

Thermoelectric and dielectric studies of Nd-doped Li-Ti ferrite

¹Lature Satishkumar, ²Kalashetty Shivanand and ²JadhavGH

¹Department of Physics, Poojya Doddappa Appa College of Engineering, Kalaburagi 585102, Karanataka, India

²Shri Chattrapati Shivaji College, Omerga 413606, Maharashtra, India

*Corresponding author Email: satishkumarlature@gmail.com

Manuscript Details

Available online on <http://www.irjse.in>
ISSN: 2322-0015

Editor: Dr. Arvind Chavhan

Cite this article as:

Lature Satishkumar, Kalashetty Shivanand and Jadhav GH. Thermoelectric and dielectric studies of Nd-doped Li-Ti ferrite, *Int. Res. Journal of Science & Engineering*, 2018; Special Issue A5: 31-35.

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ABSTRACT

Thermo-electric power measurement is carried out to determine the value of charge carrier concentration (n). Further from the calculated Fermi energy it suggested that the prepared Nd - doped Li -Ti ferrites are semiconducting material in nature. On the basis of Koop's theory a model is proposed for the variation of dielectric constant with frequency and further relaxation time for different compositions are calculated which are in agreement with reported value.

Key words: Nd-doped Li-Ti ferrite, Thermo-electric power, Dielectric, Fermi energy.

INTRODUCTION

Materials based on lithium ferrosinels involve a wide range of compositions and structural modifications, which predetermines their varied functional properties and quite extensive application areas [1, 2]. Most promising for the microwave electronics are those lithium-titanium ferrites that are prepared by the methods of ceramic engineering [1, 2]. Ferrites have higher resistances than metals by several orders of magnitude and they are also regarded as very structure-sensitive materials [3]. Several methods can be used to prepare ferrites such as solid-state reaction, mechanical milling, reverse micelle, and citrate precursor [4]. Hall effect and thermoelectric power studies are widely used in the interpretation of the conduction mechanism in semiconductors. The interpretation of Hall Effect results is more straight

forward and it gives precise results. However, in the case of low-mobility semiconductors such as ferrites, it is somewhat difficult to measure the Hall effect, in such cases the thermoelectric measurement is the only alternative. Moreover, the measurement of thermo e.m.f. is simple and its sign gives vital information about the type of conduction in semiconductors, i.e. whether they are n-type or p-type. In our earlier communication [5] Nd-doped Li-Ti nano-ferrites were synthesized by combustion method and magnetic properties were reported. Now in the present paper we report the non-magnetic behaviour viz thermoelectric studies of Nd-doped Li-Ti nano-ferrites.

METHODOLOGY

The nano-sized Nd - doped Li -Ti ferrites with compositional formula $\text{Li}_{0.4}\text{Ti}_{0.1}\text{Fe}_{2.5-x}\text{Nd}_x\text{O}_4$ (where $x=0.0, 0.025, 0.050, 0.075$ & 0.10) were prepared at 450°C by combustion synthesis discussed earlier [5]. The experimental technique used for the determination of thermo-electric power is the differential method [6]. The temperature variation of thermo-emf has been studied over a temperature range 300-650 K using a tubular furnace capable of giving temperature gradient of 10 K.

RESULTS AND DISCUSSIONS

The values of thermo-emf (V_s), the Seebeck coefficient (S) for each composition at 350 K has been evaluated, and it confirms that all prepared Nd-doped Li-Ti ferrite samples are n-type semiconductors [5].

FERMI ENERGY

When the conduction is only due to one type of charge carriers i.e., electrons, the Fermi energy of an n-type semiconductor is related to the Seebeck coefficient (S) by a relation [7, 8]:

$$S T = E_g - E_f + 2kT \rightarrow (1) \text{ for a n-type semiconductor}$$

Where,

E_g = energy gap of ferrite semiconductor

E_f = height of Fermi-energy level from the top of the filled valence band and

$2kT$ = the term which accounts for the transfer of K.E. of the carriers

Morin [9] assumed that the conduction may take place in exceedingly narrow bands or localized levels while discussing the electrical conduction of $\alpha\text{-Fe}_2\text{O}_3$. This assumption leads to the result that the K.E. term in equations (1) may be neglected. As ferrites are also supposed to be narrow band magnetic semiconductors, the assumptions of Morin [9] can be applied. As such, $S T = E_g - E_f \rightarrow (2)$ for a n-type semiconductor.

In order to determine absolute value of Fermi energy of a semiconductor in general, Nd - doped Li-Ti mixed ferrite in particular, the value of E_g (energy gap) is essential. In the present investigation as it is not possible to obtain the value of E_g experimentally, the value of ($E_g - E_f$) has been evaluated to understand the variation of Fermi energy with temperature in these ferrites. The value of $S T (= E_g - E_f)$, for each composition at 351 K temperature have been computed in elsewhere [5] using relation (2) and are agree with the reported ones [12 - 14].

