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The conduction mechanism of Cu–Si ferrite

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ABSTRACT

The mixed ferrite Cu_{1+x} Si_x Fe_{2-2x} O_4; with $0 \leq x \leq 0.3$ in step of 0.05 were prepared by standard ceramic techniques. The X-ray analysis confirmed that the compositions are well formed in a single phase. The d.c. and a.c. electrical conductivity as a function of temperature have been studied. The relation $\ln \sigma$ vs. T^{-1} showed two regions with different slopes for all values of x except $x = 0.0$ showed one slope only. The two activation energies around a kink point called T_k were calculated. The change of the slope at T_k was attributed to a change in the nature of type of conduction mechanism (i.e. from n-type to p-type). The activation energies were greater than 0.3 eV; which clearly suggested that the conduction is due to small polaron model.

1. Introduction

Spinel ferrites are one of the most important classes of magnetic ceramic materials owing to their interesting applications. In the spinel structure, the magnetic ions are distributed among two different lattice sites, tetrahedral (A) and octahedral (B) sites. However, Cu-ferrite was considered to be a mixed ferrite where Cu and ferric ions share the A-sites as well as B-sites [1]. From the crystallographic point of view Cu-ferrite exists in two crystallographic forms of tetragonal and cubic system [1–3]. Copper ferrite is rather unique among ferrites because of the considerable variation of its structural, magnetic and electrical properties depending critically upon its thermal history, cation distribution, and ion substitution [2].

In our present work a modification of structure has been obtained by introducing Si^{4+} ions in Cu-ferrite. This addition of Si^{4+} ions to ferrite samples produces a change in the electrical properties depending on the amount of Si^{4+} ions. Mazen and Zaki [4] have studied the d.c. conductivity and thermoelectric power of $CuFe_2O_4$. The measurement showed that the copper ferrite is n-type over the whole range of temperature from about 300 up to 773 K. Also, Patil et al. [5] have studied the Seebeck coefficient for the same composition of $Cu_{1+x}Si_xFe_{2-2x}O_4$. The results reveal that the sample of $x = 0.05$ is n-type conductor, while the measurements for other samples starting from $x = 0.1$ to $x = 0.3$ show p-type conduction. Moreover, there is a need for a careful study of electrical properties of these ferrites possessing desired applications. Therefore, we have undertaken a systematic study of the electrical conductivity of Cu_{1+x} Si_x Fe_{2-2x} O_4 with $x = 0.0, 0.05, 0.1, 0.15, 0.2, 0.25$ and 0.3 as a function temperature. The results of such studies are presented in this paper.

2. Experimental technique

The investigated samples of Cu–Si ferrite having the general formula Cu_{1+x} Si_x Fe_{2-2x} O_4 (where, $x = 0.0, 0.05, 0.1, 0.15, 0.2, 0.25$ and 0.3) were synthesized by the standard ceramic technique from mixing the pure oxides (Fe_2O_3, CuO and SiO_2). The weighted materials were mixed and then grounded to a very fine powder using: ball milling machine (model Puluer Isette 6). The mixture of each composition was pre-sintered at 750 °C for 5 h. The samples were pressed in a disk-shaped form of 13-mm diameter, and 3–5 mm thickness and finally sintered at 1000 °C for 10 h and cooled to R.T. The samples were polished and coated by silver paste for the electrical conductivity measurements. The electrical conductivity (d.c. and a.c.) and dielectric loss tangent ($\tan \delta$) of the investigated compositions were measured in a wide range of temperature from room temperature 293 up to 783 K in static air by the two probes method using (AC–DC bridge type FLUKA model PM6306). Digital temperature indicator (model Pro'skit 03–9303) with resolution 1 K, connected with a standard K-type thermocouple was used to measure the temperature. X-ray diffraction analyses were used to confirm that all samples were formed in single phase spinel with the average lattice constant (a) about 0.838 nm [6].

3. Result and discussion

3.1. d.c. conductivity and mobility

It is well known that the semiconducting properties of all ferrites are described by a "hopping mechanism" of charge carriers from one ionic site to the next, rather than by "the electron-band model". According to the hopping theory the conductivity is given by [7]:

$$\sigma = \frac{ne^2d^2\nu}{kT} \exp\left(\frac{-W_H}{kT}\right) \tag{1}$$

where (e) is the electronic charge, (n) is the number of charge carriers, (d) is the jump length of charge carriers as the average distance

