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Electrical and dielectric behaviour of chromium substituted nickel ferrite

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ABSTRACT

The variation of resistivity and thermo emf with temperature for the ferrites with $NiCr_xFe_{2-x}O_4$ has been studied. The conduction process in the first region < 450 K is attributed to impurity conduction while for second region > 450 K due to polarons. The samples show p-type and p-n transition takes place at 450 – 500 K. Abnormal dielectric behavior is observed for the samples with x = 0.8 and 1.0 at 10 MHz and 2 MHz respectively. This is due to increase of P-type carriers.

INTRODUCTION

Nickel ferrites are considered among the most versatile ferrite families. They have found widespread applications in microwave devices because of their high electrical resistivity, low eddy current and dielectric loss. A number of investigators have studied the influence of divalent and tetravalent substitution on the various properties of Nickel ferrites [1]. Electrical properties of these ferrites are of great interest to many workers [2]. Discussion on the electrical properties of ferrites is usually based on the band structure or carrier hopping model. However, until now no conclusive theory has been formulated for the conduction mechanism in ferrites. In this paper, we report the electrical resistivity and thermoelectric power as a function of temperature of Cr substituted $NiFe_2O_4$. The dielectric behavior in the frequency range of 1 KHz to 10 MHz is also studied to supplement the understanding of conduction phenomenon in these samples.

MATERIALS AND METHODS

Samples of the system $NiCr_xFe_{2-x}O_4$ (where x varies from 0.2 to 1 in steps of 0.2) were prepared by usual ceramic method. The completion of solid state reaction and confirmation of spinel phase was checked by X-ray diffraction [3]. The variation of resistivity (ρ) with temperature in the range of 300 to 700 K was measured by two probe method. For better ohmic contact silver paste was applied on both surfaces of the pellet. Seebeck coefficient (α) was measured in the same range of temperature by keeping the temperature difference of about 15 K across the pellet. The dielectric measurement was carried out on the pellet in the frequency range of 1 KHz to 10 MHz with the help of HP 4192 LF impedance analyzer.

RESULTS AND DISCUSSION

The variation of log ρ versus 1/T is shown in Fig. 1. Ferrites are semiconductors; they obey the relation $\rho = \rho_o \exp(-\Delta E/kT)$. The plots display the linear relationship with changing slopes. The activation energies calculated by using this relation are given in Table 1. These regions are attributed to the change in conductivity mechanism from one type to the other.

The variation of Seebeck coefficient (α) with temperature is shown in Fig. 2. Initially at lower temperature all the samples show p type behavior and then p to n transition takes place at about 450-500 ^oK. The values of thermo emf at room temperature increases with increasing ‘x’. This indicates that more and more p-type carriers are generated with Cr content.

Numbers of workers have observed three regions in ferrites [4, 5]. According to their analyses conduction in first region is due to impurities (extrinsic type), in second due to polaron hopping (intrinsic type), and in third it is due to magnetic ordering. According to Verwey et al [6] the conduction mechanism in ferrites is due to exchange of electrons from cations in the same octahedral sites in the lattice. In this case conduction can possibly be attributed to change of electrons between $Fe^{2+} \rightarrow Fe^{3+}$, $Ni^{2+} \rightarrow Ni^{3+}$. These ions, along with oxygen valancies, may be formed during the sintering process [7]. The conduction due to $Ni^{2+} \rightarrow Ni^{3+}$ gives p-type character and $Fe^{2+} \rightarrow Fe^{3+}$ gives n-type character. In the present system Fe^{3+} is replaced by Cr^{3+} which ultimately reduces the Fe^{2+} formation. Therefore observation of p-type behavior at room temperature may be due to formation $Ni^{2+} \rightarrow Ni^{3+}$. The activation energy in region-I i.e. 450^o K is about 0.2 eV. The conduction in this region may be attributed to the presence of impurities.

In this region where the conduction is due to one kind of charge carriers say holes, the relation between Seebeck coefficient (α) and Fermi energy (E_F) is given by

$$E_F = e\,\alpha\,T - AkT$$

Where A is the term related to the kinetic energy of the free hole. The calculated values of E_F as a function of temperature for A = 0 and A = 2 are plotted in Fig. (3) for the sample x = 0.6. The values of E_F extrapolated to absolute zero, Fig. (3) for the sample x = 0.6. The values of E_F extrapolated to absolute zero, i.e. $E_F(O)$, are noted in the Table 1. These values agree well with ΔE_1 values obtained from resistivity at low temperature region Table 1. This point out that impurity conduction plays an important role in this region.

