

The Effect of Tellurium Content(x) on the Optical and Electrical Properties of (Se_{1-x}Te_x) Thin Films

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Abstract:

Thin films of Se_{1-x}Te_x were fabricated by co evaporating the separate elements under vacuum on glass substrate. Careful was used in order to obtain a wide range of composition i.e. (Se, Se_{0.9}Te_{0.1}, Se_{0.8}Te_{0.2}, Se_{0.7}Te_{0.3}, Se_{0.6}Te_{0.4}, Se_{0.5}Te_{0.5},.....,Te). The transmittance and the absorbance were carried out in the spectral range (400-2500nm) and the values of optical energy gap (E_g), the optical constants (refractive index(n), the extinction coefficient(k), the real (ε₁) and imaginary (ε₂) part of dielectric constant were calculated. The effect of increasing tellurium content (x) in selenium films on the optical parameters was discussed. The results for electrical properties like d.c conductivity (σ_{DC}), concentration of charge carriers (n_H) and the activation energy (E_a) were also found which support those of the optical results.

ملخص:-

تم تحضير أغشية رقيقة من Se_{1-x}Te_x بطريقة التبخير المزوج للعنصرين على أرضية زجاجية تطلبت العملية عناية فائقة للحصول على مدى واسع من المكونات بعبارة أخرى (Se, Se_{0.9}Te_{0.1}, Se_{0.8}Te_{0.2}, Se_{0.7}Te_{0.3}, Se_{0.6}Te_{0.4}, Se_{0.5}Te_{0.5},.....,Te). تم الحصول على طيفي النفاذ والامتصاص في مدى الطول الموجي (400-2500nm) وتم حساب فجوة الطاقة البصرية، معامل الانكسار، معامل الخمود، ثابت العزل الحقيقي والخيالي. تم مناقشة تأثير المحتوى من التيلوريوم على كل المعلمات البصرية. لقد تبين أن نتائج القياسات الكهربائية التي تشمل قياسات التوصيلية الكهربائية المستمرة، كثافة حاملات الشحنة و طاقة التنشيط تدعم نتائج القياسات البصرية.

Introduction:-

The structure of crystalline selenium and tellurium are hexagonal Which composed of spiral chains arranged parallel to one another and the crystallographic(c) axis lies parallel to the chains. In selenium there are a vitreous form as well as the amorphous and crystalline forms this vitreous form is detestable with respect to the crystalline form, the rate of conversion increase with increasing temperature. The vitreous form of tellurium crystallizes below room temperature. Selenium and tellurium are miscible in all proportions, forming a series of alloys. X-ray work shown that these alloys are composed of mixed chains in which the selenium and tellurium atoms are arranged at random rather than a mixture of selenium and tellurium chains [1]. Studies of semi conducting chalcogenides glasses have received much attention because of their important application in infrared region [2,3]. The most important application of chalcogenides glasses are now in the field of optics and arise chiefly from their infrared transmitting properties exhibited by them. They have potential use in integrated optics, optical imaging, optical data storage and infrared optics [4,5]. On the other hand amorphous semiconductors are most suitable for high field conduction studies and some studies have been reported by [6]. Chalcogenides glasses have been found to exhibit the change of refractive index under the influence of light, which makes it possible to use these materials to record the magnitude and the phase of illumination, the latter is especially important in holographic optical data storage and in fabrication of various integrated components and devices such as selective optical fibers, mixers, couplers and modulators [7,8]. In the present work we chose Se-Te system to overcome many problems faced most of the work has been carried out on chalcogenides thin films having Te as the major constituent, one of

the limitation is the poor resistance to oxidizations of Te ,we attempt to investigate Se as based material also Se has wide commercial application such as switching ,memory and xerography ect which made it attractive [9],on the other hand we found in the literature survey that many workers [9,10,11,12,13,14] pointed out that $\text{Se}_{1-x}\text{Te}_x$ transforms from amorphous to crystalline phase at tellurium concentration ($x > 0.5$) thus they limit their researches in the range ($x=0.0-0.45$) thus we extend this study to the hole compositions of Te content (x) and investigate the effect of it on the optical energy gap ,the spectral dependence of (n , k , ϵ_1 and ϵ_2)on the wavelength using the optical transmittance and absorbance [15,16,17] in the hope that these results may throw some light on the structure, optical and electrical properties of Se-Te system which is still not fully understood.

Experimental Part:

The substrates, which were subjected to several cleaning stages. High purity (99.999%) of selenium (Se) and tellurium(Te) elements were weighted according their atomic percentage .Thin films of $\text{Se}_{1-x}\text{Te}_x$ were

Produced by co evaporation Se and Te from separate molybdenum boats under high vacuum (10^{-5} Toor) which was provided by rotary and diffusion pump. The substrate to the boat distance was 20 cm to ensure the uniformity of the thickness of the prepared samples and also to obtain proper mixing of the vapors of the elements .The samples having almost thickness of about 5000\AA .The deposition rate was maintained $20-40 \text{ \AA}/\text{sec}$.

