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Thermo Electrical Power Studies of Nickel-Zinc Ferrites Synthesized By Citrate Gel Technique

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Abstract
The magnetic particles of nickel-zinc ferrite with chemical composition $Ni_{1-X}Zn_XFe_2O_4$ where (x= 0.0, 0.2, 0.4, 0.6, 0.8, 0.9 & 1.0) were synthesized successfully by citrate gel auto-combustion method using high purity nitrates and citric acid as chelating agent. The prepared powder of nickel –zinc ferrites was sintered at 1000^0C for 1 hr to obtain good crystalline phase and was used for further study. The X-ray diffraction technique was employed to confirm the single phase formation of nickel ferrite. The X-ray diffraction pattern shows the Bragg's peak which belongs to cubic spinel structure. The Seebeck coefficient of the samples was obtained from thermo e.m.f measurements by keeping constant temperature difference of 10 K across the sample. The Seebeck coefficient of the sample found to be negative showing majority charge carriers are electrons. The value of charge carrer concentration has also been computed from the observed values of Sebeck coefficient. The variation of thermo electric power and charge carriers concentration has been discussed as a function of composition and temperature. On the basis of these results an explanation for the conduction mechanism in the ferrite is suggested

Keywords: Nickel –Zinc ferrite, lattice parameter, X-ray density, bulk density, porosity, Seebeck coefficient, carrier concentration.

I. Introduction

Spinel ferrites have high electrical resistivity and low eddy current loss at high frequency and they are widely used for cores of high frequency electromagnetic devices [1 2]. Thermo electrical properties are very useful for understanding the conduction mechanism in case of magnetic materials such as ferrites. These properties depend upon the method of preparation technique, chemical composition, substation of doping and distribution of cations among tetrahedral (A) and octahedral sites (B). As ferrites have low conductivity this is one of the reason for their major applications [3 4]. Particularly no information is available on thermo electric power studies of Ni-Zn ferrites prepared by citrate gel technique in the literature. Moreover, there is a need for a thorough study of thermo electric power studies Ni-Zn ferrites. In the present study we are reporting the results of thermo electrical power parameters like Seebeck coefficient and charge carrier concentration when non magnetic zinc doped to nickel ferrite.

II. Experimental

Seven different compositions of mixed Ni-Zn ferrite with chemical composition of $Ni_{1-x}Zn_xFe_2O_4$ with X=0.0, 0.2, 0.4, 0.6, 0.8, 0.9 and 1.0 are synthesized by citrate gel method, which is a chemical route. The citrate process [5, 6] is easy technique with minimum experimental set up and capable of producing homogenous mixture of constituent ions. It is a green and simple method, which cans offers significant in saving time and energy.

The raw materials used in this preparation are nickel nitrate(Merc, India), zinc nitrate (Merc, India), iron (III) citrate (Merc, India), citric acid (Merc India) and ammonia (AR grade).These chemicals were weighed according to the required stoichiometric proportion. Initially Iron (III) citrate is dissolved in distilled water heated to 40^0 C with constant stirring. Nickel nitrate, zinc nitrate and citric acid were dissolved in distilled water separately and stirred for about 30 minutes by using magnetic stirrer until to get clear solution .These solutions are added to iron citrate solution under continuous stirring and ammonia is added drop by drop to the resultant solution until to get pH=7. Finally this mixture was heated slowly to evaporate the water; by increasing the temperature up to 200^OC self ignitation takes place results a homogenous uniformly brown colored powder which is the desired Ni-Zn ferrite. This powder is subjected to calcination using microwave scintillation furnace at 1000^0C for 30 minutes. Afterwards the powder was pressed into pellets of thickness 3 mm and a diameter of 10 mm with press by applying a pressure of 2 tons/in2. The final sintering was done at 1000°C, the pellets were coated with silver paint for better electrical contact to measure the thermo electrical power studies properties.

The X – ray diffraction patterns of the ferrite powder was taken on a Rigaku Dmax-II diffractometer.

Thermo e.m.f in the Ni-Zn ferrite can be determined by using hot probe method [7-8] the experimental method for this measurement is shown in the Fig 1. In this method ferrite sample (C) is kept on metal base (D) acts as cold junction. A point of contact probe is kept on the surface of the sample which acts as hot junction (A). To provide good electrical contacts with the base, stainless steel foil is provided at bottom of the sample. With help of electrical heater provided on the probe, the temperature of the probe (A) is increased to maximum of temperature $200^{O}C$. The thermo e.m.f developed across the sample due to stainless steel foil was also taken into consideration.

III. Results and Discussions

The x-ray diffractogram of all zinc substitute nickel ferrites of the various com positions shown in the fig. 2. The sharp peaks showed all-crystalline nature of single phase ferrite. The lattice parameter of individual composition was calculated by using the formula

$$d = a(h^2+k^2+l^2)^{1/2} \quad \text{-------------1}$$

Where,

a = lattice constant;

d = inter planar distance;

(*h*, *k*, l) = the Miller indices.

The variation of lattice parameter with zinc composition is shown in **Fig 3**. The lattice parameter is found vary linearly with increasing zinc concentration, there by indicating that the Ni-Zn ferrite system obeys Vegard's law [9]. The variation in lattice constant with zinc content can be explained on the basis of the ionic radii of Zn^{2+} (0.82 Å) ions is higher than that of Ni^{2+} (0.78 Å)

The thermo e.m.f of the samples is measured during cooling cycle because during cooling the samples attain thermal stable than heating.

Seebeck coefficient can be calculated by the relation

$$S = \frac{\Delta E}{\Delta T} \quad \text{-------------2}$$

Where,

'ΔE' is thermo e.m.f produced across the sample

'ΔT' is change in temperature.

