

Optical Investigations of CdSe_{1-x}Te_x Thin Films

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Abstract:

The alloys of CdSe_{1-x}Te_x compound have been prepared from their elements successfully with high purity (99.9999%) which mixed stoichiometry ratio (x=0.0, 0.25, 0.5, 0.75 and 1.0) of (Cd, Se and Te) elements. Films of CdSe_{1-x}Te_x alloys for different values of composition with thickness(0.5μm) have been prepared by thermal evaporation method at cleaned glass substrates which heated at (473K) under very low pressure (4×10⁻⁵mbar) at rate of deposition (3Å/s), after that thin films have been heat treated under low pressure (10⁻²mbar) at (523K) for two hours.

The optical studies revealed that the absorption coefficient (α) is fairly high. It is found that the electronic transitions in the fundamental absorption edge tend to be allowed direct transition. It was also found that the optical energy gap vary non-linearly with composition (x) and have a minimum value at x=0.5 and increases after heat treatment.

It is found that the optical constants vary non-linearly with composition, and the behavior inverse at x=0.5, and affected by heat treatment. The behavior of ε₁ is similar to the behavior of n, while the behavior of ε₂ is similar to the behavior of k.

Key words: Chalcogenide glasses, thin films, Refractive index, Optical band gap.

Introduction:

The CdSe_{1-x}Te_x thin films have been prepared by several methods such as three source elemental evaporation [1], two source evaporation of CdSe and CdTe powder [2], electroplating [3-5], slurry painting [6-8], spraying and sintering suspension mixture of CdSe and CdTe powder [8], vacuum evaporation of pre-reacted CdSe_{1-x}Te_x material [9-10], and electron beam evaporation [11].

The optical properties of a semiconductor can be defined as any property that involves the interaction between electromagnetic radiation or light and the semiconductor, including absorption, diffraction, polarization, reflection, and scattering effects.

The optical properties of semiconductors are often subdivided into those that are electronic and those that are lattice in nature. The electronic properties concern processes involving the electronic states of the semiconductor, while the lattice properties involve vibrations of the lattice (absorption and creation of phonons). Lattice properties are of considerable interest, but it is the electronic properties which receive the most attention in semiconductors because of the technological importance of their practical applications [12].

The fundamental energy gaps of most semiconductors span the energy range from zero to about 4eV.

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Photons of sufficient energy can excite electrons from the filled valence bands to the empty conduction bands.

The fundamental absorption refers to band-to-band or to excitation transitions(i.e. the excitation of an electron from the valence band to the conduction band). It manifests itself by a rapid rise in absorption, which can be used to determine the energy gap of the semiconductor [13].

High absorption region observed for most semiconductors at $\alpha \geq 10^4 \text{cm}^{-1}$, the absorption is due to the transitions between extended states in both bands. The imperial formula that governs this transition have been found by Tauc [14].

$$\alpha(\nu)h\nu = B(h\nu - E_g^{\text{opt}})^r \dots(1)$$

Where ν is the frequency of the incident radiation, B is constant, which depends on density of state (DOS) of conduction and valence bands, E_g^{opt} is the optical energy gap [15], r is a constant depend on the nature of the transition.

In the transparent region[16] the absorption coefficient $\alpha = 0$ or(the absorbance) $x^- = 1$, the maxima of the interference fringes are function of S (the substrate refractive index) only and coincide with T_S . When the maxima depart from T_S denotes the onset of absorption. The refractive index can be calculated in this region from Swanepoel technique{which is using envelope technique for maxima transmission(TM) and minim transmission (Tm) [full curve in Fig (1)]} :

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

where S, is the substrate refractive index ,and

$$M = \frac{2S}{T_m} - \frac{S^2 + 1}{2} \dots(3)$$

T_m (the minimum transition) is thus a function of both n and S.

In the region of weak and medium absorption the refractive index (n) is calculated by the equation:

$$N = [N + (N^2 - S^2)^{1/2}] \dots(4)$$

Where N is given by:

$$N = 2S[(T_M - T_m)/T_M T_m] + (S^2 + 1)/2 \dots(5)$$

Once (n) is known, all the constants known and the absorbance x^- can be calculated.

The interference fringes disappear in the region of high absorption ,which is used to determinate the energy gap only by calculating the absorption coefficient and the energy of the incident photon.

When the light of intensity (I_0) incident on the film of thickness (d), the transmitted intensity can be given as[17]:

$$I = I_0 \exp(-\alpha d) \dots(6)$$

where d: is the thickness of the film.(I/I_0) represents the transmittance (T), since the absorbance (x) represents the logarithm of the reciprocal (T),

$$x = \log(I_0 / I) \text{ or } x = \log(1/T) \dots(7)$$

$$\ln(I_0 / I) = \alpha d \text{ , } 2.303 \log(1/T) = \alpha d$$

$$\alpha = 2.303 (x/d) \dots(8)$$

The energy of the incident photon can be calculated from:

$$E(\text{eV}) = h\nu = \frac{1240}{\lambda(\text{nm})} \dots(9)$$

where h: Planck's constant, ν : frequency of incident light, λ : the wavelength of incident light. By plotting the $(\alpha h\nu)^2$ against the photon energy, the straight line portions are extrapolating to zero and the value obtained represented the optical energy gap for direct transition.