The transmittance (T) and the absorbance (A)spectra are obtained using double UV/VIS/NIR spectrometer as function of wavelength of incident photon in the range (400-2500 nm).

According to Swanepoel method [18] the envelope of the interference maxima and minima occurs in the spectrum. The extinction coefficient (k) neglected in the region of weak and medium absorption ($\alpha \neq 0$).Therefore, this approximation is valid over most spectrum. The presence of maxima and minima of the transmittance spectrum at the same wavelength confirmed the optical homogeneity of the deposited samples and referred that there was no scattering or absorption occurs at long wavelength .The optical constants were deduced from the fringes patterns in the transmittance spectra. In the transmittance region where the absorption coefficient $\alpha = 0$, the refractive index (n)is given by:-

$$n = [N + (N^2 - S^2)^{1/2}]^{1/2} \dots\dots\dots(1)$$

Where

$$N = 2S/T_m - (S^2 + 1) \dots\dots\dots(2)$$

Where T_m is the envelope function of the transmittance minima and S is the refractive index of the substrate .In the region of weak and medium absorption ,where $\alpha \neq 0$ the transmittance decreases mainly due to the effect of the (α) , the refractive index is given by equation (1) :-

Where N is given by :-

$$N = [2S(T_M - T_m)/T_M T_m] + (S^2 + 1)^2 \dots\dots\dots(3)$$

T_M is the envelope function of the transmittance maximum.

While in the region of strong absorption the transmittance deceases drastically due to the influence of α and (n) can be estimated by extrapolating the value in other region .Because the thickness of our films is uniform interference give rise to the spectrum shown in Fig (1 A,B) and the fringes can be used to calculate (n) of the samples using equation (1&3) .

The extinction coefficient (k) is obtained using the following equation:-

$$k = \frac{\alpha \lambda}{4\pi} \dots\dots\dots(4)$$

the absorption coefficient α can be calculated using the well known equation:-

$$\alpha = 2.303 \frac{A}{d} \dots\dots (5)$$

where d is the thickness of samples ,the present system $\text{Se}_{1-x}\text{Te}_x$ obeys the indirect transition and the relation between the absorption coefficient and the photon energy (hv) is given by[19]:-

$$(\alpha \text{ hv})^{1/r} = \hat{A} (\text{ hv } - E_g) \dots\dots(6)\dots\dots$$

Where \hat{A} is constant, r=2 for indirect allowed transition, the straight line portion of the $(\alpha \text{ hv})^{1/2}$ versus (hv) plot to $\alpha = 0$ yield the optical energy gap(E_g).

while (ϵ_1 and ϵ_2) were determined using equation the following equations:-

$$\epsilon_1 = n^2 - k^2 \dots\dots\dots(7)$$

$$\epsilon_2 = 2nk \dots\dots\dots(8)$$

D.C electrical conductivity of semiconductor (σ) is given by the formula[20]:-

$$\sigma = e(\mu_n.n + \mu_p.p) \dots\dots\dots (9)$$

Where μ_n and μ_p are the mobility of electrons and holes respectively in units of ($\text{cm}^2/\text{V}.\text{sec}$).The n and p are the concentrations of electrons and holes and are measured in units of (cm^{-3}) and e is the charge of electron .The change of electrical conductivity with temperature of semiconductors are given by the equation [21]:-

$$\sigma = \sigma_0 e\left(-\frac{E_a}{k_B T}\right) \dots\dots\dots (10)$$

Where E_a is the thermal activation energy ,T is the absolute temperature , k_B is the Boltzmann constant and σ_0 is the minimum metallic conductivity (the value of σ when $T \rightarrow \infty$) Hall effect results from applying magnetic field (B_z) along a rectangular sample normal to the direction of current (I),the charge carriers will tend to be deflected to one side ,then building up potential gradient perpendicular to magnetic field and current ,this effect was used to determine the type and the density of charge carriers(n_H).Hall coefficient (R_H) is given by:-

$$R_H = \frac{V_H}{I} \frac{t}{B_z} \dots\dots\dots (11)$$

Where t is the film thickness, the relation gives the density of charge carriers:-

$$R_H = -\frac{1}{n_H e} \dots\dots\dots (12)$$

The mobility of Hall (μ_H) is given by the relation [22] :-

$$\mu_H = \sigma |R_H| \dots\dots\dots (13)$$

Vacuum evaporated aluminum electrodes at the bottom of substrates were

Previously done for electrical properties measurements.D.C electrical conductivity measurements were carried out at the temperature range (303-500) K using the electrical circuit which is consists of Oven

type Herease ,Power supply and Ameter .The resistivity (ρ) of $\text{Se}_{1-x}\text{Te}_x$ samples deposited at room temperature were obtained using the following equation :-

