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Effect of Thickness on Structural, Morphological, and Optical Properties for Nanocrystalline Thin Films of Cd1-xSnxS, for Optoelectronic Applications

Lamia K.Abbas*

Department of Physics, College of science, University of Baghdad, Baghdad, Iraq.
*Corresponding Author.

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Abstract

Thin films of Sn1-xCdxS nanocrystals (X=0,0.7, and 1) with different thicknesses 200, 300, and 400 nm were prepared on glass bases at 300 C by the participation method and chemical spray pyrolysis method (CSP). The effect of the concentration of Sn and thickness on the structural and optical properties of the prepared films was studied. The films were characterized to evaluate the structure, transmittance, and optical energy band gap. X-ray diffraction (XRD) studies showed that the films were polycrystalline with hexagonal and orthogonal structures. XRD and atomic force measurement results showed that the grain size increases with its thickness. The optical band gap energies and optical transmission and absorption types of the membranes were determined from the optical transmittance spectra. Optical absorption studies in the wavelength range 200-1100 nm showed that the energy gap values of the films decrease from 2.48-1.95 eV, 2.41-1.92 eV, and 2.3- 1,89 eV with increasing Sn concentration and with increase the thickness 200, 300, and 400 nm respectively. The best properties of the prepared material were determined for its possible use in the manufacture of solar cells

Keywords: Cd1-xSnxS nanocrystallain, CdS nanoparticle, Chemical spray pyrolysis, Sn, Thin films.

Introduction

There is considerable interest in the field of transparent semiconducting materials such as CdS, SnS, etc. for use in a variety of applications, including architectural windows, solar cells, heat reflectors, light transparent electrodes, thin-films photovoltaic devices, and many other optoelectronic devices[1]. CdS and SnS are both promising materials for solar cells. CdS, which belongs to II-VI compound semiconductors [2,3], is an n-type semiconductor with a direct band gap of (2.4eV), CdS is used as a window material for heterojunction thin films solar cells [4-8]. It has also applications in light-emitting diodes (LED), gas detectors photovoltaic cells, nonlinear optics, and thin film transistors [7,9,10]. SnS is one of the Tin chalcogenide layered semiconductors in group IV-VI, SnS, SnSe is a promising material for solar energy conversion [11, 12]. SnS films are highly suitable for any application in several of solid-state devices, such as photovoltaic, photoelectrochemical(PEC), photoconductive cells, and intercalation battery systems[13,14]. In addition, SnS thin films have a large optical absorption coefficient (> 104 cm-1) [13-15]. It

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is a p-type window layer heterojunction device [16- 18]. SnS materials have an optical energy gap for direct transitions of 1.6 eV.

Various methods employed for the deposition of CdS and SnS films are chemical bath deposition[19, 20], chemical vapour deposition[1], electrochemical deposition[21-23], rf sputtering, vacuum evaporation[1], and spray pyrolysis method [24, 25]. Amongst all the deposition methods, the spray pyrolysis (SP) method is a simple, convenient, and low-cost method for large-area deposition of many binaries, ternary, and quaternary semiconducting films with varying anion and cation concentrations. In this study, thin films of Cd1-xSnxS nanocrystals were prepared using the thermal chemical spray method and their structural and optical properties were studied by studying the effect of changing thickness on their properties to obtain the best properties, which we were able to use in the manufacture of solar cells.

Materials and Methods

Cd1-xSnxS nanocrystalline thin films were prepared on glass substrates at 300°C substrate temperature by the CSP method using aqueous solutions. The spray solution (prepared using the participant method) consisted of (by volume) 0.05M cadmium chloride ($CdCl_2.H_2O$), thiourea (H_2NCSNH_2), and 0.05 tin chloride ($SnCl_2.2H_2O$) solutions and sodium hydroxide (NaOH) to ensure maximum growth of medium alkaline. Tin solution or cadmium solution or together Tin and cadmium acetate solutions were heated up by 45°C with magnetic stirring principle. The alkaline NaOH solution was then added drop by drop to reach 10 pH. The color of the CdS solution in 30 minutes was light yellow. Then its color grew darker as the reaction time rose until it changed completely from dark yellow to orange. The composition of Cd1-xSnxS films changed from pure SnS to pure CdS (x=0, 0.7, and1). The glass substrates were soaked in chromic acid, cleaned in isopropyl alcohol, rinsed in distilled water at each step, and dried in air. The chemical spray-head-to-substrate distance was fixed approximately at 30 cm. Nitrogen was used as the carrier gas during spraying. The glass substrates were heated by an electrical heater and control of substrate temperature was done using a chrome – alumen thermocouple. The thickness of the films was measured using the weighted method. The structural properties of the films were studied using XRD analysis and it was performed by a Rigaku X-ray diffractometer system using CuKα radiation with the wavelength of λ=1.5406A^o. The morphological properties of the films were investigated using CSPMAA3000 AFM. Optical transmittance spectra of the films were carried out using the Shimadzu UV-160 (UV-Visible-NIR Spectrophotometer) system covering the spectral range from 200 to 1100 nm.

