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Effect of Annealing On the Structural and Optical Properties of Cdsexs1$_{-X}$thin Films

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Abstract: *In this work , $CdSe_xS_{1-x}$ thin films at x equal to (0 , 0.4 , 0.8 & 1) , with thickness (0.5)µm , had been prepared by thermal evaporation method on glass substrates at RT. These films were then annealed under low pressureof (10^{-2}) mbar at (373&473)K for one hour . XRD investigations showed that all the films were polycrystalline in nature and had a hexagonal structure with preferred orientation along (002) plane . It also showed that the increase in Se concentration and annealing temperature led to an increase in the intensity of (002) plane and the grain size , while the dislocation density and the strain of the films decreased with the increase in Se concentration and annealing temperature .The transmittance and absorbance spectra had been recorded in the wavelength range (480 - 1100) nm in order to study the optical properties .It was found that these films had direct optical band gap which decreases from (2.44) eV to (1.76) eV with the increase in Se concentration , while it increases with the increase in the annealing temperature at (x=1)and was not affected by annealing at (0&0.8). Moreover it was almost constant at (x=0.4) after annealing .*
Keywords:*$CdSe_XS_{1-X}$Thin Films ,Grain Size , Dislocation Density , Strain , Optical Energy Gap , Optical Constants.*

I. Introduction

Recently , group II–VI compound semiconductors , in general , and cadmium chalcogenides , in particular , have attracted intense scientific and technological interest. II–VI compound semiconductors with energy gaps between (1-3) eV covering the visible spectral range are promising candidates for different optoelectronic devices like thin film transistors , photovoltaic solar cells , photo detectors , electroluminescent layers , light emitting diodes , filters , photo-resistors and surface acoustic wave devices , and so on [1-7]. Cadmium chalcogenides form a technically important class of materials owing to their wide speared utility in a variety of electronic and optoelectronic devices [7] .The addition of cadmium selenide to cadmium sulphide has resulted in very interesting properties related to photo electrochemistry and optoelectronics [8] because the ternary materials provide a possibility of tailoring their properties as per requirements and hence project themselves as important semiconducting materials for the applications in the field of device fabrication [9]. $CdSe_XS_{1-X}$ as a ternary material has received recently much attention because it is a highly photosensitive material and has excellent properties such as large nonlinear susceptibilities , good photo-conduction , high quantum efficiency , narrow band edge and fast response time [10– 14]. Besides ,$CdSe_XS_{1-X}$ films could be used to form p-n junction solar cells with other suitable thin film materials for photovoltaic generation of electricity as a window layer [15] and as a buffer layer [9].

Annealing is a heat treatment of materials at elevated temperatures to improve their properties . Material annealing can lead to phase transitions , re-crystallization , homogenization , relaxation of internal stresses , annihilation and rearrangement of defects , and so on . The results of annealing depend significantly on the rate of heating and cooling and the time of exposure at a given temperature [16].Therefore , the main aim of this work is studying the effect of annealing on the structural and optical properties of $CdSe_XS_{1-X}$ thin films in order to fabricate films with high stability and transmittance that can be used in optoelectronic applications , solar cells and optical filters.

Experimental

$CdSe_XS_{1-X}$ thin films , with thickness (0.5) µm , were deposited on cleaned glass substrate (type corning , China) with dimensions (7.5 x 2.5 x 0.1) cm by thermal evaporation technique (Edwaed coating unit model 306 A) under high vacuum with pressure of (8×10^{-6}) mbar with deposition rate of about 9.25 A/sec . A molybdenum boat was used as a source for the evaporation of the material . The distance between the boat and the substrate was (15) cm. $CdSe_XS_{1-X}$ thin films were then annealed under a vacuum of about (10^{-2}) mbar in a furnace of type (Precision GCA) for one hour at (373 & 473) K . Films thickness was determined by a multiple beam interferometry (Fizeau fringes in reflection) .The films were characterized by X-ray diffraction technique using (Philips X-ray diffractometer) with CuK^{α} radiation at wavelength (1.5406)A'.A(UV-160A UV-visible recording) spectrophotometer supplied by Japanese company (Shimadzu) was used to record the optical absorbance and transmittance spectra of $CdSe_XS_{1-X}$ thin films at wavelength range (480-1100) nm .

