Electrical and dielectric properties of $Se_{100-x}In_x$ Chalcogenide glasses

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Abstract: *Glasses of the system $Se_{100-x}In_x$ (where x= 0, 6, 16, 21 at %) were prepared by the usual melt quenching technique. The amorphous nature of this system was identified using x-ray diffraction.*
The a.c conductivity (σ) of this system has been investigated in the frequency range 200 Hz – 10 kHz, and in the temperature range 292 – 374 K. The temperature dependence of (σ) for all tested values of x was reasonable and interpreted using correlated hopping model(CHM). The density of states N (E) was calculated and found to be 10^{27} m^{-3}. Using Austin and Mott model. The dielectric constant and the dielectric loss decrease as the frequency increases at constant temperature. On the other hand the dielectric constant and the dielectric loss were increased as the temperature increased at a constant frequency.

I. Introduction

Several measurements were carried out on chalcogenide glasses [1, 2, 3, 4], because of the great importance of their physical properties [5,6]. The electrical properties of the disordered materials are dominated by the deep defect levels lying in the mobility gap of these materials. Many theoretical explanations [7, 8] for the ac conduction in amorphous materials have been proposed to explain the temperature and frequency dependence of the dielectric constant and dielectric loss factor. The a.c conduction gives more information about the density of states N (E) near the Fermi level in the amorphous semiconductors; therefore we use it in this paper to explain the conduction mechanism in the Se- In system.

II. Experimental technique

2.1 Preparation of the samples:

Glassy samples of the system $Se_{100-x}In_x$ (where x = 6, 11, 16, 21, at %) were prepared by the melt quenching technique [9]. Spectrally pure Se, and In powders were weighed well mixed and sealed in evacuated silica glass tubes. These tubes were introduced into an electric furnace maintained at 400 C^o for 5 hours. During the melting process, the tubes were frequently agitated in order to intermix the constituents and ensure homogeneity of the melt. At the end of this time the tubes containing the melts were quenched in ice water. The ingot samples were obtained and prepared for a.c measurements.
The amorphous nature of the samples was verified by means of a shemadzu X-ray difractometer [model XD-3]

2.2 Conductivity measurements

The a.c conductivity measurements were carried out by means of a complex impedance technique. A constant voltage was applied on the samples and the current passing through the samples was determined by measuring the potential difference across an ohmic resistor using a lock-in amplifier (Stanford Research System SR 510). The lock-in amplifier simultaneously measured the voltage across the resistor and the phase difference between this voltage and the applied voltage.

III. Results and discussion

3.1 A.c conductivity

The effect of frequency and temperature on the a.c conductivity $\sigma_{ac}(\omega)$ of the system Se_{100-x} In_x (where x =6, 11, 16, 21 at %) at different constant frequencies and at constant different constant temperatures were studied. Figure (1) and (2) represent the temperature and frequency dependence of a.c conductivity of the sample Se_4In_6. Figure. (1) Shows very good fitness to the equation:
$\sigma_{ac}(\omega) = A. \omega^s$ (1)
where ω is the angular frequency, A and s are constants. The exponent (s) was determined for all samples, (x = 6, 11, 16, 21 at %) and given in table (1).
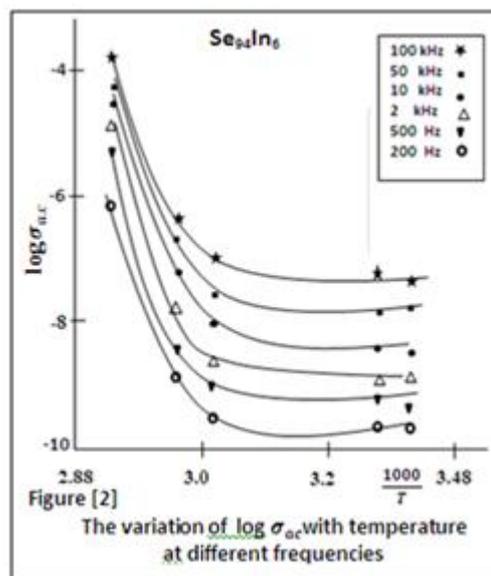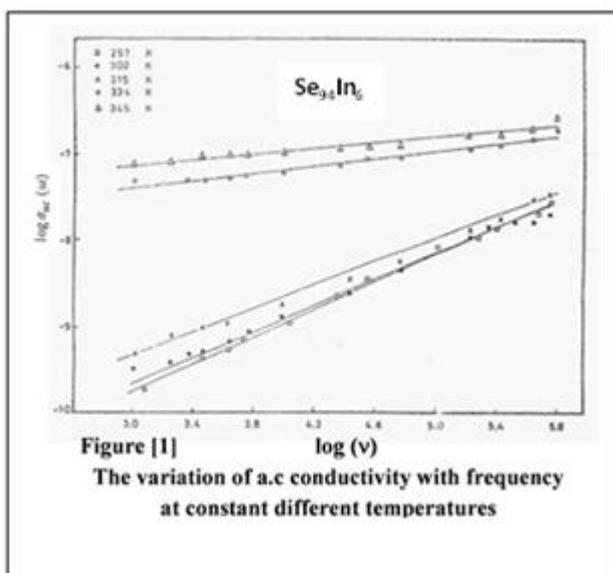
Figure [1]
The variation of a.c conductivity with frequency at constant different temperatures