The charge carrier concentration of the samples can be calculated by using Morin F.J, Gebella T H [10] relation with by using Seebeck coefficient and tabulated in the table 8.1

$$n = N \exp(-Se/K) \quad \text{-------------3}$$

Where

n = concentration of electrons

N= density of state factor or concentration of electronic levels in the conduction process

e = charge of electron

K= Boltzmann's constant

As ferrites are low mobility semi conductor then 'N' can be as 10^{22} cm^{-3} [11-13]

3.1. Variation of Seebeck coefficient with composition:

The values of Seebeck coefficient (S) at 310 K computed from the measured values of thermo e.m.f which are given in Table 1. It can be seen from the Table that the values of Seebeck coefficient (S) continuously decreases from –457to –203 µV/K up to zinc concentration X=0.9 and then increase to -393 µV/K for X=1.0. The variation of Seebeck coefficient with composition is show in the Fig 4

3.2. Variation of Seebeck coefficient with temperature:

The Seebeck coefficient as function of temperature is shown in the Fig 5. It can be observed from the graphs Seebeck coefficient is strongly -dependent on temperature. Seebeck coefficient decreases very rapidly, while beyond $150^{O}C$ the decrement in Seebeck coefficient is slow. This same behavior is observed by Kh. Roumaih [14].

It can be seen from the table that the sign of Seebeck coefficient is negative i.e all the ferrites under investigations are n-type semiconductor. The pre dominant conduction mechanism in Ni-Zn ferrite is due to hopping electrons from Fe^{2+} to Fe^{3+} ions [15-16]

$$Fe^{2+} + e^- \Longleftrightarrow Fe^{3+} \quad \text{---------4}$$

3.3. Variation of charge carrier concentration with temperature:

It can be seen from the graphs that the charge carrier concentration for all composition decreases gradually as shown in the figure 6, this same variation can be observed by [17]

3.4. Variation of charge carrier concentration with composition:

From the table 8.1, it can be observed that charge carrier concentration of the Ni-Zn ferrites are decreases gradually from X=0.0 to X=0.9 and then increase for X=1.0 composition

The variation of charge carrier concentration with composition is represented in the Fig 7

IV. Conclusions

It may be concluded that a series of Ni-Zn ferrite with compositional formula $Ni_{1-x}Zn_xFe_2O_4$ where *x* = 0.0, 0.2, 0.4, 0.6, 0.8, 0.9 & 1.0 are prepared by citrate gel method. The lattice constant was found to be increases with zinc composition. It is observed that, Seebeck coefficient decreases from -457 µV/K to -203 µV/K up to x=0.9 and then increases to -393 µV/K for x=1.0. As Seebeck coefficient is negative the ferrites under the observation is n-type semiconductors. The predominant conduction mechanism in Ni-Zn ferrite is due to hopping electrons from Fe^{2+} to Fe^{3+} ions and the charge carrier concentration decrease from 3.093 $X10^{22}$ cm^{-3} to 3.470 $X10^{22}$ cm^{-3} and then increases to 3.786 $X10^{22}$ cm^{-3} for x=1.0

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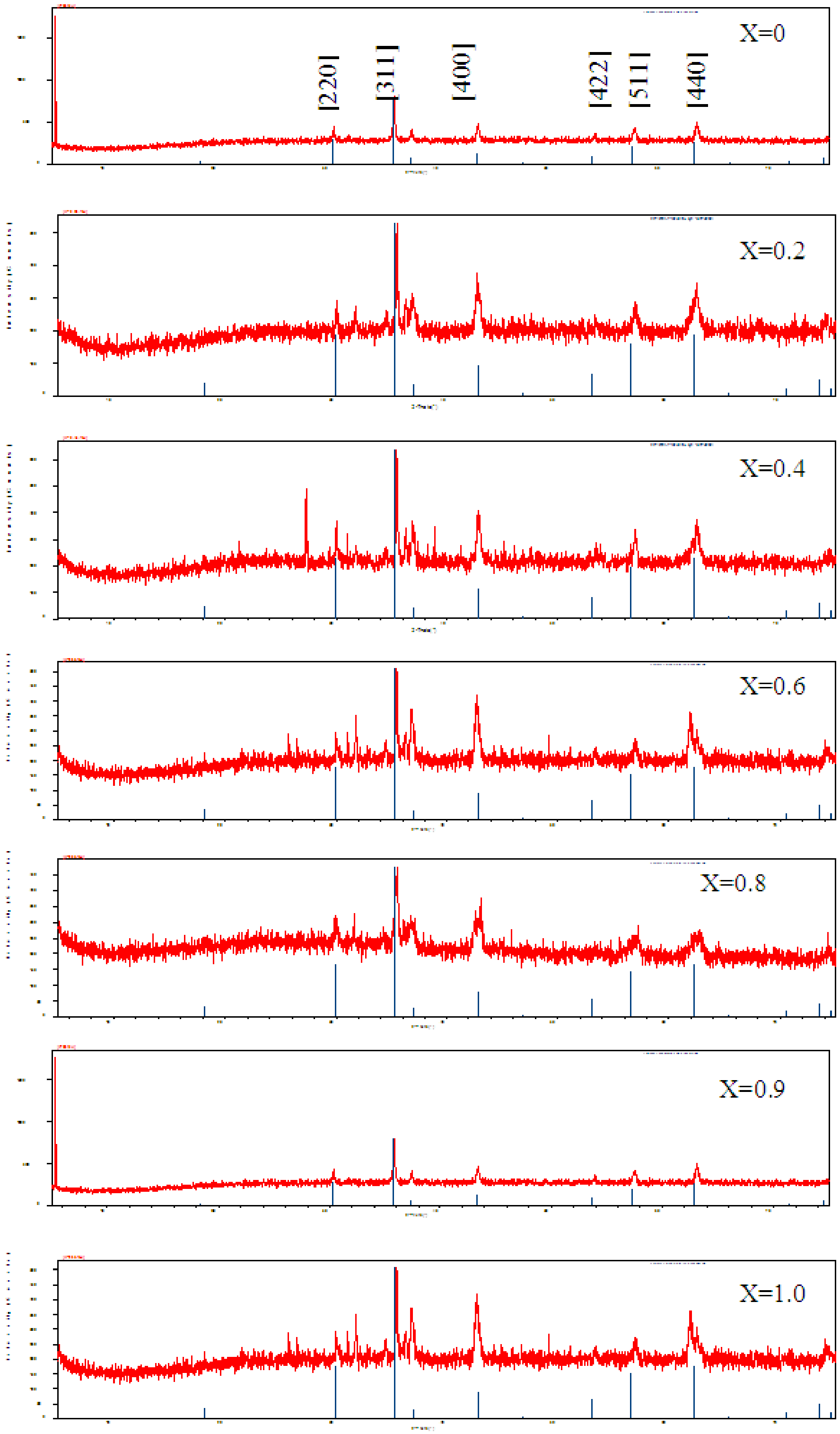