If radiation absorption occurs in media then the intensity of radiation is generally attenuated in an exponential form of the type $e^{-\alpha d}$ where

α is related to the imaginary part k of the refractive index called extinction coefficient[18], which can be calculated by the relation:

$$k = \frac{\alpha\lambda}{4\pi} \dots(10)$$

The complex index of refraction n_c is defined as[18]:

$$n_c = n - ik \dots(11)$$

is related to the velocity ($v = c/n_c$) of the radiation propagation where n is the refractive index. The dielectric constant can be introduced as follows[19]:

$$\varepsilon = \varepsilon_1 - i\varepsilon_2 \dots(12)$$

And from Maxwell equations result that[20]:

$$(n - ik)^2 = \varepsilon_1 - i\varepsilon_2 \dots(13)$$

So that ,

$$\varepsilon_1 = n^2 - k^2 \text{ (real part) } \dots(14)$$

$$\text{and } \varepsilon_2 = 2nk \text{ (imaginary part) } \dots(15)$$

The aim of this research is a preparation of $\text{CdSe}_{1-x}\text{Te}_x$ as-alloys and thin films where ($x = 0.0, 0.25, 0.5, 0.75$ and 1.0) and studying its optical properties. Indeed, the main task was studying the effect of the heat treatment and composition on optical properties of $\text{CdSe}_{1-x}\text{Te}_x$ thin films which were prepared by using the thermal evaporation technique.

Materials and methods:

In the present work, $\text{CdSe}_{1-x}\text{Te}_x$ alloys has been synthesized using high purity elemental cadmium, tellurium and selenium is about (99.9999%) with different x content where ($x = 0.0, 0.25, 0.5, 0.75$ and 1.0). Stoichiometric amounts of the elements are placed in a quartz ampoule, The quartz ampoule was cleaned carefully with water and alcohol respectively, to remove dust, grease, and other possible contaminants, which is evacuated to a vacuum of 10^{-4} Torr and then sealed. The sealed ampoule is placed in a furnace and then heated at a rate of 60°C per hour in steps up to 1173 K . The

ampoule is maintained at this temperature for about ten hours and then allowed to cool slowly to room temperature. The vacuum unit system, which is used to prepare thermally evaporated $\text{CdSe}_{1-x}\text{Te}_x$ films on the corning glass substrate was Edward Coating unit model 306A.

The molybdenum boat which were used for films preparation were covered with pin holes cover. All the prepared films heat treated under vacuum of 10^{-2} Torr at annealing temperature (523K) for two hour.

The thickness of the prepared films ($0.5\mu\text{m}$) has been determined using three different techniques: 1- The Weight Method 2- Optical Interference Fringes 3- Swanepoel Method

The transmission spectra were obtained over the range ($500-2500\text{nm}$) by a double-beam, ratio recorder UV-VIS-NIR computer controlled spectrometer (Shimadzu model UV-probe-3150).

The absorption coefficient $\alpha(\lambda)$ in the weak and medium absorption region is used only to calculate the extinction coefficient $k(\lambda)$. In the strong absorption region from the transmission spectra, the absorption coefficient $\alpha(\lambda)$ was obtained also from eq.(8) but the absorbance in this region calculated from eq.(7).

The optical energy gap was determined using the tauc formula eq.(1) for allowed direct transition, by plotting $(\alpha h\nu)^2$ against the photon energy which is calculated from eq.(9) and best fit line intersect the energy photon axis at $(\alpha h\nu)^2$ equal to zero represents the value of optical energy gap.

Refractive indices are measured by using eqs. (2) and (3), while the extinction coefficient is determined using eq. (10).

The real and imaginary parts of the dielectric constant $\varepsilon_1(\lambda)$ and $\varepsilon_2(\lambda)$

were calculated by using equations (14 & 15) respectively.

Results and Discussion:

1- The Transmission Spectrum

The experimental transmission spectrum for as-deposited and heat treated CdSe_{1-x}Te_x films, respectively, in the spectral range (500 – 2500)nm at room temperature are shown in Figs.(2) to Fig.(6).

All spectra show good transparency ($T > 85\%$) and reveal very pronounced interference effects for photon energies below the fundamental absorption edge by exhibiting interference pattern in the spectral region between (900-2500)nm. Such behavior of the spectra is evidence of the thickness uniformity of the films. Otherwise, the interference fringes would have been destroyed, resulting in smooth transmission curves. They also display a clear explicit absorption edge interrelated to the optical band gap. It is clear from these figures that spectral characterization are affected by heat treatment. For heat-treated samples the transmission coefficient is greater than before the heat treatment. This fact is due to the increase in the crystallite size observed for heat-treated samples[21].

