

Curie Points and Direct Current Electrical Conductivity for Inverse *Li*-Spinel Ferrite Replaced by Zn^{2+} Ion

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Abstract

The conventional ceramic double sintering method was used to prepare a series of mixed *Li*-*Zn* spinel ferrites (SFs) Direct electric (DC) electrical conductivity (σ) and inductance (L) were studied for the prepared series of samples from room temperature to well beyond the Curie point (T_C). σ was measured as a function of temperature, the relation between $\ln\sigma$ with $1/T$ indicated that σ was increased with increasing of the temperature, which proves that the prepared samples have a semiconductor behavior. The maximum value of the σ was reported for the sample of $x = 0.8$. The σ results were interpreted in terms of hopping model. The relation between $\ln\sigma T$ and $1/T$ showed a change in the slope which is attributed to the change of samples from ferrimagnetic to paramagnetic at T_C . The values of L increased exponentially from room temperature up to T_C , and then they decreased sharply. The calculated activation energy E_a in the ferromagnetic region was less than that in paramagnetic region. The calculated activation energy (E_a) decreased with increasing of *Zn* content. The values of the T_C for the prepared samples were determined from σ and L measurements, which decreased with increasing of *Zn* content.

Keywords

Spinel Ferrite, DC Electric Conductivity, Curie Point, Inductance, Activation Energy, Defect, Vacancies

1. Introduction

Spinel ferrites have received immense attention due to their novel magnetic, optical, electric, dielectric and catalytic properties [1-5], chemical and thermal stability, mechanical hardness, memory storage capacity, easy to synthesize and reasonable cost [5-9]. Spinel ferrites have wide extended applications encompass an impressive in different fields due to their interesting electrical properties [10, 11]. The electrical properties of spinel ferrites are sensitive to their composition and microstructure, which in turn are sensitive to their processing conditions. Lithium and substituted lithium ferrite are gaining importance because of their technological applications ranging from microwave to radio frequency. It was reported that *Li*-*Zn* ferrites showed a reversible loss of oxygen, on the other hand irreversible volatilizations of the *Zn* and the *Li* above approximately

1000°C [12]. The loss of *Zn* and *Li* imply to appearance of α - Fe_2O_3 , which is linked to the excess *Fe* content. As a result, this leads to a cation-non-deficient spinel phase, therefore, the reduction of Fe^{3+} to Fe^{2+} increases. Therefore, for the most part, the conductivity increases with sintering temperature. Furthermore, the high-temperature spinel phase is maintained by quenching (fast cooling) [12]. Where during slow cooling, oxygen atoms can return to the cubic lattice and re-oxidize most of the Fe^{2+} and Fe^{3+} but not all. In addition, the conductivity of the precipitated α - Fe_2O_3 has extremely high conductivity. The results give an account of electrical conductivity which is decreased not only an account of electrical conductivity which is decreased not only by the oxygen reintegration and the re-oxidation of Fe^{2+} ions during slow cooling, but also by the nature of precipitation of above approximately 1000°C [12]. In this study, the composition and cation distribution of the mixed *Li*-*Zn* spinel ferrites exhibits various electric properties. Therefore, a

systematic study of the σ of the mixed *Li-Zn* spinel ferrite system from room temperature to well beyond T_C was undertaken. The aim of this study is to investigate the effect of Zn^{2+} ions on σ and to determine the values of the T_C of mixed *Li-Zn* spinel ferrites.

2. Experimental

The standard double sintering ceramic technique was used to synthesize the mixed *Li-Zn* spinel ferrites that have the chemical formula $Li_{0.5(1-x)}Fe_{0.5(1-x)}Zn_xFe_2O_4$, where x is the percentage increment of Zn^{2+} ions on the compound over the range $0.0 \leq x \leq 1.0$, by mixing the stoichiometric quantities of pure Li_2CO_3 , Fe_2O_3 and ZnO . The procedures to prepare the powder were described in details in our previous work [13-17]. Some powder was pressed in a disc shape. Other powders were formed in a toroidal shape. After that, samples were sintered at 1150 ± 20 °C for soaking time of 5 h's. After sintering process, the samples were slowly cooled down to room temperature. The σ of the prepared samples was estimated at different temperature by two-probe method using a digital multimeter (model FLUKE -177). Pellets of a disk-shape were pasted with the silver paste on their two faces to get a good conducting surface. Two purity silver wires were used as electrodes. For inductance measurements, LCR meter model (GW- instek LCR-821) is used.