Fig.2 X-ray difractogram of Ni-Zn ferrites

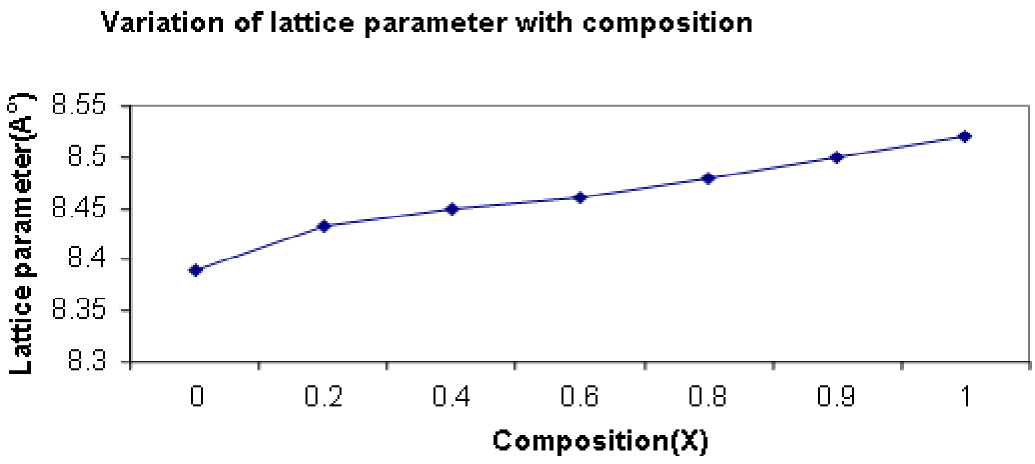

Fig 3. Variation of lattice parameter with composition

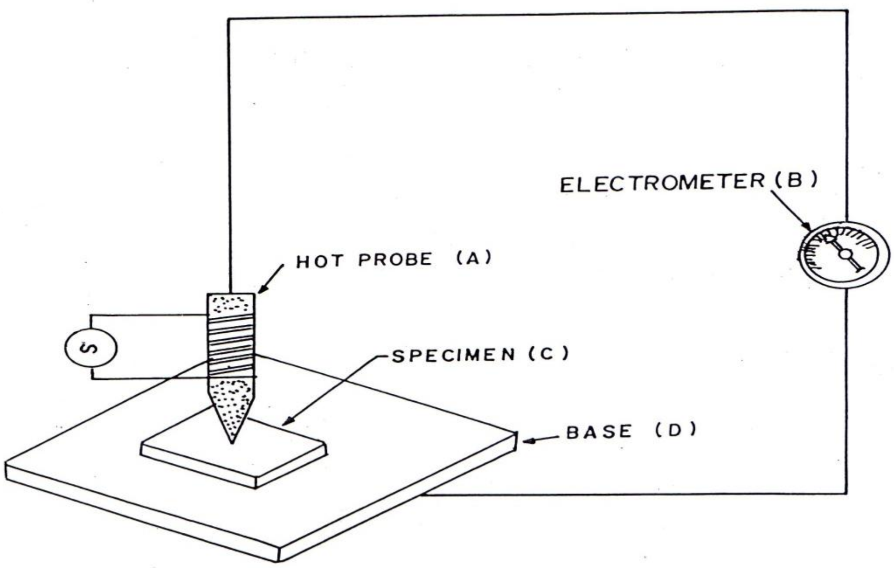

Fig. 1 Schematic diagram to find thermo electric power studies

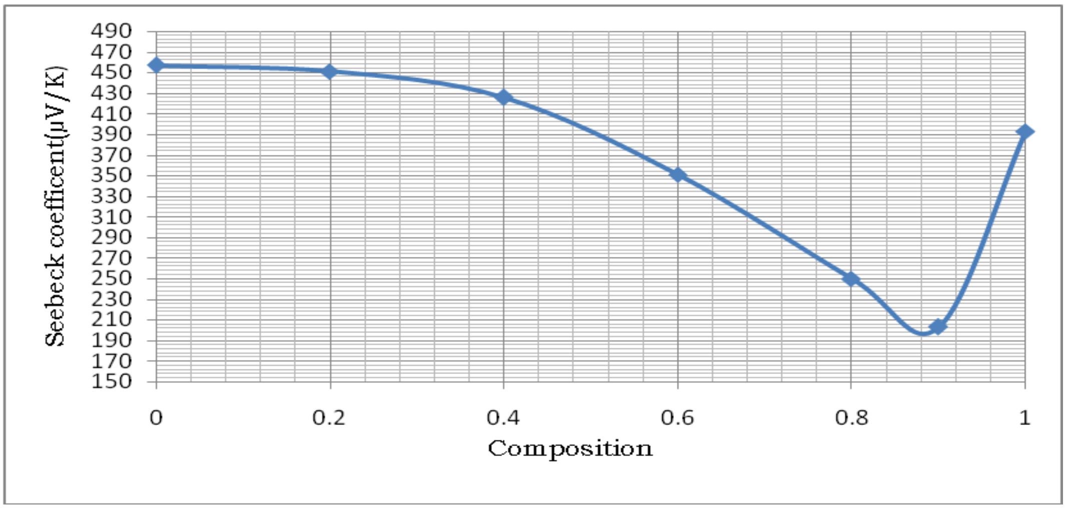

Fig. 4. Variation of Seebeck coefficient with composition

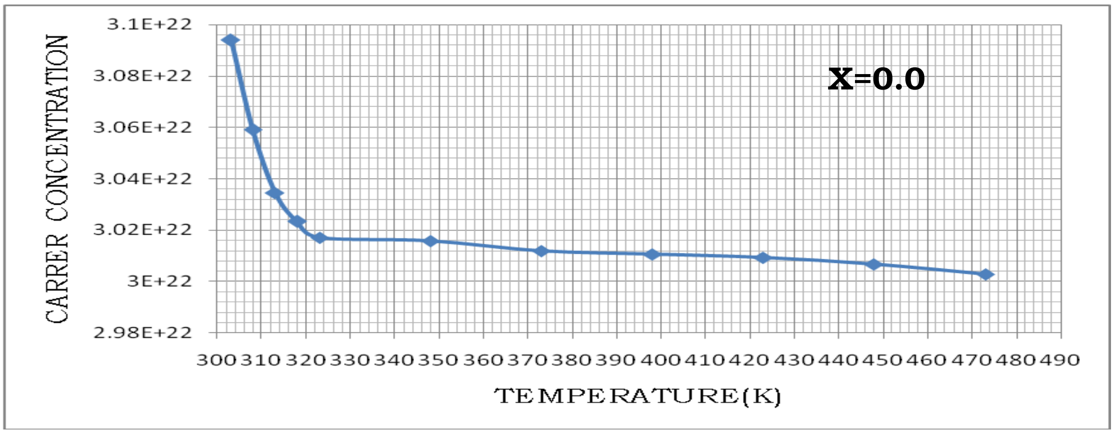