$$\rho = \frac{Rwt}{L} \dots\dots\dots (14)$$

Where L and w are the length and the width of sample respectively and R and t are the resistivity and thickness of the film, the conductivity of the mentioned samples were obtained from the relation:-

$$\sigma = \frac{1}{\rho} \dots\dots\dots (15)$$

The activation energy (E_a) of the $\text{Se}_{1-x}\text{Te}_x$ samples can be deduced by applying the following relation:-

$$E_a = \text{slop} \times 0.08652 \dots\dots\dots (16)$$

Where:-

$$\text{slop} = \frac{d \ln \sigma}{d(1000/T)} \dots\dots\dots (17)$$

The measurements of the density of charge carriers and the Hall mobility were provided from applying equations (12 and 13) respectively.

Results and Discussion:-

The Structure and Optical Properties:-

X-ray diffraction studies of $\text{Se}_{1-x}\text{Te}_x$ films for ($x=0.0-1.0$) were made ,the pattern of Te declared sharp diffraction peak indicated the polycrystalline structure ,while Se films were completely amorphous (i.e. declare no features in the diffraction patterns) similar to those samples ($\text{Se}_{0.9}\text{Te}_{0.1}$,

$\text{Se}_{0.8}\text{Te}_{0.2}$, $\text{Se}_{0.7}\text{Te}_{0.3}$, $\text{Se}_{0.6}\text{Te}_{0.4}$ and $\text{Se}_{0.5}\text{Te}_{0.5}$),however the films with higher Te concentrations ($x > 0.5$) showed relatively less amorphous nature and the degree of crystallinity increased with increase Te content ,moreover the diffraction patterns of ($\text{Se}_{0.3}\text{Te}_{0.7}$, $\text{Se}_{0.2}\text{Te}_{0.8}$) samples declared sharp peaks similar to these produced by pure (Te)film ,similar results pointed by Agrawl et al[23].

The variation of $(\alpha \text{ hv})^{1/2}$ with photon energy (hv) for $\text{Se}_{1-x}\text{Te}_x$ films for ($x=0.0-1.0$) are shown in Fig.(2 A,B).It is clear that (E_g) for pure Se film was (1.910 eV), while the Te film has (E_g) value (0.495 eV) which is identical with that reported by[24,25], this means that the increase of Te content from (0.0-1.0) reduces the (E_g) value from (1.910 to 0.495 eV) which can be estimated from Fig.(1,A,B) as a shift in the absorption band edge to longer wavelength (low energy values)., this is ascribed to Te addition to Se which greatly reduces the Se_8 rings and increases the chain fraction and the chain length which in turn enhanced the tailing of localized states consequently reduced the (E_g) values, on the other hand the decrease of E_g values is not appreciable for ($x=0.0-0.5$) but then an abrupt decrease take place at $x=(0.6-1.0)$, A.Dawar et al [26] referred this observation at Te content($x > 0.5$) and attributed to direct optical transition which accrued at($x > 0.5$) while at($x=0.0-0.5$) the gap is presumably indirect. Our explanation about the abrupt decrease in (E_g) at the mentioned Te concentrations belonged to conversion in structure from amorphous to crystalline ,moreover it is well known that the energy gap value for amorphous exceeded that of crystalline material.

Fig(3A,B)declared the variation of (n) in the wavelength range (400-2500nm).The (n)values of Se and Te are (2.605 and 5.310)at $\lambda=1800\text{nm}$,these values are similar to these obtained by [26],on the other hand (n) increases with λ and (x) but slowly for $x=0.0-0.5$ while above this concentration (i.e $x > 0.5$) the increase becomes clear .Our (n)values for ($x=0.0-0.5$) is similar to these values reported by H.Adachi and K.Kao [13] for $\text{Se}_{1-x}\text{Te}_x$ for($0.45 > x > 0$),they found that (n) increased from (3.1 to 3.6) when tellurium concentration increased from (0.0 to 0.45) at $\lambda=5461\text{nm}$, while our (n) values increased from(2.605 to 3.895) when (Te) increased in same range. The increase in the (n) values is due to the decrease in the transmittance of $\text{Se}_{1-x}\text{Te}_x$ films as shown in Fig .(1A,B)(i.e. the samples

became less transparent to the incident light with the increase in (Te) content, this result is expected from the energy gap values.

Fig.(4A,B) showed the variation of extinction coefficient (k) for $\text{Se}_{1-x}\text{Te}_x$ thin films deposited at room temperature, one can observe that (k) values increased with Te content in the films, moreover (k) increases from (0.01 to 1.246) when Te content changed from (0.0 to 1.0) at $\lambda=650\text{nm}$ for ($x=0.0-0.5$) and, at $\lambda=1800\text{nm}$ for ($x>0.5$) this increase indicated that the absorbance affected by Te content in the films, our (k) values are similar to that published by H.Adachi and K.Kao [13] and higher than that reported by A.Dewar et al [27], the difference attributed to thickness difference as well as that (k) were calculated at different wavelength of the spectrum.

