PRINT ISSN 1119-8362
Electronic ISSN 1119-8362

Full-text Available Online at
https://www.ajol.info/index.php/jasem
https://www.bioline.org.br/ja

J. Appl. Sci. Environ. Manage.
Vol. 26 (3) 385-392 March 2022

Optoelectronic Properties of Chemically Synthesized Copper Cadmium Sulphide Thin Films

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ABSTRACT: Copper cadmium sulphide (CuCdS) thin film (TFs) is one of the most important ternary semiconductors for the development of various modern optoelectronic devices. In this study CuCdS TFs were deposited on soda-lime glass substrates utilizing chemical bath deposition (CBD) procedure at normal room temperature. The deposited materials were characterized by UV-Vis spectrophotometer and four-point (4-point) probes for its optical and electrical (O&E) properties. The band gap (BG) energy was determined to be 2.36 eV using the absorption spectrum fitting (ASF) method. Optical constants such as refractive index (n), extinction coefficient (k), real (ε_r) and imaginary (ε_i) dielectric constants, dielectric loss (tan ϕ), electrical susceptibility (X_c) have been calculated at a wavelength (λ) of 900 nm. The film also demonstrates an optical transmittance of above 75% at wavelength greater than 900 nm. The electrical resistivity and conductivity were estimated to be 2.5 Ω/cm and 0.4 $(\Omega/\text{cm})^{-1}$ respectively. These determined properties confirmed CuCdS TFs as a potential ternary material for various optoelectronic applications.

DOI:https://dx.doi.org/10.4314/jasem.v26i3.3

Impact factor: **http://sjifactor.com/passport.php?id=21082**

Google Analytics: https://www.ajol.info/stats/bdf07303d34706088ffffbc8a92c9c1491b12470

Keywords: Chemical bath deposition; Conductivity; Resistivity; Optoelectronic applications; Optical constants

The increase in world population as well as commensurate demand for energy is ever growing. To meet this quest for eco-friendly energy, the conventional means of energy such as natural gas, coal, fossil fuel etc. are used. The finite and polluting nature of these conventional sources of energy, have necessitated an urgent need of an alternate source of energy that is clean and renewable (Jagadale *et al*., 2016; Emegha and Ukhurebor, 2020; Emegha *et al.*, 2023). Solar energy is a renewable and more efficient alternate source of energy that has been fully developed and evolved by researchers to convert sunlight (photons) into useful or beneficial electrical energy which are often applied in several optoelectronic applications such as advanced calculators, digitalized watches, etc. In recent times, thin films (TFs) based-solar cells have been of great interest to researchers due to their low cost of production, tunable band-gap as well as the ease of forming electron-hole pairs to convert photons to electricity (Choge, 2006; Jagadale *et al*., 2016; Emegha and Ukhurebor, 2020; Emegha *et al.*, 2023). Pure and doped cadmium sulphide (CdS) TFs has attracted substantial interest from scientists and industrialists globally for the development of window materials for the production of solar cells (Hasanzadeh *et al.*, 2014). Copper cadmium sulphide (CuCdS) TF is one of the doped cadmium sulphide chalcogenides TFs with flexible properties that are beneficial for solar cell fabrication. Additionally, the energy band structure varies continuously with the elemental compositions within the ternary matrix; hence, influencing the p-n junction (Hasanzadeh *et al*., 2014). This is due to the fact that the optoelectronic properties

**Corresponding Author Email: jjjemegha@yahoo.com* of CuCdS TFs as well as the dielectric constants are greatly dependent on the changing elemental constituents; and thus, offers great flexibility in optimizing higher efficiency in solar cells production (Ngalu, 2006; Hasanzadeh *et al.*, 2014; Contraras-Rascon *et al.*, 2020).Various chemical and physical deposition techniques have been employed or utilized for the synthesis of CuCdS TFs, viz.; chemical spray pyrolysis (Abbas and Ahmed, 2013; Ahmed *et al*., 2019), spin and dip coating (Samarasekaran *et al.,* 2017; Ghediya *et al*., 2020), pulse laser deposition (Mahdavi *et al.*, 2008), electro-deposition method (Fulari *et al*., 2014), SILAR (Pradhabhan and Sakthivelu, 2019), metal organic chemical vapour deposition (MOCVD) (Emegha *et al.*, 2020) and chemical bath deposition (CBD) (Ganesh *et al.,* 2014; Sharma *et al*., 2016; Diaz-Grijalva *et al*., 2019). However, in the present study, CBD technique was used to prepare the crystalline CuCdS TFs. This technique (CBD) was chosen because of its simplicity, low cost, low temperature of operation and suitability for large scale deposition (Anuar *et al.,* 2010; Alwan and Jabbar, 2010). Additionally, the CBD procedure and its related parameters such as the concentration and the pH of bath, the deposition time as well as the temperature influence the nucleation and the growth of the films. Up-to-date, the CBD technique has been effectively utilized for the deposition of CuCdS TFs for different applications and characterizations. Hence, the intention of this present research study is to complement and improve on the earlier study on the development and characterization of CuCdS thin films by Emegha *et al* (2021a), and to also investigate some other optoelectronic properties of semiconducting CuCdS thin films that are necessary for device applications.