Fig 5 Variation of charge carrier concentration with temperature for X=0.0

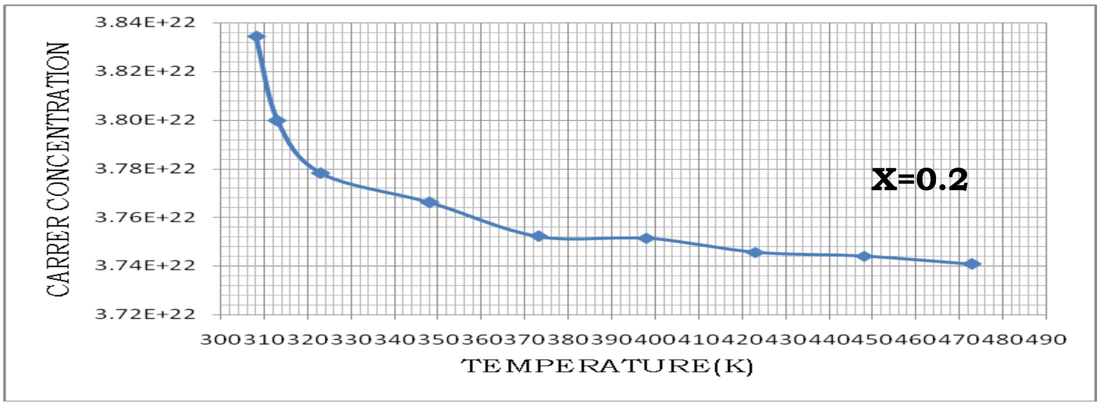

Fig 5 Variation of charge carrier concentration with temperature for X=0.2

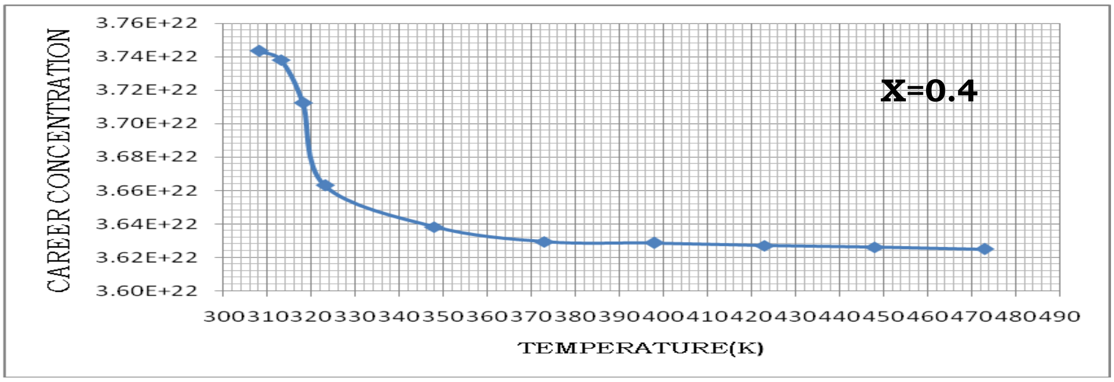

Fig 5 Variation of charge carrier concentration with temperature for X=0.4

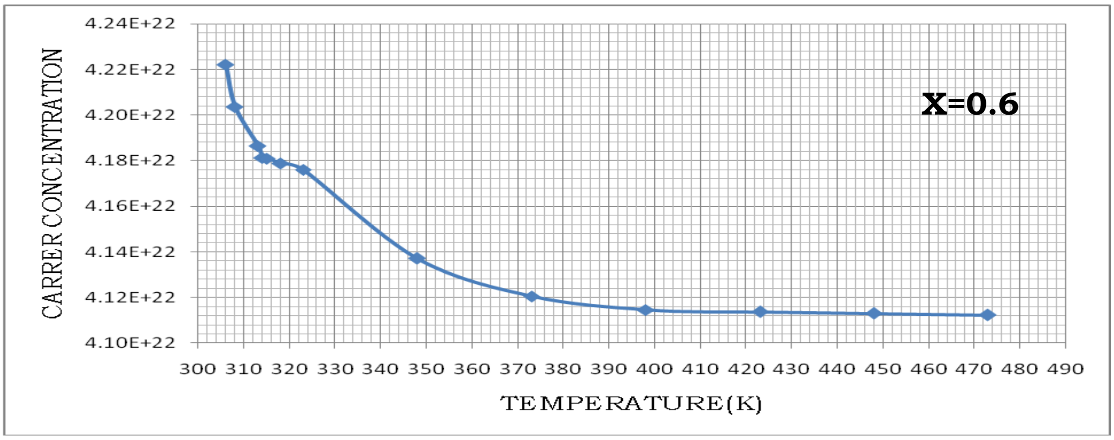

Fig 5 Variation of charge carrier concentration with temperature for X=0.6

