

A Study of the Optical Parameters of Cds Thin Films Prepared by Thermal Evaporation

*1 Shadia J. Ikhmayies

* Jabal El-Hussain, Amman 11121, Jordan.

ORCID: 0000-0002-2684-3300

Research Article

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*Corresponding author: shadia.ikhmayies@yahoo.com

Abstract

Cadmium sulfide is an interesting material for the use in solar cells as a window layer and in the manufacture of optoelectronic devices. The optimization of the optical parameters of this material is an important step in developing such uses. In this work polycrystalline cadmium sulfide (CdS) thin films with the same thickness (300 nm) are prepared on glass substrates at ambient temperature using the thermal evaporation technique. X-ray diffraction (XRD) revealed that the films mainly display a cubic (zinc blind) structure. The transmittance of the films is measured at room temperature in the wavelength range (290–1100 nm) and used to infer the optical parameters. The extinction coefficient (k), refractive index (n), the real and imaginary parts of the dielectric constant (ε_r and ε_i), and the dielectric energy loss ($\tan \delta$) are investigated. The dispersion parameters such as E₀ (single-oscillator energy) and E_d (dispersive energy) of the deposited films are determined too.

Key Words: Thin films, Cadmium Sulfide, Solar cells, Optical parameters.

Özet

Kadmiyum sülfür, güneş pillerinde pencere katmanı olarak ve optoelektronik cihazların üretiminde kullanım için ilginç bir malzemedir. Bu malzemenin optik parametrelerinin optimizasyonu, bu tür kullanımların geliştirilmesinde önemli bir adımdır. Bu çalışmada, aynı kalınlıkta (300 nm) polikristalin kadmiyum sülfür (CdS) ince filmler, termal buharlaştırma tekniği kullanılarak ortam sıcaklığında cam substratlar üzerinde hazırlanır. X ışını kırınımı (XRD), filmlerin esas olarak kübik (çinko kör) bir yapı sergilediğini ortaya çıkarmıştır. Filmlerin geçirgenliği, dalga boyu aralığında (290-1100 nm) oda sıcaklığında ölçülür ve optik parametreleri çıkarmak için kullanılır. Yok olma katsayısı (k), kırılma indisi (n), dielektrik sabitinin gerçek ve hayali kısımları (\mathcal{E}_r ve \mathcal{E}_i) ve dielektrik enerji kaybı ($\tan \delta$) araştırılmıştır. Yatırılan filmlerin E₀ (tek osilatör enerjisi) ve E_d (dağıtıcı enerji) gibi dağılım parametreleri de belirlenir.

Anahtar Kelimeler: İnce filmler, Kadmiyum Sülfit, Güneş pilleri, Optik parametreler.

1. Introduction

Heterojunction solar cells with a wide band gap window and a narrow band gap absorber region have undergone extensive investigations due to their beneficial features. Cadmium sulfide (CdS) of band gap energy of 2.42 eV at room temperature is one of the most promising thin film materials for application as a window layer [Naseem et al, 1996]. This material is also promising for nonlinear optical devices, photo-conducting cells, and other optoelectronic devices in the blue-to-ultraviolet spectral region [Kim et al., 2008]. There are several techniques for producing CdS thin films, such as spray pyrolysis (SP) [Ikhmayies & Ahmad-Bitar, 2008; Ikhmayies & Ahmad-Bitar, 2010; Ikhmayies & Ahmad-Bitar, 2011; Ikhmayies & Ahmad-Bitar, 2012c; Ikhmayies & Ahmad-Bitar, 2013a; Ikhmayies et al., 2013b; Ikhmayies, 2014a; Ikhmayies & Ahmad-Bitar, 2014b; Ikhmayies, 2015; Ikhmayies, 2016; Ikhmayies, 2017a; Ikhmayies, 2017b; Ikhmayies, 2017c; Ikhmayies, 2019], chemical bath deposition (CBD) [Metin & Esen, 2003; Ugwu & Onah, 2007], successive ionic layer adsorption and reaction (SILAR) [Ninomiya & Adachi, 1995], rf sputtering [Tsai et

al., 1996], pulsed laser deposition (PLD) [Perna et al., 2004], and thermal evaporation [Naseem et al., 1996; Sahay et al., 2007; Singh et al., 2007; Ikhmayies, 2012d; Ikhmayies, 2013c; Ikhmayies, 2014c]. The thermal evaporation technique for thin-film deposition of CdS was chosen because it is simple compared to other new and sophisticated techniques [Sahay et al., 2007]. Besides, the deposition of the films by thermal evaporation does not present any compositional problems.