Figure [2]
The variation of log σ_{ac} with temperature at different frequencies

Table [1]: The calculated value of s for all samples at different temperatures

x = 21 at%		x = 16 at%		x = 11 at%		x = 6 at%	
s	T(K)	s	T(K)	s	T(K)	s	T(K)
0.100	298.0	0.830	309.0	0.509	292	0.750	297.0
0.250	307.0	o.720	318.0	0.380	312	0.708	302.5
0.130	339.0	0.690	333.0	0.250	324	0.700	325.0
0.100	343.0	0.430	338.0	0.115	335	0.192	334.0
0.100	347.0	0.163	343.0	0.100	342	0.142	345.0

This is in good agreement with the values previously reported [9].

Austin and Mott [10] adapted the analysis to the situation likely to be appropriate for the amorphous semiconductors, and then the a.c conductivity is given by:

$$\sigma_{ac}(\omega) = \frac{\pi}{3}.e^2 kTN^2.\alpha^{-2}.\omega.[\ln(\frac{v}{\omega})]^4 \quad (2)$$

Where $\alpha^{-1} = 8A^o$, N is the density of states and $v = 10^{13}$ s^{-1}

The correlated barrier hopping model proposed by Elliot [11] has been applied to the chalcogenide glassy semiconductors. In this model the electrons in charged defect state hop over columbic barrier whose height is given by the formula:

$$W = W_M - \frac{4ne^2}{\varepsilon.R} \quad (3)$$

Where W_M is the maximum barrier height of the energy band, ε is the dielectric constant, e is the electronic charge, n is the number of electrons that hop (in case of chalcogenide glass n = 2) and R is the distance between the hopping sites. The relaxation time τ for electrons to hop over a barrier of height W is given by the formula:

$$\tau = \tau_0 e^{(\frac{W}{Kt})} \quad (4)$$

Where τ_0 is of the order of 10^{-13} s. in the range of an atomic vibrational period, and k is Boltzmann constant. Plotting the relation between the relaxation time and temperature, the barrier height W_M can be deduced.

Substituting into equation (3), the values of W_M were estimated and given in table (2). The final expression for a.c conductivity proposed by Elliot [11] is given as:

$$\sigma_{ac}(\omega) = \frac{\pi^2.N^2.\varepsilon}{24}[(\frac{8e^2}{W_M.\varepsilon})^6 x(\frac{\omega^5}{\tau.\beta})] \quad (5)$$

Where N is the density of the localized states while β is given by :

$$\beta = \frac{6KT}{W_M} \quad (6)$$
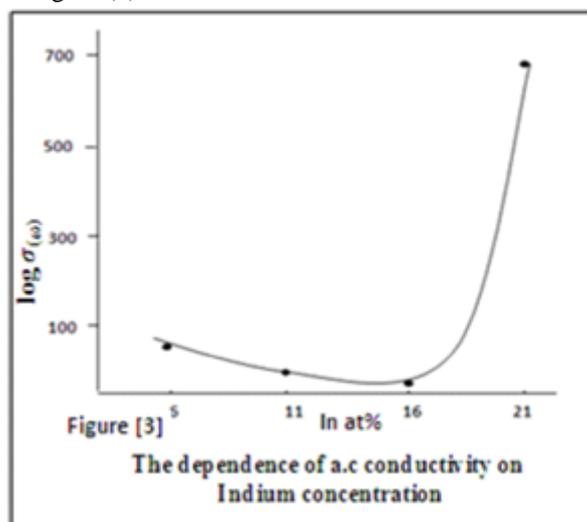
And $\quad s = 1 - \beta \quad (7)$

From equations (5, 6, 7) the density of states N (E) could be calculated at room temperature and frequency 10 kHz. The results are given in table (2).