In ferrites, the transport properties have been explained on the basis of polaron model just as in the case of transition metal oxides [8] Kilinger et al have reviewed the charge transport phenomenon in ferrites. Accordingly, conduction by hopping process is taken over by polarons which are characterized by large effective mass and low mobility carriers. The temperature dependence of electrical conductivity in such cases involves less temperature dependant concentration of carriers and is mostly associated with temperature dependant mobility. For the present study it is noted that the activation energies (ΔE_2) in the region of > 450 ^oK are greater than 0.7 eV which is much higher than the energy required for hopping, (0.2 eV). Therefore, the conduction due to polarons is favoured in this region. The mobilities are calculated for these samples by using resistivity and thermo-emf values. The activation energy from mobility ($\Delta E\mu$) is noted in the same Table 1. From the Table 1 it is observed that ΔE_2 and $\Delta E\mu$ are same which itself confirms the conduction process in this region is due to polarons. Therefore it is reasonably concluded that conduction in the region < 450 K is due to impurities whereas it is due to polarons for region > 450 K. The variation of dielectric constant with frequency is shown in Fig. 4. From the plots it is observed that the dielectric constant decreases rapidly at lower frequencies and remains steady for higher frequencies remains steady at higher frequencies. The similar behavior is observed for other ferrites [9,11]. The dielectric behavior can be explained on the basis that the mechanism of polarization process in ferrites is similar to that of conduction process.

The dielectric constant ($\in'$) decreases continuously with increase in frequency which is natural because any species contributing polarizability is bound to show lagging behind the field at higher and higher frequencies. The sensitivity of the spices like Fe^{2+}, Ni^{3+}, oxygen, grain boundary, pores to the composition and frequency would give rise to large dispersion at lower frequencies. As the ionic polarization lags behind, only the electronic part gets reflected in$\in'$ at higher frequencies showing constancy at larger frequencies.

The dielectric loss tanδ with frequency is shown in Fig. 5. The samples with x = 0.8 and 1.00 show peak at 10 MHz and 2.0 MHz respectively. An abnormal behavior is observed in Ni ferrite [10]. The abnormal dielectric behavior in ferrites is due to a collective contribution of p-and n-type carriers to the polarization [12,13]. Therefore, local displacement of p-type carriers takes part in the polarization in an opposite direction to that of an external field. In addition, the mobility of p-type carriers is lower than that of n-type carriers and their contribution to polarization decrease more rapidly at lower frequencies. Therefore the position of peak depends upon majority of charge carriers. In the present system the value of α increases with increase in Cr content and all the samples are p-type at room temperature. Therefore increases in p-type carriers decreases the maximum frequency to lower frequency side, hence the observed behavior.