3. Results and Discussion

3.1. Electric Properties

3.1.1. Temperature-Dependent of DC Electric Conductivity

The spinel ferrites that have a general formula DFe_2O_4 (D is divalent metal ions) [15-17], are widely used in several electronic devices due to noticeable electrical resistivity (from 10^{-5} to 10^9 ohm-m) [2] and low eddy current [6]. The high and low resistivity of spinel ferrites are, mainly, demonstrated according to cations distribution in tetrahedral (T_d) and octahedral (O_h) sites of the spinel ferrites structure, space charge polarization (SCP) and hopping mechanism. The conduction takes place when electrons move from Fe^{2+} to ferric Fe^{3+} ions. The high value of σ in spinel ferrites is due to simultaneous existence of Fe^{2+} and Fe^{3+} into T_d and O_h sites. The low resistivity in spinel ferrites links to the occupation of O_h sites by different Fe^{2+} and Fe^{3+} ions. The conduction mechanism of spinel ferrites, also, depends on temperature. It is reported in [18, 19] that, the electric conduction at lower temperature (below T_C) is due to hopping electron between Fe^{2+} and Fe^{3+} ions, whereas at a higher temperature (above T_C) is due to different conduction mechanisms such as the polarons. In the hopping process, the additional electron of Fe^{2+} ions requires little energy to move to an adjacent Fe^{3+} on O_h sites. The thermal dependence of σ for the mixed *Li-Zn* Spinel ferrites with different *Zn* concentrations was investigated from room temperature to fit beyond the transition temperature i.e. T_C . The variation of $\ln \sigma$ versus the reciprocal of temperature ($10^3/T$) is depicted in Figure 1. Such Figure illustrates that,

$\ln \sigma$ increases continuously with increasing of the temperature. This could be attributed to the increase in the drift mobility of electric charge carriers, which are thermally activated on increasing the temperature [20]. At relatively high temperature, some of the electrons are thermally activated and leave holes in the covalent bonds, which become partially ionic intrinsic carriers (electron hole). In this process, an electron moves by thermally activated hopping from one localized state to another. The hopping can be considered as an energy exchange between electrons and phonons leading to electron-phonon interaction, which decreases the electron mobility as well as the conductivity of the system [21]. The obtained results confirms that the spinel ferrites under investigation have the same behavior of various spinel ferrites systems [22, 23]. The observed behavior of increasing conductivity with increasing temperature clearly indicates that the present zinc substituted lithium ferrites have semiconductor like behavior [20]. The change in the Fe^{2+} ion content in the spinel ferrites lattice and/or the distance between them is crucial to the intrinsic resistivity of the mixed *Li-Zn* spinel ferrites grains, including the intrinsic grain boundaries. The σ is seen to increase much more rapidly with increasing temperature as the samples undergo a ferrimagnetic to paramagnetic transition. This behavior may be attributed to the increase in drift mobility of the charge carriers [14]. Figure 2 presents the $\ln \sigma T$ versus ($10^3/T$). As in this Figure, the values of $\ln \sigma T$ for the samples with $x \leq 0.6$ increased linearly with increasing of temperature up to T_C at which a slope changed. Several researchers for different ferrite systems [13, 14] reported similar behavior. Generally, the change of slope was attributed to existence of different conductivity mechanism [24] in which the sample moving from ferrimagnetic to paramagnetic. It was proved theoretically that at T_C , a change must be occurred in the gradient of the straight line. In addition, the magnitude of the change depends on the exchange interaction between the outer and the inner electrons, which alter the value of T_C . It was stated that the activation energy and the effective mass of current carrying excitons in ferrimagnetic semiconductors depend on the spontaneous magnetization because of a "magnetizing" exchange interaction between the outer and inner electrons. This led to an additional temperature dependence of the electrical resistance, which is especially strong near the Curie temperature. The kink will be larger for cases in which there is a stronger exchange interaction between the outer and inner electrons. Furthermore, the decrease of the spins in the adjacent incomplete shells of the metallic ions in the system will be accompanied by an increase in the rate of migration of charge carriers and an increase in the electrical conductivity of the paramagnetic region [21]. The dependence of the DC electrical conductivity (σ) on temperature, which is demonstrated in the Figure 2 fulfills the Arrhenius relation [25]

