DC AND AC CONDUCTIVITY MEASUREMENTS OF Se$_{60}$Te$_{30}$Ge$_{10}$ FILMS

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Abstract

D.c. $\sigma(0)$ and a.c conductivity $\sigma(w)$ and dielectric parameter (ε and $\tan\delta$) of Se$_{60}$Te$_{30}$Ge$_{10}$ glassy thin films which were prepared by thermal evaporation were investigated. Measurements of a.c. conductivity over frequency range (100Hz-100kHz) and temperature range (303-363K) showed that the a.c. conductivity follows the empirical formula $\sigma_{a.c}(w)=Aw^s$, where (s) lies between (1.03-0.60).In the glasses of Se$_{60}$Te$_{30}$Ge$_{10}$ thin films the d.c. conductivity along localized states prevails. A.c conductivity is explained by hopping mechanism of charge transfer between two defect centers.

Introduction

For many years, it was believed that the physical properties of Chalcogenide glasses can not be modified by foreign atoms. Doping could not be achieved with conventional impurities. Recently however, P to n transition has been reported [1-4] in binary Ge-Se chalcogenide alloy, when a third element is introduced in this alloy.

The common feature of these glasses is the presence of localized states in the mobility gap. This is due to the presence of short range order as well as various inherent defects. In Chalcogenide glasses, it is assumed that the localized states in the mobility gap are the charge defects D^+ and D^- with negative effective correlation energy [5]. Selenium based Chalcogenides have attracted much attention due to their device applications such as rectifiers, photocells, xerography, switching and memory devices[6].

Chalcogenide glasses are vitreous materials having one or more of the Chalcogenide elements (group VI):Sulfur(S), Selenium(Se), and Tellurium(Te). The addition of the network formers (group IV and V) such as silicon(Si), germanium(Ge), tin(Sn), phosphorus(P), arsenic(As), and antimony(Sb) establishes cross-linking between the tetrahedral and pyramidal units which facilities stable glass formation[7]. Depending on the composition the Chalcogenide glasses are stable against crystallization and are chemically inert. They are excellent thermal stability, and are relatively easy to fabricate. Optical properties for Ge$_{10}$Se$_{90-x}$Te$_x$ (x=0, 30)

thin films are calculated by Sharma et al.[8] who found that the optical band gap is a bond sensitive property so the decrease in average bond energy of the system leads to decrease in the optical band gap. The glass transition temperatures for Ge-Se-Te glasses depend on both heating rate and composition [9].

We shall focus in this paper to the study of the electrical as well as dielectrical properties of Ge-Se-Te pure glasses.

Experimental

Alloy of Se$_{60}$Te$_{30}$Ge$_{10}$ glass alloy was prepared by melting the starting elements (weighed according to their atomic percentages and sealed in quartz ampoules and evacuated to pressure 10^{-2} Torr). The sealed ampoule is kept inside a furnace where the temperature is increased up to 1000^0C. The ampoule is frequency rocked for 7 hours at the highest temperature to make the melt homogeneous to avoid phase separation. the quenching is done in ice cold water. Thin films of glassy alloy are deposited on glass substrates by vacuum evaporation technique at room temperature and bas pressure of ~10^{-5} Torr using a molybdenum boat.

D.C measurements have been done using sensitive digital electrometer type kethley (616) to measure the current, also d.c. power supply (2 Volt).A.C. measurements have been achieved using HP-RLC unit model 4274A and 4275 multi-frequency LRC meter, in the range of (100-10MHz),with amplitude of 0.08 volt and zero bias, the measurements were

performed at different temperatures from 30-90 ^{0}C.

Results and Discussion
1- D.C Conductivity:

We can calculate the activation energy (E_a) according to equation ($\sigma_{dc}=\sigma_0 \exp(-E_a/k_BT)$) where k_B is Boltizman constant and T is absolute temperature, so the temperature dependence of direct electrical conductivity, dc, are plotted in Fig.(1). The activation energy E_a of the glasses decreases with increasing annealing temperature, T_a, this decrease due to the dense in the V.B. & C.B., the effect is shown by the improvement the structure during annealing process. These results are shown in Table (1).
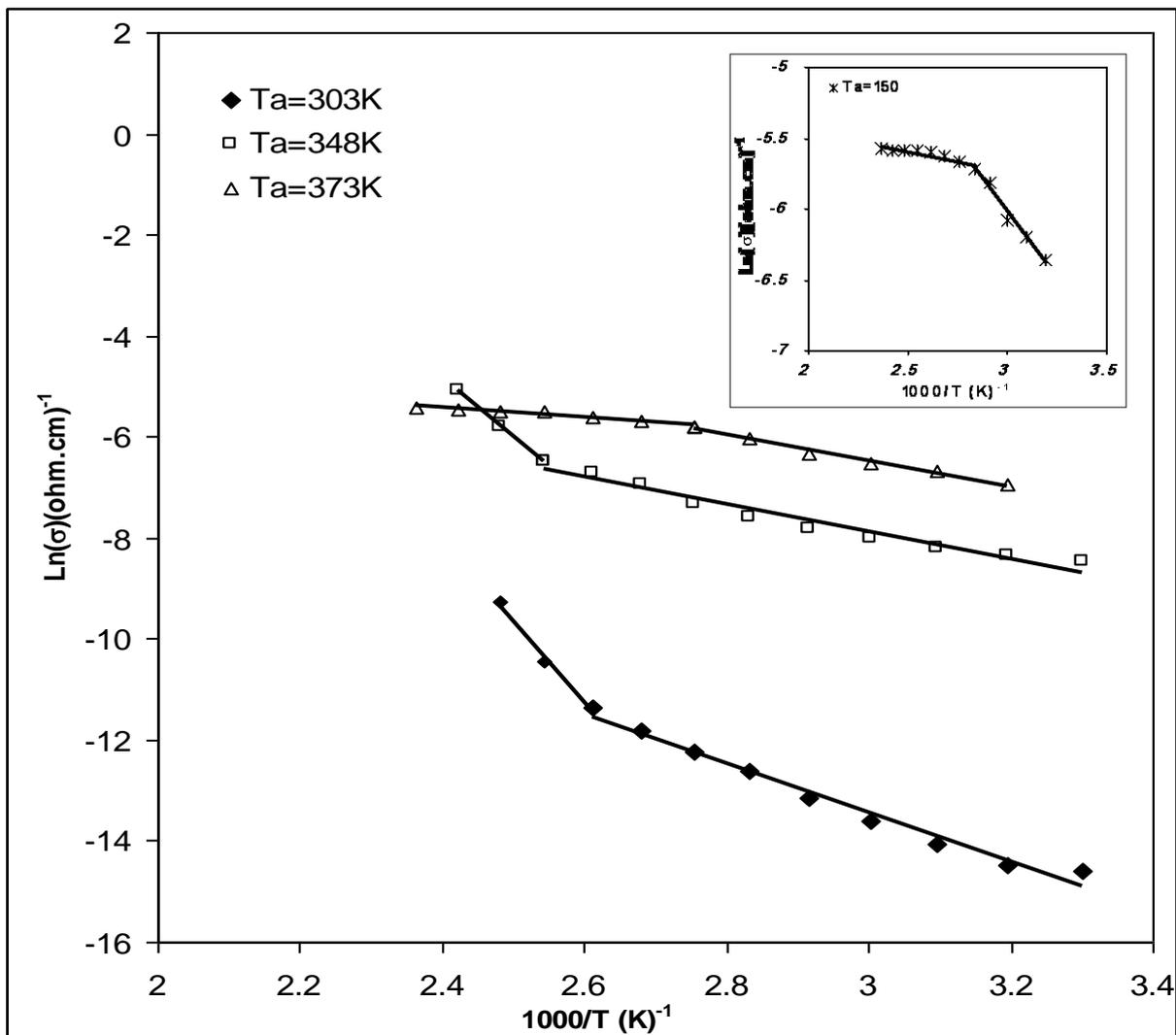

Fig.(1): Temperature dependence of σ_{dc} at different annealing temperature of $Se_{60}Te_{30}Ge_{10}$ thin films.

This figure also illustrates that at $T_a=373K$, the E_a at higher temperatures is less than that at lower temperatures, this behavior occurs in the Chalcogenide semiconductor when these material reach near glass temperature, T_g, this is due to that the concentration of defect centers quenched from the glass transition temperature could be small enough to give the previous behavior [10].