Comprehensive investigation of the optical properties of CdS is very interesting for their use in the design of solar cells and optoelectronic devices. Even many workers investigated these characteristics, but most of this work was done to assess the energy band gap [Ninomiya & Adachi, 1995; Perna et al., 2004; Sahay et al., 2007; Singh et al., 2007; Kim et al., 2008; Lohitha et al., 2020]. Little work has been done on various optical parameters such as the refractive index, extinction coefficient, absorption coefficient, and the real and imaginary parts of the dielectric constant, and loss tangent, where some results in this field are found in references [Al-Zahrani et al., 2015; Rao & Ashith, 2015; Yahia et al., 2018; Shkir et al., 2019, Manthrammel et al., 2020], and the author of this work have some other investigations [Ikhmayies, 2014c; Ikhmayies, 2015; Ikhmayies, 2017a]. This work is a continuation in the same line, and its objective is to deduce and discuss the optical parameters of thermally evaporated CdS thin films prepared at ambient temperature. The optical transmittance of the films is measured in the wavelength interval 290-1100 nm and used to deduce the reflectance from which the optical parameters, such as the refractive index, extinction coefficient, real and imaginary dielectric constants and dielectric energy loss and the dispersion parameters are all deduced.

2. Material and Methods

Thin films of cadmium sulfide (CdS) of thickness 300 nm are produced by thermal evaporation on glass substrates of dimensions $2.5 \times 6 \times 0.1$ cm³ which are cleaned with acetone then rinsed in distilled water and finally dried by air. The films are prepared in high vacuum ($\sim 10^{-5}$ mbar) using a Turbo pump. Evaporation rate is about 10 Å/s and it is measured using a cooled quartz crystal monitor. The substrate temperature is the ambient temperature, and the source–substrate distance is about

30 cm. The film transmittance at room temperature is measured using a double beam Shimadzu UV 1601 (PC) spectrophotometer with respect to a glass microslide similar to the substrates in the wavelength range 290-1100 nm. X-ray measurements are performed using a Philips PW1840 Compact X-ray diffractometer system with Cu K_{α} (λ = 1.5405 Å) at a diffraction angle 2 θ from 2° to 60°.

3. Results and/or Discussion

Figure 1 displays the X-ray diffractogram for one of the films under study. From the figure, the film appears to be polycrystalline with a predominant zinc blind structure. Miller indices for the lines belonging to the cubic phase are indicated in the figure as C(hkl) while those belonging to the hexagonal (wurtzite) phase are referred as H(hkl). This pattern is also accompanied by a hump associated with the amorphous glass substrate.





Figure 2 depicts the transmittance of three thermally evaporated CdS films with a thickness of about 300 nm measured at room temperature in the wavelength range 290–1100 nm. As the figure shows the transmittance is high and reaches a maximum of 100%. The presence of interference fringes is evidence on the smoothness of the surfaces of the films. The differences in the positions of the

maxima and minima seen in the figure are due to slight differences in thickness of the films. The transmittance measurements shown in Figure 2 are used to deduce the reflectance which is used to deduce the other optical parameters for the films such as the refractive index, extinction coefficient, real and imaginary parts of the dielectric constant, a



Figure 2. The plot of the transmittance of three CdS films of the same thickness (300 nm) against wavelength

Knowledge of the refractive index of the semiconductor in the energy region below or near the fundamental absorption edge is important in the design of the optoelectronic device design. To find the refractive index and the extinction coefficient, the definition of the reflectance of a film for a light wave incident normally from air, with refractive index $n_0 = 1$, on a medium of complex refractive index is used. The following relationship is considered [Babkair et al., 2011].

$$R = \frac{\left(n^* - 1\right)^2}{\left(n^* + 1\right)^2} = \frac{\left(n - 1\right)^2 + k^2}{\left(n + 1\right)^2 + k^2} \tag{1}$$