Results and discussion

Structural characterization

The structural properties of the Cd1-xSnxS films have been investigated by XRD patterns. The XRD patterns of the samples are given in Fig. 1. The spectra have been obtained by scanning angle 2θ in the range from 20° to 60°. The existence of multiple eight diffraction peaks, and sulphide phases in the diffraction patterns indicates the polycrystalline nature of the Cd1-xSnxS. It is seen that the crystallinity of the CdS films is better than that of other films. It should be noted that the XRD patterns exhibit clear dependence on Sn concentration. The CdS film has been crystallized in a hexagonal (JCPDS Card no:96-101-1055) with the preferential orientation of(002) as shown in Fig. 1a. The intensity of the peak corresponding to the CdS phase decreases as SnS concentration increases as shown in Fig.1.c The SnS phase becomes dominant with lower Cd content. The SnS film has been crystallized in an orthorhombic structure (JCPDS Card no: 96-900-8296) with the preferential orientation of (021) as shown Describes the spectrum X-ray diffraction of films (SnxCd1-xS) thin when changing thickness, as evidenced by the way that the increase in thickness leads to increase the height of the peaks and increase the intensity of which indicates that the change in thickness affects the installation of the film, and this is due to the difference in the number of atoms forming the film from one area to another.
The crystallite size of all prepared samples was determined for three peaks with the greatest intensity based on the following Scherrer equation [26]:

$$D= 0.9\lambda /\beta \cos\theta, \text{ --- } 1$$

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Where λ is the wavelength of the x-ray (Å), β is FWHM (radian) is the intrinsic full width at Half Maximum of the Peak and θ is the Bragg's diffraction angle of the respective XRD Peak. The dislocation density (δ) is calculated using[27] δ=1/D2. It is interesting to note that the crystallite size of $Cd_{1-x}Sn_xS$ thin films improves and the defects like dislocation density decrease with the increase of film thickness. This may be due to the improvement in crystallinity in the films with the increase in film thickness. The crystallite size (D) and dislocation density (δ) for different thicknesses are shown in Table 1.

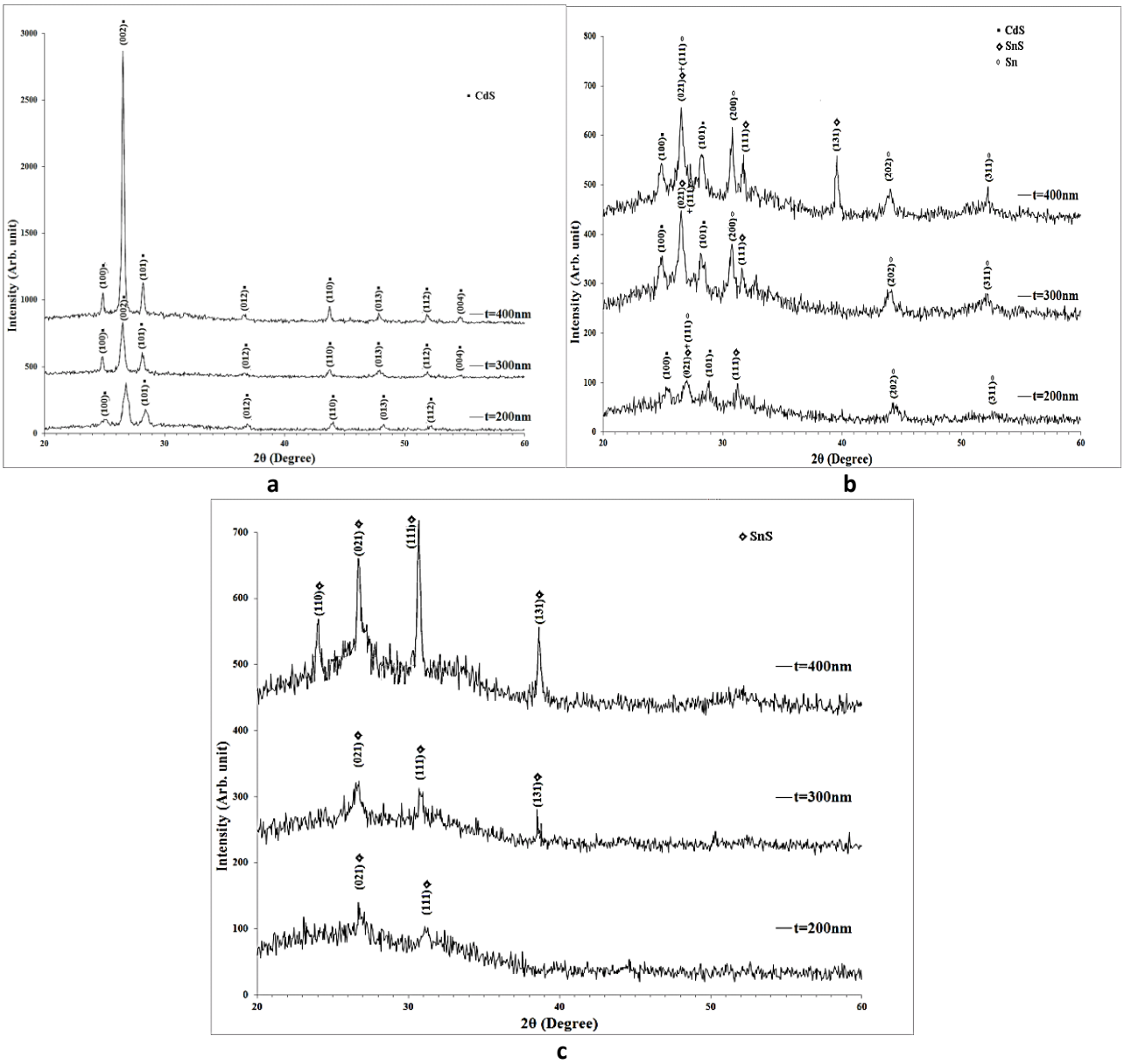

Figure1. a: XRD patterns of the CdS nanocrystall thin films of different thicknesses, b: XRD patterns of the $Sn_{0.7}Cd_{03}S$ nanocrystall thin films,c: XRD patterns of the SnS nanocrystall thin films of different thicknesses.