Table(1)
D.C conductivity and activation energy of Se₆₀Te₃₀Ge₁₀ thin films.

| T_a(K)	E_{a1}(eV)	σ_{01}(ohm.cm)$^{-1}$	E_{a2}(eV)	σ_{02}(ohm.cm)$^{-1}$	$\sigma_{R.T}$(ohm.cm)$^{-1}$
303	0.422	3.56	1.10	$1.71*10^{13}$	$4.67*10^{-7}$
348	0.232	1.246	0.986	$0.659*10^{10}$	$2.13*10^{-4}$
373	0.218	3.07	0.086	0.049	$9.55*10^{-4}$
423	0.159	0.614	0.023	0.0073	$17.33*10^{-4}$

2- AC- Conductivity:
A) Frequency Dependence:
1. Capacitance:

Fig.(2a) shows the variation of capacitance with angular frequency at room temperature. At low frequency there is a sharp decrease of capacitance with increasing frequency, this can be appropriately explained on the basis of decreasing in space charge region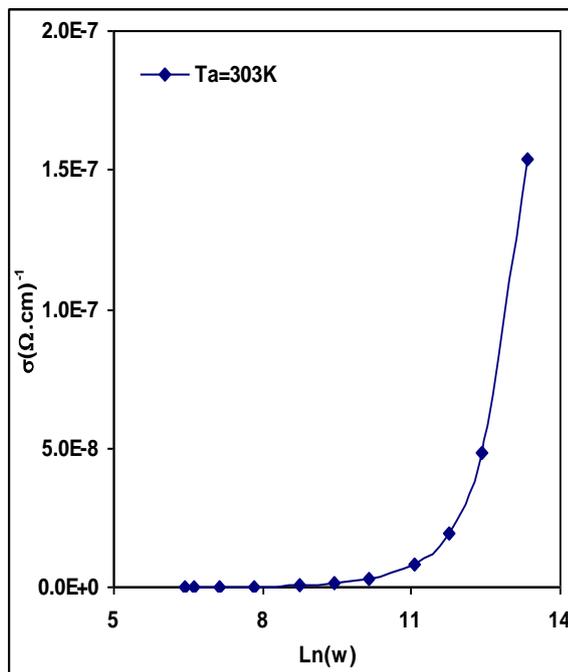 at the electrodes. At high frequency region, there is a slight decrease of capacitance, this can be attributed to the partially blocked charge carriers near the electrodes [11]. Fig.(2b) shows the same capacitance behavior at different temperature values (323, 343 and 363K), which can attributed to the same mechanism, but it is clear that the capacitance decreases rapidly as the temperature decreases.
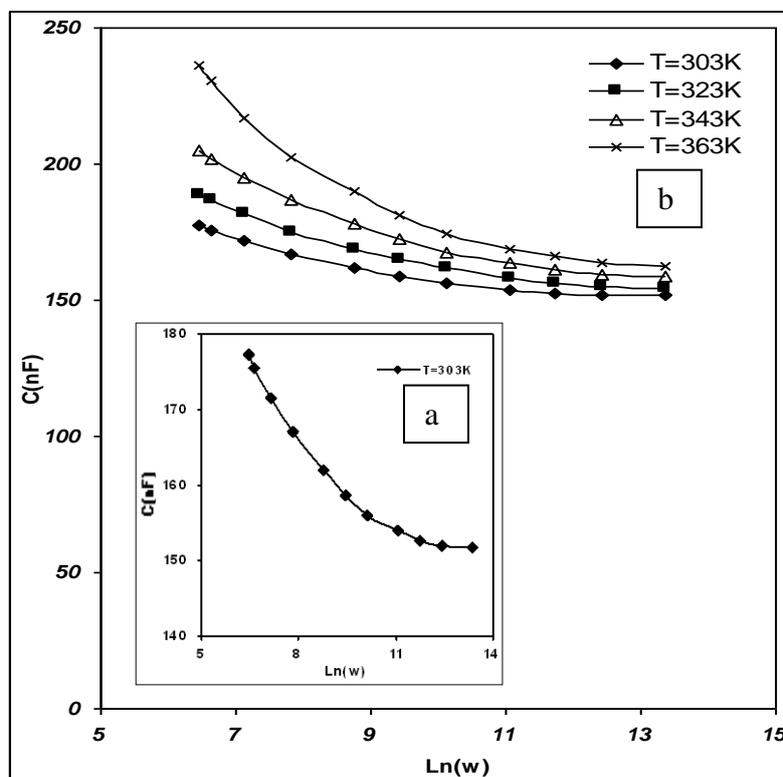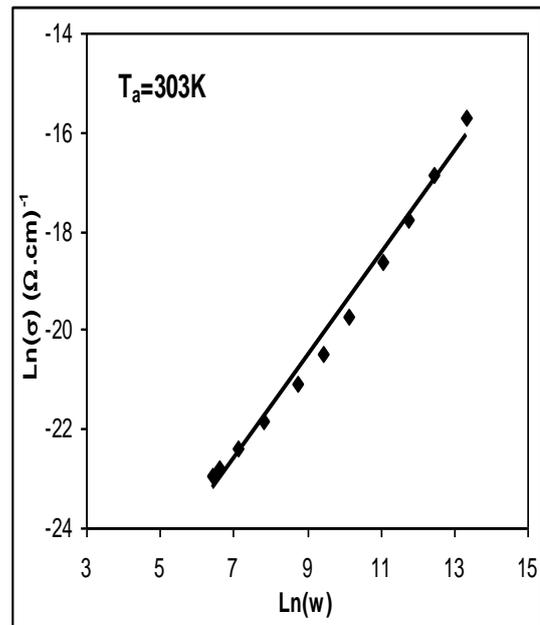

Fig.(2): Variation of capacitance (c) with Ln(w) at different temperature.