The complex refractive index of the film is given by;

$$n^* = n + ik \tag{2}$$

where *n* is the refractive index and, *k* is the extinction coefficient of the film. If the value of the absorption coefficient α is known, then the extinction coefficient can be calculated using the following relationship

$$k = \frac{\lambda \alpha}{4\pi} \tag{3}$$

where λ is the wavelength in free space. Solving Eq.(1) for the refractive index n gives;

$$n = \frac{(1+R) + \left[(1+R)^2 - (1-R)^2 (1+k^2) \right]^{1/2}}{1-R}$$
(4)

The values of the extinction coefficient are calculated using Eq.(3) and plotted against photon's energy (Figure 3). As the figure shows, the extinction coefficient has an oscillatory behavior in the transparent region and its value in this region is small (<0.1) but not zero. In the case of polycrystalline films, extra light absorption occurs at the grain boundaries. This results in a non-zero value of for photon energies smaller than the fundamental absorption edge [Sahay et al., 2007].

Comparing these curves with the curve obtained by [Sahay et al., 2007] for the film of thickness 250 nm which is the closest to the thickness of the films under study, it was found that they [Sahay et al., 2007] have larger values of . The difference can be attributed to many factors such as, the difference in film thickness, deposition temperature, and the ratio of Cd:S in the films. At the absorption edge rapidly increases with photon's energy then the increase becomes slower. This behavior is similar to the behavior of the absorption coefficient and also it is similar to behavior of for the film of thickness 250 nm obtained by [Sahay et al., 2007]. Lower values of in the transparent region are better for using CdS as a window layer in thin film solar cells.



Figure 3. The plot of the extinction coefficient of three thermally evaporated CdS thin films of the same thickness against photon's energy

The refractive index n is calculated from Eq.(4) and plotted against the photon's energy hv (Figure 4), where the inset shows the same plot in a narrower range (1 -2.5 eV) to clarify the variations of n in the transparent region. The refractive index shows an oscillatory behavior in this region and its value is restricted in the range 0.96–2.04. These values are smaller than the refractive index of bulk CdS which is ~2.3. Also these values are smaller than the values obtained by [Gordillo et al., 2003] for CdS_xTe_{1-x} films that are thermally evaporated at a thickness of 2.2 µm prepared at substrate temperature 250 °C when x = 0, where n values in the range 2–2.6 were obtained in the wavelength range 500–1000 nm. The smaller values of n obtained for the films under study can be attributed to disorder [Naseem et al, 1996], where disorder increases with the decrease in film thickness and there is more disorder in the polycrystalline form of a material than its monocrystalline form. Additionally, in the case of thin films, the values of the refractive index depend on the deposition rate [Naseem et al, 1996], deposition parameters, and the elemental ratios along with thickness. In the the absorption edge region and beyond up to 3.5 eV, the refractive index increases rapidly with the photon energy.

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Figure 4. The plot of the refractive index of three thermally evaporated CdS thin films of the same thickness against photon's energy

Below the absorption edge, refractive index dispersion can be analyzed by the single oscillator model. Therefore the data obtained from the refractive index (*n*) is also analyzed to yield the long wavelength refractive index (n_{∞}) together with the average oscillator wavelength (λ_{θ}) of the CdS thin films using the following relationship [Tüzemen et al., 2009]

$$\frac{n_{\infty}^2 - 1}{n^2 - 1} = 1 - \left(\frac{\lambda_0}{\lambda}\right)^2 \tag{5}$$

where λ_0 and n_∞ are evaluated from the plots of $(n^2 - 1)^{-1}$ against λ^{-2} which is shown in Figure 5. The values of λ_0 , n_∞ , and $n_\infty^2 = \varepsilon_\infty$ are shown in Table 1. From these values, $E_0 = hc / \lambda_0$ the average excitation energy for electronic transitions (single oscillator energy) and $E_d = E_0 (n_\infty^2 - 1)$ the dispersion energy, which is a measure of the strength of interband optical transitions, are calculated. These parameters are also included in Table 1. The obtained values of E_0 are smaller than the value obtained by [Ravangave & Biradar, 2013] for Cd_{1-x}Zn_xS thin films prepared by chemical bath deposition when x =0 where they got $E_0 = 3.72 eV$. Also the values of E_d are smaller than the value obtained by [Ravangave & Biradar, 2013] where they got $E_d = 23.1 eV$. This difference is most likely due to the fact that the dispersion parameters change with deposition parameters as found by [Ilican et al., 2007].