II. Results And Discussion

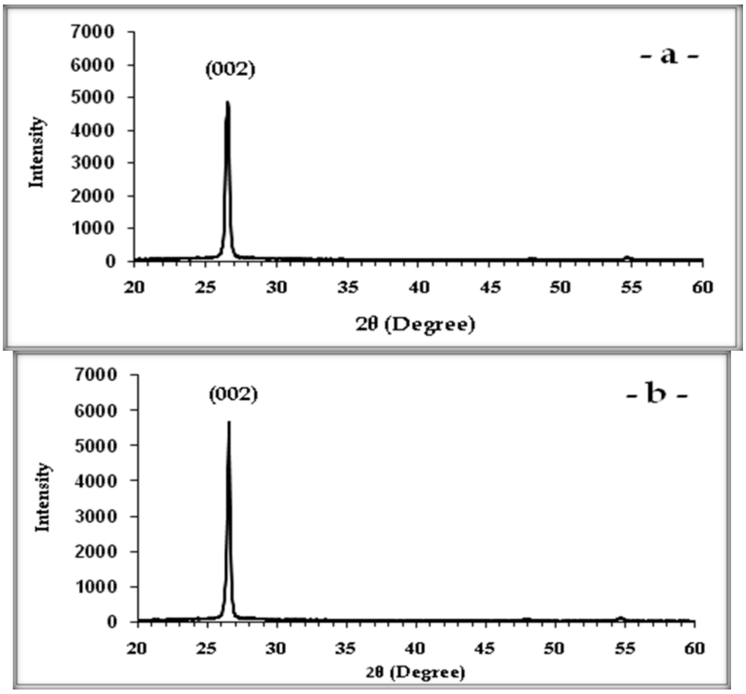
Structural properties

Figures (1, 2 , 3 & 4) illustrate the effect of annealing at (373 & 473) K for one hour on the X-ray diffraction patterns of $CdSe_xS_{1-x}$ thin films at x equal to (0 , 0.4 ,0.8 & 1) respectively . The XRD patterns for all cases reveal a polycrystalline in naturewith a hexagonal structurehaving one peak corresponding to the reflection from (002) plane except for $CdSe_{0.4}S_{0.6}$ having another peak corresponding to the reflection from (103) plane . It is clear from these figures that annealing leads to an increase in the peaks' height for all values of x , indicating that increasing the annealing temperature improves the crystallinity of $CdSe_xS_{1-x}$ thin films and also leads to the propensity of the films' atoms toward their preferential orientation along (002) plane with their stable structure (hexagonal) .As well as , it is observedfrom these figures that there is a slight shift toward higher value of 2θ with the increase in annealing temperature. The grain size (D) of $CdSe_xS_{1-x}$ thin films was calculated from the relation [17] :

$$D = \frac{0.9\lambda}{\beta\cos\theta} \quad (1)$$

Where , β: is the full width at half maximum (FWHM) in radian and λ : is the X-ray wavelength (1.5406 A°).

From table (1) , it is obvious that there is an increasing in the grain size of $CdSe_xS_{1-x}$thin films with the increasing annealing temperaturefor all values of x which can be also deduced from the decreasing in the (FWHM) with the increasing annealing temperature . Muraliet al.[18] , who had annealed these films in an argon atmosphere at different annealing temperatures in the range (623 - 723) K , had mentioned that the annealed films exhibited the same peaks as in the case of the as-deposited films corresponding to (002 , 100 , 101 , 102 , 110 , 103 , 200 , 112 , 201 , 202 , 203 , 210 &211) planes for all the compositions of x where $(0 \leq x \leq 1)$. He also mentioned that the crystallinity of the X-ray diffraction patterns improved and the (FWHM) of the peaks decreased after annealing .



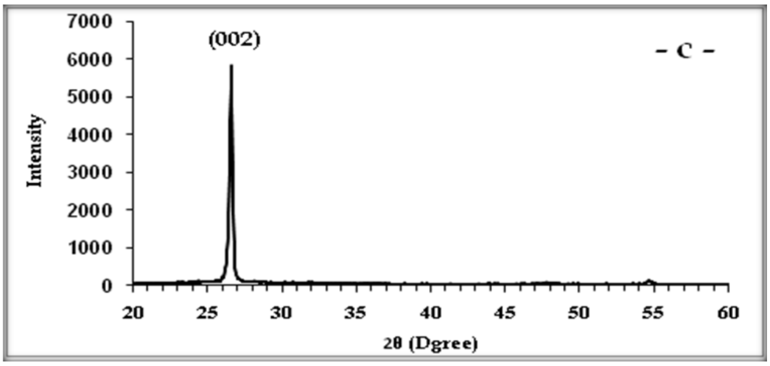


Fig.(1): XRD patterns of $CdSe_xS_{1-x}$ thin films at x=0 with different annealing temperature
(a) as-deposited , (b) 373 K , (c) 473 K

