Physical Science International Journal



16(1): 1-11, 2017; Article no.PSIJ.35738 ISSN: 2348-0130

Influence of Thickness on the Optical Properties of Cadmium Sulphide Thin Film Deposited by Chemical Bath Deposition Technique

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Authors' contributions

This work was carried out in collaboration between all authors. Author OO designed and prepared the manuscript. Authors ADAB and AMR analyzed and interpreted the manuscript. All authors read and approved the final manuscript.

Article Information

DOI: 10.9734/PSIJ/2017/35738 <u>Editor(s):</u> (1) Samin Femmam, Strasbourg University of Haute Alsace, France and Safety Systems of Polytechnic School of Engineering "L3S", France. (2) Bheemappa Suresha, Department of ME, The National Institute of Engg, Mysore, India. (3) Christian Brosseau, Distinguished Professor, Department of Physics, Université de Bretagne Occidentale, France. <u>Reviewers:</u> (1) Raunak Kumar Tamrakar, Bhilai Institute of Technology (Seth Balkrishan Memorial), India. (2) En-Chih Chang, I-Shou University, Taiwan. (3) Kiritkumar Siddhapara, S. V. National Institute of Technology, India.

> Received 27th July 2017 Accepted 18th September 2017 Published 21st September 2017

Original Research Article

ABSTRACT

Cadmium Sulphide (CdS) thin films were deposited onto glass substrates by chemical bath deposition (CBD) technique from a bath containing Cadmium acetate, ammonium acetate, thiourea and ammonium hydroxide and the bath was maintained at 90° C using 78H - 1 hot plate magnetic stirrer. The CdS thin films were deposited simultaneously at different dip times of (10, 20, 30, 40, 50, and 60) minutes to achieve different film thickness of (14.13, 17.61, 18.20, 20.84, 25.93 and 27.21) nm respectively in order to investigate the influence of the thickness on the optical properties of the films. The optical absorbance was measured directly using Single – Beam Helios Omega UV – VIS spectrophotometer in the wavelength range of (280 – 920) nm. Other optical parameters which include; transmittance, reflectance, refractive index, extinction coefficient, and the optical band gap were calculated using the optical absorbance data. Results of the study show that the optical properties of the CdS thin film were largely influenced by the thickness of the films.

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Keywords: Chemical bath deposition; thin films; transmittance; absorbance; reflectance; band-gap; optical conductivity.

1. INTRODUCTION

In the recent years, II - VI semiconducting materials including the chalcogenide compounds are extremely and highly researched because of their interesting properties which include - wide band gap, high absorption coefficients, binding energy, high chemical stability, environmental friendly application and low cost of fabrication [1]. There is also the possibility of adjusting and tailoring their electrical properties especially in the optoelectronics, nonlinear optics and photoelectrochemical solar cell applications. These attributes have made them so attractive to researchers technologists and [2-4]. Microstructure of thin films have been observed to differ from the bulk material of similar composition and can vary depending on the growth conditions [5]. CdS thin film is in the category of these semiconductor materials and is considered one of the best buffer materials for Culn(Ga)Se₂ solar cells [6,7].Low series resistance, high transmittance and optimum band gap are very important requirements for the buffer material. So the CdS film should be less resistive, thin to allow high transmission and uniform to avoid short circuit effects [6]. Also good quality at the interface of CIGS/CdS, well lattice matching. less pin hole are needed in the usage of CdS thin film as a window material [6,7]. For high transmission and optimum band gap to be achieved the film must be thin enough [8]. Many researchers have studied the effect of thickness of CdS thin films on its optical properties [9,10].

Several methods have been extensively employed in the fabrication of CdS thin films such as electrodeposition [11], spray pyrolysis [12], molecular beam epitaxy [13], successive ionic layer adsorption and reaction [14], screen printing [15] sputtering [16,17], pulsed laser deposition [18], physical vapour deposition [19] sol -gel deposition [20], thermal evaporation [21,22], and chemical bath deposition [23-34]. Chemical bath deposition (CBD) technique has been adjudged to be the most widely used method of preparing thin film materials because it is cheap, does not require any complex machine, it is very simple and can be adjusted easily to achieve desired result. The chemical bath deposition of CdS consists of preparing a chemical bath of a salt which contains Cadmium cations and anions of either sulphates, nitrates,

chlorides or acetates [28]. The properties of CdS thin films obtained by CBD depend on the preparative parameters such as; pH of the solution, concentration of the solution, deposition temperature, and deposition time [32]. Therefore, by optimizing these parameters it is possible to tune the optical, structural, and electrical properties of CdS films. The Optical properties of semiconductor films depend on the film thickness for their preparations and thus, optical properties of CdS thin films can be modified to achieve desired result by preparing thin films having different thickness. In this paper, CdS thin films were prepared by chemical bath deposition (CBD) technique and the influence of film thickness on the optical properties were investigated.

