature by two probe method. From these studies, it is observed that all samples with  $\text{Sn}^{4+}$  show a cusp like minimum. The samples with  $x = 0.05, 0.2$  and  $0.3$  exhibit n-type conduction within the entire range of temperatures. The compositions  $x = 0.1$  and  $0.15$  initially show p-type conduction up to 425 K while the conduction is n-type above 475 K.

Compositional and temperature dependence of  $\alpha$  in the present ferrite system has been discussed. On the basis of these results a conduction mechanism for Cu-Sn ferrite system is suggested in different temperature region.

The variation of ac conductivity with temperature was studied from these studies it can be seen that the  $\sigma_{ac}$  increase slowly up to 350 k, and then the frequency dependence increases rapidly. The conductivity is found to be high for higher frequency.

**Key Words:** Ferrites, Spinel, X ray diffraction, Thermoelectric Power, Seeback Coefficient.

## 1. Introduction

Many physical and chemical properties of solids can be understood from the measurement of electrical conductivity, thermoelectric power, Hall coefficient and magnetoresistance. Ferrites exhibit both semiconducting and magnetic properties, and could be used in electronic devices. Ferrites as a class of materials show a very wide range of resistivity from 10 ohm cm (magnetite) to 10 M ohm cm (magnesium ferrite) [1]. The low electrical conductivity of ferrites in comparison with that of

magnetic metals or alloys has been the main aspect that makes them less lossy and therefore superior in magnetic applications. For applications at microwave frequencies, ferrites with conductivity lower than  $10^{-8}$  ohm<sup>-1</sup>cm are required since the dielectric loss tangent depends on conductivity [2]. With suitable preparation techniques and chemical compositions, sufficient control of the conductivity of the ferrites [3], allows tailor making of these for various applications.

The d.c. and a.c. conductivity studies provide valuable information about the conduction mechanism in ferrites. The electrical properties mainly depend on the method of preparation, presence of impurities, amount of iron present in the lattice in ferrous state, which can be investigated by carrying out the above said experiments. The conduction in ferrites has been explained as due to exchange of 3d electrons, localized at the metal ions from  $\text{Fe}^{3+}\text{-Fe}^{2+}$  [4]. Jonker [5] derived an expression for mobility from hopping conduction model, based on localized levels for electrons. Dogoade et al. [6] suggested Band polaron model based on electron transition between localized cells. Small polaron model has been introduced by Haubenreisser [7]. Lorentz and Thle [8] have explained the electrical properties on the basis of thermally activated motion of electrons and Srinivasan [9] has extended it to polaron induced tunneling. Verway et al. [10] have shown that the conduction can be increased by mixing a small amount of foreign oxides as impurities in high resistivity ferrites. Koops has studied the variation in the resistivity and dielectric constant, which fall by large amount at higher frequencies in inhomogeneous material. Jefferson [11] has also studied the conductivity and Seeback coefficient for copper containing ferrites. Thermoelectric properties are widely used in the interpretation of the conduction mechanism in semiconductors. In ferrites, for which mobility is low, thermoelectric power measurements play an important role in determining the type of charge carrier concentration. This helps in determination of

various regions of conduction viz. impurity conduction, impurity exhaustion, and intrinsic conduction regions of semiconductors. Nanba and Kabayashi and Rezlescu and Rezlescu [12,13] have reported unusual thermal magnetic and dielectric properties of  $\text{Cu}^{2+}$  ferrite and  $\text{Cu}^{2+}$  containing ferrites. In general the behaviour of these ferrites has been attributed to the Jahn-Teller distortion occurring under certain conditions due to  $\text{Cu}^{2+}$  ions in these ferrites. Phase transition from tetragonal to cubic for copper ferrite has been reported at certain critical temperature and composition by Otari et al. [14]. Abnormal behavior in magnetic, dielectric and thermal properties of Cu containing ferrites also has been studied by many workers [15,16]

## 2. EXPERIMENTAL

In order to measure the variation of Seebeck coefficient  $\alpha$  with temperature of the sample, the experimental set up consist of electrically heated furnace, a temperature controller, two digital d.c. micro voltmeter (VMV-15, Vasavi electronics), a specially designed sample holder and a digital multimeter. For the measurement of thermoelectric power, the samples in the form of pellets coated with thin layer of silver paste for good electrical contact were used. A pellet was put into the sample holder, with necessary connections the sample holder was put into the regulated furnace. For the measurement of thermoemf, the thermal gradient ( $\Delta T = 20$  K) across the sample was obtained with the help of micro furnace attached to one of the electrodes of the sample holder. The thermal gradient was measured by differential Chromel-Alumel thermocouple. The thermoemf ( $\Delta v$ ) developed across the pellet was measured on digital micro voltmeter by silver electrodes. Sufficient time was allowed after applying the thermal gradient, to obtain stabilization before recording the thermoemf. The sample was then heated by establishing a constant thermal gradient ( $\Delta T = 20$  K) and the developed thermoemf was measured at different ambient temperatures. The hot end of the sample is connected to positive terminal of micro voltmeter. If the micro voltmeter shows the +ve voltage, then the charge carrier is of n-type. This

situation is reverse when the charge carriers are of p-type. The thermoelectric power was determined by using the relation