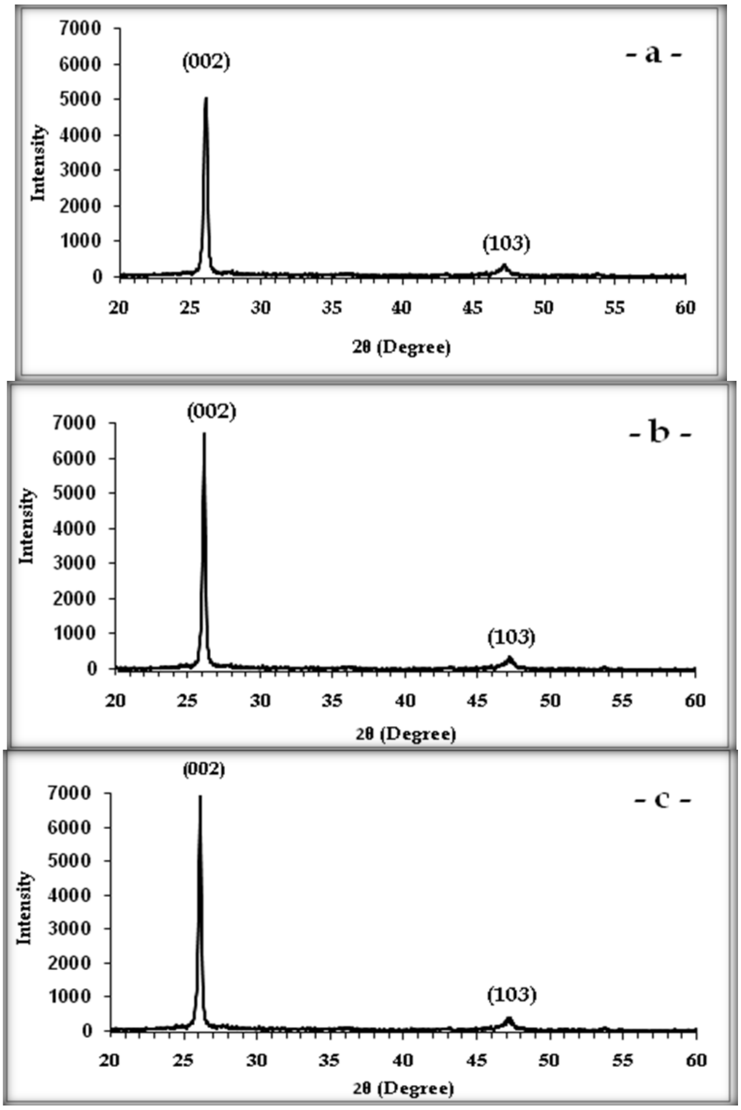


Fig.(2): XRD patterns of $CdSe_xS_{1-x}$ thin films at x=0.4 with different annealing temperature
(a) as-deposited , (b) 373 K , (c) 473 K

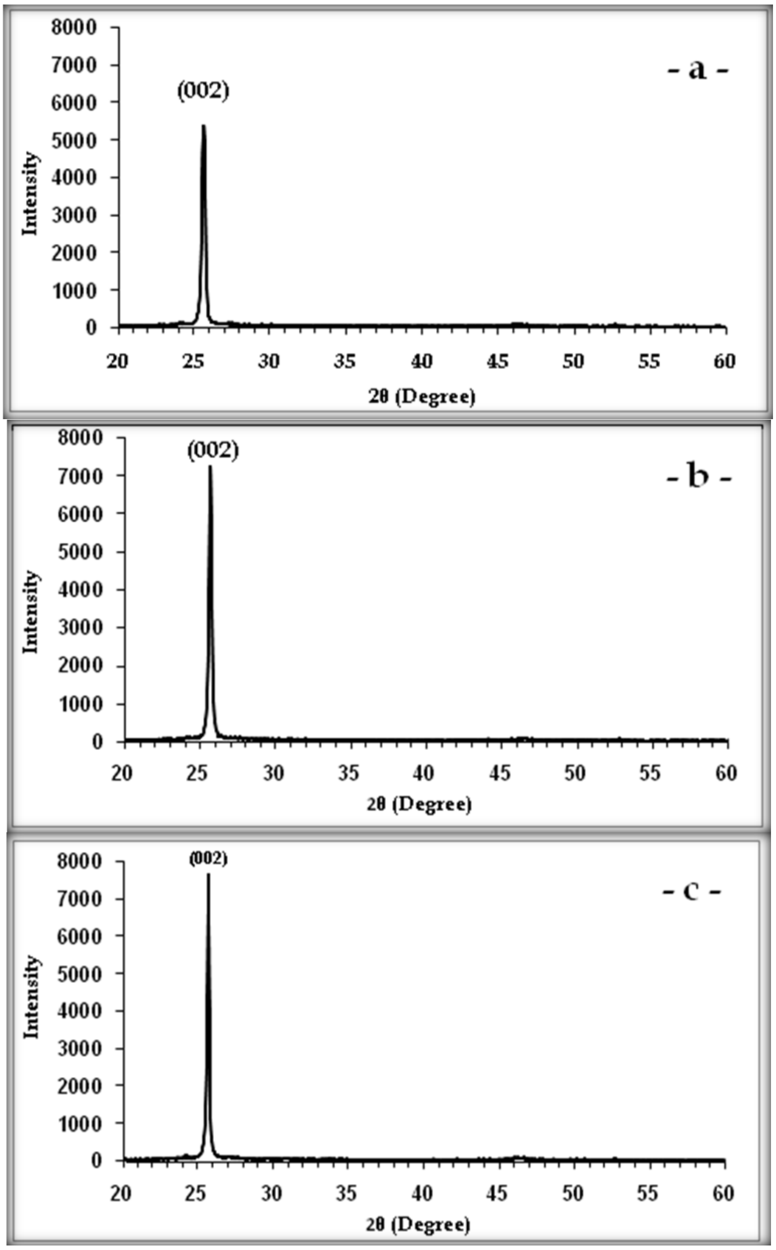
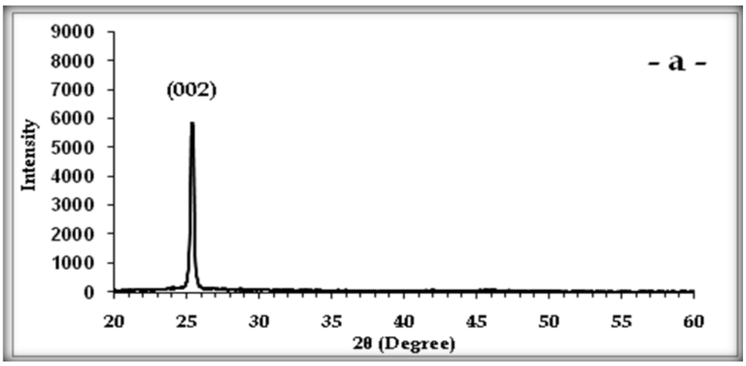


Fig.(3): XRD patterns of $CdSe_xS_{1-x}$ thin films at x=0.8 with different annealing temperatureas-deposited , (b) 373 K , (c) 473 K



