

Effects of Concentration on the Properties of Zn-Doped Cadmium Sulphide Thin Films

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Abstract: *Thin films of cadmium sulphide (CdS) were grown using the SILAR (successive ionic layer and reaction) technique. The films were then doped using Zinc (Zn) impurities at different concentrations. The effects of the different concentrations on the properties of the films are reported. The films were characterised using a X-ray diffractometer to investigate the structural properties of the layers. The results show that an increase in the doping concentrations resulted in a corresponding increase in the crystallinity of the layers. The layers crystallised with the cubic structure. Data extracted from the XRD analysis were used to deduce other important structural parameters such as the grain size, strain, and the number of crystallites in the films. These structural parameters deduced from the XRD studies were found to decrease with an increase in the doping concentrations. The film thickness measured using the gravimetric methods were found to increase with an increase in the concentration of the dopants. The film thickness was in the range 0.414 nm to 0.546 nm. The refractive index varied between 1.0 to 1.6, increasing with an increase in the photon energy at the different concentrations up the "critical wavelength" and then decreased.*

Keywords: Cadmium Sulphide, SILAR, doping concentration, XRD, refractive index.

1. Introduction

Cadmium sulphide has been widely used as thin films in the most advanced thin film solar cells such as CdTe (cadmium telluride) and CIGS (copper indium gallium selenide) based devices. It is generally known that Cadmium Sulphide belongs to the II-VI group of semiconductor compounds and that it has been widely utilized in making heterojunction thin film solar cells [1-5]. CdS is a wide band gap semiconductor and has been found to exhibit very good properties needed for materials to be used as window layers in thin film solar cell fabrication. Cadmium Sulphide exhibits good thermal stability and can be used as light dependent resistors sensitive to visible and near infrared light [6]. In the literature, Cadmium Sulphide thin films has been grown using different range of deposition methods such as thermal evaporation [7] – [8], chemical bath deposition technique [9] – [12], spray pyrolysis [13], SILAR [14] – [15], electrodeposition [16] and RF magnetron sputtering [17]. It has been established in the literature that cadmium sulphide can be doped using different element such as tellurium [18] – [20]. Cadmium sulphide has been widely used in different optoelectronics applications and the objective of this study is to; enhance the properties of the Cadmium Sulphide thin films by using a low cost deposition technique, doping the films with cheap and more environmental friendly materials, and to characterise the layers using standard characterisation technique in order to establish their suitability in various optoelectronic-related devices. The work reported herein is a fundamental step toward establishing a more advanced but simple way of obtaining high quality Cadmium Sulphide thin films for applications in the optoelectronic and photonic devices such as solar cells, and in the electronic industry.

2. Literature Review

2.1 Cadmium Sulphide as Window Layers in Solar Cell Devices

Several research groups as earlier indicated have reported on the use of Cadmium sulphide thin films for windows/buffer layers in solar cell devices mostly because of its wide energy bandgap property.

2.2 Cadmium Sulphide in Electronic Devices

In the literature, Cadmium Sulphide finds very wide applications in the electronic industry [21] – [23]. The films are mostly grown using the thermal oxidation method, lithography, epitaxial growth, and chemical vapour deposition techniques among others [21], [24] – [26]. CdS is one of the most promising materials for use in photoelectric conversion in some photonic devices [27]. CdS thin films also find applications in so many electronic devices such as transistor (TFT) [28], nonlinear optics [29], semiconductor lasers [30], and in flat panel display [31]. Such films are mostly grown using molecular beam epitaxy (MBE) [32], chemical vapor deposition [33] or other advanced deposition techniques.

3. Research Methodology

The Cadmium Sulphide thin films were deposited on cleaned glass substrates. The glass substrates were cleaned ultrasonically. The CdS films were deposited using the SILAR technique. The films are grown at different concentrations of 0.00, 0.010 and 0.050 mol respectively using a substrate temperature of 363 K. Details of the deposition procedure, and the materials used has been reported previously [34].

The structural properties of the CdS films were characterised using the PANalytical (XPRT=PRO) D8 advance X-ray

diffractometer with a Cu K α radiation source ($\lambda = 1.5406 \text{ \AA}$) while the optical characterisation was done using the UV spectrophotometer to investigate the transmittance (T) and reflectance (R) versus wavelength measurements.