$$\sigma = \frac{S}{T} e^{(-E_a/kT)} \quad (1)$$

where S is a constant, k is the Boltzmann constant and E_a is the activation energy.

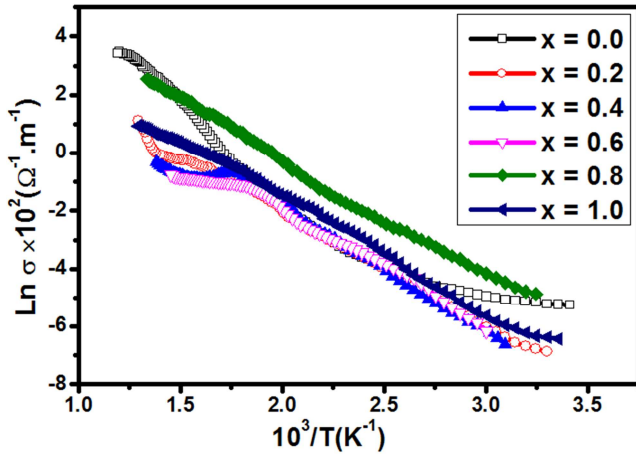


Figure 1. Variation of $\ln \sigma$ with $(10^3/T)$ for all prepared samples.

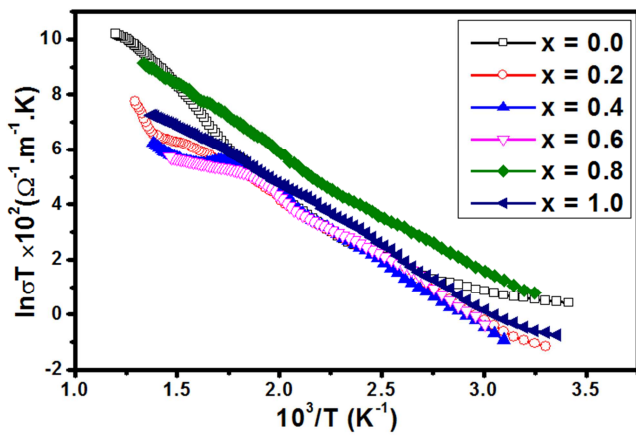


Figure 2. Variation of $\ln \sigma T$ with $(10^3/T)$ for the samples with different compositions.

3.1.2. Composition Dependent of Activation Energy

The E_a of both regions before and after T_C transition i.e. ferrimagnetic (E_f) and paramagnetic (E_p) for the studied compositions were calculated using the two different slopes in Figure 2 and Eq. (1), which confirms two parallel conductivity mechanisms with differing activation energies. The calculated values of E_f , E_p and $\Delta E = E_p - E_f$ for the given spinel ferrites system were listed in Table 1. As showing in Table 1, with increasing of Zn^{2+} ions concentration in the spinel ferrites matrix, the values of E_f and E_p are decreased where the values of $\Delta E = E_p - E_f$ are changed from 0.10-0.13 eV. This is related to the decreasing of the resistivity of the samples because activation energy behaves in the same way as resistivity [26, 27]. It is noticed that, the values of E_f vary from 0.51 eV to 0.29 eV for the samples with $x \leq 0.6$ and the values of E_p vary from 0.62 eV to 0.42 eV. While the E_a cannot be determined for the samples with $x = 0.8$ and 1.0 since they are paramagnetic at room temperature. It is noticed that, the values of E_p are greater than the values of E_f . This is, also, was found by [28, 29]. This may be attributed to the existence of small number of oxygen vacancies [24]. The presence of oxygen vacancies in all cases leads to the formation of Fe^{2+} ions. The Fe^{2+} cations appear on the sites adjacent to the oxygen vacancy [30]. The values of E_f and E_p were given in Table 1 are greater