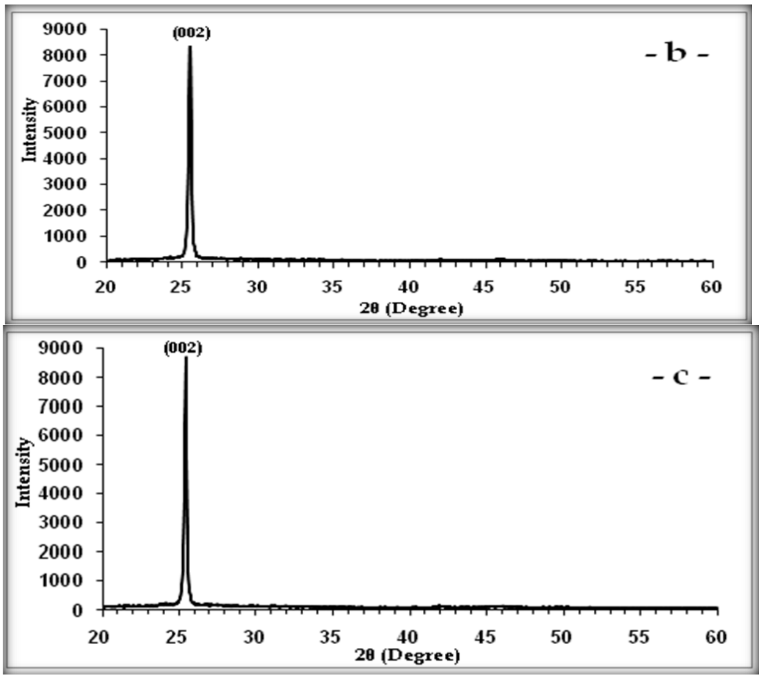


Fig.(4): XRD patterns of $CdSe_xS_{1-x}$ thin films at x=1 with different annealing temperature
(a) as-deposited , (b) 373 , (c) 473 K

This increase in the grain size of the films after annealing is evidence on the improvement of the films' crystallinity . Our result agree with Metinet al.[19] and Babanet al.[20], who found that the grain size of the films at (x = 0) and(x=1) respectively increases after annealing .The strain (ξ) [21]and the dislocation density (δ)[22] developed in $CdSe_xS_{1-x}$ thin films were calculated from the relations (2)&(3)respectively :

$$\xi = \frac{\beta \cos\theta}{4} \quad (2)$$

$$\delta = \frac{1}{D^2} \quad (3)$$

It is clear from table (1) that the strain and the dislocation density of $CdSe_xS_{1-x}$ thin films decrease with the increase in annealing temperature for all values of x . This indicates that annealing imparts the atoms of the films the sufficient energy to diffuse more easily to find their proper location in the lattice .Therefore , when these atoms take their proper location as a result of annealing , surface energy increases and a state of equilibrium is achieved causing reduction in the strain and the dislocation density after annealing . Our results agree with Metinet al.[19] , who found that strain and the dislocation density of the films at (x = 0) decrease after annealing .

Table(1): Variation of the full width at half maximum , grain size , dislocation density and strain of $CdSe_xS_{1-x}$ thin films with composition (x) at different annealing temperature.

x	(T_a)K	(FWHM) rad	Grain size (nm)	Dislocation density (lines.m^{-2}) x 10^{14}	Strain (rad) x 10^{-4}
0	RT	0.00457	31.41	10.31	11.13
	373	0.00436	32.63	9.388	10.62
	473	0.00420	33.91	8.695	10.22
0.4	RT	0.00441	32.25	9.609	10.74
	373	0.00427	33.27	9.032	10.41
	473	0.00419	33.93	8.681	10.21
0.8	RT	0.00365	38.92	6.598	8.904
	373	0.00363	39.15	6.522	8.852
	473	0.00361	39.29	6.477	8.822
1	RT	0.00340	41.78	5.726	8.295
	373	0.00326	43.48	5.288	7.971
	473	0.00316	44.97	4.944	7.707

Optical properties:

Figure (5) illustrates the transmittance spectrums of $CdSe_xS_{1-x}$thin films at (RT , 373 & 473) K for x equal to (0 , 0.4 , 0.8 & 1) . It is clear from this figure that the transmittance of $CdSe_xS_{1-x}$thin films increases with the increase in the annealing temperature for all values of x . This indicates that annealing imparts the film's atoms the sufficient energy to diffuse more easily through the crystal structure of the film and leads to reach the stability state . Our results agree with Jafariet al. [23] andBabanet al.[20] , who found that the transmittance increases with the increase in the annealing temperature at x=0 . Our resultsalsoagree withZaidunn[24]at x=0.5 . Furthermore , it is obvious from figure (5) that the transmittance values of $CdSe_xS_{1-x}$thin films annealed at (473) K were more than 90 % at x equal to(0 , 0.4 & 0.8) and more than 80 % at x=1 . On the other hand , at (373) K the maximum transmittance values of $CdSe_xS_{1-x}$thin films reach about (87 , 83 , 81 & 79) % for x equal to (0 , 0.4 , 0.8 & 1) respectively . According to this it can be deduced that annealing temperature at (473) K was more effective temperature than (373) K on $CdSe_xS_{1-x}$thin films . Our results agree withJafariet al. [23] , who found that the best optical transmittance was observed at annealing temperature (473) K for x=0 .