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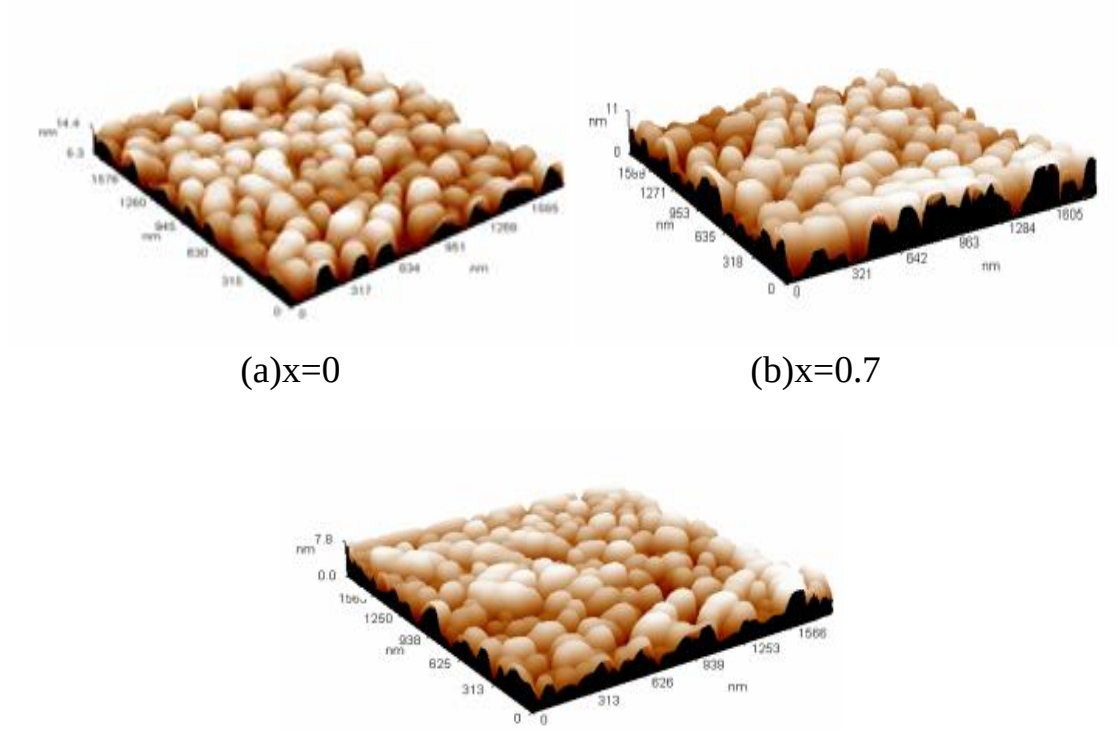
Table 1. Calculated values of the structural parameters of $Cd_{1-x}Sn_xS$ thin films of different thickness.

Thin films	2θ (Deg.)	FWHM β(deg.)	crystallite size (nm)	δ $*10^{-3}$ m^{-2}	Thickness (nm)
CdS	26.785	0.531	15.38	4.22	200
	26.558	0.354	23.06	1.88	300
	26.574	0.214	38.12	0.688	400
$Sn_{0.7}Cd_{03}S$	26.959	0.643	12.70	6.20	200
	26.474	0.468	17.43	3.29	300
	26.515	0.468	17.44	3.28	400
SnS	26.842	0.819	9.97	10.06	200
	26.649	0.936	8.72	13.15	300
	26.339	0.526	15.51	1.44	400

AFM technology provides digital images that quantitatively evaluate surface characteristics, including grain size (nm) and roughness average (nm). Fig. 2 demonstrates the spherical shapes for all samples studied. The images also show a non-compact surface which is not smooth. The grain size (average diameter) obtained from AFM measurements are listed in Table 2. The sizes of nanoparticles obtained from the AFM images appear bigger than the values obtained from XRD measurements [28]. Those results can be interpreted for several reasons; the first explanation is that the nanoparticles tend to form aggregates on the surface during deposition. The second explanation is related to the shape of the tip AFM which may cause misleading cross-sectional views of the sample. The results show that the grain size of CdS is larger than other samples which are consistent with X-ray results.

Table2. Variation of grain size and average diameter of $Cd_xSn_{1-x}S$ Nanocrystalline thin films with thickness of 400 nm

nanocrystall films	Grain size(nm)	Avg. Diameter(nm)
CdS	38.12	112.06
$Cd_{0.3}Sn_{0.7}S$	17.44	117.03
SnS	15.51	99.59



(a)x=0 (b)x=0.7

(c)x=1

Figure 2. AFM micrographs of (a)CdS ,(b) $Sn_{0.7}Cd_{03}S$,(c)SnS Nanocrystalline thin films.

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The optical properties of all films with different thicknesses 200,300 and 400 nm have been determined by using the transmittance (T) and absorbance (A) spectrum in the region (220-1100) nm.