than 0.31 eV while the transition energy between Fe^{2+} and Fe^{3+} is 0.2 eV [31]. According to the ref. [31], it was suggested that if the value of E_a is lower than 0.2 eV then the conduction mechanism is, predominantly, due to the electron hopping than small polaron. It was subjected that the effect of composition x on ΔE is greater if the substituted cations occupy O_h sites but almost no change in ΔE is observed when the substitution is made on T_d sites without disturbing the O_h sites [32].

Table 1. Values of the E_f , E_p and $\Delta E = E_p - E_f$ for the mixed Li-Zn spinel ferrites (SFs).

x	Activation Energy		$\Delta E = E_p - E_f$ (eV)
	E_f (eV)	E_p (eV)	
0.0	0.51	0.62	0.11
0.2	0.37	0.49	0.12
0.4	0.34	0.44	0.10
0.6	0.29	0.42	0.13
0.8	-	-	-
1.0	-	-	-

3.1.3. Composition Dependence of DC Electrical Conductivity

The cations distribution is important to explain the electric conduction mechanism. Herein, the expected cations distribution of the Li-Zn spinel ferrites is given by the following $(Zn_x^{2+}Fe_{(1-x)}^{3+})_T\{Li_{0.5(1-x)}^{1+}Fe_{0.5(1+x)}^{3+}Fe_1^{3+}\}_O O_4^{2+}$. According to the given cations distribution, the change of the σ with increasing of the Zn^{2+} ions is explained for the Li-spinel ferrites. It is, obviously, from cations distribution that, with increasing of the Zn^{2+} ions in T_d sites implies to decrease the Fe^{3+} ions in the T_d sites and to decrease of the Li^+ ions in O_h sites. This may be disturbing the bond $Fe^{2+} - O^{2-}$ of T_d and O_h sites in the spinel ferrites [23]. The $Fe^{2+} - O^{2-}$ bond exerted by the Fe^{2+} ions that is formed in the samples during the oxidation process ($Fe^{2+} \leftrightarrow Fe^{3+} + e^-$) in air at the time of the sintering process. On the other hand, the electronic distribution of the $Fe^{2+} - O^{2-}$ bond is, greatly, affected when the Zn^{2+} ions are increased. The decrease in the electrical conductivity with increasing Zn^{2+} ions concentration may be attributed to the decreasing relative concentration of Fe^{2+} at O_h sites. The magnitude of exchange depends up on the concentration of Fe^{2+}/Fe^{3+} ion pairs present on O_h sites. For the sample of $x = 0.0$, the cations distribution becomes $(Fe_{(1)}^{3+})_T\{Li_{0.5}^{1+}Fe_{1.5}^{3+}\}_O O_4^{2+}$. It is clearly; that the two cations, i.e. Li^{1+} and Fe^{3+} , occupied the O_h sites are responsible for electrical conduction in Li-spinel ferrites, which can be described as the following $Fe^{3+} + e^- \leftrightarrow Fe^{2+}$. It has been assumed that the electrons, which participate in the formula (4) exchange process, are strongly coupled to the lattice and tunnel from one site to other due to a phonon-induced transfer mechanism. Electrical conductivity of single crystals of Li ferrite in the temperature range 300 to 1100 K, discontinuous decrease can be produced at the order-disorder transition temperature (1000 K). However, at 1175 °C oxygen loss can occur even if the firing is in an atmosphere of oxygen. Otherwise, for the sample with $x = 1.0$, the cations distribution can be reduced to the following $(Zn^{2+})_T\{Fe_2^{3+}\}_O O_4^{2-}$. Thus, only the Fe^{3+} cation occupies the O_h sites, which is responsible for electrical conduction in

Zn-Spinel ferrites. This leads to increase the number of electrons in the O_h sites, which implies to increase the electric conductivity. The variation of conductivity of Li-Zn spinel ferrites with Zn composition was shown in Figure 3.