The real (ϵ_1) and imaginary (ϵ_2) part of dielectric constant were calculated and plotted in the spectral range (400-2500nm) in Figs.(5A,B) and (6A,B). The real part showed maximum values at values at $\lambda=560, 652, 688, 826, 1234, 1297, 1372, 1447$ and 1647 nm when (x varied from 0.0 to 1.0) which corresponded to energies (2.214, 1.901, 1.802,

1.501, 1.501, 1.004, 0.956, 0.903, 0.856 and 0.752 eV), noticeable remark is the peak occurred at energies little higher than the energy gap values, moreover the location of the peaks shift to lower energy or to longer wavelength with increase Te content. The variation of imaginary part (ϵ_2) of dielectric constant in Fig.(6A,B) declared the same behavior of that of (k) thus it can give the same explanation, indeed (ϵ_2) increased from (0.0632 to 13.05) at $\lambda=650\text{nm}$ for ($x=0.0-0.5$) and at $\lambda=1800\text{nm}$ for $x>0.5$).

The Electrical Properties:-

Our data for Hall effect measurements referred that $\text{Se}_{1-x}\text{Te}_x$ thin films in the whole composition ($x=0.0-1.0$) were p-type (R_H is positive), i.e. there is direct relation between the current (I) and Hall voltage (V_H), thus the created electric field [permits the passage of the charge carriers (holes)] consequently the output current will be increased with increasing the applied electrical field. Fig(7A,B) shows the temperature dependence of d.c electrical conductivity ($\sigma_{d.c}$) for $\text{Se}_{1-x}\text{Te}_x$ thin films for ($x=0.0-1.0$) deposited at room temperature. It can be observed that it is characterized by two stage of conductivity through out the heating temperature, this is in agreement with that pointed out by H.Loynset [1] and K.Kokenyesi et al [11], they indicated that the conductivity takes place through out one mechanism, on the other hand the activation energies in the low and high temperature range declared to decrease with increase (Te) content in the $\text{Se}_{1-x}\text{Te}_x$ samples. Table (2) illustrates the values of ($\sigma_{d.c}$) at room temperature (R.T), the activation energies (E_{a1}, E_{a2}) in the high and low range of temperature respectively, also it is well known that the activation energies reduced with increasing ($\sigma_{D.C}$), moreover ($\sigma_{d.c}$) increased by a factor of 10^3 when (Te) increased from (0.0 to 1.0) this increase comes from increase in concentration of charge carriers (n_H) which increase by a factor of 10^2 as well as to the increase in the mobility (μ_H) which increase by a factor of 1.04×10^{-2} with increase (x) content, (see table (2)).

Conclusion:-

According to the above observations, the following conclusion are drawn :- (1) the introducing (Te) to Se at concentration $x>0.5$ regards as agent factor to nucleation, i.e. obtain crystalline film. (2) the optical energy gap decreased drastically for ($x>0.5$) when the structure of the samples convert from amorphous to crystalline phase, indeed this shift in the energy gap values indicates that the $\text{Se}_{1-x}\text{Te}_x$ samples for ($x<0.6$) behave as selenium like while the $\text{Se}_{1-x}\text{Te}_x$ samples for ($x>0.5$) behave as tellurium like. (3) the optical constant increase in systematic manner with increase tellurium concentration. (4) The increase in (Te) content in $\text{Se}_{1-x}\text{Te}_x$ films increased charge carrier concentrations resulting in the decrease of activation energy.

Table(1) Illustrates the values of (n) at $\lambda=1800$ nm, and (k, ϵ_1 and ϵ_2) at $\lambda=650$ nm for (x=0.0-0.5), and at $\lambda= 1800$ nm for (x=0.6-1.0) for $\text{Se}_{1-x}\text{Te}_x$ thin films deposited room temperature.

Tellurium content(x)	(n)	(k)	(ϵ_1)	(ϵ_2)
0	2.605	0.01	9.86	0.0632
0.1	2.820	0.009	10.61	0.058
0.2	3.182	0.0425	13.31	0.309
0.3	3.388	0.0533	14.05	0.397
0.4	3.515	0.12	12.64	0.873
0.5	3.759	0.415	15.03	3.237
0.6	3.895	0.5305	15.02	4.366
0.7	4.008	0.624	15.71	5.077
0.8	4.427	0.694	19.15	6.089
0.9	4.746	1.094	21.4	10.47
1.0	5.310	1.258	26.72	13.05

Table (2) The values of ($\sigma_{D.C}$) at R.T ,(E_a), (n_H) and(μ_H) for $\text{Se}_{1-x}\text{Te}_x$ thin films with different (Te) content deposited at room temperature.