MATERIALS AND METHOD

The CuCdS TFs was prepared using the CBD technique. The reacting solution comprise of copper chloride dehydrate, cadmium chloride and sodium sulphide. The bath pH was kept at 9.0 using potassium hydroxide. Details of the growth procedures are highlighted in a recent reported study by Emegha *et al*.(2021a).

The optical characterizations were gotten from the standard λ ranging from 300 to1500 nm utilizing the UV-Vis-NIR spectrophotometer (Shimadzu 1800). Prior to obtaining the absorption data of the TFs, a clean (blank) substrate was employed as the reference. After the baseline modification was attained, the TFs were then place vertically on the holder of the sample and illuminated with a monochromatic light. The attained absorption data were utilized for the computation of the other optical constants such as n, k, ε_r and ε_i constants as well as the thickness of the TFs. The electrical studies were done using afour-point(4-point) probe (Old JANDEL-TY242MP) formation at normal room temperature.

RESULTS AND DISCUSSION

Optical studies: The optical characterization of CuCdS thin films is one of the most important steps that are needed for the consideration of the optoelectronic nature of a material which can be interpreted in terms of its interaction with the incident photons. When a beam falls on a material, the optical properties are modified by the intensity of the incident beams. So, some photons are absorbed, reflected or transmitted (Hassanien, 2016; Emetere Moses *et al.*, 2021). Figure 1 illustrates the plot of the optical transmittance of CuCdS TFs against the wavelength. As observed from the curve, the transparency was increasing with increase in the wavelength. This therefore means that the TFs have good absorbency which could be due to the surface irregularity within the deposited CuCdS TFs (Ozutok *et al.,* 2012). It was also observed that the TFs had a very high transmittance greater than 75.0% at λ = 900 nm. Figure 2 shows the plot of reflectance of the deposited CuCdS TFs as a function of the wavelength. It was noted that the reflectance decreases with the wavelength for the material. Generally, all the film has low reflectance values that lie below 20%. However, at wavelength of 900 nm, the average value was about 11.0%, which may possibly be as a result of the decreasing tendencies of the TFs' density with rise in the wavelength. The high transmittance and low reflective properties of the film makes it a good material for several electronic applications such as solar cells, LEDs, thermal controlled window coatings for moderate climates as well as anti-reflective coatings (Emegha *et al.*, 2021b).

Optical absorption coefficient was determined by employing the transmittance and reflectance data according to the relationship shown in Eqn. 1 (Dongol *et al.*, 2012):

$$\alpha = \frac{1}{d}\ln\left(\frac{(1-R)^2}{T}\right) \qquad (1)$$

Here, d is the thickness of the film (36.43 nm) and T and R are the transmittance and reflectance respectively. The value of the absorption coefficient (α) was found to be 31.45×10^4 $(cm)^{-1}$ at λ = 900 nm. Since, the α is within the spectra ranging from1×10^4 to $1\times10^6 (cm)^{-1}$, the TFs could find application as absorber materials in solar cells production as reported by Dongol *et al.* (2012) and Tariq *et al.* (2014).As long as ternary CuCdS TFs is a direct band-gap material, the estimation of the optical band gap (BG) was based on the transition (electronic) between the conduction band and the valance band (Emegha *et al.*, 2021a). Similarly, the $\alpha(\nu)$ and the photon energy (represented by $h\nu$) is related by Eqn. 2 (Ghobadi, 2013):

$$\alpha(v)hv = B(hv - E_g)^q \qquad (2)$$

E_g, $h\nu$ and B are the optical band gap (BG), the photon energy and constant respectively; where B depends on the transition probability. Also, from Eqn. (2) the index (*q*) known as the power factor of the electronic transition mode assumed values between 0.5 (or ½) and 3; these values depend on the nature of the transition of the constituent. As reported by Emegha *et al* (2019a), if q = 0.5 it allows direct transitions, q = 2 it allows indirect transitions, q = 3/2 it is forbidden direct transitions and if q = 3 it is for forbidden indirect transitions. Employing the Beer–Lambert's law, Eqn. (2) can be re-arranged as a function of wavelength as follows (Souri and Shomalian, 2009):

$$A(\lambda) = K_1\lambda\left(\frac{1}{\lambda} - \frac{1}{\lambda_g}\right)^q + K_2 \qquad (3)$$

Where, Beer-Lambert's function, K_1= [B $(hc)^{q-1}$ d/2.303] and $K_2$is a constant that is related to the reflectance of the material (Souri and Shomalian, 2009). Upon Equation (3), one can estimate the BG of the deposited CuCdS TFs in electron volts without considering the thickness.