2. EXPERIMENTAL DETAILS

Thin films of cadmium sulphide were deposited glass onto commercial slides $(25.4 \times 76.2 \times 1)mm$ using the chemical bath deposition (CBD) technique. All the chemicals used were of analytical grades.Full details of the deposition technique was already reported in [10, 33]. Prior to the deposition, the glass slides were first washed with an alkali free detergent and a piece of gauze sponge, then rinsed with distilled water many times. The slides were ultrasonically cleansed using dilute HCI and rinsed several times with de - ionized water and then dried in air. This process was carried out to ensure clean surface essential for the formation of nucleation centers that is required for thin film deposition. The deposition was carried out when $0.264 \ adm^{-3}$ cadmium acetate in 30 ml was placed in a 250 ml glass beaker, as a Cd^{2+} precursor, 0.158 $gdm^{-3}NH_{A}OH$ in 15 ml was added with constant stirring, which reduced the concentration of Cd^{2+} , 0.153 gdm^{-3} Thiourea in 30 ml was added as a source of sulphur, while 1.542 gdm^{-3} Ammonium Acetate in 20 ml was added to the mixture as a complexing agent this brought the volume of the resulting solution to 95 ml. The pH of the mixture was maintained at 11.18. Keeping the alkalinity of the bath constant. Clean glass slides were inserted into the bath vertically with plastic rubber clips attached to a cover. The bath temperature was maintained at 90°C using 78HW – 1 constant magnetic stirrer. The deposition was allowed to take place at time intervals of 10, 20, 30, 40, 50 and 60 minutes respectively after which the slides were withdrawn from the bath simultaneously and rinsed with de - ionized water and then annealed in an oven at a temperature of 400°C for 60 minutes. The gravimetric (Micro Balance) technique was employed in calculating the thickness of the films grown in this work. This method is based on the direct determination of the mass of the film which was deposited onto a glass substrate. The glass substrate was weighed before and after deposition of film on it and then, the thickness determined using relevant equation. Also the samples were characterized for optical properties using spectrophotometer (single - beam UV-VIS Helios Omega) by measuring the absorption within the wavelength range of (280 - 920) nm. The transmittance and other optical parameters were calculated.

3. THEORETICAL CONSIDERATION

3.1 Deposition Mechanism

The deposition of CdS thin films occur when the ionic products of Cd^{2+} and S^{2-} exceeds the solubility product of CdS ($K_{sp} = 1.4 \times 10^{-27}$) [34]. At this point CdS precipitate is obtained either in the bulk of the solution with the formation of colloids or at the surface of the substrate immersed in the solution leading to the formation of the layer.

The chemical reaction leading to the layer formation may be represented as follows [35];

A). decomposition of the tetramine complex ion;

$$[Cd(NH_3)_4]^{2+} \leftrightarrows Cd^{2+} + 4NH_3 \tag{1}$$

The instability constant of $[Cd(NH_3)_4]^{2+}$ is given as

 $K_i = 7.56 \times 10^{-8}$

B) Hydrolysis of thiourea in alkaline solution with S^{2-} ions generation:

$$(NH_2)_2CS + 2OH^- \rightarrow CH_2N_2 + 2H_2O + S^{2-}$$
 (2)

This reaction consumes hydroxide ions tending to decrease the starting pH value of the reaction mixture.

C) The CdS synthesis:

$$Cd^{2+} + S^{2-} \to CdS \tag{3}$$

When the ionic product of Cd^{2+} and S^{2-} exceeds the solubility product of CdS ($Ks = 1.4 \times 10^{-27}$) one obtains CdS precipitate either in the bulk of the solution, with the formation of colloids, or at the surface of the substrate immersed in the solution, leading to the formation of the layer.