The optical transmittance spectra of the prepared samples are shown in Fig. 3. The figure shows that the permeability decreases with increasing thickness for all prepared samples. The transmittance shows two distinct regions, first at short wavelengths of less than 500 nanometers, where the transmittance suddenly increases with increasing wavelength. This phenomenon is attributed to the pack-pack transition, and the behavior of transitions appears directly in this region, while the second area, is larger than 500 nanometers. We notice that the curve tends to saturate. This is agreed upon by the researcher [29], as well as the researcher [30], while Fig. 4 shows the absorption spectrum.

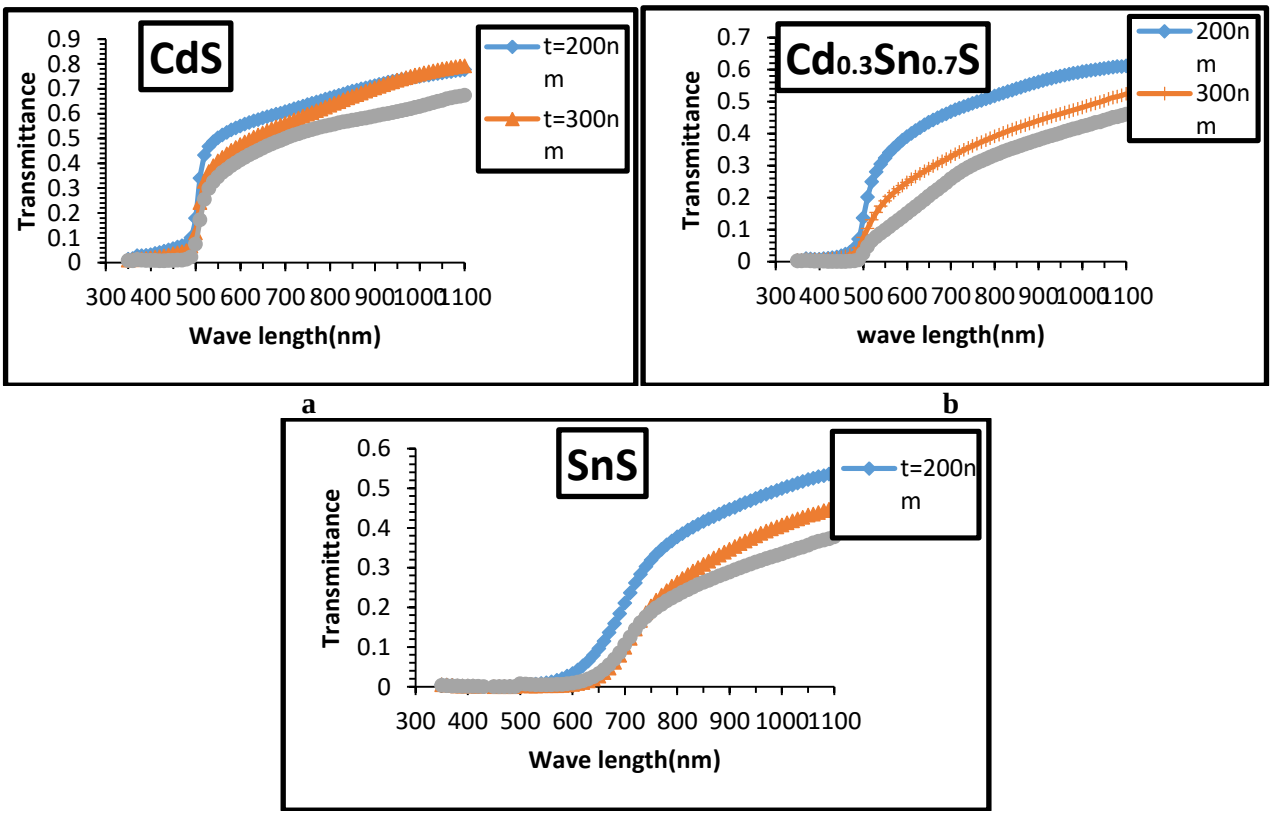
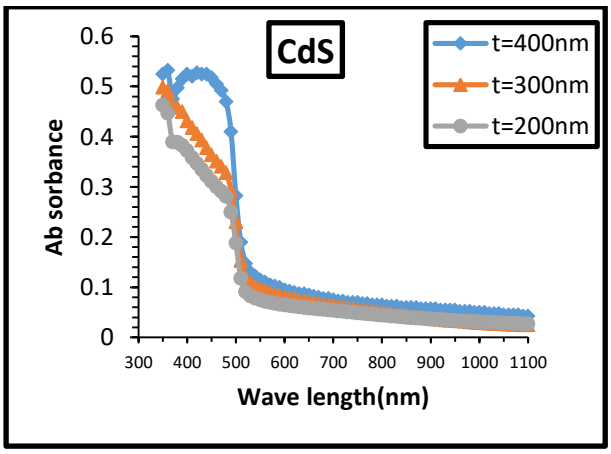


Figure 3. optical transmittance spectra of (a) CdS ,(b) $Cd_{0.3}Sn_{0.7}S$,(c) SnS Nanocrystalline thin films with different thicknesses

The absorption spectra of $Cd_{1-x}Sn_xS$ nanocrystalline thin films are shown in Fig. 3. It was observed that the absorption edge shifts towards longer wavelengths with increasing thickness, and this indicates a decrease in the energy gap with increasing thickness. Absorption increases with increasing thickness, and this is because increasing thickness increases the number of atoms, which provides absorption instances for many photons. It has been observed that after (400-500) nanometers there are no absorption peaks, and this stems from the high permeability of the membranes in the visible spectrum region, to observe the behavior. The opposite behavior of permeability. Agreed with the researcher [29].