Tellurium content(x)	($\sigma_{D.C}$) ($\Omega.cm$)⁻¹ at R.T	(E_{a1}) (eV)	Temp.Range (K)	(E_{a2}) (eV)	Temp.Range (K)	n_H (cm⁻³)	μ_H (cm²/V.sec)
0	0.603×10^{-7}	0.802	(500-416.6)	0.071	(416.6-303)	4.50×10^{16}	0.837×10^{-5}
0.1	1.952×10^{-7}	0.767	(500-416.6)	0.054	(416.6-303)	4.60×10^{16}	0.265×10^{-4}
0.2	9.338×10^{-7}	0.756	(500-416.6)	0.050	(416.6-303)	7.60×10^{16}	0.767×10^{-4}
0.3	10.430×10^{-7}	0.593	(500-370.3)	0.042	(370.3-303)	2.30×10^{17}	0.280×10^{-4}
0.4	13.805×10^{-7}	0.580	(500-370.3)	0.041	(370.3-303)	3.34×10^{17}	0.258×10^{-4}
0.5	6.602×10^{-6}	0.535	(500-384.6)	0.037	(384.6-303)	3.56×10^{17}	0.115×10^{-3}
0.6	13.764×10^{-6}	0.491	(500-400.0)	0.032	(400.0-303)	3.83×10^{18}	0.223×10^{-4}
0.7	18.982×10^{-6}	0.378	(500-370.3)	0.030	(370.3-303)	3.98×10^{18}	0.298×10^{-4}
0.8	8.801×10^{-5}	0.277	(500-384.6)	0.028	(384.6-303)	4.23×10^{18}	0.129×10^{-3}
0.9	4.808×10^{-4}	0.257	(500-434.0)	0.021	(434.0-303)	5.36×10^{18}	0.506×10^{-3}
1.0	12.211×10^{-4}	0.160	(500-434.0)	0.020	(434.0-303)	8.70×10^{18}	0.876×10^{-3}

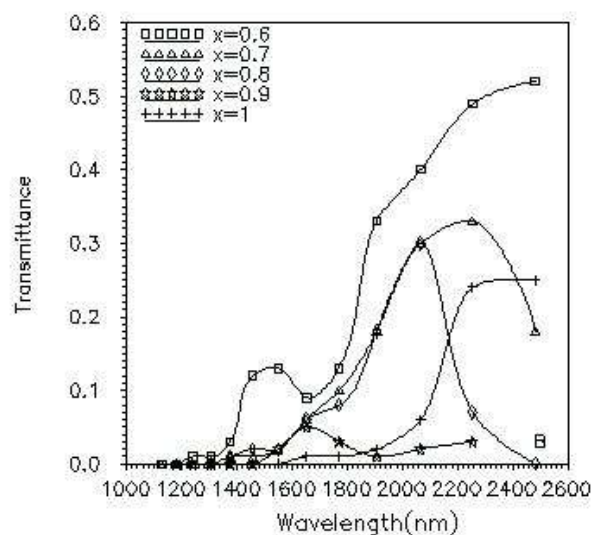
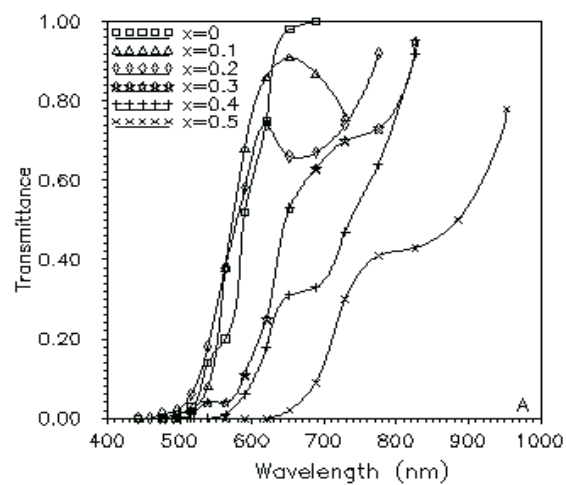


Fig.(1 A ,B) Transmittance spectra as function to wavelength for $\text{Se}_{1-x}\text{Te}_x$ thin films with different (Te) content deposited at room temperature.