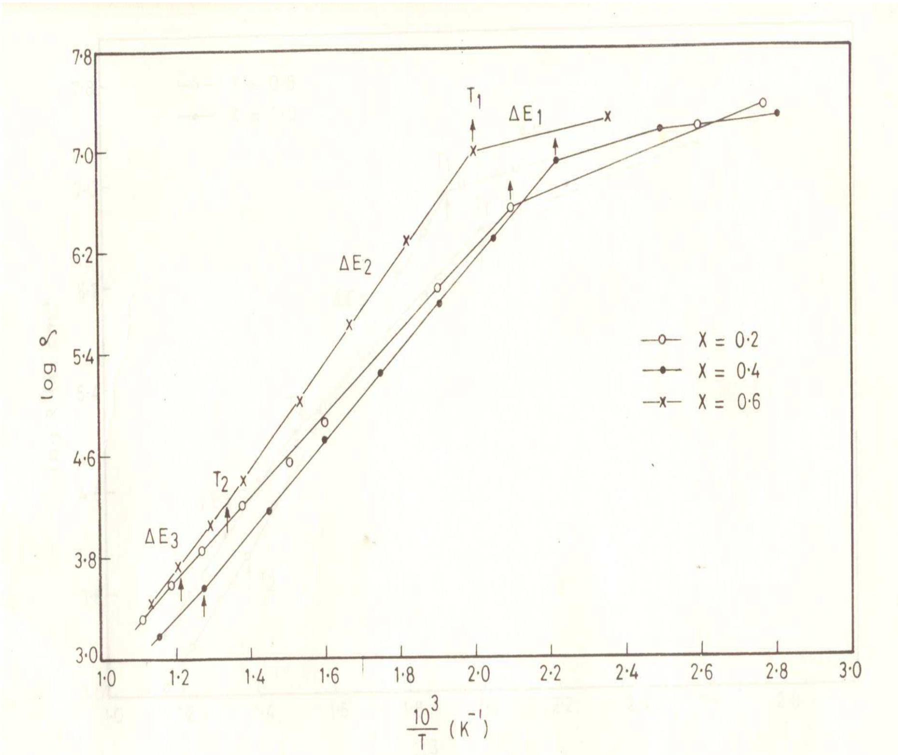

Fig 1- Variation of Log ρ with 1/T for $NiFe_{2-x}Cr_xO_4$

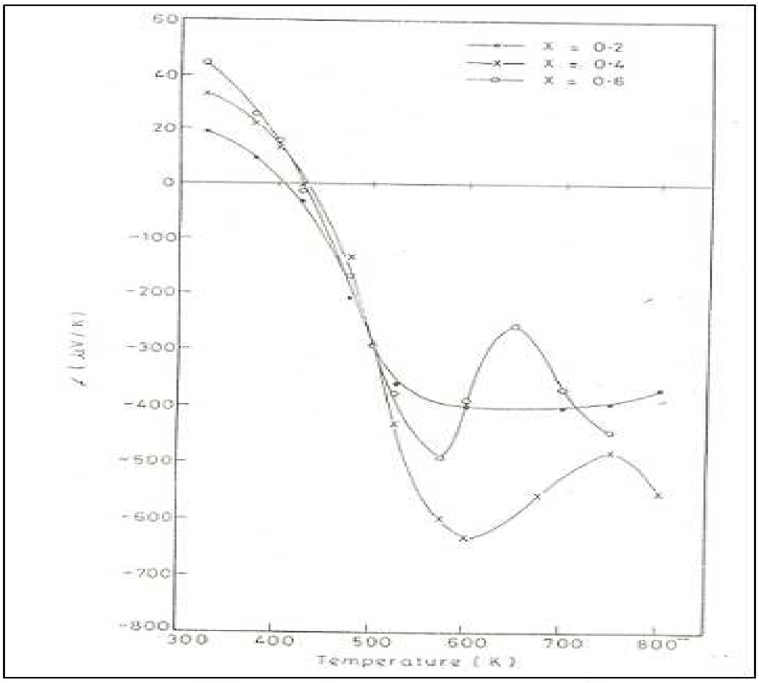

Fig 2: Temperature variation of thermoelectric power for $NiCr_xFe_{2-x}O_4$

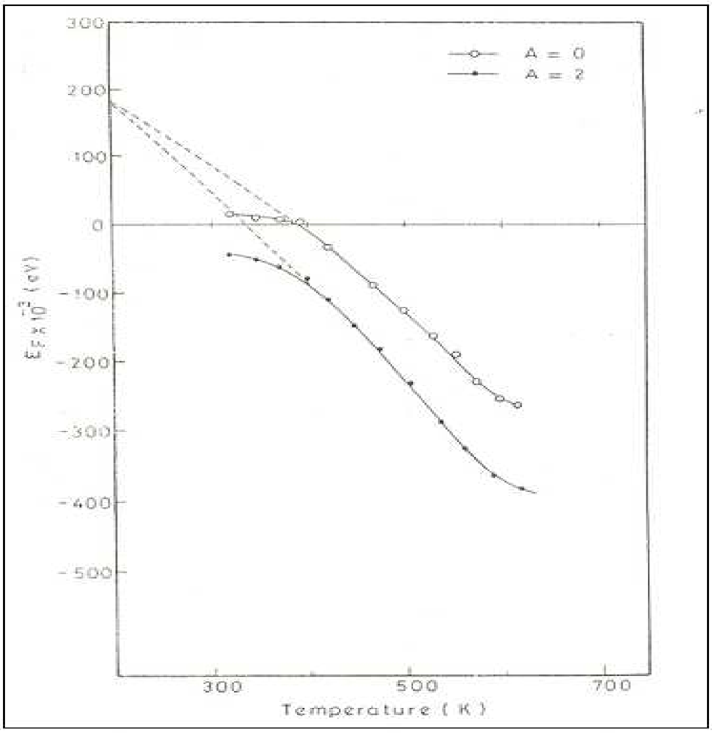

Fig.3: Fermi energy (E_F) as a function of temperature calculated from the Seebeck coefficient (α) for x = 0.6