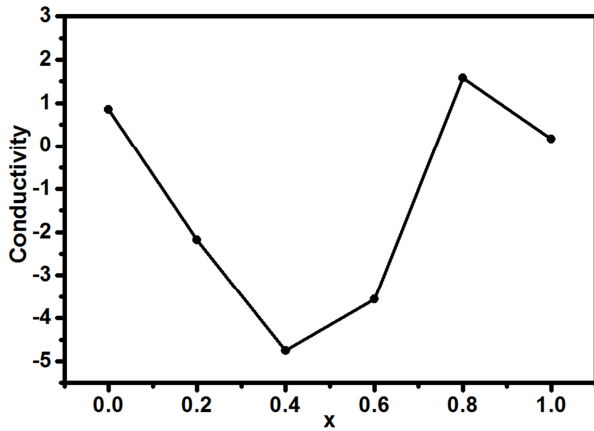


Figure 3. Variation of conductivity with different compositions.

It is observed that the conductivity decreases with increasing Zn²⁺ ions up to $x=0.4$, beyond which it increases again up to $x=0.8$, because at high temperature the rate of charge carrier generation increases and reaches the limit at which their motilities, as well as the mean free path, decrease [21]. Some researchers [13] also observed the same anomalous behavior. In Zn²⁺ ions doped ferrites prepared by conventional solid-state method, there may be a loss of Zn²⁺ ions by evaporation caused by high sintering temperatures and longtime treatments. This results in non-stoichiometric (reorganization) ferrite [33]. Such reorganization would so lead to expensive crystallographic network reorganization from an energetic point of view. It was observed that the conductivity decreased with increasing concentration of Zn²⁺ ions in the ferrite. This behavior was explained in terms of an increase in Fe²⁺ ions concentration caused by the evaporation of Zn²⁺ ions during sintering and cooling process [34]. The 1:3 order of Li⁺ and Fe³⁺ ions can be disturbed either by quenching the samples from 1000 K or by Zn²⁺ ions doping. The decrease in conductivity with Zn content may be due to the decrease in the Li loss at the lower Li content. Hence, the possibility of Fe²⁺ ions and enhancement of conductivity is more probable for higher Zn²⁺ ions. Whereas, in the lower Zn content, the effect of Li volatilization is more significant in explaining the conductivity data. It is well known that the preparation of Li containing spinel is very critical because of the Li oxide volatility [35].

3.2. Inductance and Curie Point

The relation between magnetization (M) and the L of a closed packed coil (toroid) knitted around a substance is given by [13, 14]

$$M = \left[\frac{L}{2\mu_o N^2 l \ln(D_o / D_i)} - \frac{1}{4\pi} \right] H \quad (2)$$

where μ_o is the permeability of the free space, N is the number of the turns, l is the thickness of the toroidal sample, D_o and D_i

are the diameter of outer and inner of the toroidal sample, respectively. As in Eq. (2) magnetization (M) is directly proportional to L . The thermal spectra of L - T curve can be taken as a test function of homogeneity of the ionic structure of the sample [13]. Figures 4 and 5 depict the variation of inductance versus temperature from room temperature to fit beyond Curie point (T_C). It is noticed that, L is constant up to transition happens at which a sharp drop of L is occurred for $x=0.0$ but for $x=0.2, 0.4$ and 0.6 it was increased exponentially up to a certain value and then decreased sharply at T_C . The sample of $x=0.8$ and 1.0 have no transition since they are paramagnetic at room temperature as in Figure 5. The same behavior was shown for the Zn-spinel ferrite, which was found as the paramagnetic substance at room temperature [3, 36]. It is found that the transition temperatures decreased as the Zn content increased (Table 2). This is attributed to the addition of the non-magnetic Zn²⁺ ions that replaced the magnetic Fe³⁺ ions at the T_d sites, thus; the number of the Fe³⁺ ions decrease at the T_d sites. This tends to decrease the strength of T_d and O_h exchange interactions of the type $Fe_T^{3+} - O^{2-} - Fe_O^{3+}$, apart from decreasing number of bonds or linkages between the magnetic ions [37].