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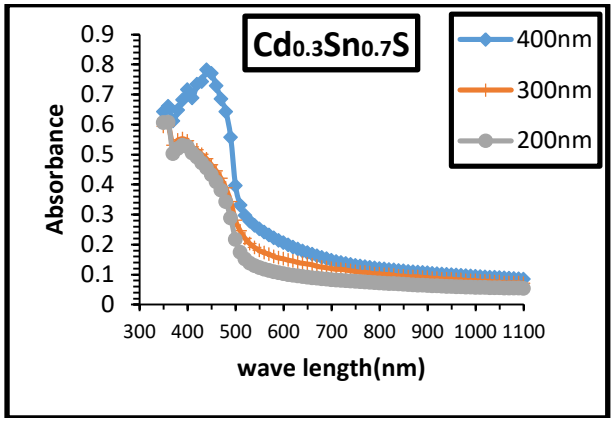

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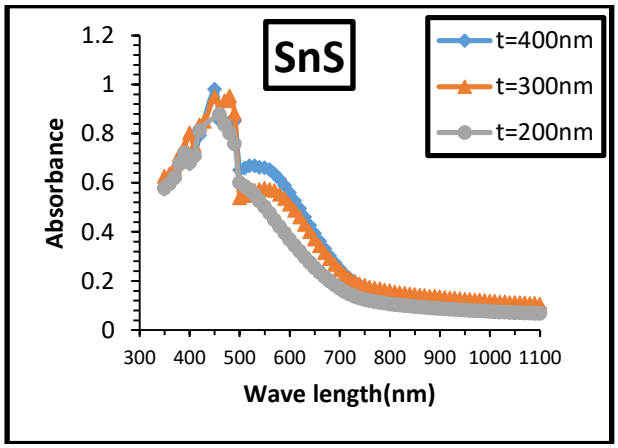

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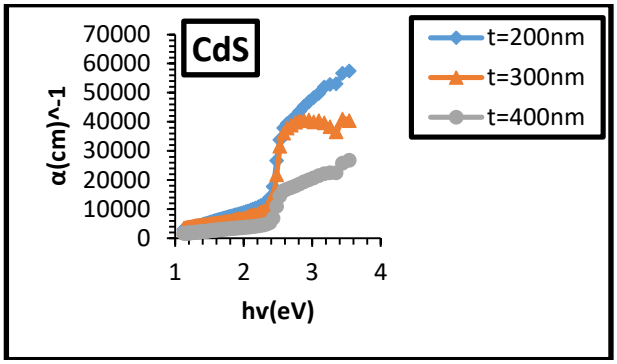
Figure 4. Optical absorption spectra of (a) CdS, (b) $Cd_{0.3}Sn_{0.7}S$, (c)SnS Nanocrystalline thin films with different thicknesses.

The absorption coefficient (α) associated with the strong absorption region of the films was calculated from absorbance (A) and the film thickness (t) using the relation: α = 2.3026 A/t. The absorption coefficient of the Cd1-xSnxS nanocrystalline films for the different thickness films increases with thickness as shown in Fig. 5.

The figure notes the value of the absorption coefficient was ($\alpha > 10^4$) cm^{-1}. This indicates that the transition takes place between the extended levels in the valence band and the extended levels in the conduction band. We also notice an increase in the values of the absorption coefficient with decreasing thickness at high optical energies, at low optical energies we notice a great convergence in the values of the absorption coefficient, and this is what we also observed in the absorbance. Likewise, the absorbance values converge greatly at high wavelengths. This is what researchers agreed upon [29]and[31]. Likewise, the researcher [32].



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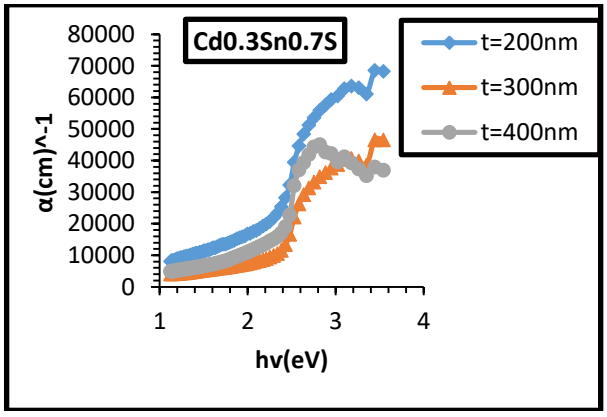

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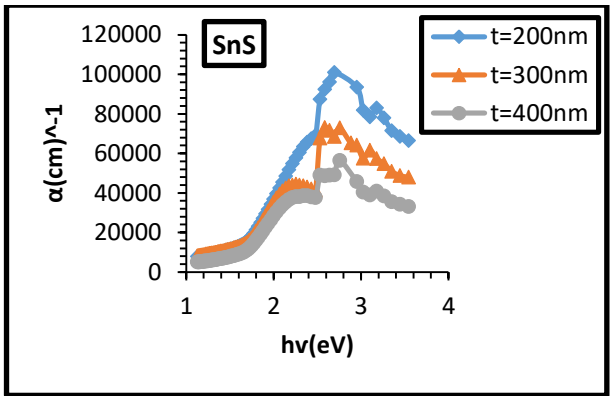

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Figure 5. absorption coefficient spectra of (a)CdS ,(b)$Cd_{0.3}Sn_{0.7}S$,(c)SnS Nanocrystalline thin films with different thicknesses.