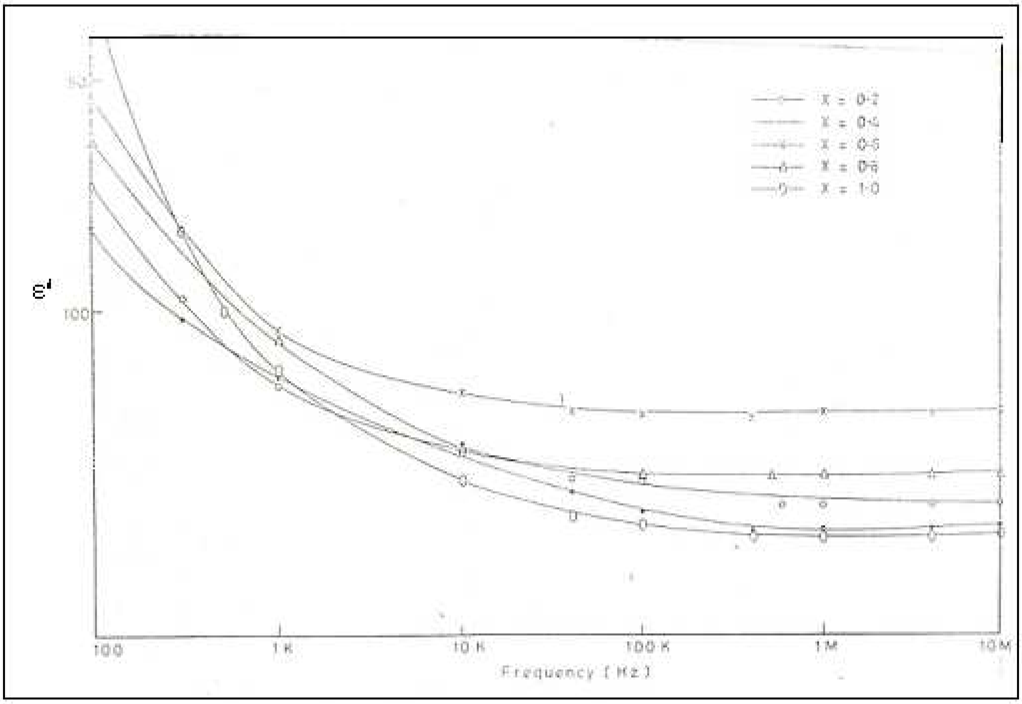

Fig.4: Variation of dielectric constant (∈') with Frequency for $NiCr_xFe_{2-x}O_4$ system

Table 1: Values of activation energy (ΔE), transition temperature (T), for $NiCr_xFe_{2-x}O_4$ system

Content x	Activation energy (eV)				Transition Temperature T_1(k)
	From Resistivity		From Mobility ΔEμ	From Band Theory E_F(O)	
	ΔE_1	ΔE_2			
0.2	0.22	0.86	0.86	0.20	473
0.4	0.20	0.70	0.72	0.21	448
0.6	0.16	0.76	0.83	0.19	498
0.8	0.15	0.80	0.82	----	510
1.0	0.15	0.65	0.54	0.13	498

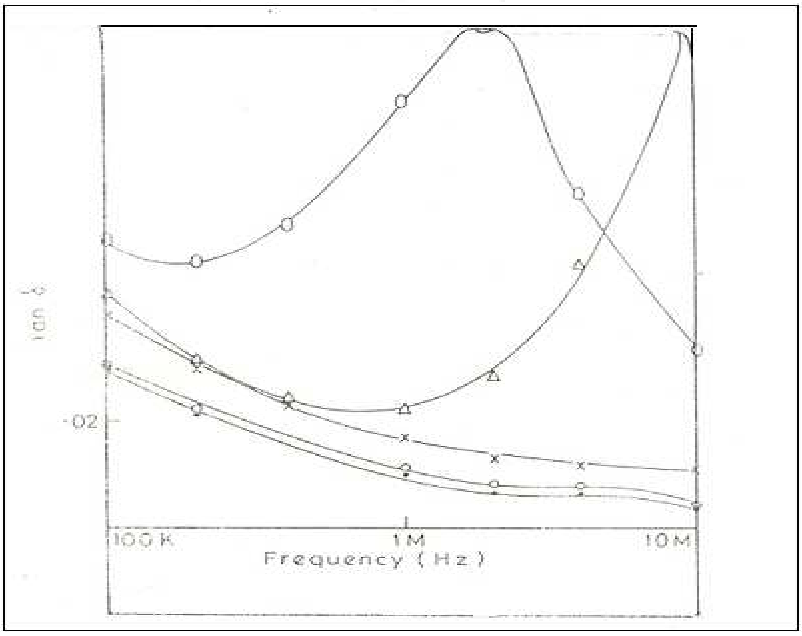

Fig 5: Variation of dielectric loss tangent (tan δ) with frequency

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