The BG is determined by plotting $(A/\lambda)^2$ against $(1/\lambda)$ as indicated in Figure 3.Byequating the linear portion of the curve at $(A/\lambda)^2$= 0, $(1/\ \lambda_g)$ can be estimated which is then used to calculate its BG by using; $E_{g\,(ASF)} = 1239.83/\lambda_g$ as reported by Moghadam *et al.*(2018). The value of the BG was estimated to be 2.36 eV using the ASF method. The observed BG value of 2.36 eV was very close to the bulk BG of CdS (2.42 eV) (Qutub and Sabri, 2012).

Moreover, the value indicates that CuCdS TFs is a ternary material whose BG can be engineered by varying his binary constituents of CdS (2.44 eV) and CuS (1.70 eV), therefore, obeying the Vegard's rule of mixture (Emegha *et al.*, 2021a). Similar values have been reported in some other studies (Samarasekara *et al.*, 2017; Ghediya *et al.*, 2020; Emegha *et al.*, 2021a).

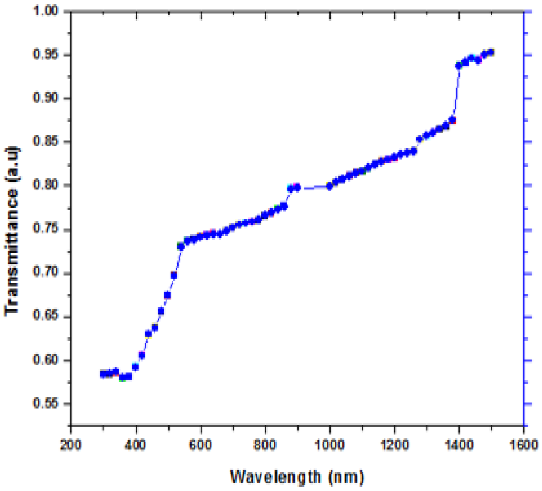

Fig 1: The transmittance plot of CuCdS TFs

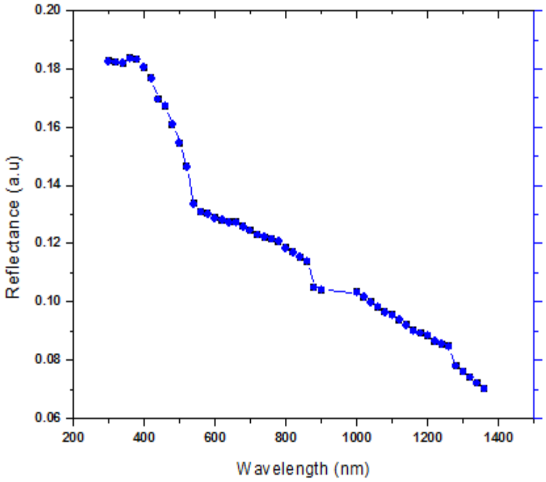

Figure 2: The reflectance plot of CuCdS TFs

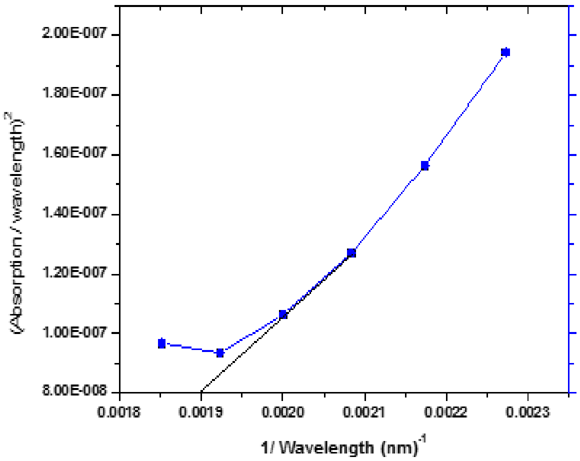

Fig 3: The ASF plot of CuCdS TFs

The refractive index (n) is a crucial parameter for optical TFs and device applications due to its close association with the polarization (electronic) of the ions as well as the local field in the optical material (Hassanien, 2016). Thus, it is consequential in determining the optical constants of the TFs. The refractive index of the material was determined from the following formula (Emegha *et al.,* 2021b):

$$n = \frac{-(R+1) \pm \sqrt{3R^2 + 10R - 3}}{2(R-1)} \quad (4)$$

Where R is the reflectance, the refractive index was estimated to be 1.95 at λ = 900 nm. Similar range has been reported in the literature for CuCdS TFs. Furthermore, the range of the refractive index makes the deposited material beneficial for optoelectronic applications (Emegha *et al.,* 2021b).The extinction coefficient(*k)* which corresponds to the quantity of absorbed energy in the material defines the attenuation of the electromagnetic wave that is travelling within the films (Ngalu, 2006). It depends basically on the density of free electrons as well as the structural defects within the film's matrix. The extinction coefficient was obtained via the Eqn. 5 (Emegha and Ukhurebor, 2020; Emegha *et al.,* 2021b):