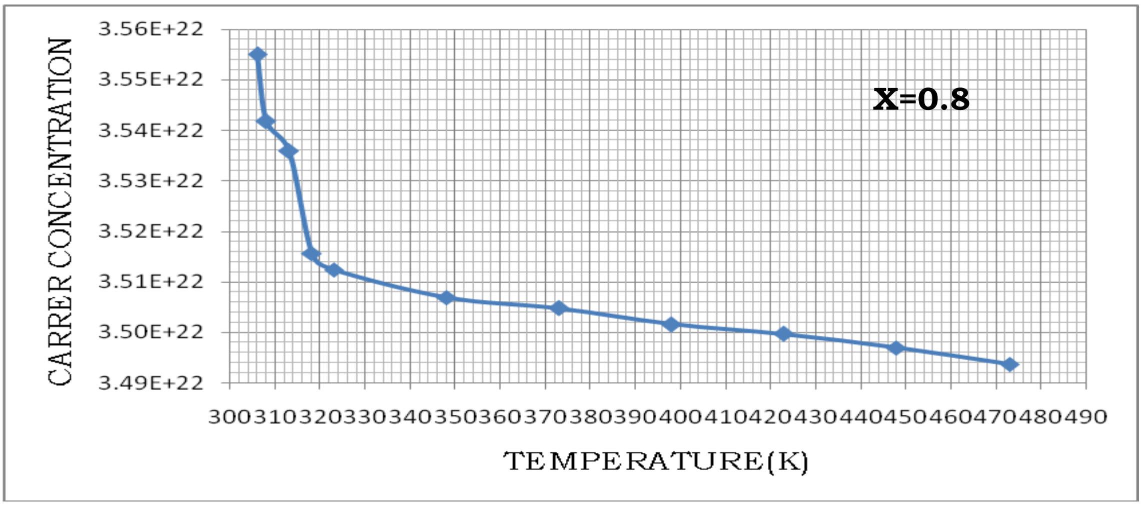

Fig 5 Variation of charge carrier concentration with temperature for X=0.8

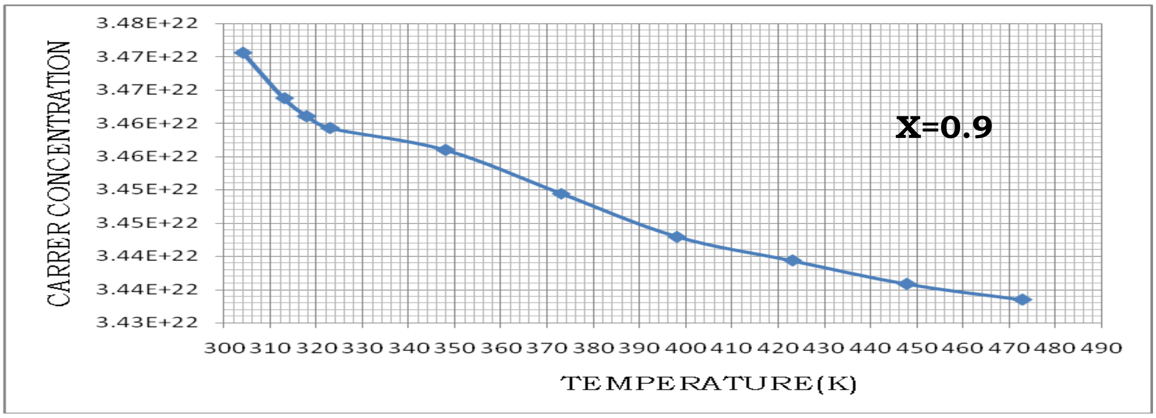

Fig 5 Variation of charge carrier concentration with temperature for X=0.9

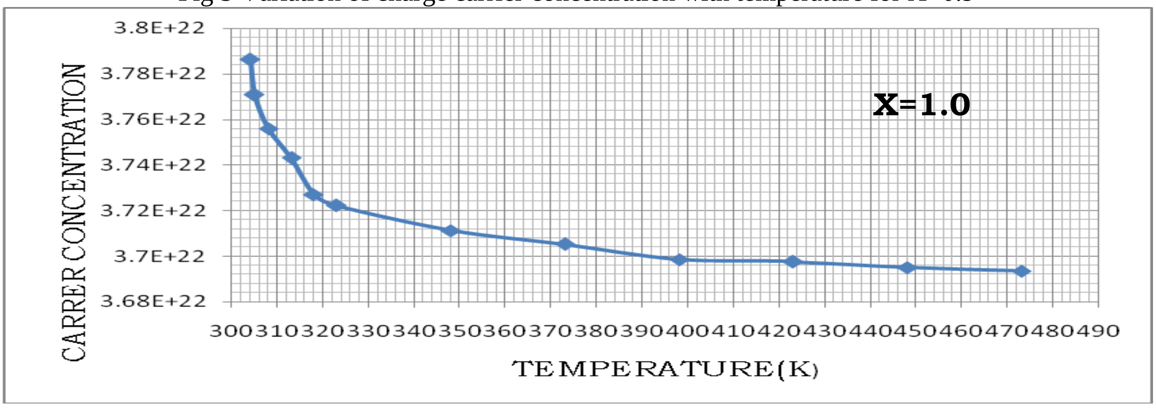

Fig 5 Variation of charge carrier concentration with temperature for X=1.0

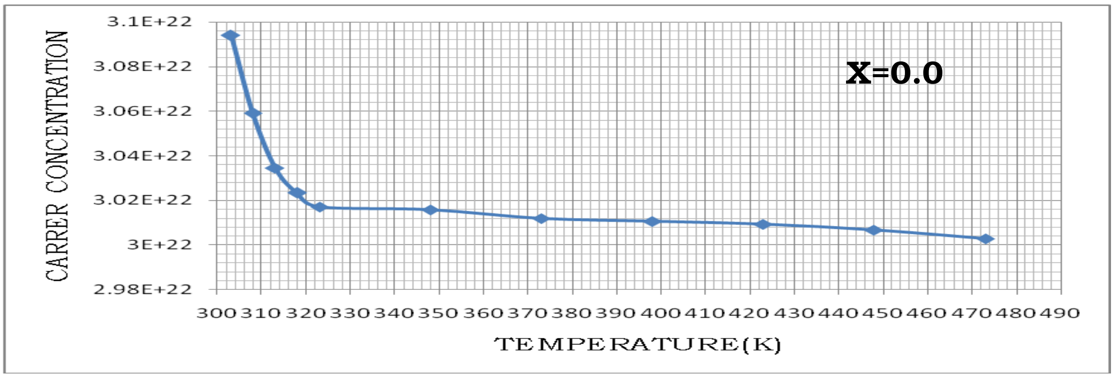

Fig 6 Variation of charge carrier concentration with temperature for X=0.0