Table 1: Thermoelectric data at 351 K

Composition	Seebeck Coefficient S ($\mu\text{V/K}$)	(Eg - Ef) X 10 ⁻³ eV	Carrier Concentration (n) X 10 ²²	Relaxation time in 10 ⁻⁴ S			
				First region		Second region	
				Observed	As per equ(50)	Observed	As per equ(50)
$\text{Li}_{0.4}\text{Ti}_{0.1}\text{Fe}_{2.5}\text{O}_4$	100.00	35.10	4.00	1.49	1.52	1.57	1.62
$\text{Li}_{0.4}\text{Ti}_{0.1}\text{Fe}_{2.475}\text{Nd}_{0.025}\text{O}_4$	112.22	39.39	3.78	1.06	1.11	0.79	0.95
$\text{Li}_{0.4}\text{Ti}_{0.1}\text{Fe}_{2.45}\text{Nd}_{0.05}\text{O}_4$	145.14	50.94	3.26	2.01	2.11	1.50	1.49
$\text{Li}_{0.4}\text{Ti}_{0.1}\text{Fe}_{2.425}\text{Nd}_{0.075}\text{O}_4$	146.74	51.51	3.23	1.57	1.69	1.07	1.15
$\text{Li}_{0.4}\text{Ti}_{0.1}\text{Fe}_{2.4}\text{Nd}_{0.1}\text{O}_4$	113.80	39.94	3.76	3.98	3.72	2.98	3.11

CHARGE CARRIER CONCENTRATION

It is known that the Fermi-distribution given by

$$f(E) = \frac{1}{1 + \exp\left(\frac{E - E_f}{kT}\right)} \rightarrow (3)$$

where $f(E)$ is the probability that a state of energy E is occupied, E = energy of an electron, E_f = Fermi energy, k = Boltzmann's constant and T = temperature

The occupancies of the various allowed electronic states depend upon the number and distribution in energy of the allowed states $f(E)$, the position of the Fermi level, and the temperature. To illustrate how these quantities are related to the occupancy of states, we will consider a semiconductor with a certain density of donor levels, N_D , all at the same energy, and a smaller concentration of acceptor levels, N_A , also at one energy [15].

According to the condition of charge neutrality,

$$n + (N_A - P_A) = P + (N_D - n_D) \rightarrow (4)$$

where N_A = total concentration acceptor centres, P_A = the concentration of holes on acceptor centres, n = number of charge carriers in the conduction band. N_D = density of donor centres and n_D = density of electrons on donor centres.

The total number of electrons in the conduction band and in donor centres is determined by the position of the Fermi level.

$$\text{Thus, } n + n_D = \int f(E) N(E) dE, \rightarrow (5)$$

where $f(E)$ is the Fermi distribution function $N(E) dE$ is the number of quantum levels in the energy range dE , and is given by

$$N(E) dE = \frac{4\pi}{h^3} (2m^*)^{3/2} E^{1/2} dE \rightarrow (6)$$

The number of electrons in the conduction band is given by

$$n = \int_{E_g}^{\infty} f(E) N(E) dE \rightarrow (7)$$

After substituting the values of $N(E)$, $f(E)$ and B by substituting the value of $(E_g - E_f)$ from equation (2) and integrating we obtain,

$$n = 2 \left(\frac{2\pi m^* kT}{h^2} \right)^{3/2} \exp\left(\frac{-S}{k}\right) \rightarrow (8)$$

Where m^* = effective mass of the electron and h = Planck's constant.

The values of carrier concentration (n) have been calculated for each composition using equation (8). The values of carrier concentration (n) at 350 K are given in table (1). It can be observed from the table that the carrier concentration decreases with increase in Nd-content. The variation of ' n ' with temperature for all Nd-doped ferrites is shown in figure (1) and it is evident that ' n ' is decreasing continuously with increasing temperature.

FREQUENCY DEPENDENCE OF DIELECTRIC CONSTANT

Figure 1 show the variation of dielectric constant (ϵ') with frequency for $x = 0$ to 0.1. Dielectric constant decreases as frequency increases. The decrease is rapid at lower frequencies and slower at higher frequencies. Similar results were observed for other systems of ferrites [16 - 18]. The decrease in dielectric constant with frequency can be explained on the basis of Koops theory [19], which considers the dielectric structure as an inhomogeneous medium of two layers of the Maxwell-Wegner type [20].