4. Results and Discussion

4.1 Structural Analysis

The colours of the films were generally yellowish from physical observation. The films had no cracks and showed no trace of pinholes. Figure 1 gives the X-ray diffraction profiles of Cadmium Sulphide thin films at different concentrations. The XRD analysis indicates that the films were polycrystalline because of the presence of X-ray diffraction peaks as indicated in Figure 1. The results from the XRD investigations show that the layers crystallized in the cubic structure.

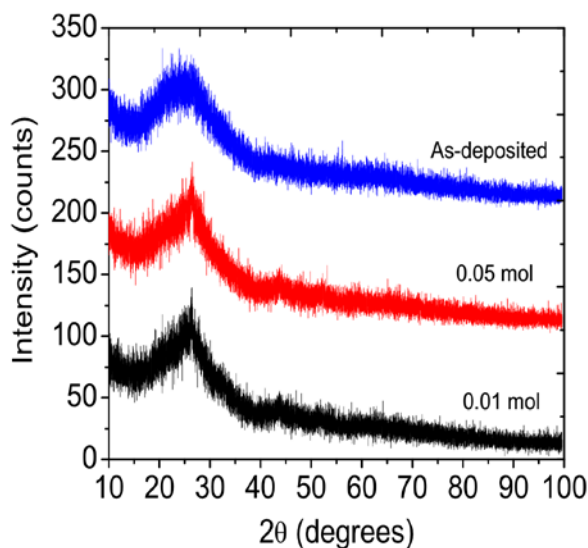


Figure 1: X-ray diffractograms at different concentrations.

The crystallinity of the grown films was observed to increase with an increase in the concentration. This is clearly evidenced in the emergence of the [hkl] peaks as the concentration increases, implying more crystallinity of the layers. Other research groups have observed similar behavior for Cadmium Sulphide thin films grown by the chemical bath deposition technique [35]–[37] or other methods [38]–[41]. Data extracted from the XRD diffractograms were used to calculate the average size of the crystallites by employing the Scherer's formula [42]–[44] given as;

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

In equation 1, D is the average crystallites, K = 0.89 is the shape factor, λ ($\lambda = 1.5406 \text{ \AA}$) is the wavelength of the CuK α radiation, β is the full width at half maximum of the peak and θ is the Bragg's angle. The average crystallite size was found to be in the range 0.0002 nm to 0.004 nm with the higher values corresponding to the layers deposited with the addition of higher concentration of the zinc impurities. Increase in the crystallites size as the concentration increases

could be attributed to the increase in crystal ordering or other associated effects. It is possible that the increase in the crystallite size is due to the agglomeration of the smaller particles into larger ones probably due to the potential energy difference between the small and large particles. This can also occur through solid state diffusion. Nwofe et al [45]–[47] reported an increase in the crystallite size caused by an increase in the substrate temperature for thermally evaporated SnS thin films and explained it on the basis of the increased surface mobility of the films. Devika et al [48] observed an improvement in the crystallites size induced by annealing effects and explained it on the basis of the Hutting and Tamman concepts. Other research groups [44] also reported a similar observation for Cadmium Sulphide thin films grown by the chemical bath deposition method and attributed it to the annealing effects. For the chalcogenides thin films, Kumar et al [42] reported on bismuth sulphide thin films and observed that the crystallite size increased with an increase in substrate temperature and attributed it to the Oswald ripening effects.

4.2 Optical Analysis

The optical transmittance and reflectance measurement, and the absorbance versus wavelength were taken using the UV spectrophotometer. Figure 2 gives the absorbance versus wavelength measurements while Figure 3 shows the reflectance versus wavelength behaviour.

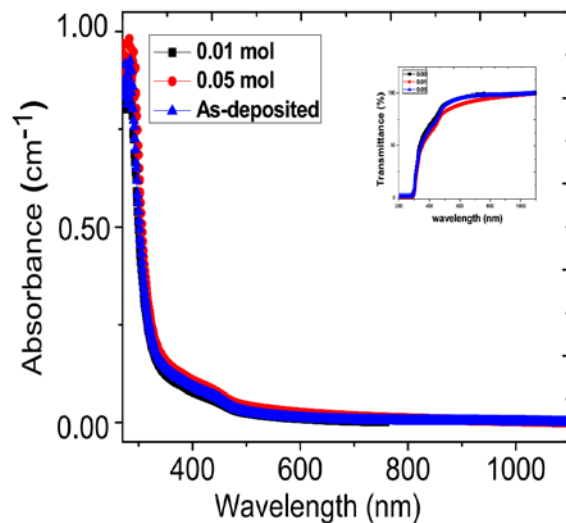


Figure 2: Absorbance vs wavelength at different concentrations.