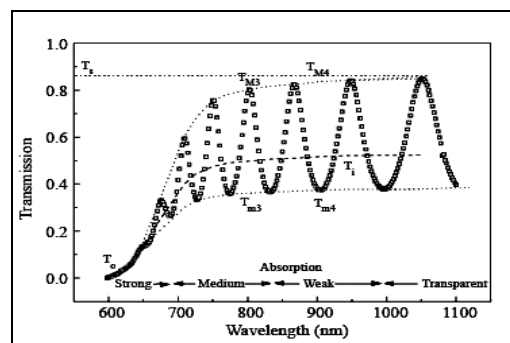


Fig.(1): Typical transmission spectra

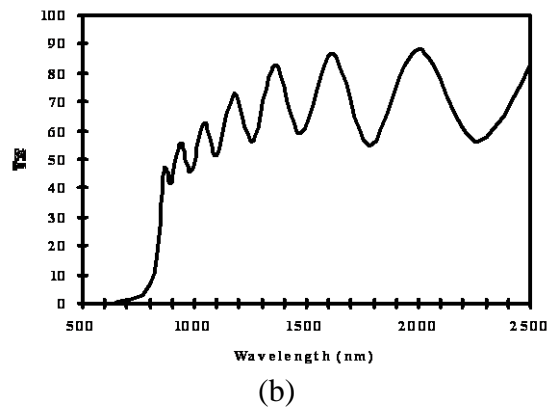
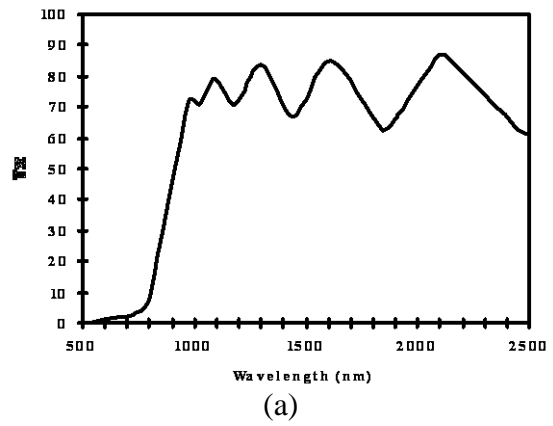


Fig.(2) Transmission spectrum of CdTe (a) as-deposited, (b) annealed

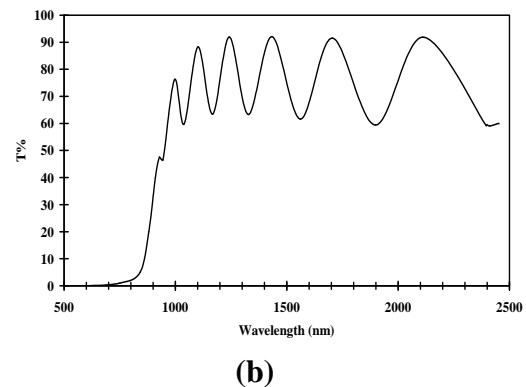
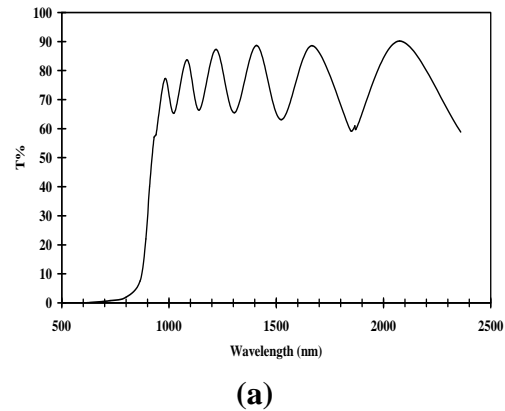
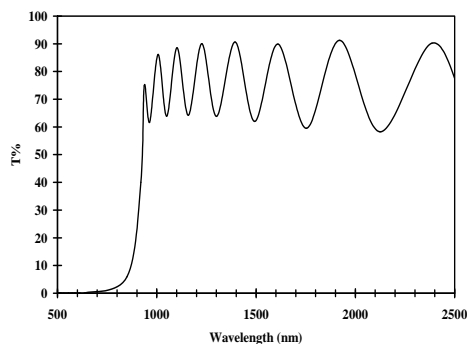
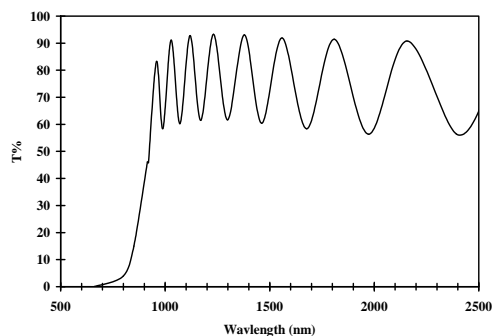