3.2 Thickness

The thickness of the films can be calculated using gravimetric method according to equation (4), below [36]. In this method, a microscopic glass slide substrate was weighed before and after the film was deposited on it. With mass M_1 for the substrate before deposition and M_2 after deposition, the average thickness t was calculated using the density of the coated substance.

$$t = \frac{M}{2AD} \tag{4}$$

Where

 $M = M_2 - M_1$ A = Area covered by the film D = Density of the CdS thin film (4.82 gcm⁻³).

3.3 Optical

The transmittance (T) can be calculated from the relationship in (5) below [37,38]. The absorbance (A) is the fraction of radiation absorbed from the radiation that strikes the surface of the material. Alternatively, A is the logarithm to base 10 of the transmittance, hence knowing one, the other can be calculated.

$$T = \frac{1}{10^A} \tag{5}$$

Where A is the absorbance.

From (5) above

$$A = \log_{10} T \tag{6}$$

The absorption coefficient (α) can be calculated from the observed absorbance data using Beer Lamberts law which relates the transmittance (T), absorbance (A), and thickness (d) of the film in the form [9].

$$T = e^{\alpha d} \tag{7a}$$

Such that

$$\alpha = 2.303 \left(\frac{A}{d}\right) \tag{7b}$$

Where d is the thickness of the film.

The energy band gap E_g is related to the absorption coefficient α and can be calculated using this relation [24, 39].

$$(\alpha h\nu) = A (h\nu - E_a)^m \tag{8}$$

Where A is a constant, E_g is the band gap corresponding to a particular transition occurring in the film, ν is the transition frequency, h is Planks constant given by $6.63 \times 10^{-34} m^2 kg s^{-1}$, m is the characteristic nature of the band transition.

The reflectance (R), is the fraction of the incident radiation of a given wavelength that is reflected when it strikes a surface. A relation between transmittance (T), absorbance (A) and reflectance (R), according to the law of conservation of energy is given is calculated from the relation (9b) below [37,38,40].

$$A + R + T = 1 \tag{9a}$$

Or

$$R = 1 - (A + T) \tag{9b}$$

The boundary between vacuum (or air) and an absorbing layer specified by refractive index (n), the extinction coefficient (k) and thickness (d) at normal incidence, yields the reflectance in terms of the optical constants of layer as [38, 41].

$$R = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2} \tag{10a}$$

For semiconductors, where $(K^2 << n^2)$, then the relationship between R and n [38, 41] reduces as given in (10b) below.

$$R = \frac{(n+1)^2}{(n-1)^2}$$
(10b)

Where R is the reflectance, and n is the refractive index.

From (10b) above, it is found that the refractive index n is given as;

$$n = \frac{1 + \sqrt{R}}{1 - \sqrt{R}} \tag{11}$$

Also the relationship between the extinction coefficient K and the absorption coefficient α [40] is given as;

$$k = \frac{\alpha\lambda}{4\pi} \tag{12}$$

The optical conductivity σ is the optical response of a transparent solid. Hence σ is the conductivity at the optical frequency concerned and is not generally equal to the direct current (DC) or low frequency conductivity. It is related to absorption coefficient α and refractive index η as shown in (13) below [41].

$$\sigma = \frac{\alpha \eta c}{4\pi} \tag{13}$$

where c is the speed of light.

4. RESULTS AND DISCUSSION

4.1 Determination of the Thickness

The thickness of the samples was calculated using equation (4) and the result is as shown in Table 1. As observed from the table, there was a gradual and steady increase in the thickness of the thin film samples as the time of deposition increases this result, which is in agreement with other works [24,29,31,34].

4.2 Optical Properties

Fig. 1 shows the variation of the transmittance with the wavelength, and the performance of each film according to their thickness is clearly established as the spectrum shows the transmittance in this order A > B > C > D > E > F. The optical transmittance at visible region decreased more when increasing the thickness, this result may cause weak absorption when used as a window layer material [7]. This property makes the films with higher thickness good materials for antireflection [7].