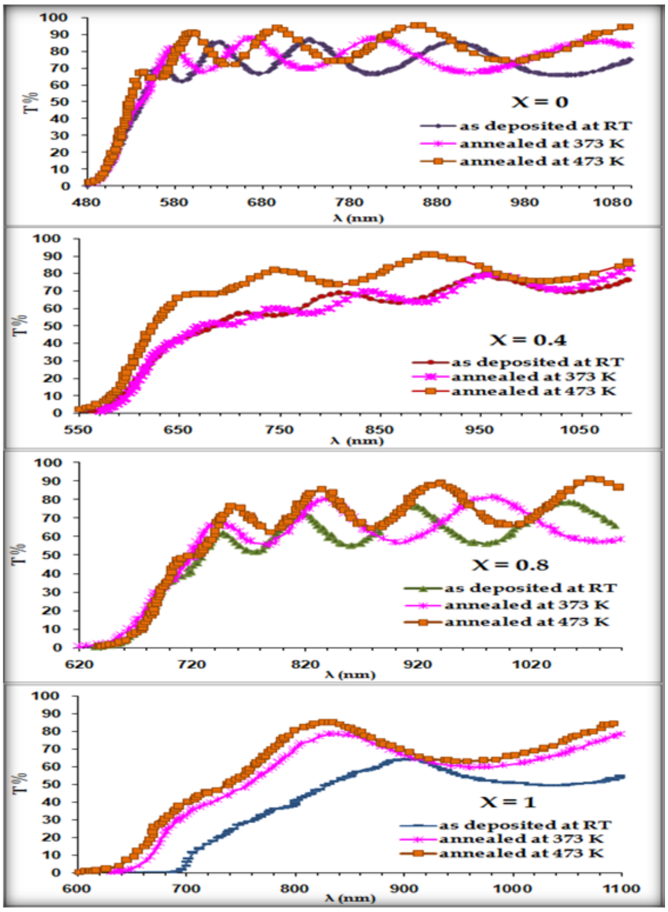


Fig.(5): Transmittance spectrums of $CdSe_xS_{1-x}$ thin films at different annealing temperature and different composition of (x)

The absorbance spectrums of $CdSe_xS_{1-x}$thin films at (RT , 373 & 473) K for x equal to (0 , 0.4 , 0.8 & 1) are illustrated in figure (6) . It can be observed from this figure that the absorbance of $CdSe_xS_{1-x}$thin films decreases with the increase in annealing temperature for all values of x . Our resultsalso agree with Zaidunn[24]at x=0.5 . Also it is clear from figure (6) that the absorption edge shifts to the shorter wavelengths as the annealing temperature increases at x equal to (0.4 & 1) . This shifting in the absorption edge indicates an increase in the optical energy gap at these values of x . A similar observation was also reported byZaidunn[24]for x=0.5 .Figure (7) illustrates the absorption coefficient of $CdSe_xS_{1-x}$thin films at (RT , 373 & 473) K for x equal to (0 , 0.4 , 0.8 & 1) . It is clear from this figure that the absorption coefficient of $CdSe_xS_{1-x}$thin films decreases with the increase in annealing temperature for all values of x . This decrease is attributed to the decrease in the absorbance of $CdSe_xS_{1-x}$thin films with the increase in annealing temperature where the relation between the absorbance and the absorption coefficient is proportional at constant thickness . Our results agree withZaidunn[24],who found that the absorption coefficient decreases with the increase in annealing temperature at x=0.5 .

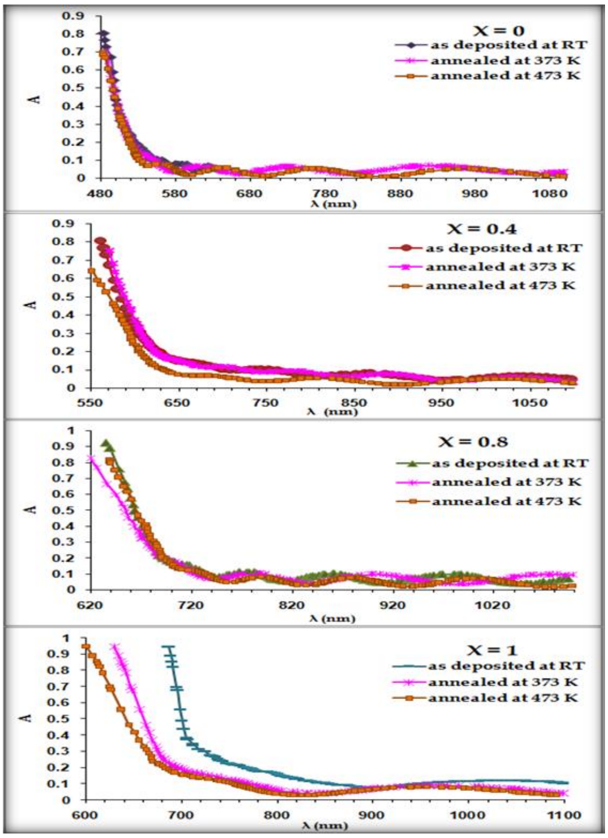


Fig.(6): Absorbance spectrums of $CdSe_xS_{1-x}$ thin films at different annealing temperature and different composition of (x)