Table [2]: The calculated W_M and N€ values for all compositions
At room temperature and at a frequency 10 kHz .

N (Elliot)	N (Mott)	$\sigma_{ac}(\omega)$	W_M(eV)	β	s	In at%
1.95×10^{16}	2.03×10^{27}	0.90×10^{-7}	0.6137	0.250	0.75	6
1.19×10^{15}	15.08×10^{27}	0.27×10^{-7}	0.3070	0.490	0.509	11
5.50×10^{17}	1.48×10^{27}	8.70×10^{-9}	0.9380	0.170	0.803	16
3.32×10^{16}	34.60×10^{28}	7.40×10^{-7}	0.2050	0.750	0.250	21

The calculated values of N (E) according to Elliot model were convenient and reasonable to be applied for our chalcogenide glasses, since it is comparable to those previously reported [12].

Figure (2), shows the temperature dependence of $\sigma_{ac}(\omega)$. It can be noticed that $\sigma_{ac}(\omega)$ increases as the temperature increase, the rate of increment was slow at first then increases as the temperature increase, this may be attributed to the hopping of electrons due to the effect of the applied electric field only in the slow rate region, and due to the applied electric field in addition to the thermal excitation energy in the fast region. The effect of increasing Indium content in the system $Se_{100-x}In_x$ on the a.c conductivity in the hopping region at 10 kHz and at 303 k, is shown in Figure (3).

Figure [3]
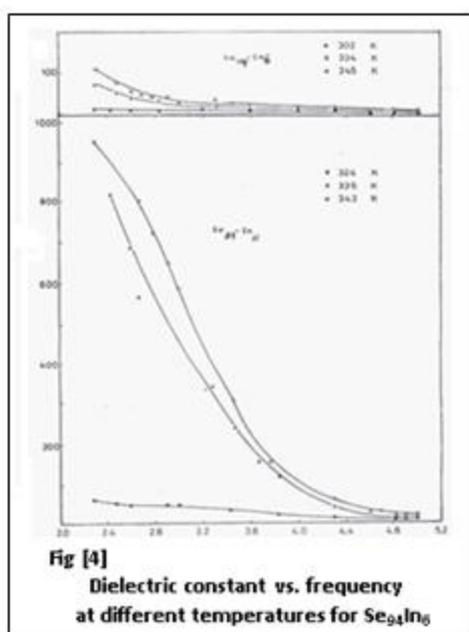The dependence of a.c conductivity on Indium concentration

It shows that $\sigma_{ac}(\omega)$ has a minimum value at(Se 16 at% In) and a maximum value at (Se 11 at% In). In the hopping region, $\sigma_{ac}(\omega)$, decreases smoothly as In content increases from 6 % up to 16 %, then it increases with a faster rate from 16% to 21% In content. This may be attributed to the increase of the weak (In-In) bonds concentration, on the expense of the strong (In-Se) bonds. This in turn, increases the band tail width and enhances the conduction through a narrower energy barrier [13, 14].
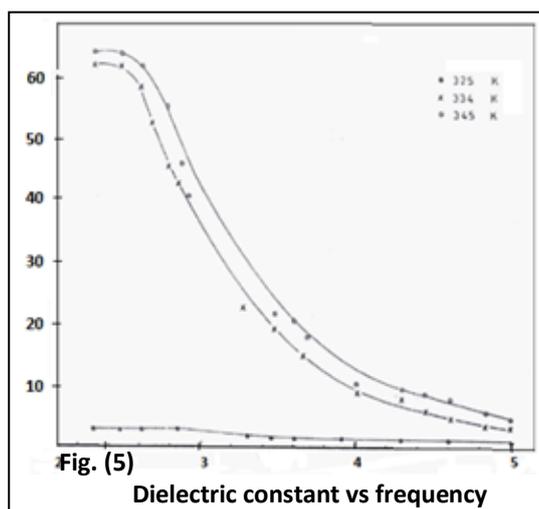
3.2. Dielectric constant and dielectric loss
3.2.1. Frequency dependence of (ε) and tan (δ)
The frequency dependence of the dielectric constant (ε) and the dielectric loss tan (δ) at different isotherms for all samples of the system ($Se_{100-x}In_x$), where (x=6, 11, 16, 21 at%) have been studied within the frequency range 200Hz – 100 kHz as indicated in figures (4, 5).