The transmittance versus wavelength measurement is shown as an inset in the absorbance versus wavelength plots (Figure 2) since the inverse of absorbance measurements gives the transmittance. The measurements were performed in the wavelength range of 300 nm to 1100 nm. The reflectance of the films at the different concentrations all exhibits similar trends. The transmittance plots exhibits a sharp fall at the cut-off wavelength. This could be attributed to a uniform doping of the films. Other research groups have observed similar behaviour at the cut-off wavelength for Cadmium Sulphide

thin films grown by different method or in other related compounds [49] – [51].

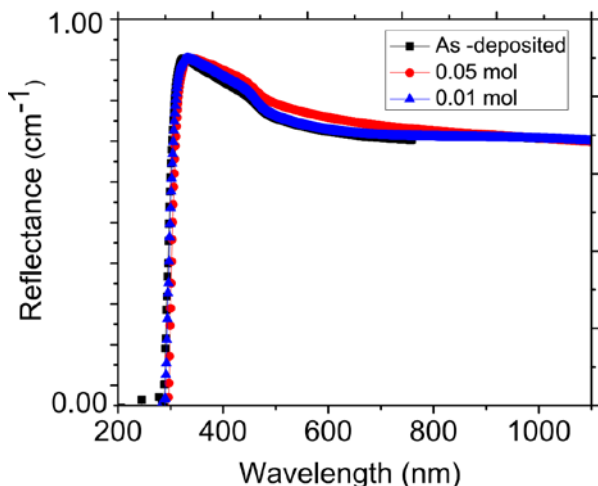


Figure 3: Reflectance vs wavelength at different concentrations.

Information extracted from the reflectance and transmittance versus wavelength measurements were used to deduce the important optical constants such as the optical absorption coefficient (α), the refraction index (n), and then generate the data that enabled a plot for $(\alpha h\nu)^2$ vs $h\nu$. Figure 4 gives the plot for $(\alpha h\nu)^2$ vs $h\nu$. The plot of $(\alpha h\nu)^2$ vs $h\nu$ was calculated using the relation [52] – [54];

$$\alpha(h\nu) = B(h\nu - E_g)^n \quad (2)$$

In equation 2, α is the optical absorption coefficient, h is the Planck's constant, ν is the frequency of the radiation, E_g is the energy bandgap, B is an energy independent constant, and n is an integer which depends on the nature of the transition. It is usually 0.5 for the case of allowed direct transitions and 1.5 for forbidden direct transitions. However, it has been established in the literature [52], [55] that B depends on the effective masses of the holes and electrons, and on the refractive index of the material under investigation. The energy bandgap obtained in the study was in the range 3.7 eV to 3.8 eV. The higher values of the energy bandgap was obtained from the layers formed by the addition of the lower concentration of the zinc impurities. Increase/decrease of the energy bandgap induced by doping effects is commonly observed in polycrystalline thin films due to the overlap of wave functions or quantum size effects. Some research groups [56] – [58] have observed similar effects for core-shell thin films grown by the solution growth technique and attributed it to the quantum size effects. For thermally evaporated SnS thin films subjected to post-deposition heat treatments after a doping with tin (iv) chloride in methanol ($\text{SnCl}_4/\text{CH}_3\text{OH}$), Nwofe et al [59] observed that an increase in the film thickness for the as-deposited layers could lead to a reduction of the energy bandgap. Similar trend has been reported for Cadmium Sulphide films grown using the chemical bath deposition technique by other authors in the literature [60] – [61]. Studies done by Ashour et al [37] on Cadmium Sulphide thin

films prepared by a physical vacuum deposition technique indicate that the deposition rate could induce some change on the energy bandgap. From Figure 4, it could be observed that the plots of $(\alpha h\nu)^2$ vs $h\nu$ exhibits the so called “Urbach tails” rather than show $(\alpha h\nu)^2 = 0$ at the $h\nu$ axis for photons with energies lower than the energy bandgap. This could be attributed to the increase in the doping concentrations. Similar behavior has been observed by other researchers for CdS thin films or other chalcogenides materials in the literature [5], [37] – [39], [62]. Figure 5 gives the variation of the refractive index with photon energies at the different concentrations investigated in this study. The refractive index was calculated using the relation [63] defined as;

$$n = \frac{1 + R^{1/2}}{1 - R^{1/2}} \quad (3)$$

In equation (3), R is the reflectance of the Cadmium sulphide films.