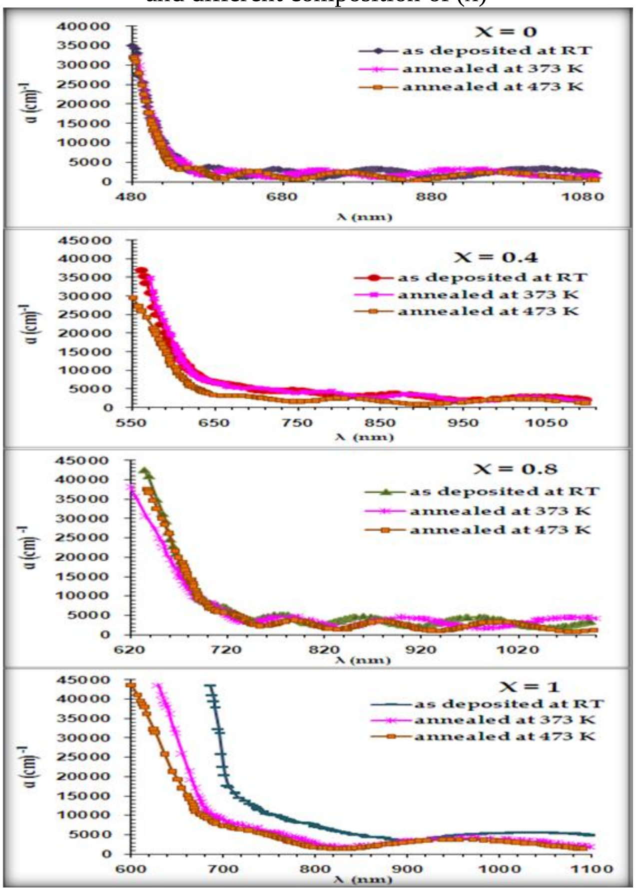


Fig.(7): Absorption coefficient of $CdSe_xS_{1-x}$ thin films at different annealing temperature and different composition of (x)

The optical energy gap values (E_g) for $CdSe_xS_{1-x}$ thin films prepared by thermal evaporation method have been determined from the region of the high absorption at the fundamental absorption edgeof these films by using Taucequation[25] :

$$\alpha h\upsilon = B_o(h\upsilon - E_g)^r \tag{4}$$

Where α :the absorption coefficient , is $h\upsilon$: incident photon energy in eV , is a coB_o :ant depends on the nature of the material (properties of its valence and conduction band) [26]and is a r :stant depends on the nature of the transition between the top of the valence band and bottom of the conduction band [21].

This equation is used to find the type of the optical transition by plotting the relations $(\alpha h\upsilon)^2$, $(\alpha h\upsilon)^{2/3}$, $(\alpha h\upsilon)^{1/2}$ and $(\alpha h\upsilon)^{1/3}$ versus photon energy ($h\upsilon$) and select the optimum linear part. It is found that the first relation yields linear dependence, which describes the allowed direct transition , then E_g was determined by the extrapolation of the portion at (α=0) .The variation of the optical energy gap (E_g) of $CdSe_xS_{1-x}$thin films with annealing temperature is listed in table (2) . It is obvious from this table that$E_g$at x equal to (0 &0.8) remains constant at (2.44 & 1.84) eV respectively and it is not affected by increasing annealing temperature . Our results agree with letinrgüet al.[27] , who found that the optical energy gap of the films at x=0 annealed between (323 & 523) K dose not differ much from the values obtained for as-deposited films approximately (2.45) eV . However , annealing at (573) K and above do result in the optical energy gap decreasing slightly to a value of about (2.37) eV . Also it is obvious from this table that$E_g$at (x = 0.4) is almost constant ,while$E_g$at (x =1) increases with the increase in annealing temperature which can be also deduced from the shift in the absorption edge to the shorter wavelengths with the increase in annealing temperature . This increase in the E_g is attributed to the annealing role in improving the crystallinity of the films by decreasing the strain and the dislocation density that happened in the films .

Table (2):Variation of the optical energy gap (E_g) of $CdSe_xS_{1-x}$thin films with annealing temperature.

X	(E_g) eV		
	(RT)K	(373)K	(473)K
0	2.44	2.44	2.44
0.4	2.10	2.11	2.12
0.8	1.84	1.84	1.84
1	1.76	1.86	1.90

The refractive index $(n_\circ)$ of $CdSe_xS_{1-x}$ thin filmswas determined by using the following equation [26]:

$$n_\circ = \sqrt{\frac{4R}{(R-1)^2} - k_\circ^2} - \left(\frac{R+1}{R-1}\right) \tag{5}$$

Where , R : is the reflectance of the films and k_o : is theextinction coefficient.

Figure (8) illustrates the variation of the refractive index with the wavelength for $CdSe_xS_{1-x}$thin films at(RT , 373 & 473) K for x equal to (0 , 0.4 , 0.8 & 1) . It can be observed from this figure that the refractive index of $CdSe_xS_{1-x}$thin films decreases with the increase in annealing temperature for all values of x . Thisdecrease is very pronounced at (473) K . The decrease in the refractive index is associated with the fundamental band gap absorption. Our results agree with Zaidunn[24], who found that the refractive index decreases with the increase in annealing temperature for x=0.5 .

The extinction coefficient () $k_\circ$as determined by using the following equation [26]:

$$k_\circ = \frac{\alpha\lambda}{4\pi} \tag{6}$$

Where: , $\alpha$the absorptioncoefficient and λ: the wavelength of the incident photon .

It is clear from this equation that$k_\circ$depends on α and has a similar behavior to α .The variation of the extinction coefficient of $CdSe_xS_{1-x}$thin films with the wavelength at (RT , 373 & 473) K for x equal to (0 , 0.4 , 0.8 & 1) is illustrated in figure (9) . From this figure it can be noted that the extinction coefficient of $CdSe_xS_{1-x}$thin films decreases with the increase in annealing temperature for all values of x . Because annealing leads to the overcoming of some of local states and then decreases the absorbance and increases the transmittance . Hence the number of charge carriers produced from the deviation of the ideal structure of the film will be less . The extinction produced from the absorption of radiation by charge carriers then decreases , which is attributed to the decrease in the absorption coefficient with the increase in annealing temperature , since the extinction coefficient depends on the absorption coefficient and has a similar behavior to it according to equation (6) . Our results agree with Zaidunn[24], who found that the extinction coefficient decreases with the increase in annealing temperature for x=0.5 .