The optical band gap energy values of the $Cd_{1-x}Sn_xS$ films have been evaluated using the relation between absorption coefficient (α) and incident photon energy (hv) by using Tauc eq.[33] :

$$\alpha h\nu = B_o(h\nu - E_g)^r \quad \ldots\ldots 2$$

Fig. 6 and Table 3 show the variation of energy band gab of $Cd_xSn_{1-x}S$ Nanocrystalline thin films with thickness and concentration of Sn

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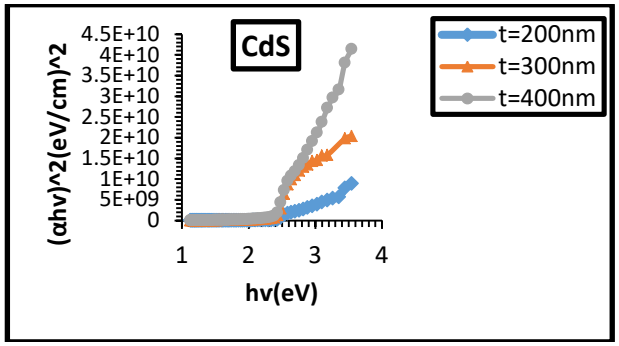

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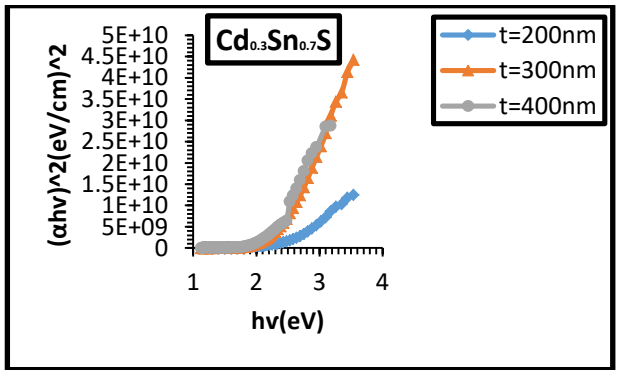

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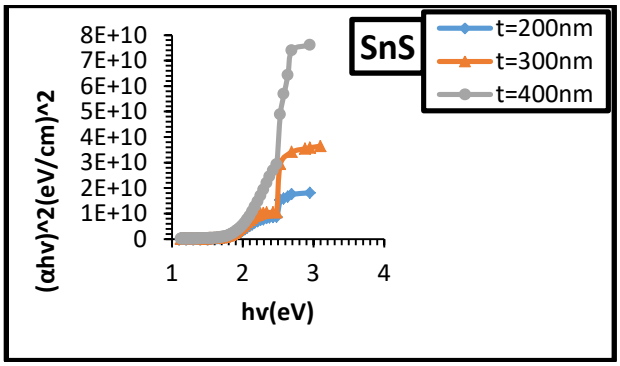

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Figure6. plots of $(\alpha hv)^2$ versus photon energy(hv) of (a)CdS ,(b)$Cd_{0.3}Sn_{0.7}S$,(c)SnS Nanocrystalline thin films with different thicknesses.

Table 3. Variation of energy band gab of $Cd_xSn_{1-x}S$ Nanocrystalline thin films with thickness and concentration of Sn

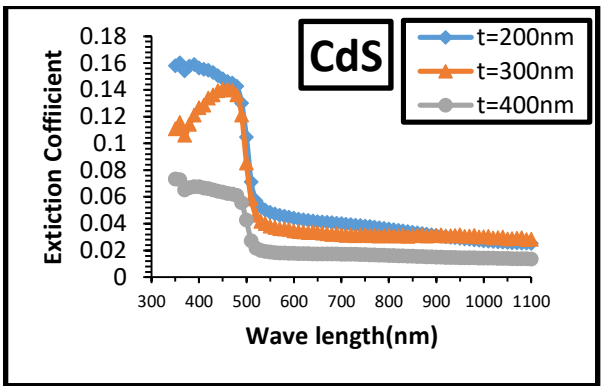
X	Thickness t(nm)	Band gaps Eg (eV)
0	200	2.48
	300	2,41
	400	2,3
0,7	200	2.26
	300	2,15
	400	1.92
1	200	1.95
	300	1.89
	400	1.87

Fig. 6 and Table 3 show the variation of the energy band gap of CdxSn1-xS Nanocrystalline thin films with the thicknesses of the films and concentration of Sn. It was found that the optical energy gap values decrease with increasing thickness and also with increasing Sn concentration.

The extinction coefficient (k_o) has been determined by using the following equation[34]:

$$K_o=\alpha\lambda/4\pi \quad \ldots\ldots\ldots 3$$

Where, α:is the absorption coefficient and λ:is the wavelength of the incident photon.
It is clear from this equation that ko depends on α and has a similar behavior to α. Fig. 6, illustrates the variation of the extinction coefficient of Cd1-xSnxS thin films with the wavelength for x (=0,0.7,1). From the figure, it is noted that the extinction coefficient changes with the change in thickness. The figure notes that the relationship is inverse between the extinction coefficient and the thickness, as the extinction coefficient increases with the increase in thickness, because the extinction coefficient behaves the same way as the absorption coefficient, and this is consistent with the results of the researcher [29]. Likewise, the researcher[31], and also the researcher[32].