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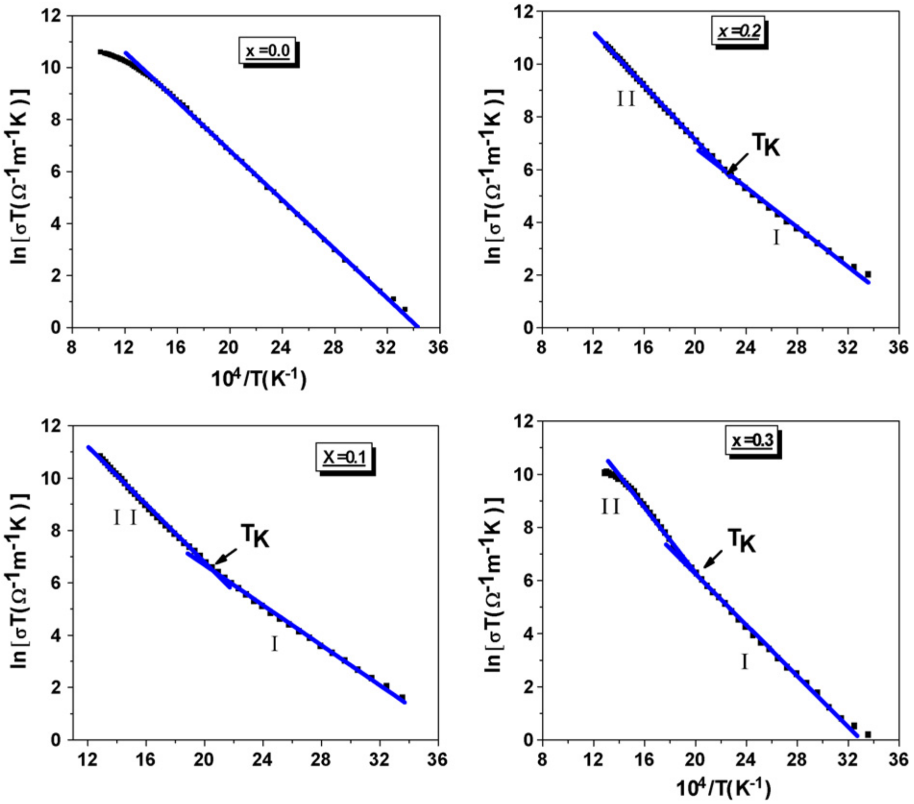

Fig. 1. The variation of $\ln \sigma T$ vs. reciprocal temperature for Cu_{1+x} Si_x Fe_{2-2x} O_4 (where, $x = 0.0$, 0.1, 0.2 and 0.3).

between neighbouring octahedral sites (≈0.296 nm) which calculated from the following relation: $d = \frac{a}{4}\sqrt{2}$ (where a is the lattice constant) [8], (k) is the Boltzmann's constant, (ν) is the lattice frequency active in the jumping process and (W_H) is the activation energy of hopping.

The logarithm of conductivity "$\ln \sigma T$" has been studied as a function of reciprocal temperature T^{-1} over a wide range of temperature from room temperature 293 up to 783 K in static air for $Cu_{1+x}Si_xFe_{2-2x}O_4$ (where $x = 0.0$, 0.05, 0.1, 0.15, 0.2, 0.25 and 0.3). Fig. 1 shows this relation for $x = 0.0$, 0.1, 0.2 and 0.3, as representative examples. The two activation energies for the two regions (I&II) around a kink point called T_k are tabulated in Table 1, but the composition of $x = 0.0$ shows one region only. The different regions with changing the slopes have been observed from conductivity studies in many ferrites [5,7,9]. Such anomalies may arise due to various reasons such as: changing from ferrimagnetic to paramagnetic; distortion in some spinel structure (e.g. transformation from tetragonal to cubic system [10–11] or a change in the conduction mechanism. The kink point T_k can not be attributed to Curie point or structural phase transformation, because the Curie points were found at high temperature (708–717 K) [12]. Also, the X-ray diffraction detected that all samples are in cubic phase. So that, the change of the slope at T_k from region I to region II, may be due to a change in the type of conduction (i.e. from n-type to p-type). It was found from previous work [4,13] that $CuFe_2O_4$ is n-type semiconductor over the whole range of elevated temperature. Moreover, the cation distribution will play an important role in the conduction process. Therefore the cation distribution could be suggested according to this formula:

$$(Cu_t^{2+}Fe_{1-t}^{3+})^A(Cu_{1-t}^{2+}Fe_{1+t}^{3+})^B O_4^{2-}, \quad x = 0.0$$
$$(Si_x^{4+}Fe_{1-0.5x}^{3+})^A(Cu_{1+x}^{1+}Fe_{1-1.5x}^{3+})^B O_4^{2-}, \quad 0.05 \le x \le 0.2$$

where the value of (t) does not exceed 0.1 for $x = 0$ [3]. According to this distribution the iron ions Fe^{3+} are replacement with Cu^{2+} and Si^{4+} ions according to:

$$2Fe^{3+} \leftrightarrow Cu^{2+} + Si^{4+}$$

Thus, the conduction mechanism can be discussed as follows:

For the composition with $x = 0.0$ ($CuFe_2O_4$), the kink point T_k disappeared and the continuity in the logarithm of conductivity with T^{-1} can be attributed to one type of conduction mechanism.