Fig.(3) Transmission spectrum of CdSe_{0.25}Te_{0.75} (a) as-deposited, (b) annealed

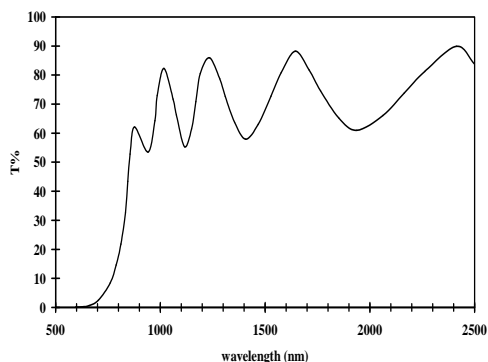


(a)

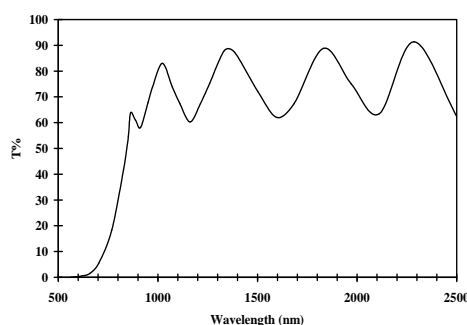


(b)

Fig.(4) Transmission spectrum of CdSe_{0.5}Te_{0.5} (a) as-deposited, (b) annealed

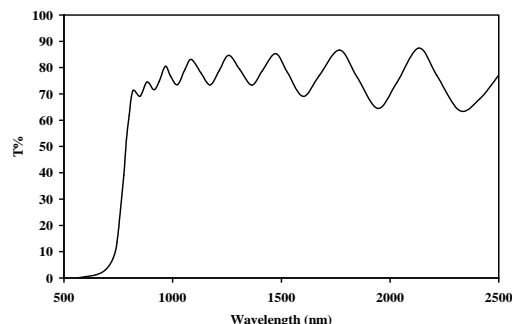


(a)

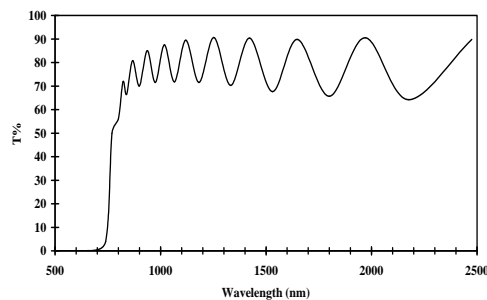


(b)

Fig.(5) Transmission spectrum of CdSe_{0.5}Te_{0.5} (a) as-deposited, (b) annealed



(a)



(b)

Fig.(6) Transmission spectrum of CdSe_{0.5}Te_{0.5} (a) as-deposited, (b) annealed.

2- Absorption Coefficient Results:

The absorption coefficient α were determined from the region of high absorption i.e. at the fundamental absorption edge of the films using the equation (8). Fig.(7a and b) shows the dependence of the absorption coefficient on the component x value as a function of wavelength for as-deposited and annealed CdSe_{1-x}Te_x films of thickness (0.5 μ m) with different x content (0.0, 0.25, 0.5, 0.75 & 1.0). From this figure we can observe that the optical absorption coefficient for all films were fairly high values reached above 10⁴cm⁻¹, and found that the α decreases with heat treatment for all samples as shown in (6b), and this is due to the increasing value of energy gap after annealing process. This result agrees with the result of Najeeb[2].

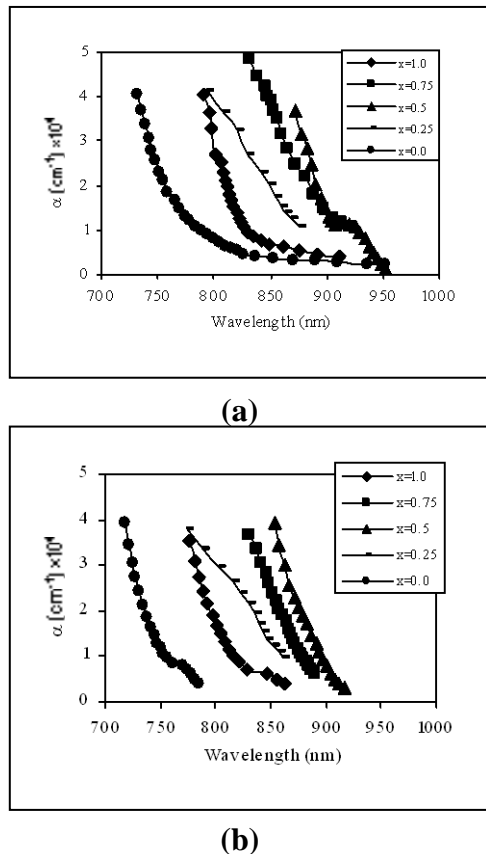


Fig. (7) The absorption coefficient vers wavelength for CdSe_{1-x}Te_x thin film at different composition (a) as-deposited (b) annealed.