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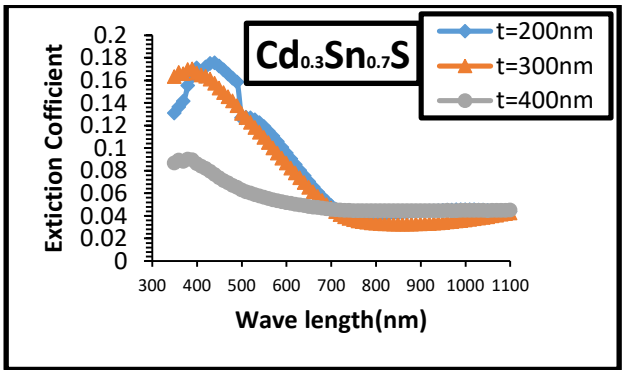

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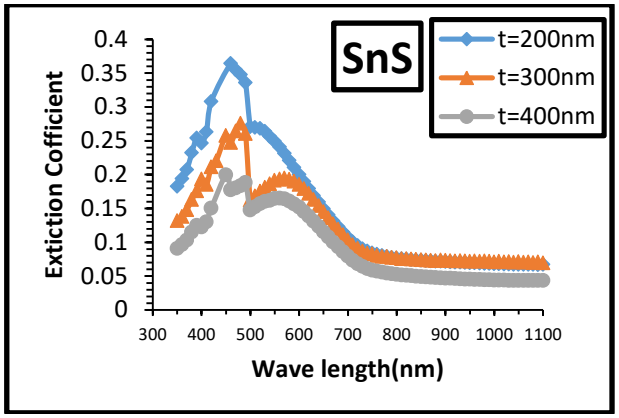

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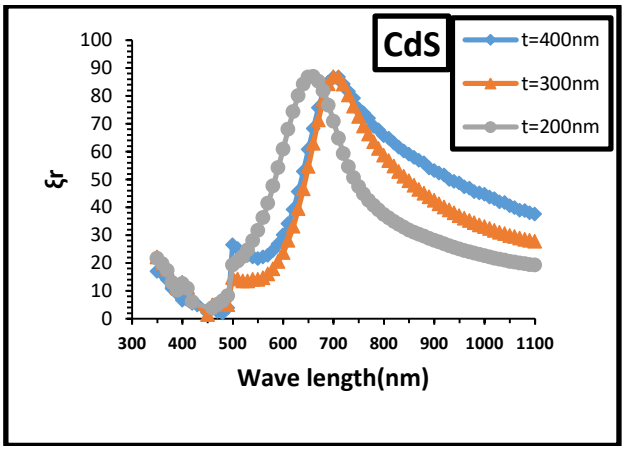
Figure 7. Extinction coefficient as a function of of (a)CdS ,(b)$Cd_{0.3}Sn_{0.7}S$,(c)SnS Nanocrystalline thin films with different thicknesses.

The variation of the real (Ɛr) and imaginary (Ɛi) parts of the dielectric constant values versus wavelength for Cd1-xSnxS films deposited at R.T with different thicknesses are shown in Figs. 8, 9.



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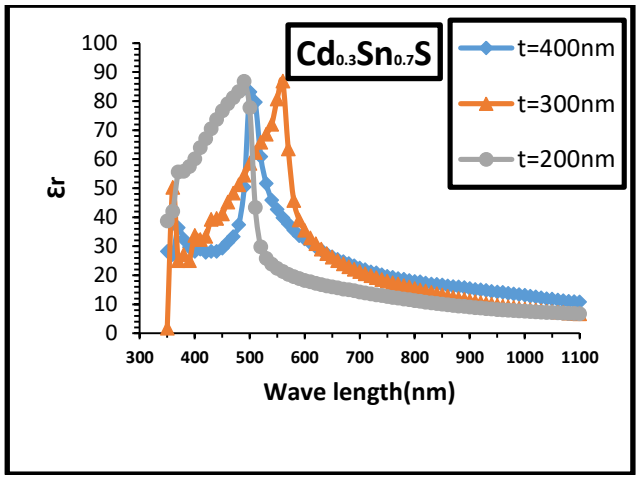

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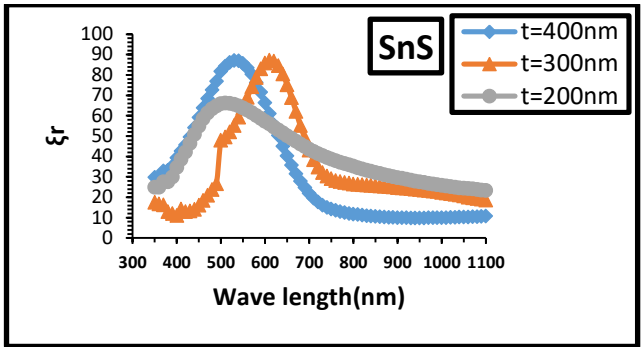

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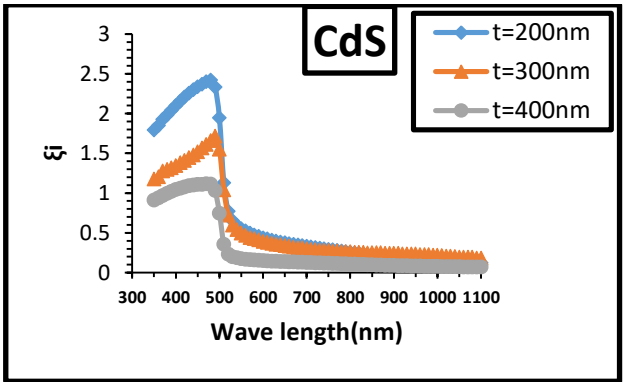
Figure 8. optical real constant(Ɛr) of (a)CdS ,(b)$Cd_{0.3}Sn_{0.7}S$,(c)SnS Nano crystalline thin films with different thicknesses.