Table 1
The values of activation energy from (d.c. and mobility) of all samples of Cu–Si ferrite.

x	E_σ (eV)		W_H (eV)		E_μ (eV)		W_μ (eV)		T_k
	I	II	I	II	I	II	I	II	
0.0	0.364	–	0.414	–	0.357	–	0.4	–	–
0.05	0.405	0.438	0.48	0.51	0.395	0.44	0.46	0.49	512.5
0.1	0.383	0.41	0.443	0.49	0.36	0.421	0.42	0.47	493.1
0.15	0.36	0.39	0.393	0.466	0.34	0.397	0.38	0.45	511
0.2	0.339	0.38	0.338	0.423	0.319	0.379	0.32	0.4	480
0.25	0.337	0.362	0.329	0.419	0.312	0.35	0.315	0.395	463
0.3	0.333	0.36	0.329	0.418	0.298	0.347	0.315	0.38	438

The activation energy (W_H) of $CuFe_2O_4$ is equal 0.414 eV, which is in good agreement with the result obtained in our previous work [14]. The addition of silicon leads to increase W_H from 0.41 eV to 0.48 eV at $x = 0.05$, after that the activation energy decreases with increasing Si content, as shown in Table 1. In general, the activation energy is greater than that of the transition energy between $Fe^{3+} \leftrightarrow Fe^{2+}$ which is about 0.2 eV. These values are due to narrow-band conduction and formation of small polaron [15]. It is known that Si^{4+} ions have strong preference for A-sites [5]. Despite the fact that Si^{4+} ions do not participate directly in the conduction process, the substitution of Si^{4+} ions on A-sites tends to increase the copper content on B-sites and decrease the iron ions on both sites (A&B), the over lapping of d-orbital of Fe^{3+} ($4S^2$ $3d^5$) and Cu^{2+} (4 S^2 $3d^9$) with the 2p of the oxygen has an indirect effect on the hopping process at B-sites. When the interaction between the electron and phonon is strong, a small polaron will be formed. As a result, the electrical conductivity in Cu–Si ferrite can be given by:

$$\sigma = \sigma_n + \sigma_p, \tag{2}$$

where σ_n and σ_p are the conductivities due to electrons and holes, respectively. The holes can be produced by Cu^+ formation through the sintering process due to the reduction of Cu^{2+} at high temperature (900–950 °C) [16]. According to Gardner et al. [17], the formation of each pair of Cu eliminates an oxygen atom. The existence of Cu^+ ions tend to initiate hopping conduction with Cu^{2+} and a hole e^+ is involved as following:

$$Cu^{2+} \Leftrightarrow Cu^+ + e^+$$

Since, the possibility of Cu^+ formation increases with the increase of x and the positivity increases. So that, T_k (the transformation point from n-type to p-type) decreases with increasing silicon content as shown in Fig. 2. For further increase of x, the Si^{4+} ions which locate at the A- sites push Cu^{2+} ions to the B-sites. Hence, this migration tends to increase the possibility of the hopping $Cu^{2+} \Leftrightarrow Cu^+$ and the activation energy tends to decrease with the increase of x as shown in Table 1. Since the hole transfer between, Cu^{2+} and Cu^+ due to p-type conduction and the n-type conduction is due to electron hopping between Fe^{3+} and Fe^{2+}, as a result the electrical conductivity can be expressed by:

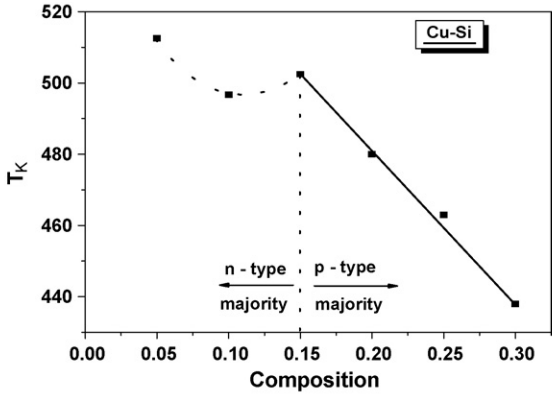

Fig. 2. The effect of composition on the kink point temperature.

$$\sigma = e(n_e \mu_e + n_h \mu_h). \tag{3}$$

where e is the electronic charge, μ is the mobility of charge carriers for electron (e) and hole (h), respectively and (n) is the total number of charge carriers per unit volume and usually for the narrow-band material (as ferrites) can be taken as 10^{22} cm^{-3} [18]. The drift mobility (μ) of the carriers was calculated over a wide rang of temperature employing the data of the d.c. conductivity according to the relation:

$$\mu = \sigma / n_e \tag{4}$$

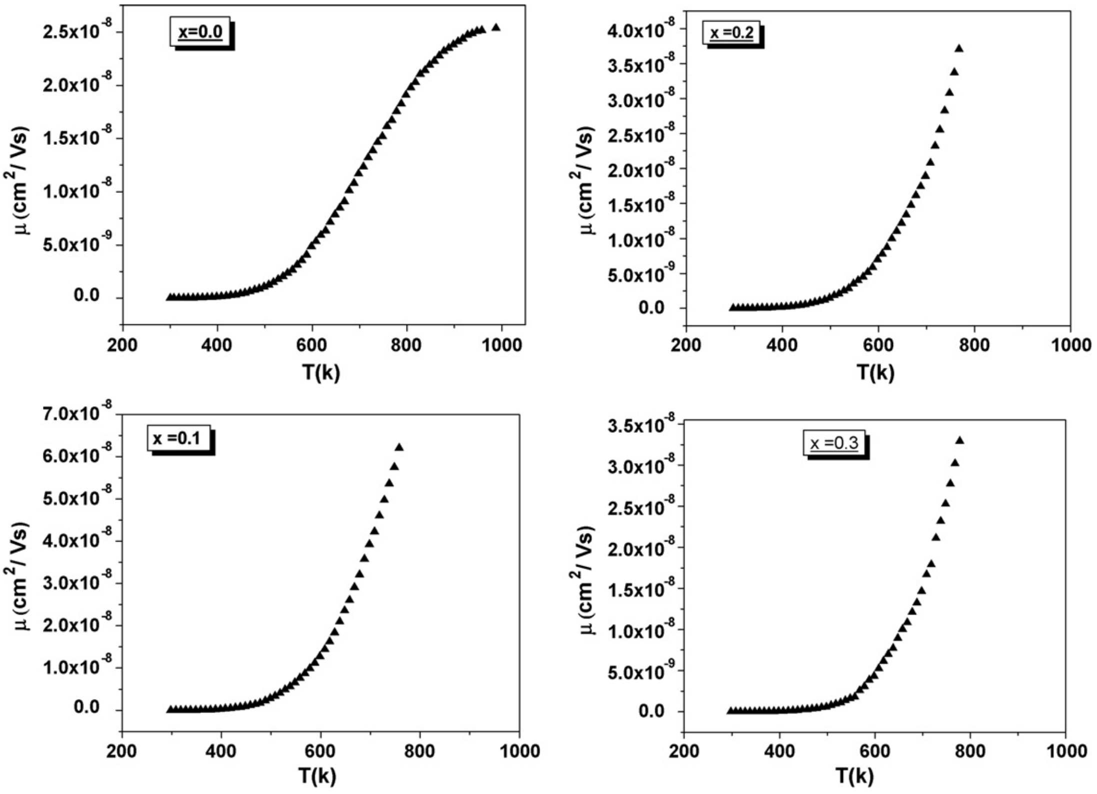

Fig. 3. Dependence of the drift mobility (μ) vs. the absolute temperature for $Cu_{1+x}Si_xFe_{2-2x}O_4$.

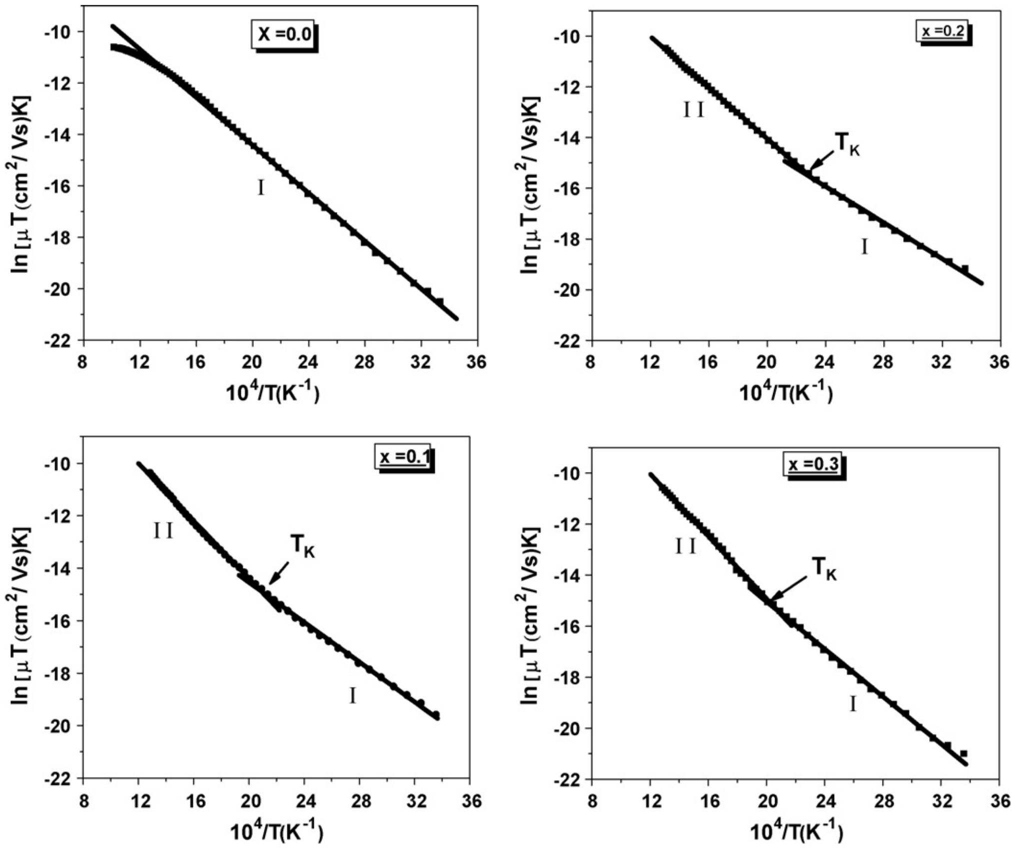

Fig. 4. The variation of ln μT vs. reciprocal temperature for Cu_{1+x} Si_x Fe_{2-2x} O_4; $x = 0.0$, 0.1, 0.2 and 0.3.