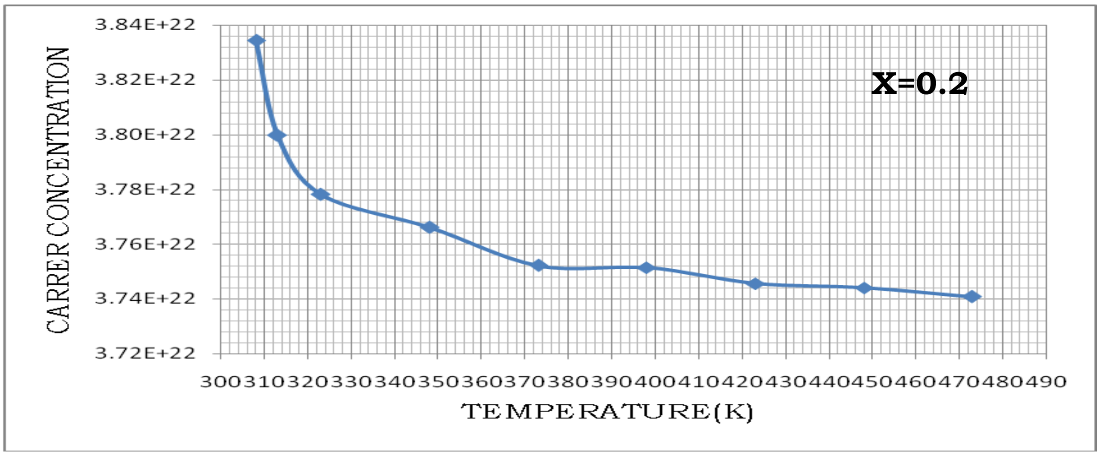

Fig 6 Variation of charge carrier concentration with temperature for X=0.2

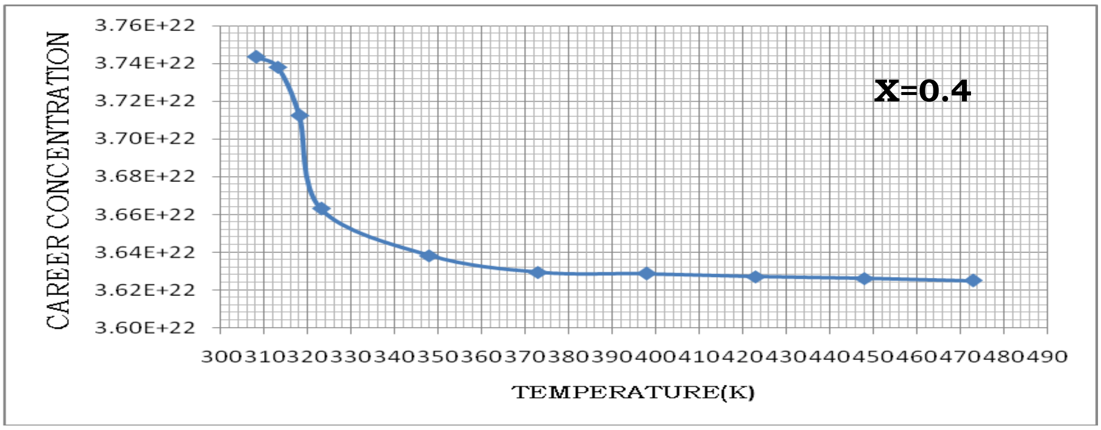

Fig 6 Variation of charge carrier concentration with temperature for X=0.4

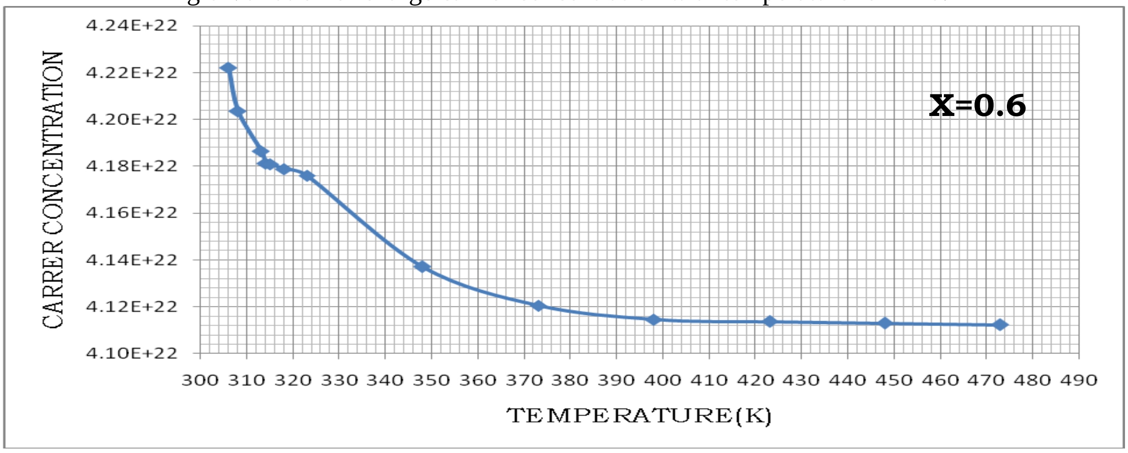

Fig 6 Variation of charge carrier concentration with temperature for X=0.6

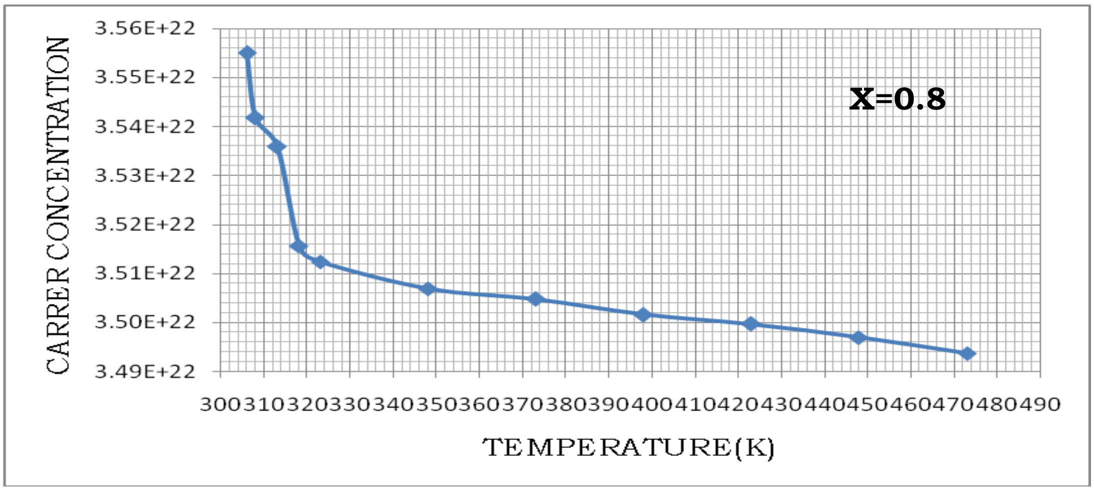