The absorbance spectra are analyzed into two regions. In Fig. 2, they are $\lambda > 340 \ nm$ and $\lambda <$ 340 nm. In region for $\lambda < 340 nm$, it was observed that the films indicates a good absorber behavior as there was almost total absorption in that region. Whereas, in region for $\lambda > 340 \ nm$ the films exhibit good transparent behavior due to low absorbance values. Also the absorbance increases with increase in thickness of the film as a result of scattering losses [24]. This increase in the absorbance cause large variations in the optical edge of the films. The thickness of the films caused a shift in the optical absorption edge as can be observed from Fig. 2. This behavior, therefore can cause a change in the band structure of the films [42].

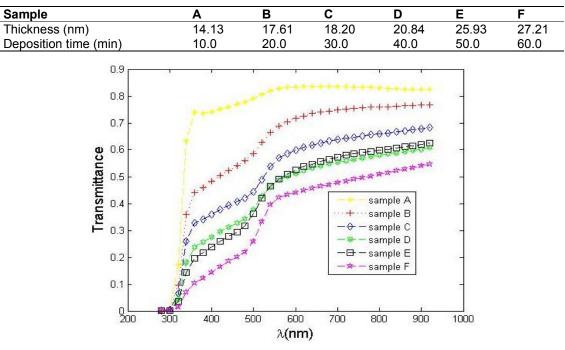


Table 1. Show thickness of the samples with the deposition time

Fig. 1. Variation of Transmittance with Wavelength

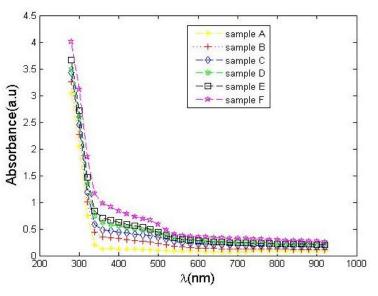


Fig. 2. Variation of absorbance with the wavelength

In semiconductor materials, the absorption due to the band to band transitions that determine the optical band gap E_g is expressed by equation (8) where A is a constant called the band tailing parameter, E_g is the energy of the optical band gap and m is the power factor of the transition mode, which depends on the nature of the

material, (whether it is crystalline or amorphous; $m = \frac{1}{2}$ for allowed direct transitions and m = 2 for allowed indirect transitions). Results of the optical band gaps from the plot of $(\alpha h \nu)^2$ against photon energy, is as shown in Fig. 3 and the values are in the range of 3.75 - 3.85 eV which

show a shift in the energy band gap of the films from that of the bulk material. Higher values of energy band gap of thin films are associated with the quantum confinement effects [9,30], and the values obtained in this work are very similar with the values reported by some researchers [9,30,43,44]. This effect is due to the crystallite size and the very thin films obtained for short deposition times [45,46]. Some researchers reported that the energy gap of films approaches the energy gap of bulk for crystalline grain width higher than 50 nm, otherwise, the energy gap of the film takes values considerably higher than that of the bulk material. This behavior is due to the breaking down of the continuum density of states into discrete levels, such that band gap energy widens compared with the band gap of the bulk [45,46,47].

Fig. 4 shows the variation of reflectance of the CdS samples with the wavelength. It is observed that the CdS thin films exhibit low reflectance for almost all wavelengths. However, there was a strong variation of the reflectance in relation to the thickness of the films. This property of the deposited films makes them suitable as antireflecting coating materials for thin film solar cell [43,48].

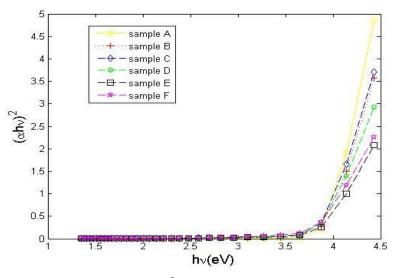


Fig. 3. A plot of $(\alpha hv)^2$ vs hv of CdS thin film of different thickness

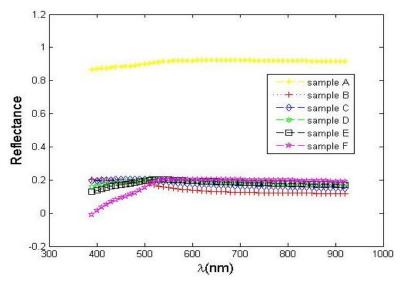


Fig. 4. Variation of reflectance with wavelength

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The refractive index of the films was calculated from reflectance spectrum [42]. The refractive index n of the films as a function of photon energy is shown in Fig. 5a & b. It is seen that after 3.5 eV, the n values decreases with increasing photon energy and the refractive index curves indicate the normal dispersion behavior. The refractive index of the films changes with film thickness. The n curves show a peak at visible region and the intensity and position of the peak change with film thickness. The presence of this peak is due to optical transition from valence band to conduction band. The shifting of the peak position is indicative of variation in the optical band gap of the thin films [24]. This suggests that the thickness of the CdS

thin films can be used to modify the refractive index of the material for use in communication devices and in designing devices for spectral dispersion [49].