Fig. 3 represents the variation of μ with temperature for $x = 0.0$, 0.1, 0.2 and 0.3. The mobility behaviour indicates two regions, low temperature region $T < 500$ K where the mobility is independent on temperature and for $T > 500$ K the mobility is strongly dependent on temperature and increases exponentially with the increase of temperature. Such a trend has been observed in other ferrites [19,20]. The carriers mobility (μ) have values between 10^{-12} and 10^{-8} cm^2/V s. If a small polaron hopping conduction is applied, then the exponential increase of mobility could by associated with the localization of the charge carriers at ions or vacant sites. Due to the lattice vibrations, the ionic carriers are hopping from one site to another, thus leading to an exponential temperature dependence of the conduction. As a result, the mobility associated with carriers tends to increase exponentially with temperature according to the relation [21]:

$$\mu = \frac{\text{Const}}{T} \exp\left(\frac{-W_\mu}{kT}\right); \tag{5}$$

where W_μ is the activation energy for the mobilitization of charge carriers. The constant was mentioned in Eq. (1). Fig. 4 shows the relation between $\ln(\mu T)$ vs. $10^4/T$ and gives a linear relationship with two slopes before and after the kink point T_k. It is predicted that the temperature T_k lies in the range of $T \leq 500$ K. Comparing the values of W_H and W_μ for the corresponding region (either region I or region II), one finds that the both energies are nearly equal. This means that the energy used in moving the electrons (or holes) is not for generation of further charge carriers.

3.2. a.c. electrical conductivity

Over a wide range of temperature from 300 up to 783 K, the a.c. conductivity ($\sigma_{\text{a.c.}}$) was measured in the range of frequencies from $10^2 \rightarrow 10^6$ Hz for the above investigated composition as shown in Fig. 5. It can be seen that the studied ferrite shows a semiconducting trend, as commonly seen in most ferrites. The conductivity for all compositions between temperatures (453–573 K) seems to be frequency-independent, while dispersion was observed at the low temperature range. In the dispersion region the a.c. conductivity increases with increasing frequency. The increase in conductivity (σ) with temperature may be attributed to the increase in the drift mobility and hopping frequency of the charge carriers with increasing temperature. The experimental data of the a.c. conductivity of the investigated ferrite successfully comply with the following equation:

$$\sigma_{\text{a.c.}} = \sigma_0 \exp\left(\frac{-E_\sigma}{kT}\right) \tag{6}$$

where σ_0 is a temperature-dependent term and E_σ represents the activation energy of conduction mechanism. The behaviour of ln σ vs. T^{-1} can be divided into three main regions; the first region I [relatively low temperature region], the second region II [intermediate temperature region] and the third region III [the high temperature region] as shown in Fig. 5. In the dispersion range of region I, the activation energy E_σ decreases with increasing the frequency as shown in the Fig. 6 for all samples. In the second region II "intermediate region"; the conductivity at higher temperature seems to be frequency-independent and it is known as the d.c. part. The obtained values are tabulated in Table 2. For region III, a strange behaviour (as a metallic behaviour) has been seen at high temperature high frequency. This metallic behaviour is shown in insets of Fig. 5. According to this behaviour, the Matthiessen's rule can be applied for the total resistivity (ρ_T):