## 3. RESULT AND DISCUSSION

Klinger and Samokhavalov [17] have discussed the charge transport in ferrites and have speculated about the relative importance of disorder and polaronic contribution to such properties as resistivity and thermoelectric power. Recently Klinger has explained the conduction mechanism in magnetic like solids using two-phase polaron model. V.R.K.Murthy and Sobhanandri [18] have reviewed the conduction phenomenon in ferrites. The temperature dependence Seebeck coefficient  $\alpha$  for  $\text{Cu}_{1-x}\text{Sn}_x\text{Fe}_{2-2x}\text{O}_4$  system is shown in fig.1 In  $\text{Sn}^{4+}$  containing series the samples with  $x = 0.1$  and  $0.15$  initially shows p-type conduction, make transition to n-type at  $425$  k  $475$  k respectively, and shows cusp at  $750$  k. All other samples exhibits n-type conduction from room temperature throughout the temperature range shows cusps at  $625$ - $725$  k. The sample  $x = 0.3$  shows small peak at  $650$  k and then the value of ( $\alpha$ ) remains constant for further increase in temperature. The observed variation in Seebeck coefficient ( $\alpha$ ) with temperature obviously indicates the conduction process taking place in the simultaneous presence of acceptor and donor centers with different relative predominance. It has been already reported in the literature that copper ferrite exhibited p-type conduction attributed to the appearance of the p-carriers due to reduction of the  $\text{Cu}^{2+}$  ions to  $\text{Cu}^{1+}$  at about  $950$  °C during sintering process. The presence of certain quantity of  $\text{Cu}^{1+}$  exhibiting p-conduction via hole on oxygen ion determines an important modification of electrical and magnetic properties of ferrites containing copper [19,20]. A cusp appears near the Curie temperature. This has been explained by assuming two important conduction processes to be considered for  $\text{CuFe}_2\text{O}_4$  at lower temperature [16]-

$$\text{Cu}_A^{1+} + \text{Cu}_A^{2+} \rightarrow \text{Cu}_A^{2+} + \text{Cu}_A^{1+} \quad (1)$$

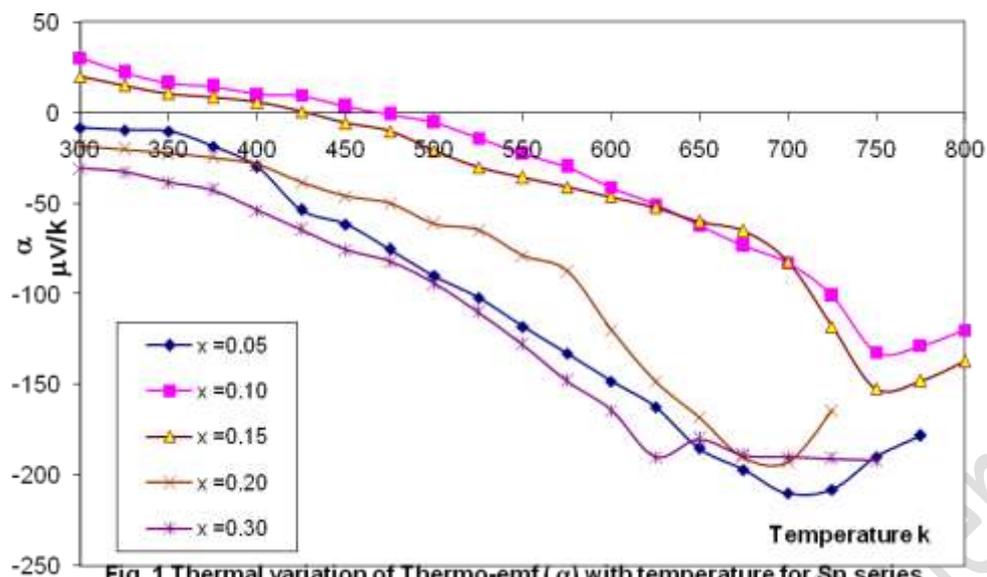


Fig. 1 Thermal variation of Thermo-emf ( α) with temperature for Sn series.