The variations of the extinction coefficient K, as a function of wavelength are shown in Fig.6 which shows that K increased with increasing film thickness. The rise and fall in the extinction coefficient is directly related to the absorption of light. In the case of polycrystalline films, extra absorption of light occurs at the grain boundaries. This leads to non-zero of K for wavelengths greater than the fundamental absorption edge [50].

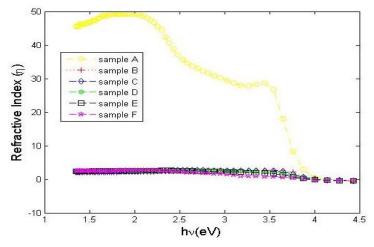


Fig. 5a. Variation of refractive index with photon energy

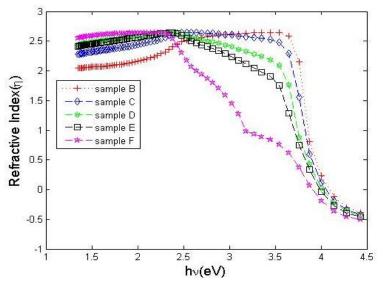


Fig. 5b. Variation of refractive index with photon energy

The optical conductivity of thin films is dependent upon many parameters, among them are; the absorption coefficient, optical band gap, refractive index, the frequency of incident photons, and the extinction coefficient. The optical conductivity can be calculated using equation (13), where c is the speed of light in air or free space. Fig. 7 illustrates the variation of optical conductivity with the incident photon energy. The behavior of the optical conductivity of all the samples followed similar pattern as is observed from the graph. It showed that conductivity increased to a peak at 3.60 eV ($\lambda = 340 \text{ nm}$), and sample A having the highest peak. This behavior is caused by the dependency of optical conductivity on the nature of refractive index and absorption coefficient which are greatest at UV region and diminishes as the wavelength increases [51]. It show electronic transition accompanying absorption of photon energy and is dependent on the thickness of the thin film [1].

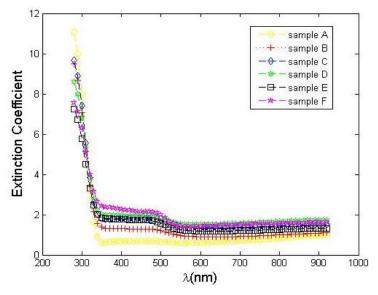


Fig. 6. Variation of extinction coefficient with wavelength

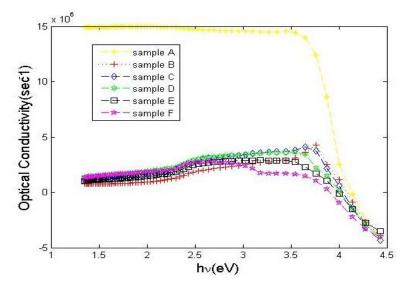


Fig. 7. Variation of optical conductivity with photon energy

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5. CONCLUSION

Good quality, adherent and uniform CdS film have been deposited by CBD technique and characterized. The influence of thickness of the film on the optical properties were investigated. The optical band gap were found to differ from the band gap of bulk CdS due to quantum size effect. The refractive index values of the films also vary from each other as a result of differences in the film thickness. Result of the optical conductivity of the films followed the same manner but less in thicker films, it rises to a peak at a certain photon energy before reducing as the photon energy increases. The film thickness were found to have modified the optical constants of the CdS thin films.

ACKNOWLEDGEMENT

The authors are thankful to the Department of Physics, University of Abuja for the support and also the Centre for Energy Research, Obafemi Awolowo University Ile Ife, Osun State for allowing us to use their laboratory facilities in carrying out this research.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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