Fig 6 Variation of charge carrier concentration with temperature for X=0.8

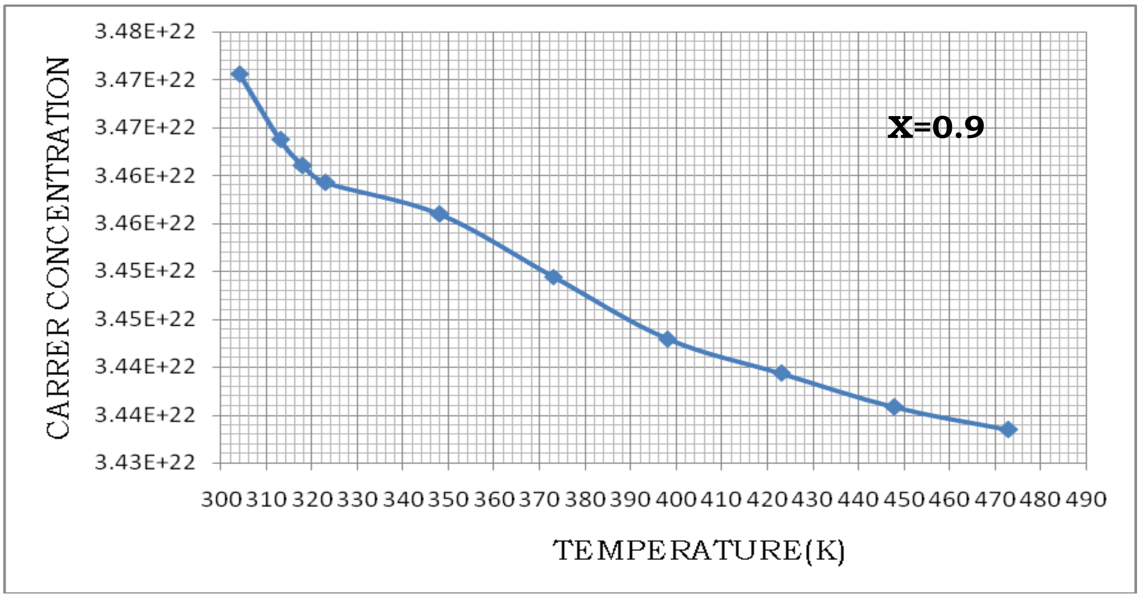

Fig 6 Variation of charge carrier concentration with temperature for X=0.9

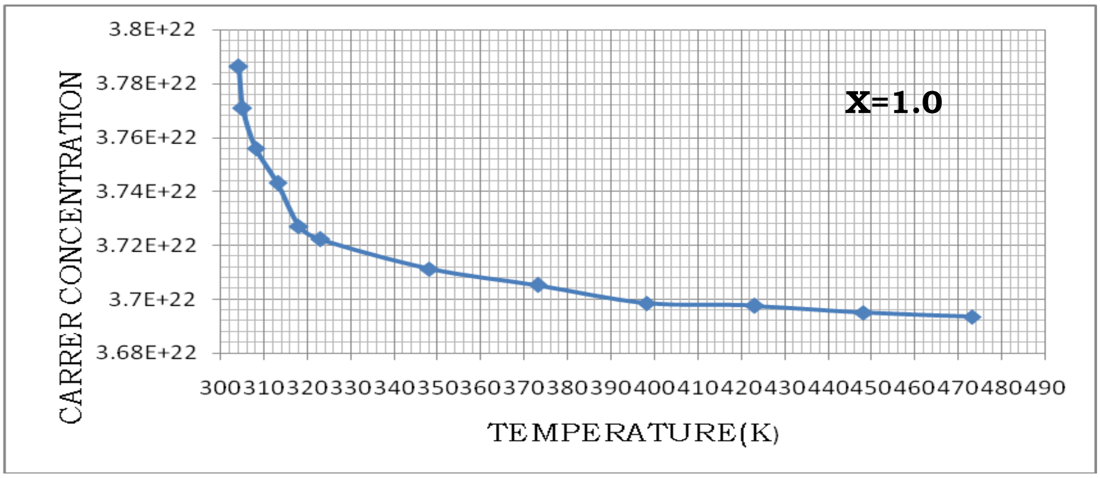

Fig 6 Variation of charge carrier concentration with temperature for X=1.0

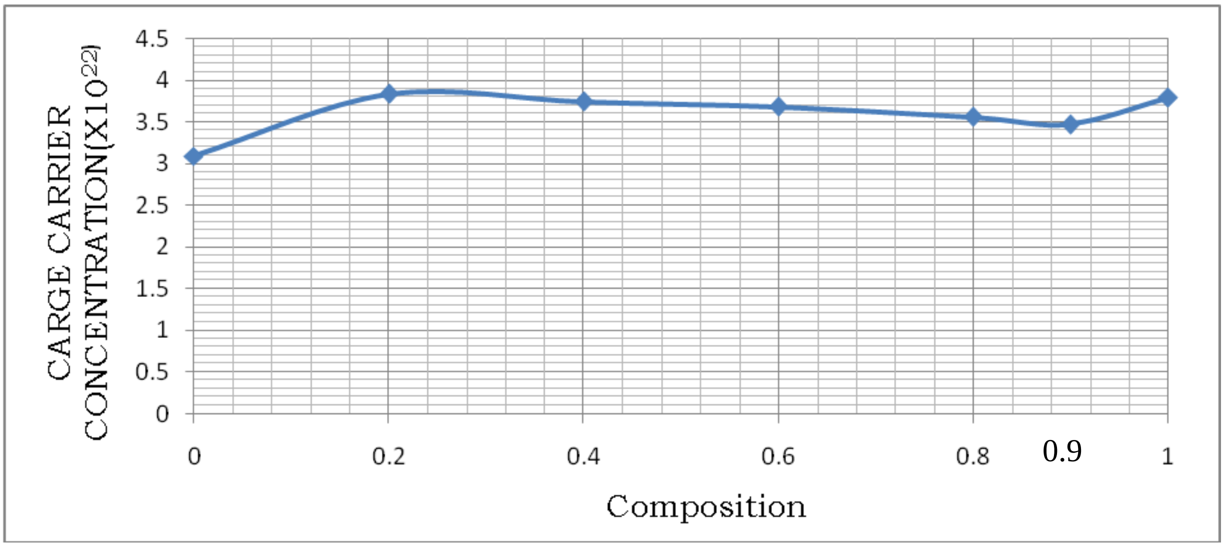

Fig 7 Variation of charge carrier concentration with composition

S.No	Composition	Seebeck Coefficient(μV/K)	Charge carrier concentration($X10^{22}cm^{-3}$)
1	$NiFe_2O_4$	-457	3.093
2	$Ni_{0.2}Zn_{0.8}Fe_2O_4$	-451	3.834
3	$Ni_{0.4}Zn_{0.6}Fe_2O_4$	-426	3.742
4	$Ni_{0.6}Zn_{0.4}Fe_2O_4$	-351	3.682
5	$Ni_{0.8}Zn_{0.2}Fe_2O_4$	-250	3.554
6	$Ni_{0.9}Zn_{0.1}Fe_2O_4$	-203	3.470
7	$ZnFe_2O_4$	-393	3.786

Table 1: Seebeck coefficient and charge carrier concentration with composition