$$\rho_T = \rho_0 + \rho_{ph}(T) \tag{7}$$

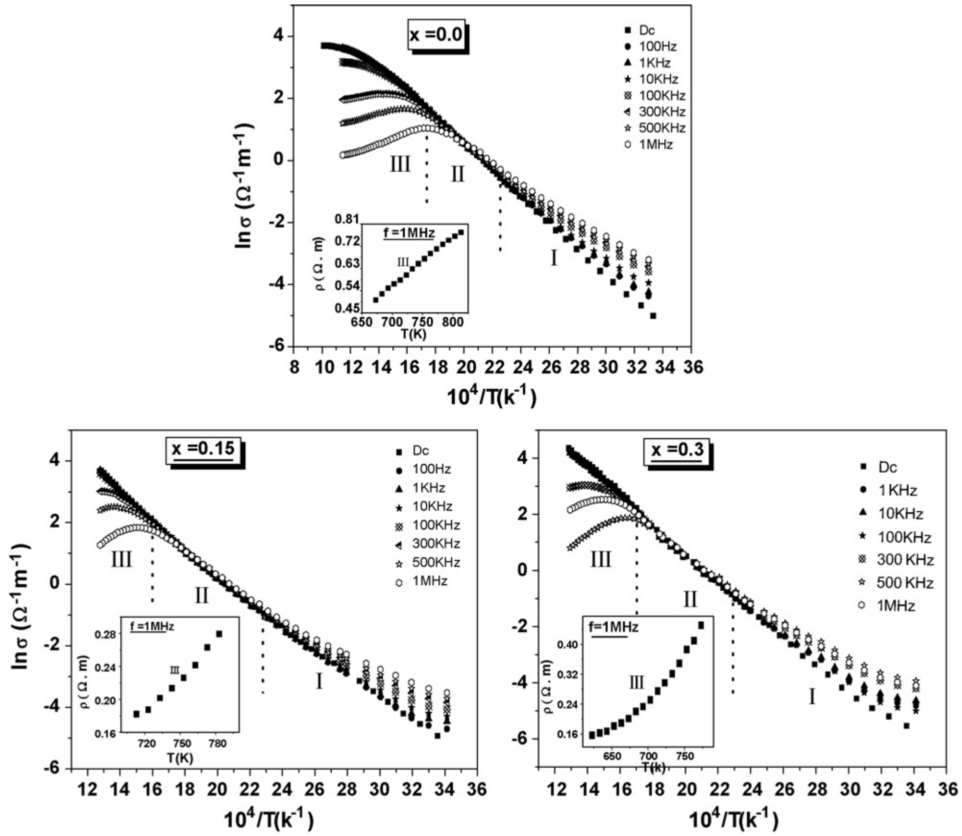

Fig. 5. The variation of ln σ as a function of 104/T (k^{-1}) at different frequencies for x = 0.0, 0.15 and 0.3.

where ρ_0 represents the impurity defects and it is predominant in the range of low temperature. While $\rho_{ph}(T)$ represents the resistivity by phonon scattering, the larger the amplitude of vibration at high temperature, the greater will be ρ_{ph}; while ρ_0 can be neglected.

The behaviour in the first region can be attributed to the a.c. hopping conduction of the localized carriers proposed by Pike [22]. This model has analyzed the a.c. conductivity for many oxides and amorphous materials. The present results are analogous to that observed by many researchers [23–25]. Also, it can be noticed that the activation energies associated with a.c. conductivity are bit lower than that of the corresponding energy involved in the d.c. conductivity as shown in Tables 1 and 2. This can be explained as follows; for the d.c. conductivity, the charge carriers choose the easiest path between ions, but these paths will be included some hops for which R "the distance between ions" is large. These are not important in a.c. conductivity. Thus rather lower activation energies may be involved in the a.c. conductivity than in the d.c. conduction.

3.3. *Dielectric loss tangent behaviour tan δ*

Fig. 7 is a set of typical curves indicating the variation of the dielectric loss tangent tan δ with the absolute temperature as a function of frequency ranging from 100 Hz to 1 MHz for the investigated composition of Cu-Si ferrite. By increasing temperature, tan δ begins to increase slowly until it attains a maximum value and then decrease with increasing the temperature. The temperature corresponding to the peaks (T_m) were found to be shifted to lower temperature with increasing the frequencies as shown inset in the Fig. 7. The electron exchange between $Fe^{2+}$and Fe^{3+}ions, and hole transfer between ($Cu^{2+} \leftrightarrow Cu^{1+}$) at octahedral sites are responsible for electric conduction in this ferrite, where the local displacements of localized electric charge carriers include dielectric polarization in ferrites. Therefore, the dielectric polarization increases causing a marked increase in tan δ as the temperature increase up to the peak values. At the resonance, where the applied frequency becomes equal to that of the ions, a relaxation peak appears, varying in position and shape depending on the Si^{4+} concentration for

Table 2
The values of activation energy from (a.c. conductivity) of all samples of Cu–Si ferrite.

x	Activation energy	10^2		10^3		10×10^3		100×10^3		5×10^5		10^6	
		I	II	I	II	I	II	I	II	I	II	I	II
0.0	E_σ	0.326	0 .351	0.324	0.347	0.315	0.34	0.305	0.33	0.295	0.32	0.29	0.31
0.05	E_σ	0.38	0.41	0.33	0.4	0.31	0.39	0.29	0.38	0.27	0.37	0.27	0.35
0.1	E_σ	0.37	0.4	0.34	0.39	0.33	0.39	0.33	0.385	0.32	0.37	0.3	0.35
0.15	E_σ	0.34	0.39	0.33	0.38	0.33	0.37	0.31	0.35	0.305	0.35	0.29	0.33
0.2	E_σ	0.33	0.37	0.32	0.37	0.32	0.35	0.31	0.33	0.29	0.31	0.28	0.31
0.25	E_σ	0.31	0.35	0.31	0.34	0.3	0.34	0.29	0.33	0.27	0.32	0.26	0.3
0.3	E_σ	0.3	0.34	0.29	0.33	0.29	0.31	0.27	0.31	0.26	0.3	0.24	0.29

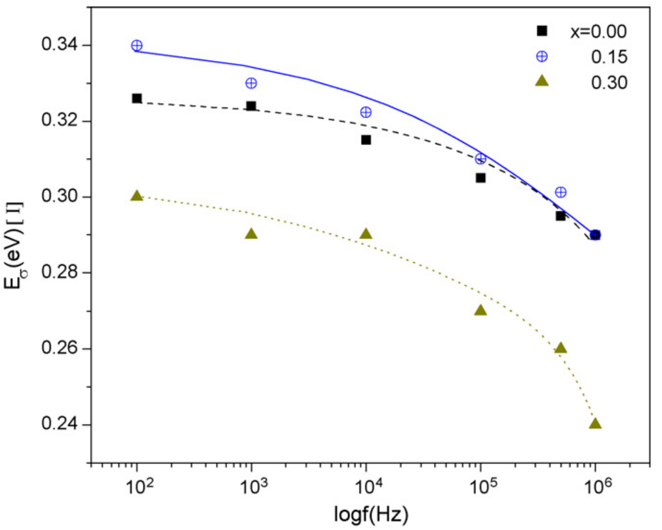

Fig. 6. The effect of frequency on the activation energy in the dispersion region (I), for $x = 0.0$, 0.15 and 0.3.