2- Complex conductivity:

The conductivity increases with increasing angular frequency as shown in Fig. (3). Ionic polarization has an effect in low frequency region where the polarization is slightly changing and $\sigma_{tot}(w)$ becomes less dependent with frequency because d.c component of conductivity dominates in this region of frequency. At high frequency region, the increase of $\sigma_{tot}(w)$ with increasing frequency is due to electronic polarization and hopping, since the space charge is fluctuating a cross the interface and thus causing intrinsic conduction [12], that means the a.c conductivity dominates at higher frequencies region. Fig.(4) shows a plot of Ln $\sigma_{tot}(w)$ versus Ln (w), at room temperature. The a.c conductivity has been found to vary according to the relation ($\sigma_{tot} \sim w^s$). From this curve we can calculate the value of the exponent (s), also we found that the exponent s decrease with annealing process which causes decreases in σ_{tot} .

Fig.(4) : Variation of Lnσ with Lnw at room temperature.

3-Complex permittivity plot (Cole-Cole Diagram):

Complex permittivity ($\varepsilon^*=\varepsilon_r$-$i\varepsilon_i$) contents dielectric constant ($\varepsilon_r=ct/\varepsilon_0A$) and dielectric loss ($\varepsilon_i=\sigma_{tot}(w)/\varepsilon_0A$),where *A* is the effective erea, ε_0 space permittivity and *t* is the thickness of the sample. The calculated points of the real and imaginary parts of the complex permittivity diagram are shown in Fig.(5). The intersect of the semicircle with ε_r axis gives the static dielectric constant (ε_s). Our experimental Cole-Cole diagram diverted from that of Debye, where the center of the semicircle is lying on a line depressed by an angle *(απ/2)* with respect to the real axis ε_r, and the same for the position of the point at top of the semicircle in which $w_p\tau_p=1$, where α is the polarizibility, w_p and τ_p are frequency and time relaxation at the top of semicircle. The values of (ε_s) and (α) are tabulated in Table (3).

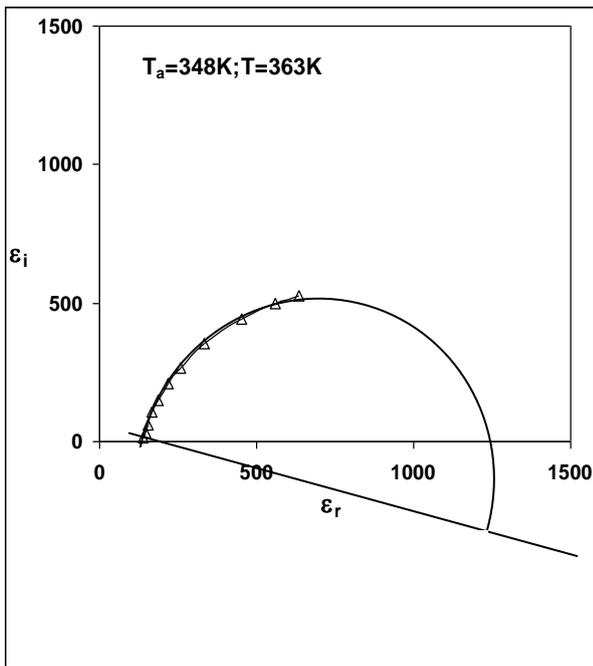**Fig.(3) : Variation of conductivity with Ln(w) at room temperature.**

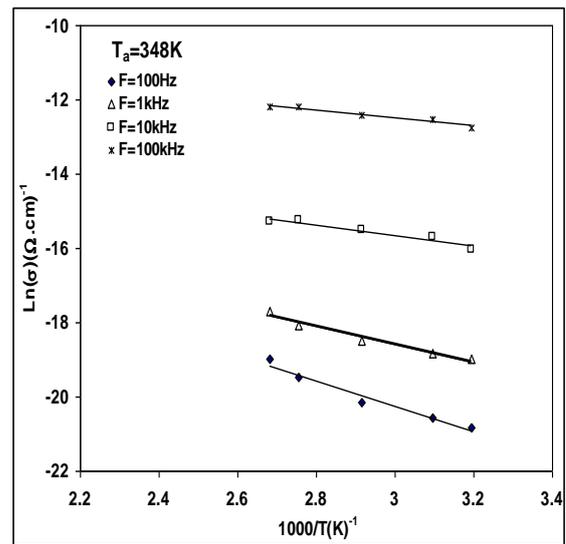*Fig.(5): Cole-Cole diagram for $Se_{60}Te_{30}Ge_{10}$ thin film.*

Fig.(6): Temperature dependence of $\sigma_{tot}(w)$ at different frequencies.