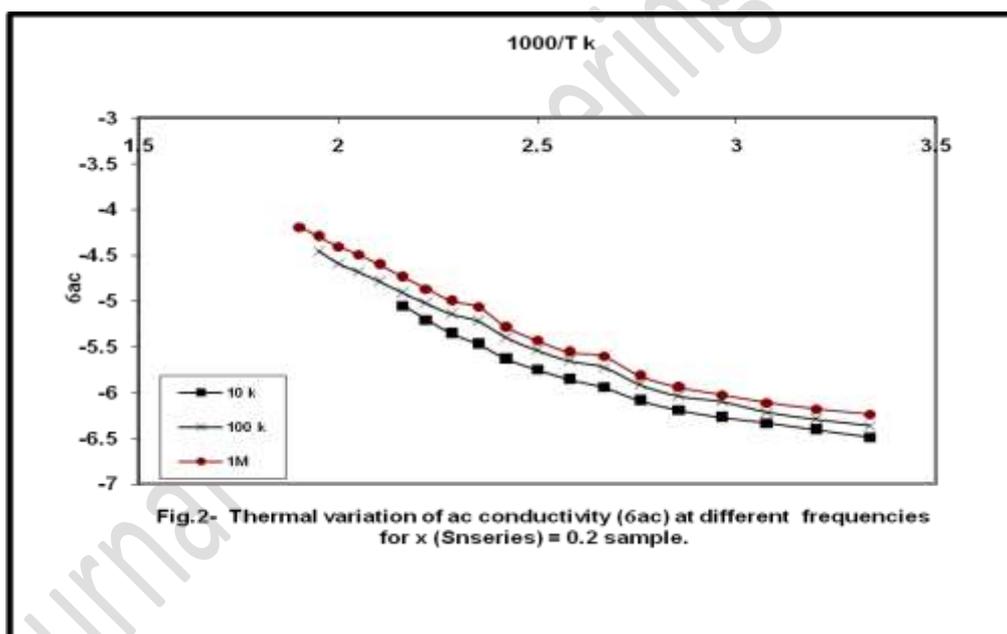
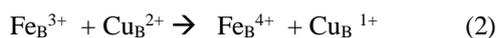
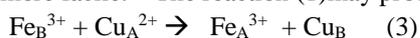


Fig.2- Thermal variation of ac conductivity (σac) at different frequencies for x (Snseries) = 0.2 sample.



The mixed valance, small polaron reaction (1) gives only p-type conduction depending on the relative concentration of  $\text{Cu}^{1+}$  and  $\text{Cu}^{2+}$  ions at A-site. Seebeck coefficient data for the present system shows p-type conduction. Therefore, the predominant conduction may be due to reaction (1). Formation of both types of centers ( $\text{Cu}^{1+}, \text{Fe}^{2+}$ ) may result from the loss of oxygen during sintering. The transition from p-

type to n-type can be explained as above. At certain temperature, the intersite cation exchange reaction is more facile. The reaction (1) may be produced as



and reaction (3) gives n-type conduction. All samples in this series show a cusp like minima near the Curie temperature. The samples  $x = 0.05, 0.2, 0.3$  exhibit n-type conduction from whole range of temperature. The compositions  $x = 0.1$  and  $0.15$  initially show p-type conduction up to 450 K and n-type above 450 K.

The results result show that the majority of charge carriers are electrons for all samples.

The plots of a.c. conductivity with temperature for  $\text{Cu}_{1.2}\text{Sn}_{0.2}\text{Fe}_{1.6}\text{O}_4$  are shown in fig 2. From these plots it can be seen that the  $\sigma_{ac}$  increase slowly up to 350 K, and then the frequency dependence increases rapidly. The conductivity is found to be high for higher frequency. This conduction can be attributed to localized charge carriers. According to localized model the electronics are strongly localized on cations. Theoretical work by several workers over the years has provided some understanding of conduction in oxides and transition metal compounds. For these materials, the interaction between electrons and optical phonons is strong and conduction is explained on the basis of polarons. The treatment of conduction by polarons is discussed by several workers. Polarons belongs to two categories, large and small polarons. In the large polaron model, the conductivity is by band mechanism at all temperatures and the a.c. conductivity decreases with frequency. The small polaron conduct in band like manner up to a certain temperature, the conductivity shows an increase with frequency. At higher temperature, the conduction is by thermally activated hopping mechanism. The localization may be attributed to electron-phonon interaction or strong magnetic interaction between carriers and magnetic sub – lattice. An additional localization  $\text{Fe}^{2+}$  ions may arise from inhomogeneous

distribution of ions over octahedral and tetrahedral sites in spinel lattice' the experimental results in present case also show a trend expected for small polaron conduction.

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