the compositions. After the relaxation peak and by increasing the temperature, tan δ, deceases until it reaches a flat region which observed clearly at high frequency. In the first region (before the peaks) the low frequency and low temperature helps in the aligning the dipoles in the field direction with the result of an increase in polarization as well as tan δ. Also, as the temperature increase the thermal energy liberate more localized dipoles and the field tries to align them in its direction either by rotational or orientational contributing to an increase in tan δ up to a peak value. In the second region (after the peaks) the temperature becomes relatively high and the diploes were distributed with the result of decease in tan δ [26]. As the temperature increases (in the flat region) the disorder becomes maximum. Moreover, the mobility of holes is smaller than that of electrons and the replacement of Fe^{3+} with Si^{4+} in Cu-Si ferrite decreases the number of charge carriers which means the more contribution of p-carriers to polarization will appear at high concentration of Si^{4+} ions and high temperatures. Also the shift of relaxation dielectric peak towards lower temperature with increasing frequency for the studied composition may be due to the decease in the hopping frequency for both types of charge carriers with increasing temperature.

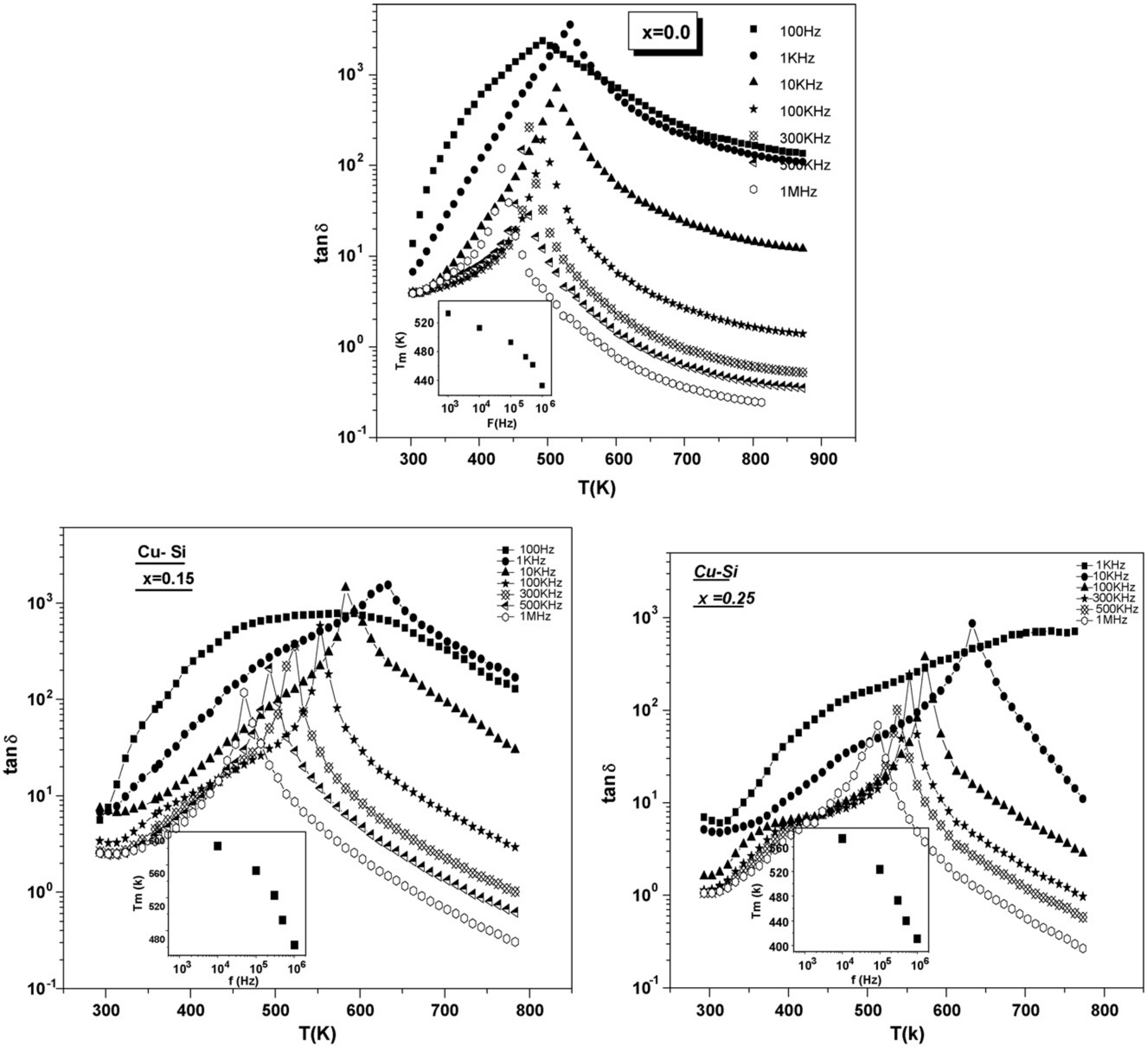

Fig. 7. Temperature dependence of dielectric loss tangent tan(δ) for $x = 0.0$, 0.15 and 0.25.