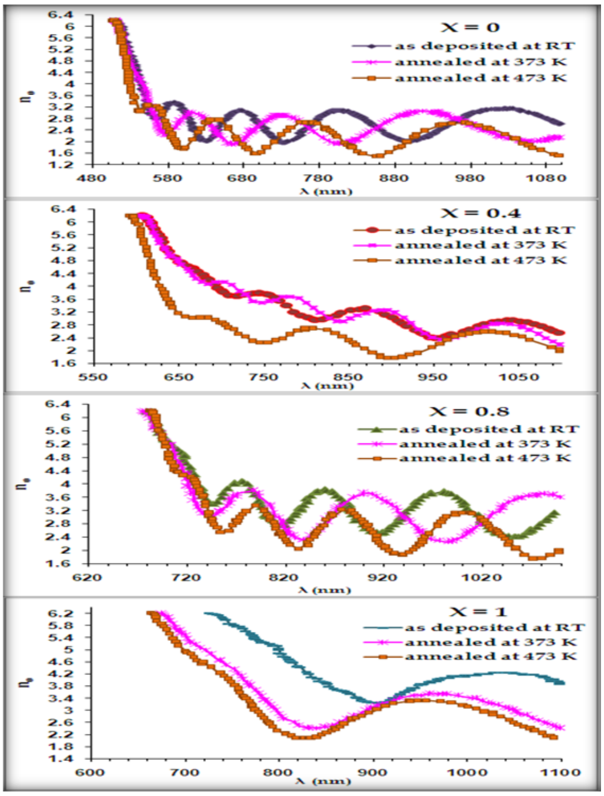


Fig.(8): Refractive index of $CdSe_xS_{1-x}$ thin films at differentannealing temperature and different composition of (x)

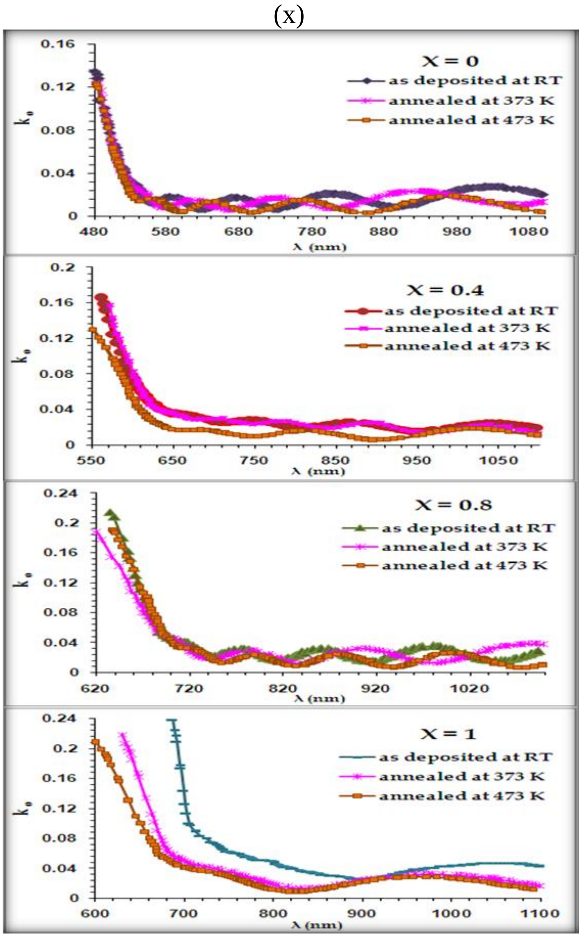


Fig.(9): Extinction coefficient of $CdSe_xS_{1-x}$ thin films at differentannealing temperature and different composition of (x)

The rea (ε_r)d imaginarypart(ε_i) the dielectric constant were determined by using equations (7)&(8)[26] respectively:

$$\varepsilon_r = n_\circ^2 - k_\circ^2 \qquad (7)$$

$$\varepsilon_i = 2n_\circ k_\circ \qquad (8)$$

According to eq.(7) ,the real part of the dielectric constant (ε_r) depends mainly on the value of$(n_\circ^2)$, because of the smaller values of $k_\circ$ comparison with $(n_\circ^2)$ whereas according to eq.(8) the imaginary part of the dielectric constant (ε_i) depends mainly on the $(k_\circ)$ values which are related to the variations of the absorption coefficient . Figures (10) and (11) illustrate the variation of the real and imaginary parts of the dielectric constant with the wavelength respectively for $CdSe_xS_{1-x}$thin films at (RT , 373 & 473) K for x equal to (0 , 0.4 , 0.8 & 1) . It can be observed from this figure that the real and imaginary parts of the dielectric constant of $CdSe_xS_{1-x}$thin films decrease with the increase in annealing temperature . This is attributed to the dependence of the real and imaginary parts of the dielectric constant on the refractive index and the extinction coefficient respectively .