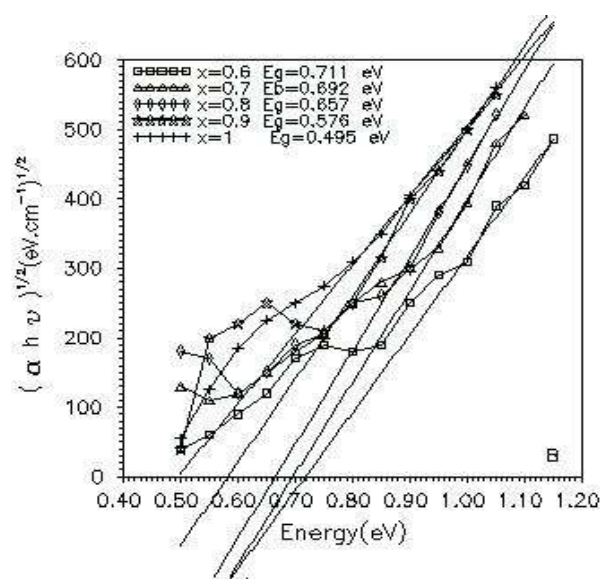
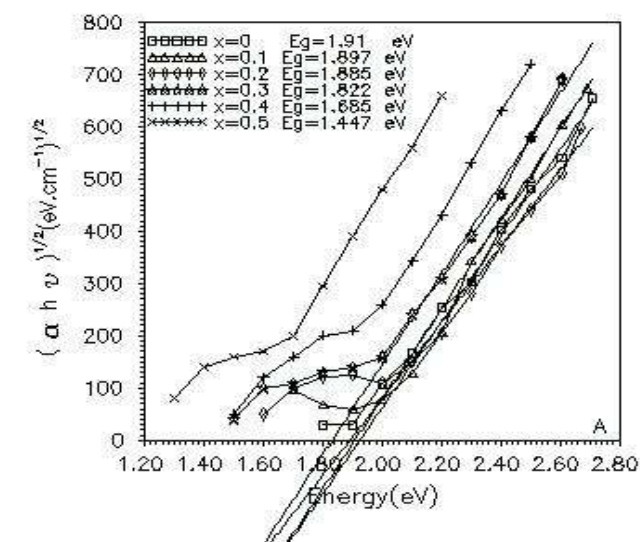


Fig.(2 A,B) The variation of $(\alpha h\nu)^{1/2}$ versus photon energy for $\text{Se}_{1-x}\text{Te}_x$ thin films with different (Te) content deposited at room temperature.

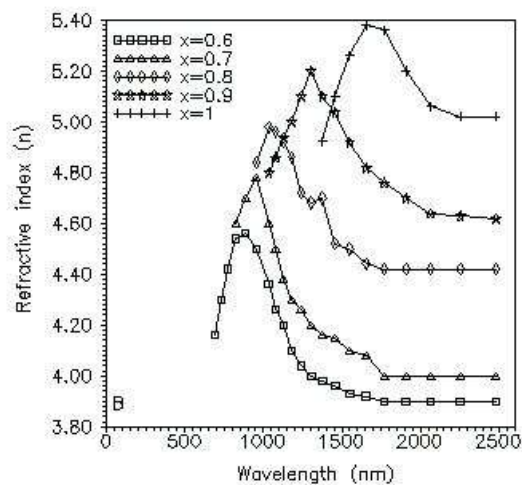
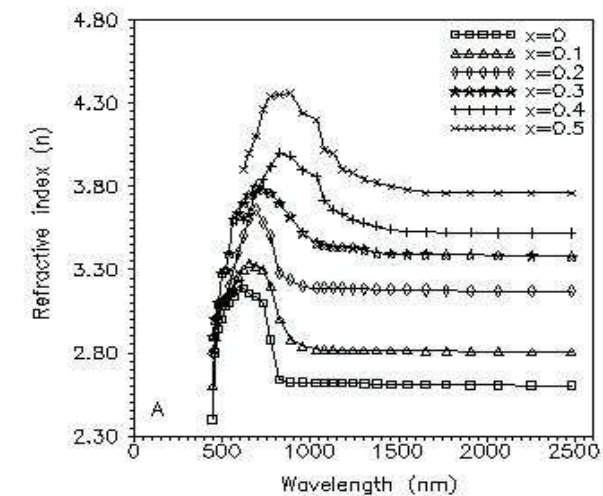


Fig.(3 A,B) The variation of refractive index (n) versus wavelength for $\text{Se}_{1-x}\text{Te}_x$ thin films with different (Te) content deposited at room temperature.