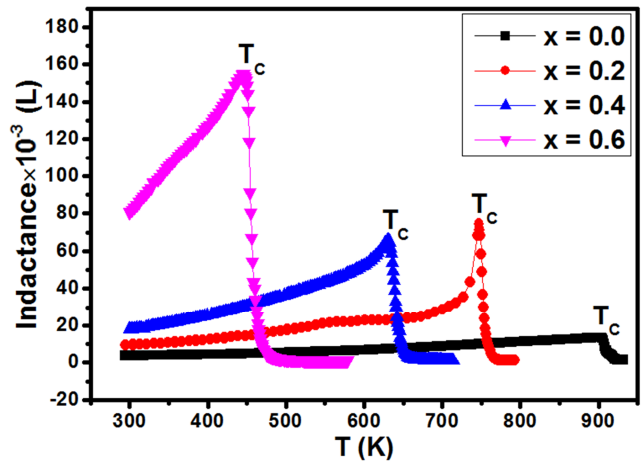


Figure 4. Variation of the L with T for the samples with $x=0.0, 0.2, 0.4$ and 0.6 .

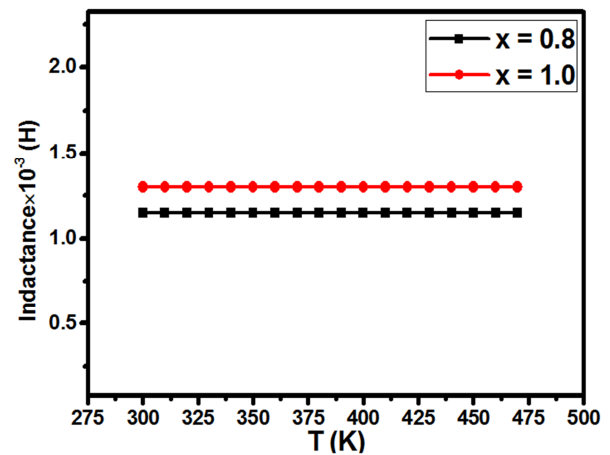


Figure 5. Variation of the L with T for the samples with $x=0.8$ and 1.0 .

3.3. Composition Dependence of Curie Point

Values of T_C are determined along with the value of x in Table 2. From this table, it is seen that the value of T_C decreases with increasing of Zn^{2+} ions up to $x = 0.6$. This is attributed to the decrease of $T_d - O_h$ interaction. As the content of Zn^{2+} ions is increased, the relative number of Fe^{3+} ions at T_d site decreases, which causes a reduction in the $T_d - O_h$ interaction [38]. Other researchers [39] also, observed the decrease in the values of T_C with increasing of Zn^{2+} ions in the different ferrimagnetic materials.

Table 2. Values of T_C , which were determined by induction and DC conductivity measurements for the mixed Li-Zn Spinel ferrites.

x	$T_C(K)$	
	Induction	DC conductivity
0.0	900	843
0.2	769	728
0.4	628	583
0.6	448	488
0.8	N/A	N/A
1.0	N/A	N/A

4. Conclusions

The mixed *Li-Zn* spinel ferrites was substitution of the non-magnetic Zn^{2+} ions in *Li*-spinel ferrites has a tremendous influence on the electric properties. So, the mixed *Li-Zn* spinel ferrites are considered as a soft ferrite material, which is proved an interest material for technological and scientific applications. The *Li-Zn* spinel ferrites were, successfully, synthesized by double sintering method. The σ increased that confirms the semiconductor behavior for the prepared mixed *Li-Zn* spinel ferrites, also, the calculated activation energy values in paramagnetic region were greater than ferrimagnetic region, which attributed to the existence of small polaron-hopping. The measured T_C from the L and σ showed decreasing with increasing of Zn^{2+} ions, as a result, it was found that transition occurred for all samples except the samples of $x = 0.8$ and 1.0 , which can be considered as a paramagnetic at room temperature.

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