The M_{-1} and M_{-3} moments of the optical spectra can be obtained from the relationships [Caglar et al., 2006],

$$E_0^2 = \frac{M_{-1}}{M_{-3}} \tag{6}$$

and

$$E_d^2 = \frac{M_{-1}^3}{M_{-3}} \tag{7}$$

The obtained values of M_{-1} and M_{-3} moments are listed in Table 1. The values of M_{-1} are smaller than the value obtained by [Ravangave & Biradar, 2013] for Cd₁₋ _xZn_xS thin films when x =0, where they got M_{-1} = 6.71. The first two values of M_{-3} in Table 1 are greater than the value obtained by [Ravangave & Biradar, 2013] for Cd₁₋ _xZn_xS at x = 0, while the last value in Table 1 is smaller than the value they got, which is M_{-3} = 0.49. Also the discrepancy is mainly due to the difference in the deposition parameters.



Figure 5. The plot of $1/(n^2-1)$ against $1/\lambda^2$ for CdS thin films of the same thickness

Sample	n∞	3 3	$\lambda_0(nm)$	E ₀ (eV)	E _d (eV)	M-1	M-3(eV) ⁻²
1	2.474	6.122	466.7	2.657	13.609	5.125	0.726
2	2.394	5.732	467.7	2.651	12.545	4.730	0.673
3	1.888	3.564	467.2	2.654	6.805	2.564	0.364

Table 1. The optical parameters of thermally evaporated CdS thin films

The complex dielectric constant is a fundamental intrinsic material property. The real part of it is associated to the refractive index and thus the speed of light in the material. The imaginary part of the dielectric constant is associated with the absorption of light into the material. The real and imaginary parts of the dielectric constant are determined using the following equations [Alnajjar et al., 2012]

$$\varepsilon_r = n^2 - k^2 \tag{8}$$

$$\varepsilon_i = 2nk \tag{9}$$

Figure 6 displays the relationship between \mathcal{E}_r and the photon's energy, where the inset shows the same relationship in the range from 1–2.5 eV. It is evident

from the figure that \mathcal{E}_r increases as the photon's energy increases. From the inset of Figure 6 it is observed that the values of \mathcal{E}_r in the transparent region of the three films vary in an oscillatory behavior between 1 and 4. In the region near the absorption edge \mathcal{E}_r sharply increases with increasing photon's energy.

Figure 7 depicts the relationship between \mathcal{E}_i and the photon's energy, where the inset shows the same relationship in the range 1–2.5 eV. The oscillatory behavior of \mathcal{E}_i also appears in the transparent region, and a sharp increase with photon's energy is observed at the absorption edge. So, the real and imaginary parts follow the same pattern, and it is noticed that the values of the real part are higher than the imaginary part.



Figure 6. The plot of the real part of the dielectric constant and photon's energy for thermally evaporated CdS thin films

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Figure 7. The plot of the imaginary part of the dielectric constant and photon's energy for thermally evaporated CdS thin films

The dielectric loss is the energy loss that goes into heating the dielectric material in a varying electric field. The variations of dielectric loss with photon's energy are represented in Figure 8. The dielectric loss is given by the relationship [Hasnat & Podder, 2012]

$$\tan \delta = \frac{\varepsilon_i}{\varepsilon_r} \tag{10}$$

where δ is the loss angle. From Figure 7, it is observed that dielectric loss has an oscillatory behavior in the transparent region and after the absorption edge it decreases with increasing of photon energies.



Figure 8. The plot of the loss tangent $(tan \delta)$ against photons energy hu

4. Conclusions

Polycrystalline CdS thin films of 300 nm thickness are produced by thermal evaporation at ambient temperature. The transmittance of the films is recorded at room temperature in the 290–1100 nm wavelength range, and used to deduce reflectance. From the relationships between reflectance and the complex refractive index the refractive index and extinction coefficient are calculated and discussed. Dispersion parameters are estimated and compared to those obtained by other authors. The real and imaginary parts of the dielectric constant are obtained using the refractive index and extinction coefficient and used to find the dielectric energy loss. The study of these optical parameters is important for the use of CdS in the design of optoelectronic devices and for the use of CdS thin films as a window material in thin film solar cells.

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