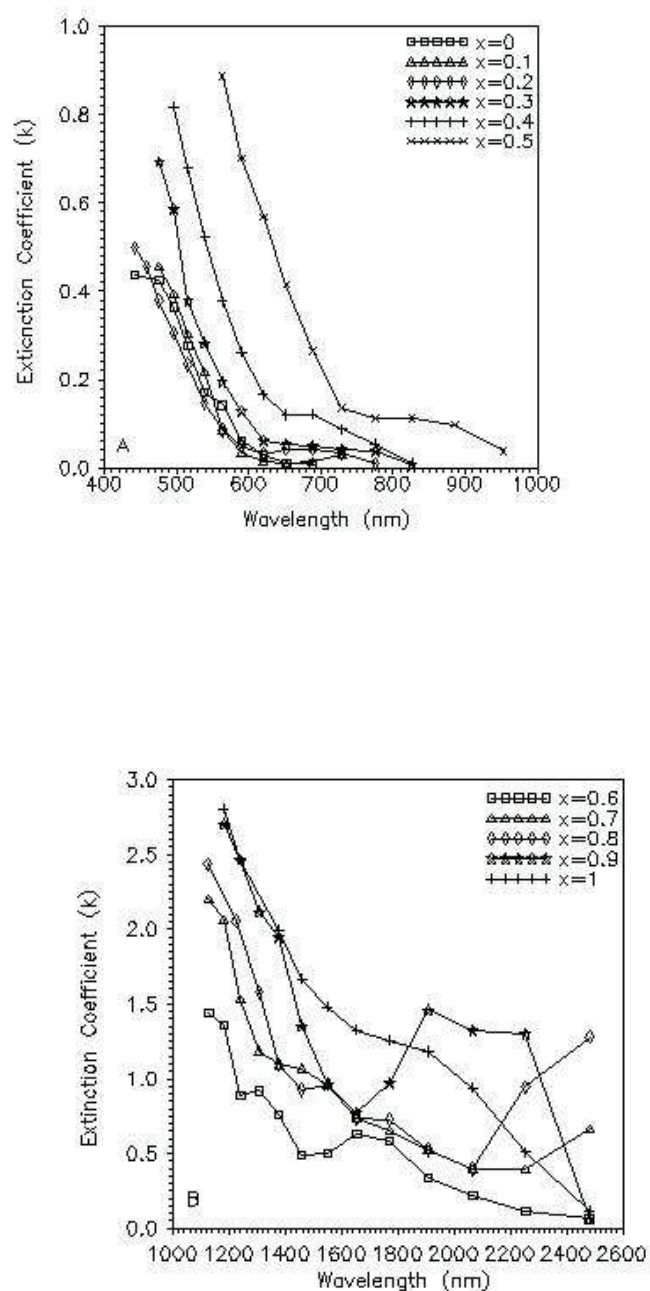


Fig.(4 A,B) The variation of extinction coefficient (k) versus photon energy for $\text{Se}_{1-x}\text{Te}_x$ thin films with different (Te) content deposited at room temperature.

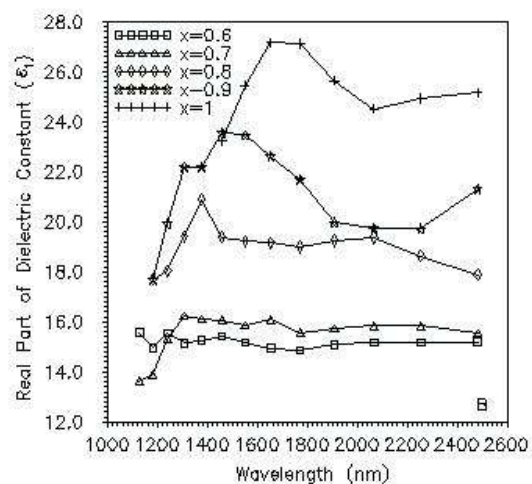
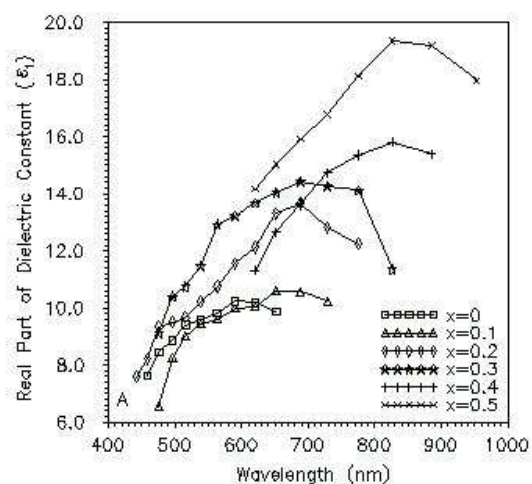


Fig.(5 A,B) The variation of real part of dielectric constant (ϵ_1) versus wavelength for Se_{1-x}Te_x thin films with different (Te) content deposited at room temperature.