B) Temperature dependence:
1. A.C conductivity and Activation energy:

Fig. (6) shows the temperature dependence of the a.c. conductivity at different frequencies for ($Se_{60}Te_{30}Ge_{10}$) thin films. The physical reason for this temperature dependence may be the necessity to provide a certain amount of energy from lattice vibration to overcome the slight difference between energy levels of the initial and final states. The difference in energy levels may be due to the applied field and the stray coulomb potential of charged centers in the lattice [11]. The values of the activation energy are calculated from the plot of log conductivity against reciprocal temperature at various frequencies. At low frequency, the a.c conductivity is small and the activation energy is large, this behavior is due to ionic conduction but at high frequencies, the a.c conductivity is large and the activation energy is low.

The physical reason for this behavior is that the ionic conduction is characterized by low mobility (i.e. low conductivity) and high activation energies, while electronic conduction is associated with relatively higher mobilities (i.e. high conductivity) and lower activation energy [13].[see Table (2)].

Table (2)
A.C activation energy for $Se_{60}Te_{30}Ge_{10}$ thin films.

T_a(K)	E_w (eV) at			
	(100Hz)	**1kHz**	**10kHz**	**100kHz**
303	0.514	0.464	0.461	0.429
348	0.296	0.207	0.120	0.092
373	0.152	0.145	0.072	0.015
423	0.141	0.118	0.087	0.01

2- Dispersion parameter (α) and static Dielectric constant (ε_s):

There is a symmetric distribution of relaxation time around a central value, broadening of the Debye curve, and a flatting of the semicircular Cole-Cole diagram, which has been occurred when the temperature is increasing. This behavior, is due to relaxation time process, with continuous distributed relaxation time. The basic relaxation time can be ascribed to the difference in the electrical properties between he electrodes, (such as variation in the density of free charge carrier with changing in temperature). For this reason (ε_s) is increasing with increasing temperature, as shown in Table (3) [14,15].

This table indicated that (α) is increasing with increasing temperature. This is because an increasing in thermal energy will weaken the forces of intermolecular interaction [15].

Table (3)
The values of s, ε_s and α for $Se_{60}Te_{30}Ge_{10}$ thin films.

T_a (K) / T (K)	303K			348K			373K			423K		
	s	ε_s	α	s	ε_s	α	s	ε_s	α	s	ε_s	α
303	1.03	24	0.6	0.846	325	0.58	0.67	405	0.50	0.64	233	0.5
323	0.98	36	0.63	0.817	475	0.62	0.65	445	0.44	0.64	300	0.55
343	0.87	110	0.45	0.79	610	0.58	0.63	500	0.36	0.64	350	0.5
363	0.69	172	0.17	0.64	1170	0.43	0.62	655	0.38	0.63	680	0.5

Conclusion

The temperature dependence of direct electrical conductivity consisted that activation energy of the glasses decreases with increasing annealing temperature. The ac-conductivity has been found to vary according to the relation $\sigma \sim w^{s(T)}$ and show less dependence on temperature at higher frequencies.

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الخلاصة

تم دراسة التوصيلية المستمرة والمتناوبة وثابت العزل لأغشية المركب$Se_{60}Te_{30}Ge_{10}$ والتي حضرت بطريقة التبخير الحراري في الفراغ. وجد ان قياسات التوصيلية المتناوبة في مدى الترددات (100Hz-100kHz) ودرجات حرارة (303-363K) تخضع للعلاقة التجريبية $\sigma_{a.c}(w)=Aw^s$ حيث (s) ان تتراوح قيمها بين (1.03-0.60) وجد ان التوصيلية الكهربائية المستمرةلأغشية المركب $Se_{60}Te_{30}Ge_{10}$ العشوائية تحدث بسبب الانتقال بين المستويات الموضعية تم تفسير نتائج التوصيلية المتناوبة وفقا لميكانيكية تنطط حاملات الشحنة وانتقالها بين مراكز العيوب.