3- The Optical Energy Gap Measurements:

The optical energy gap values (E_g^{opt}) for CdSe_{1-x}Te_x films have been determined by using Tauc equation (1) which is used to find the type of the optical transition by plotting the relations $(\alpha h\nu)^2$, $(\alpha h\nu)^{2/3}$, $(\alpha h\nu)^{1/2}$ and $(\alpha h\nu)^{1/3}$ versus photon energy ($h\nu$) and select the optimum linear part. It is found that the first relation yields linear dependence, which describes the allowed direct transition.

E_g^{opt} is then determined by the extrapolation of the portion at $(\alpha=0)$ as shown in Figs.(8a and b). The optical energy gap decreased from 1.64eV to 1.37eV when the Te content increased from zero to 0.5 whereas, its values increased to 1.51eV with increasing Te to $x=1$, and the films after heat treatment have the similar behavior, this means

that the E_g^{opt} varies non-linearly with composition as shown in Fig.(9). These results agree with the results of the energy gap versus composition studied by Brodin[22] for single crystal films, electron beam evaporated films by Mangalhora[11], elemental evaporated films and dual source evaporated films by Russak[1]. The optical energy gap was found to increase after heat treatment, this increasing is attributed to the existence of a tail below the fundamental absorption edge (near the bands) which is observed in the absorption coefficient Fig.(7a), which is a characteristic of most of the optical data on polycrystalline semiconductors which is considered to be determined mainly by the structural disorder existing at the grain boundaries[21], these tails decreased after heat treatment due to the increasing in crystallite size and the grain boundaries became more ordered than in as-deposited films. This result is observed in CdTe by Iqbal[23] and in CdSe by Baban[21].

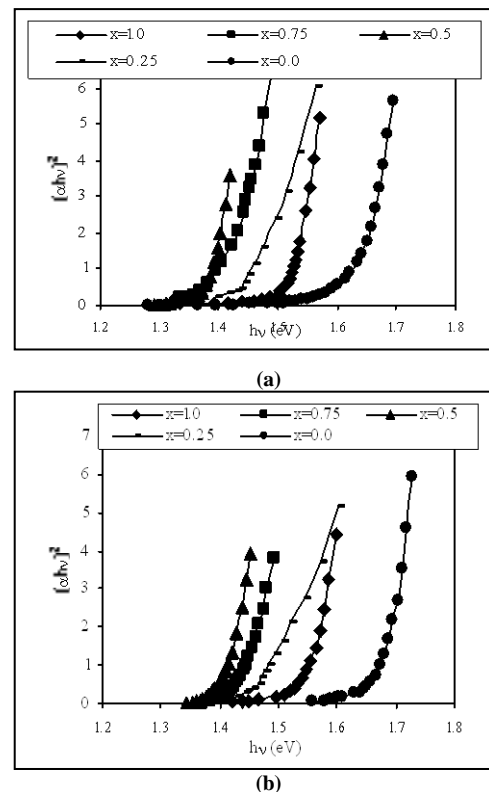


Fig. (8) The variation of $(\alpha h\nu)^2$ with photon energy for CdSe_{1-x}Te_x film at different composition (a) as-deposited (b) annealed.

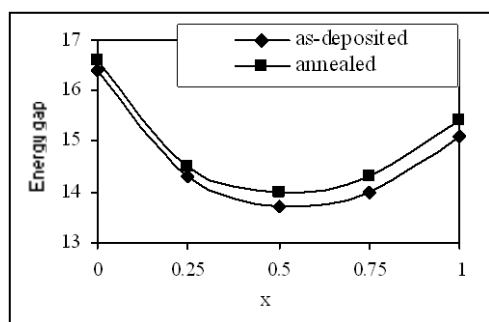
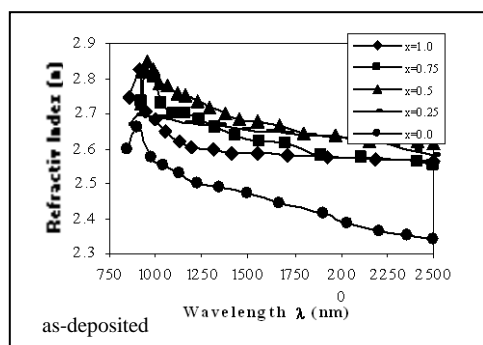