These curves show that both of **(ε) and tan**(**δ**), start as a high value and then drastically decrease with a high rate at lower frequencies, while they decrease at a slower rate as frequency increases for the dielectric constant.

Fig [4]

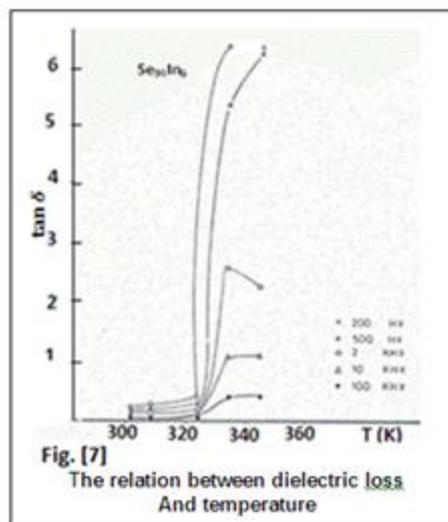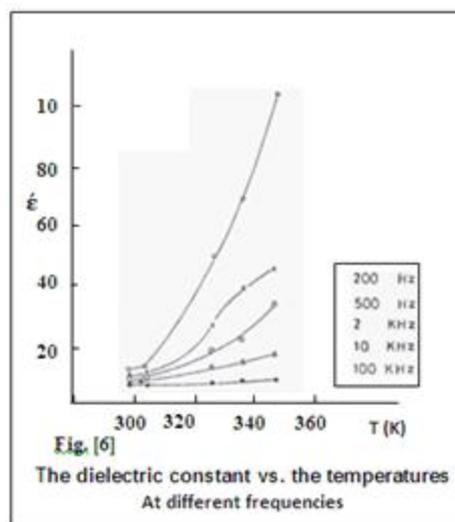**Dielectric constant vs. frequency at different temperatures for Se$_{94}$In$_6$**

This behavior may be attributed to the contribution of multi components to the polarizability of the glass semiconducting material at low frequency, and as the frequency increases, the ionic and orientational sources of polarizability decrease and finally disappear due to the inertia of the molecules and ions. This will lead to the observed gradual decrease in the dielectric constant values.

Fig. (5)

Dielectric constant vs frequency

The electronic polarization is sufficiently rapid to follow the alternative fields, at high frequencies, as well as the ionic polarization too. Dielectric constant and dielectric loss values decrease as the frequency increases. The high values of the dielectric constant and dielectric loss at low frequencies may be attributed to the strong share of all polarizing components in the process. While at higher frequencies, some components are withdrawing from the contribution and only electronic polarization keeps sharing.

and dielectric loss at different constant frequencies for all the system (Se$_{100}$-xIn$_x$) where, (x = 6,11,16,21 at %) been studied.

The temperature dependence of (ε)and$\tan\delta$:

Dielectric constant and dielectric loss at different constant frequencies for all samples were studied. Figures (6, 7) show that the dielectric constant and the dielectric loss for all samples generally increase as the temperature increase. The increase in(ε) at low temperature may be attributed to the small contribution of the electronic and ionic components in the polarization, while neglecting the orientation component.

Fig. [6]
The dielectric constant vs. the temperatures
At different frequencies

Fig. [7]
The relation between dielectric loss
And temperature

As the temperature increases the electronic and ionic polarizability sources start to increase to higher values.

Figure. (7) Shows that the dielectric loss starts with small values for all samples, then increases with accelerated rate as the temperature increases. This may be attributed to the fact that at low temperature, the relaxation loss is the main contribution and appeared to be very much greater than the conduction loss. As the temperature increases, the relaxation loss increases more and more rapidly.

IV.   Conclusion

In the system ($Se_{100-x}In_x$) where (x = 0,6, 11, 16 ,21at%), The a.c conductivity, increases with frequency and temperature. In the same time it reaches a highest value at x = 21 at%, suffering from a minimum value at x = 16 at%. This means that the best conduction ratio is 21 at%. The dielectric constant and dielectric loss have a reverse behavior to the a.c conductivity, with the frequency, but both of them, revealed the same sense as the ac conductivity, with temperature.

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