In this model, the dielectric structure is assumed to be consisting of Well-conducting grains which are separated by poorly conducting grain boundaries. It was found that for ferrites, the dielectric constant is directly proportional to the square root of conductivity [17].

Let C_1 and C_2 represent the equivalent capacity of the layers and grain respectively and R_1 and R_2 represent the resistance of layers and grains respectively. Let us also assume that C_p and R_p are the equivalent parallel capacity and the equivalent resistance respectively.

Now, the admittance Y_p of the parallel R-C circuit is given by

$$Y_p = \frac{1}{R_p} + j\omega C_p \rightarrow (9)$$

The admittance of the series parallel combination is

$$Y_{sp} = \frac{(1 + j\omega C_1 R_1)(1 + j\omega C_2 R_2)}{(R_1 + R_2) + j\omega R_1 R_2 (C_1 + C_2)} \rightarrow (10)$$

Now let us define the relaxation time as

$$\tau = \frac{(\epsilon_1 + \epsilon_2 x) / \sigma_1 \sigma_2}{\left(\frac{x}{\sigma_1} + \frac{1}{\sigma_2}\right)} \rightarrow (11)$$

The low frequency value of dielectric constant as

$$\epsilon_p^0 = \frac{\frac{\epsilon_1 x}{\sigma_1^2} + \frac{\epsilon_2}{\sigma_2^2}}{\left(\frac{x}{\sigma_1} + \frac{1}{\sigma_2}\right)^2} \rightarrow (12)$$

The high value of dielectric constant as

$$\epsilon_p^\infty = \frac{\epsilon_1 \epsilon_2}{(\epsilon_1 + x \epsilon_2)} \rightarrow (13)$$

Now according to equation (11) the relaxation constant has been defined as,

$$\tau = \frac{(\epsilon_1 + \epsilon_2 x) / \sigma_1 \sigma_2}{\left(\frac{x}{\sigma_1} + \frac{1}{\sigma_2}\right)}$$

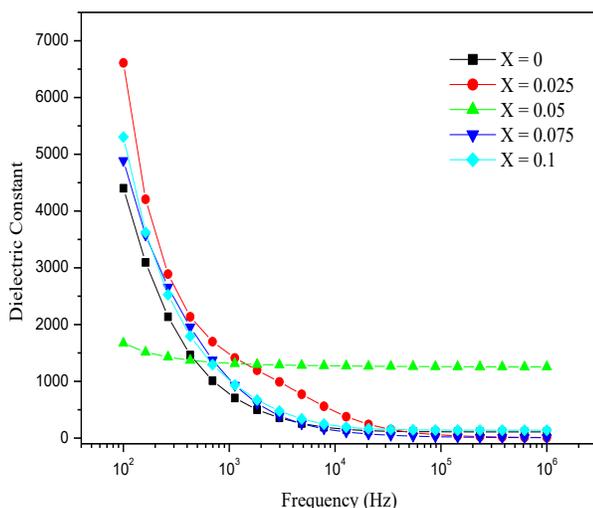


Figure 2: Dielectric constant with frequency for $x = 0$ to $x = 0.1$ in $\text{Li}_{0.4}\text{Ti}_{0.1}\text{Fe}_{2.5-x}\text{Nd}_x\text{O}_4$

Now by expressing the equation (11) in terms of absolute permittivities, we get

$$\tau = \frac{(\epsilon_1 + \epsilon_2 x)}{4\pi(\sigma_1 + \sigma_2 x)} \times \frac{1}{9 \times 10^{11}} \rightarrow (48)$$

Koops [21] has assumed that the intrinsic dielectric constant of the large domain (grains in present investigation) has approximately the same value as that of the boundary layer ($\epsilon_1 = \epsilon_2$). This will be approximately equal to the ϵ caused by the oxygen ions. It is assumed that, $R_1 \gg R_2$ we have

If we assume that the value of 'x' is very small, we have

$$\tau = 8.55 \times 10^{-14} \times \frac{\epsilon_0}{\sigma_\infty} \rightarrow (50)$$

Koops [21] reported electrical conductivity of some ferrites as a function of frequency of several ferrites and observed that the high frequency values of electrical conductivity is roughly 4 to 8 times higher than those of low frequency values. As such, it has been concluded at higher frequency (σ_∞) in the present investigation may increase approximately about 6 times (average of 4 and 8), the value at low frequency. Using the value of σ_∞ , τ for $x = 0$ to $x = 0.1$ has been calculated and is given in table (1).

CONCLUSION

The values of carrier concentration (n) have been calculated from thermo-electric studies for each composition using equation (18) and the values of carrier concentration (n) decreasing continuously with increasing temperature. Further the relaxation time obtained from dielectric data was found to be in agreement with the model proposed.

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