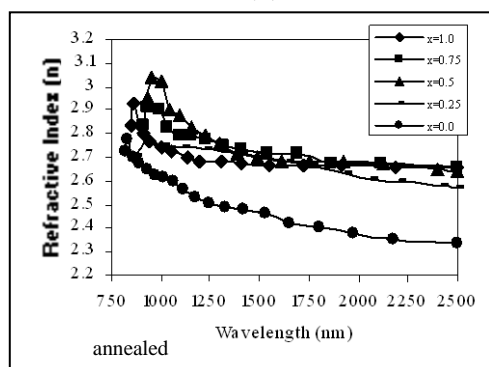
Fig. (9) The variation of energy gap ($E_g^{opt.}$) with composition for as-deposited and annealed CdSe_{1-x}Te_x films.

Optical Constants

1-Refractive Index: The wavelength dependence of the refractive index has been calculated in the range between 750nm to 2500nm on the CdSe_{1-x}Te_x films using the equations (2) and (3). Fig.(10) shows the variation of refractive index with wavelength of as-deposited and annealed CdSe_{1-x}Te_x for different values of composition. It is found from this figure that the refractive index decreases with the increasing of wavelength of the incident photon.



(a)



(b)

Fig.(10) The refractive index as a function of wavelength for as-deposited and annealed CdSe_{1-x}Te_x thin films.

And we can observed the appearance of a peak occur in dispersion of the refractive index of the films, this was attributed to the rapid change in the optical absorption coefficient in the vicinity of the absorption edge[19].

A plot of the refractive index n at $\lambda = 1500\text{nm}$ versus composition x is presented in Fig.(11) for as-deposited and annealed films. The refractive index increases for the Te content ($x=0.0$ to 0.5) and then decreases with further increases in Te. Similar behavior in refractive index has been observed in annealed films. This variation may be attributed to the existence of two phases structure in CdSe_{1-x}Te_x compound, when $x>0.5$ the structure is cubic while it has a hexagonal structure when $x<0.5$ and the packing density of hexagonal structure more than in cubic structure.

The refractive index increases after the heat treatment probably due to the increase of the compactness of the films after the heat treatment simultaneously with the increase of the crystallite size[21].

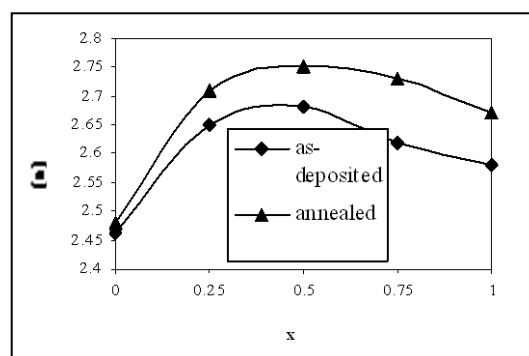
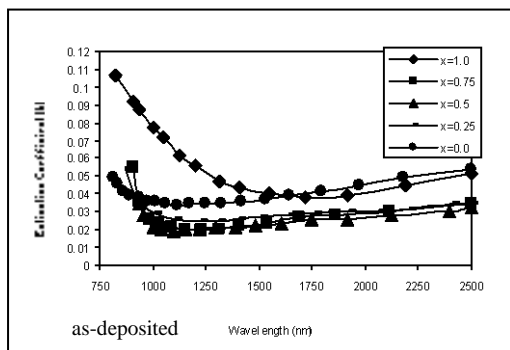


Fig.(11) The variation of refractive index with composition for as-deposited and annealed CdSe_{1-x}Te_x thin films.

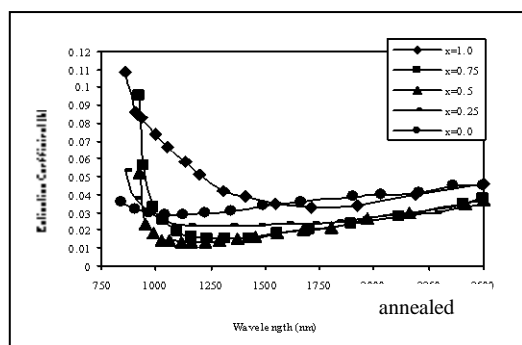
2- Extinction Coefficient

The relation between the extinction coefficient and wavelength for CdSe_{1-x}Te_x films deposited at 473K and heat treated at 523K with different values of x is shown in Fig.(11). From this figure it is found that the

extinction coefficient (k) takes the similar behavior of the corresponding absorption coefficient. One can deduce from this figure that the extinction coefficient decreased with increasing the wavelength up to $\approx 1150\text{nm}$ due to the high values of the absorption coefficient at this range of wavelength, after that the extinction coefficient increased very slightly which is associated with the increasing of the transmittance in this region and the large decreasing of the absorption coefficient at this wavelength. Fig.(12) shows the extinction coefficient dependence on composition at $\lambda = 1500\text{nm}$ for as deposited and annealed films. And we notice from this figure that the extinction coefficient is affected by heat treatment, which is decreased after annealing process. This behavior of the extinction coefficients values similar to that of the absorption coefficients for the same reasons as mentioned before.