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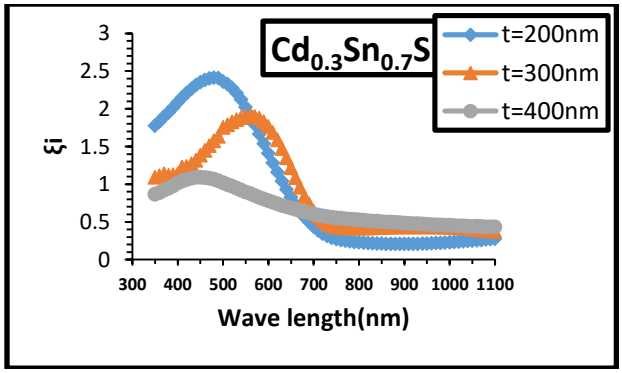

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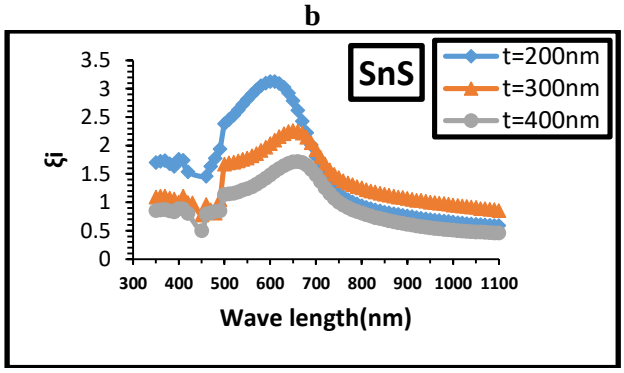

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Figure 9.optical dielectric constant (Ɛi) of (a) CdS, (b) Cd0.3Sn0.7S, (c)SnS Nanocrystalline thin films with different thicknesses

Conclusion

To prepare the $Cd_{1-x}Sn_xS$ nanocrystalline films use first the participant method then use the chemical spray pyrolysis method at 300°C substrate temperature. Films have been characterized using optical and structural measurements. XRD patterns of the films by found polycrystalline. Optical studies indicate that $Cd_{1-x}Sn_xS$ thin films exhibit a direct band gap which strongly depends on the Sn concentration and the thickness of the prepared nanocrystalline films. It was found that the optical energy gap values decrease with increasing the Sn concentrations and with thickness. From the results obtained, it is clear that the prepared material can be used to manufacture the solar systems in taking into account the efficiency of the solar system, whether it is a solar thermal collector or a solar cell which are used as building facades, as well make the other tests to study the possibility of using the prepared samples in the manufacture of the sensors and the effect of changing thickness on the parameters of the gas sensors.

Acknowledgment

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Author's Declaration

- Conflicts of Interest: None.
- I hereby confirm that all the Figures and Tables in the manuscript are mine. Furthermore, any Figures and images, that are not mine, have been included with the necessary permission for re-publication, which is attached to the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee in University of Baghdad.

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تأثير السماكة على الخصائص الهيكلية والمورفولوجية والبصرية للأغشية الرقيقة النانوية من $Cd_{1-x}Sn_xS$ للتطبيقات الإلكترونية البصرية

لمياء خضير عباس

قسم الفيزياء، كلية العلوم، جامعة بغداد، بغداد، العراق.

الخلاصة

تتم تحضير أغشية رقيقة من البلورات النانوية$Sn_{1-x}Cd_xS$ (x=0,0.7, and 1) بسماكات مختلفة (200، 300، 400) نانومتر على قواعد زجاجية عند درجة حرارة 300 درجة مئوية بطريقة المشاركة وطريقة الانحلال الحراري بالرش الكيميائي (CSP). تمت دراسة تأثير تركيز Sn والسمك على الخصائص التركيبية والبصرية للأغشية المحضرة. تم توصيف الأغشية لتقييم البنية والنفاذية وفجوة نطاق الطاقة الضوئية. أظهرت دراسات حيود الأشعة السينية (XRD) أن الأفلام كانت متعددة البلورات ذات هياكل سداسية ومتعامدة. أظهرت نتائج الأشعة السينية وقياس القوة الذرية أن حجم الحبيبات يزداد مع سمكها. تم تحديد طاقات فجوة النطاق البصري ونوع الانتقال والامتصاص البصري للأغشية من أطياف النفاذية البصرية. أظهرت دراسات الامتصاص البصري في مدى الطول الموجي (200-1100 نانومتر) أن قيم طاقة فجوة الطاقة للأغشية تتناقص من (1.95-2.48)الكترون فولت، (1.92-2.41)الكترون فولت، و(2.3- 1.89) الكترون \فولت مع زيادة تركيز القصدير ومع زيادة السمك (300,200، 400) نانومتر على التوالي. تم تحديد أفضل خواص للمادة المحضرة لإمكانية استخدامها في صناعة الخلايا الشمسية.

الكلمات المفتاحية: بلورات $Sn_{1-x}Cd_xS$، الانحلال الحراري بالرش الكيمياوي، جسيمات CdS النانوية، جسيمات SnS النانوية، الاغشية الرقيقة.