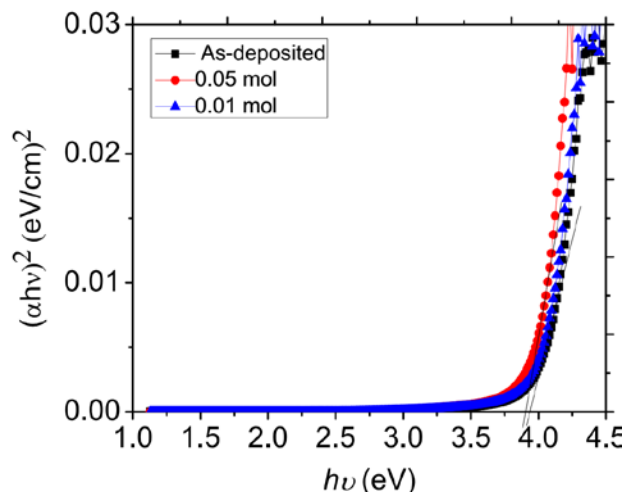


Figure 4: Plots of $(\alpha h\nu)^2$ vs $h\nu$ at different concentrations.

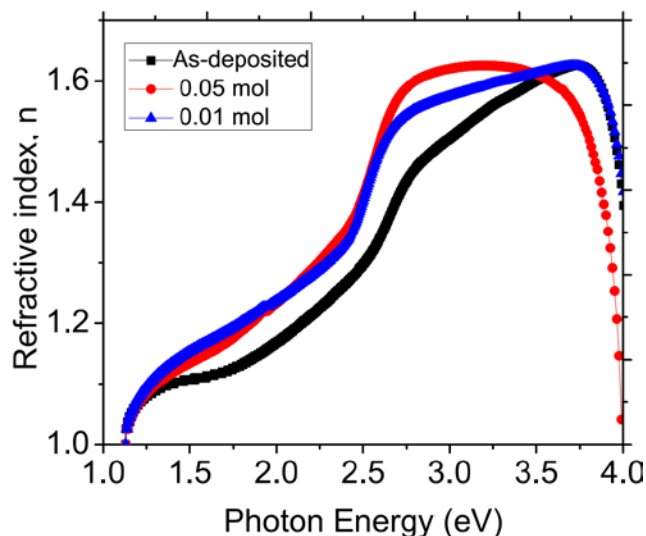


Figure 5: Plots of refractive index (n) vs photon energy ($h\nu$) at different concentrations.

As indicated in Figure 5, the refractive index increased with increasing photon energy up to a “critical refractive index” of about 1.62 and then decreased thereafter. The behaviour of the refractive index with respect to the different concentrations of the zinc impurity is relatively independent. However the undoped layer exhibits a steep fall at photon energies greater than 3.5 eV while at the concentration of 0.05M, higher values of the refractive index (1.5 -1.6) are observed up to the critical value as indicated in Figure 5. For the 0.05M concentration, it was also observed that the refractive index was relatively constant between photon energies of 2.5 eV to 3.5 eV. However, the plot for the as-deposited layer and the plot for the 0.01M exhibits similar steep fall at the same value of the photon energy as shown in Figure 5. Variation of the refractive index with wavelength/photon energy has been reported by other authors [11], [41], [44], [54].

5. Conclusion

Cadmium sulphide thin films have been grown using the successive ionic layer and reaction (SILAR) techniques and then doped with zinc impurities at different concentrations to improve the properties of the layers. Structural characterisation done using X-ray diffraction analysis indicate that the layers crystallised in the cubic structure. The crystallite size calculated from the data obtained from the XRD studies, using the Scherrer's formula was observed to increase with an increase in the concentration. The film thickness was also observed to increase with an increase in the concentration of the zinc impurities. The optical analysis show that the energy bandgap was in the range 3.7 eV to 3.8 eV which is within the range reported by other authors in the literature and also confirms the suitability of the films as window layers in solar cell devices.

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