(a)



(b)

Fig.(12) The extinction coefficient versus wavelength for as-deposited and annealed CdSe_{1-x}Te_x thin films.

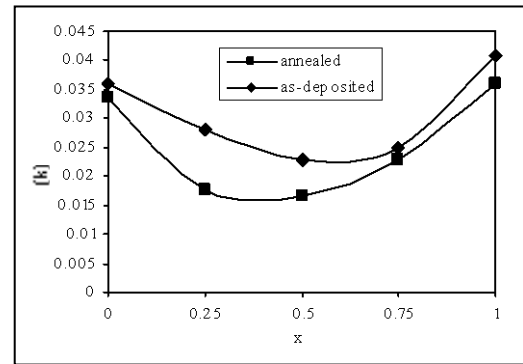
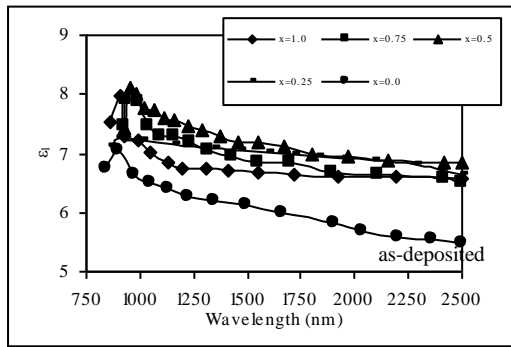


Fig.(13) The variation of extinction coefficient with composition for as-deposited and annealed CdSe_{1-x}Te_x thin films.

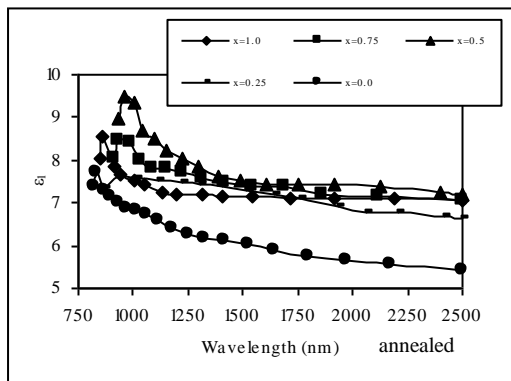
3-Dielectric Constant

Figs.(14) and (15) show the variation of the real (ϵ_1) and imaginary (ϵ_2) parts of the dielectric constant with the wavelength for as-deposited and annealed CdSe_{1-x}Te_x films. The behavior of ϵ_1 and ϵ_2 is the same as that of n and k , respectively with the variation of x values and heat treatment. This due to that the variations of ϵ_1 mainly depends on the value of (n^2), because of the smaller values of (k^2) comparison with (n^2), while the imaginary part of the dielectric constant mainly depends on (k) values which were related to the variations of absorption coefficient.

Figs.(16) and (17) represent the dependence of the real (ϵ_1) and imaginary (ϵ_2) parts of the dielectric constant on the composition and also explicit the effect of heat treatment on dielectric constant at λ equal t

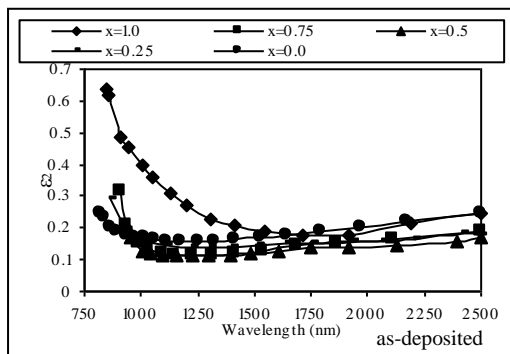


(a)

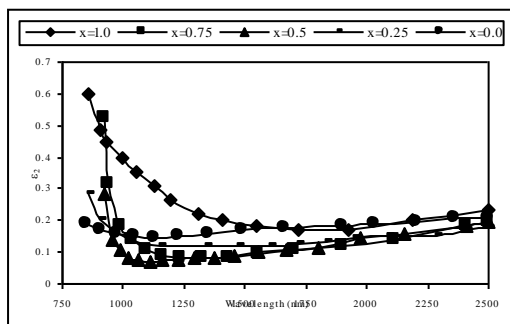


(b)

Fig.(14) The real part of dielectric constant as a function of wavelength for as-deposited and annealed CdSe_{1-x}Te_x thin films.



(a)



(b)

Fig.(15) The imaginary part of dielectric constant versus wavelength for as-deposited and annealed CdSe_{1-x}Te_x thin films.