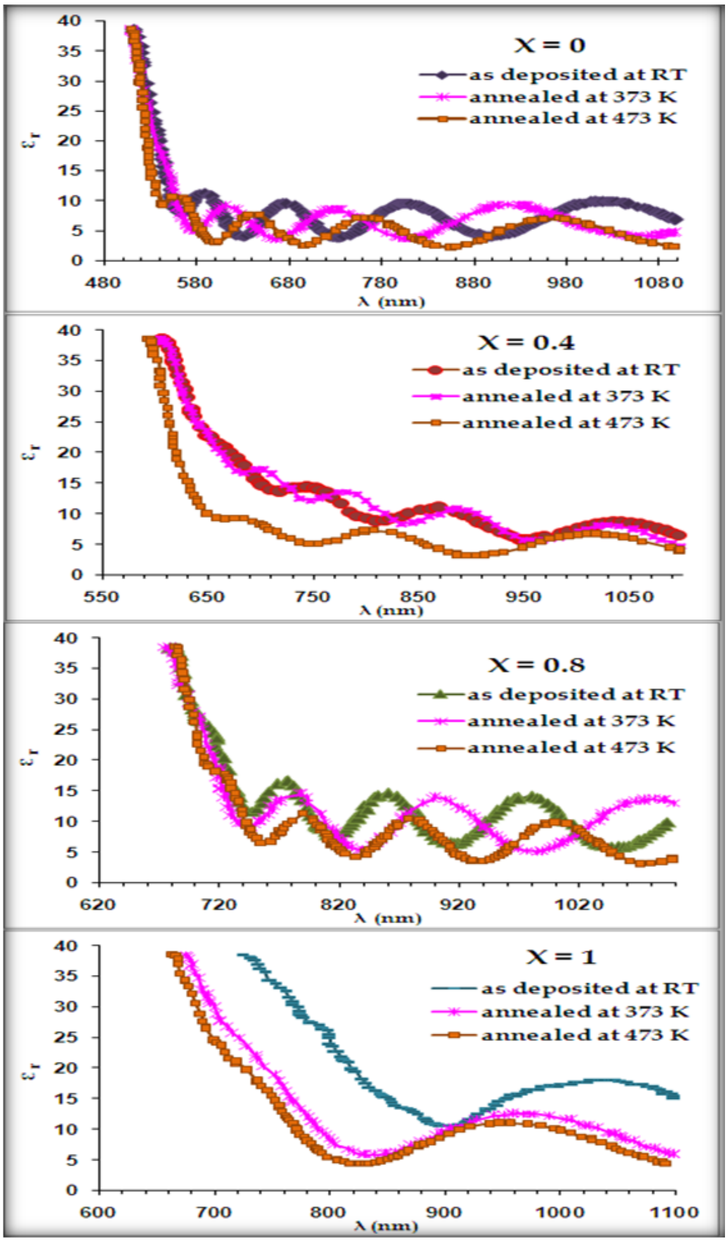


Fig.(10): The real part of the dielectric constant of $CdSe_xS_{1-x}$ thin films at different annealing temperature and different composition of (x)

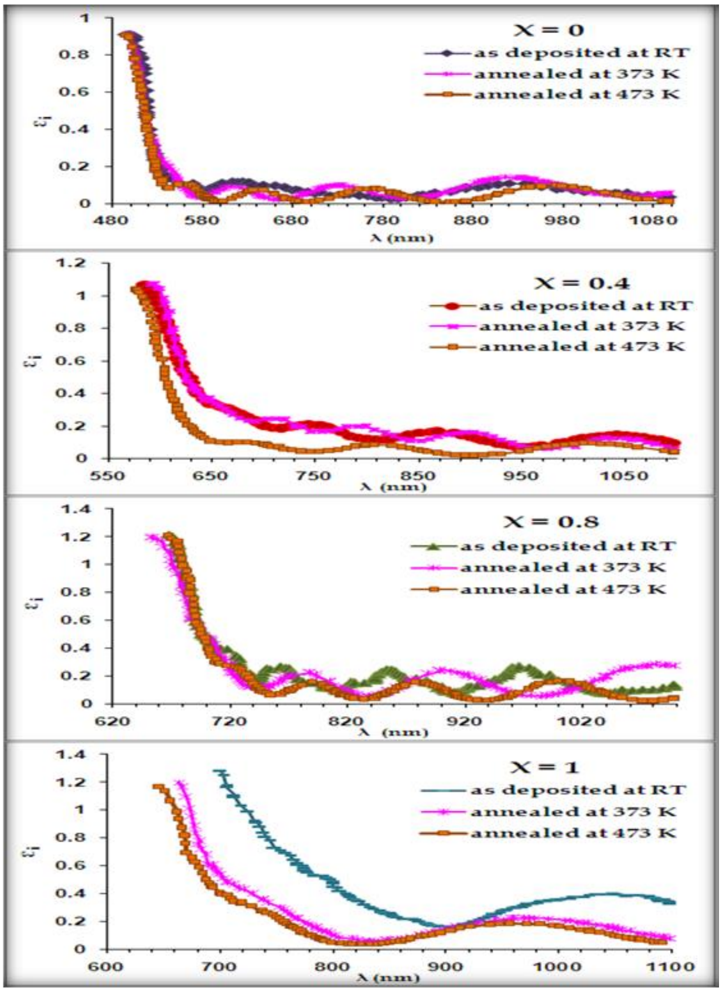


Fig.(11): The imaginary part of the dielectric constant of $CdSe_xS_{1-x}$ thin films at different annealing temperature and different composition of (x)

III. Conclusions

From the results of this research , several conclusions had been deduced as follows :

a) The crystallinity of the films has been improved by annealing in vacuum and by increasing the Se concentration which has been deduced from the decrease in the (FWHM) , dislocation density and strain and the increase in theintensity of the XRDpatterns and the grain size of the films . .

b) Thetransmittanceof the films decreases with the increase in the Se concentration , while it increases after annealing ; the maximum optical transmittance was obtained at annealing temperature (473) K .

c) $CdSe_xS_{1-x}$ thin films exhibit a direct band gap which strongly depends on the Se concentration , almost covering the entire visible spectrum which makes these films are suitable for optoelectronic devices , especially for solar cell and optical filters .

d) The films' ability to the extinction of the incident photon energy increases with the increase in the Se concentration , while it decreases after annealing .

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