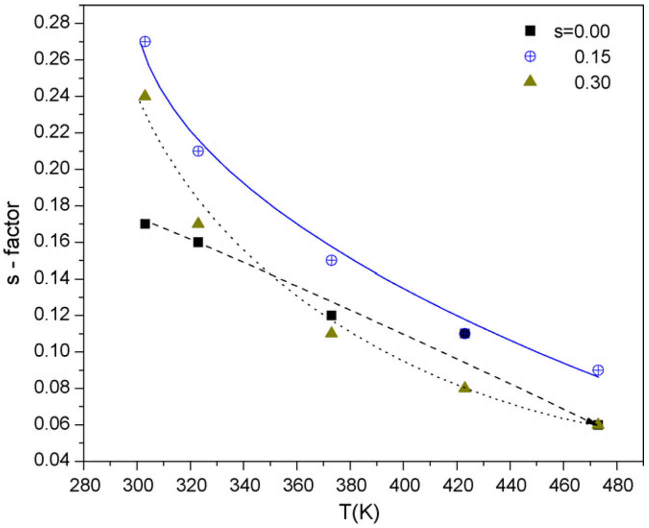

Fig. 8. The relation between (*s* vs. *T*) for *x* = 0.0, 0.15 and 0.3.

Moreover, in narrow-band semiconductors [27], the charge carriers are not free to move but are trapped causing a polarization, and also the thermal energy given at this range of temperature to the samples was not sufficient to free the localized dipoles to be oriented in the field direction. As the temperature increases, the thermal energy librates more localized dipoles and the field tries to align them in its direction either by rotational or orientation motion contributing to an increase in tan δ up to the maximum value, after which the thermal energy added to the system with electrical energy of the field decreases the internal viscosity, instantly, giving more degrees of freedom leading to a disturbance in the system and hence tan δ decreases again.

3.4. Determination of the frequency exponential factor

Because, the hopping conduction mechanism is assumed in the investigated system of Cu-Si ferrite, the a.c. conductivity $\tilde{\sigma}(\omega)$ can be represented by a power law as [22]:

$$\tilde{\sigma}(\omega) \propto A\omega^{s} \tag{8}$$

where A is little dependent on temperature, ω is the frequency at which the conductivity $\tilde{\sigma}(\omega)$ was measured and the power s, which is a weak function of frequency is to be determined for the investigated compositions. The value of s is usually, for physical convenience <1 [28]. Eq. (8) agrees quite well with the obtained results. These results may be explained by the hopping model proposed by Pike [22] which has analyzed the a.c. conductivity results obtained for many oxides and amorphous materials.

In some cases, the exponent s has a temperature dependent. In contrast with the tunneling model, temperature dependence can arise if the carrier form a polaron of binding energy $W_{p,}$ then the effective phonon frequency in this case can be written as [29]:

$$\nu_{ph} = \nu_o \exp\left(\frac{-W_p}{2kT}\right); \tag{9}$$

where ν_o is the frequency of the undistorted lattice. The exponential factor s can be taken the form [30]:

$$s = 1 - \frac{4}{\{\ln(\nu_o/\omega) - W_p/2kT}. \tag{10}$$

In the present work, Fig. 8 shows that s decreases with increasing of temperature for the investigated compositions of Cu–Si ferrite. According to quantum–mechanical tunneling model [21,28], the exponent *s* factor is temperature independent. The large overlapping polaron model [31] predicted that s decreases with increasing temperature up to certain temperature after which it begins to increase with further increase of temperature. The small polaron model and the classical hopping model over a barrier separating two sites [30] predicted that *s* decreases with increasing the temperature. This confirms the conduction mechanism which was discussed in the d.c. conductivity.

4. Conclusion

1. The variation of electrical conductivity of Cu–Si ferrite with temperature can be discussed based on a narrow-band conduction mechanism and a formation of small polaron.
2. The dielectric behaviour can be explained in terms of the electron exchange between $Fe^{2+}$and Fe^{3+}, and the hopping of a hole between Cu^{2+} and Cu^{+} ions at B-sites, suggesting that the polarization in these compositions is similar to that of the conduction process in ferrites.
3. Abnormal behaviour (peaks) was observed in tan δ curves at relatively high temperatures. Such relaxation peaks take place when the jumping frequency of localized electrons between Fe $^{2+}$ and Fe^{3+} ion equals that of the applied a.c. electric field.
4. The exponent factor *s* was calculated and found to be acceptable in the range of the reported values and decreases with increasing of temperature for the investigated composition.

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