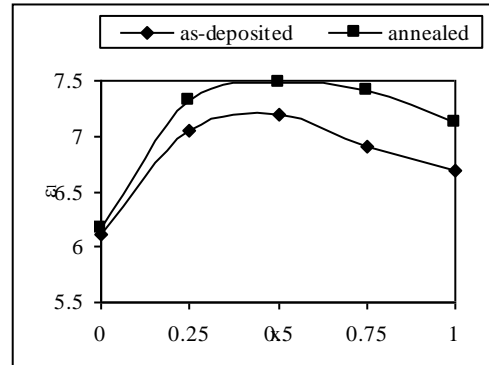


Fig.(16-a) The variation of real part of dielectric constant with composition for as-deposited and annealed CdSe_{1-x}Te_x thin films.

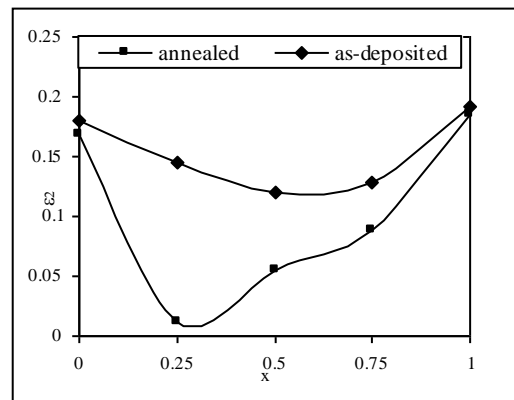


Fig.(16-b) The variation of imaginary part of dielectric constant with composition for as-deposited and annealed CdSe_{1-x}Te_x thin films.

Conclusions

1. The CdSe_{1-x}Te_x alloys for (x = 0.0, 0.25, 0.5, 0.75 & 1.0) have successfully prepared, which was used for bulks and films preparation.
2. It is observed from the transmission spectrum, the thin films surfaces and thickness was uniform because of the appearance of interference fringes clearly.
3. The refractive index in CdSe_{1-x}Te_x varies with varying the composition by non-linear relation and have a maximum value at x=0.5 which represents the point of phase transition. The refractive index increases after heat treatment due to the increasing in the films compactness.

4. The variation of real and imaginary parts of dielectric constant have similar trends as for refractive index and extinction coefficient respectively according to Maxwell's equations.

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الاستقصاء البصري لأغشية $CdSe_{1-x}Te_x$ الرقيقة

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الخلاصة:

تم تحضير سبائك المركب $CdSe_{1-x}Te_x$ بنجاح من عناصرها الأولية بنقاوة عالية (99.9999%) بخلط نسب العناصر ($x = 0.0, 0.25, 0.5, 0.75, 1.0$) من (Cd, Se, Te)، واستخدم تحليل مطياف الامتصاص الذري لمعرفة تركيز العناصر في السبيكة. وكذلك أجريت فحوصات حيود الأشعة السينية ولوحظ أن السبائك تملك تركيباً متعدد البلورات.

حضرت أغشية رقيقة من سبائك $CdSe_{1-x}Te_x$ لقيم مختلفة من التراكيز بسمك ($0.5\mu m$) بطريقة التبخير الحراري على قواعد نظيفة من الزجاج المسخن بدرجة حرارة 473K تحت ضغط واطئ جدا ($4 \times 10^{-5} mbar$) بمعدل تبخير (3 \AA/s) وبعد ذلك تمت معاملة الأغشية حرارياً تحت ضغط واطئ ($10^{-2} mbar$) بدرجة حرارة 523K لمدة ساعتين.

من الدراسة البصرية تبين أن قيم معامل الامتصاص (α) عالية بعض الشيء، ووجد ان الانتقالات الإلكترونية عند حافة الامتصاص الأساسية كانت من نوع الانتقال المباشر المسموح، وان فجوة الطاقة البصرية تتغير بشكل غير خطي مع التركيز (x) وتملك أوطاً قيمة لها عند $x=0.5$ ، وكذلك وجد أن فجوة الطاقة البصرية تزداد قليلاً بعد المعاملة الحرارية. استخدم طيف النفاذية في إيجاد الثوابت البصرية ($n, k, \epsilon_1, \epsilon_2$) التي تم حسابها بواسطة طريقة سوانبل (Swanepoel) باستخدام النفاذية العظمى T_M والنفاذية الصغرى T_m في تقنية الغلاف. وتبين ان الثوابت البصرية جميعها تتغير بشكل غير خطي مع التركيز وان انقلاب السلوك يكون دائماً عند $x=0.5$ ، وجد انها تبدي تأثيراً بعد معاملة الأغشية حرارياً. وكان تصرف الجزء الحقيقي لثابت العزل مشابهاً لتصرف معامل الانكسار، بينما الجزء الخيالي لثابت العزل كان مماثلاً لتصرف معامل الخمود.