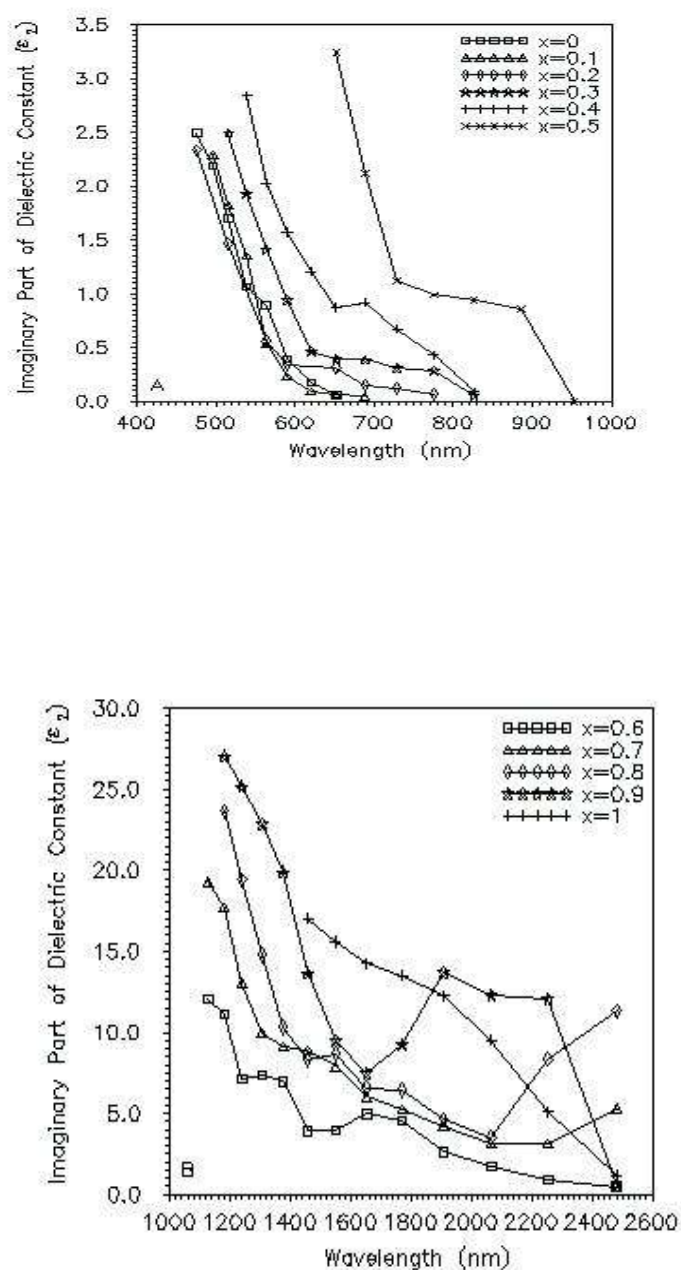


Fig.(6 A,B)The variation of imaginary part of dielectric constant (ϵ_2) versus wavelength for $\text{Se}_{1-x}\text{Te}_x$ thin films with different (Te) content deposited at room temperature.

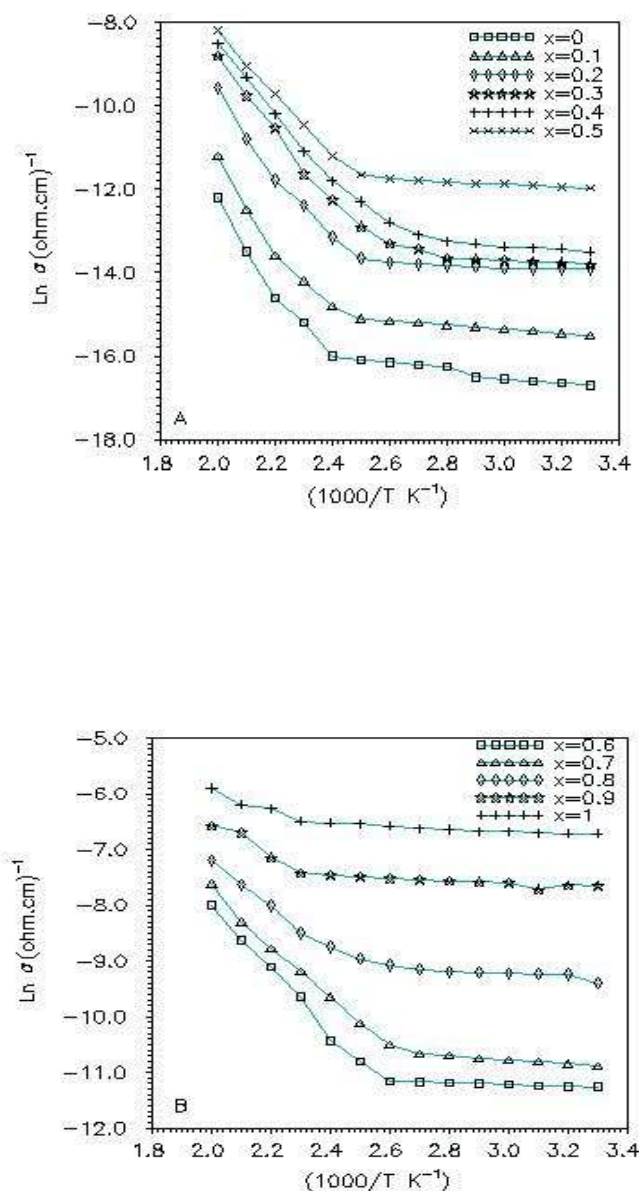


Fig.(7 A,B) The variation ($\ln\sigma$) versus $(10^3/T)$ for $\text{Se}_{1-x}\text{Te}_x$ thin films with different (Te) content deposited at room temperature.

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