$$k = \frac{\alpha\lambda}{4\pi} \quad (5)$$

However, the extinction coefficient was estimated to be 7.80 x 10^{-3}at λ = 900 nm. The optical conductivity (σ), depends on the extinction coefficient as well as the optical constants such as the refractive index and the frequency of incident photons. The parameter provides information about the optical response of a transparent material and it could be estimated via Eqn. 6:

$$\sigma = \frac{\alpha nc}{4\pi} \quad (6)$$

Here c is the velocity of free space. Appropriately, the optical conductivity at any optical frequency is generally not equal to the direct current (DC) or the low frequency conductivity. Hence, the value of the optical conductivity was 5.07 x 10^{12} (1/S) at λ = 900 nm. Similar magnitudes of optical conductivity have been reported for semiconductors TFs (Emegha *et al.* 2021b).

From the optical data of the refractive index, the optical electro-negativity (ɳ) of CuCdS TFs could be estimated. The optical electro-negativity is a principal parameter in understanding the nature of many optical properties of a material (Ahmad *et al.,* 2013). The (ɳ), which is the aptness of an atom to attract electron in an ionic crystal, can be determine using the Duffy classical model (Hassanien, 2016). Duffy suggested a model for estimating accurately the optical electro-negativity of any optical material. The model relates electro-negativity (ɳ) to the refractive index as follows (Hassanien, 2016):

$$\eta_{opt} = \left[\frac{A}{n}\right]^{1/4} \quad (7)$$

Where n is the refractive index at λ = 900 nm. *A* is a constant and is equal to 25.45 for most materials. The optical electro-negativity value at λ = 900 nm was 1.90.

To understand the possible electronic structure as well as the density of state within the BG of the deposited film; it is important to study the electronic dielectric constant (Ɛ) of the material. The Ɛ is fundamentally an intrinsic property of a material that characterizes the optical nature of the transparent solid (Ahmad *et al.,* 2013). The dielectric constant is given by Eqn.8 (Moghadam *et al.,* 2018, Emegha *et al.,* 2019b):

$$\varepsilon = \varepsilon_r + \varepsilon_i \quad (8)$$

Where, ε_r and ε_i are the real and imaginary part of the electronic dielectrics constant respectively. The real part is associated with the speed of the electromagnetic wave that travels within the material while the imaginary part is concern with energy absorbed due to the dipole movement in the electric field (Hassanien, 2016; Emegha *et al.,* 2019b). The ε_r and ε_i parts of the electronic dielectric constants depend on the refractive index as well as the extinction coefficient, and could be evaluated using the expression (Hassanien, 2016):

$$\varepsilon_r = n^2 - k^2 \qquad (9)$$

$$\varepsilon_i = 2\pi k \qquad (10)$$

The values of the real dielectric constant and imaginary dielectric constant at $\lambda = 900$ nm were 3.810 and 3.04 x 10^{-2} respectively. The nature of the electronic dielectric values of the CuCdS TFs is capable of producing induced polarization as a result of intense incident radiation (Thirumavalavan *et al*., 2015).

From the dielectric constants (real and imaginary) values obtained, the dielectric loss (tan ϕ) and the loss angle (ϕ) could be estimated. The tan $\phi$is associated with the loss of energy in a dielectric material due to temperature variation in an alternating electric field (Emegha *et al.*, 2019b). It has to do with the loss-rate of power of a mode of oscillation (electrical, mechanical or electromechanical) in any dissipative system (Nwofe *et al.*, 2012; Hassanien, 2016). It is the reciprocal of the quality factor (QF), which shows the 'quality' or 'durability' of oscillation (Hassanien, 2016). The tan ϕ tends to be higher in materials with higher dielectric constants and depends on the frequency of the applied electromagnetic field as well as the temperature, composition and structure of the material. The dielectric loss (tan ϕ) and the loss angle (ϕ) were calculated using the relation (Emegha *et al.*, 2019b):

$$\tan\phi = \frac{\varepsilon_i}{\varepsilon_r} \qquad (11)$$

And loss angle;

$$\phi = \tan^{-1}\left(\frac{\varepsilon_i}{\varepsilon_r}\right) \qquad (12)$$

Moreover, the values of the tan ϕ and loss angle at λ = 900 nm were estimated to be 7.979 x 10^{-3} and 0.457 respectively. Since the loss angle is less than unity, the material will experience a small loss when used in device applications.

The electrical susceptibility, X_c is a dimensionless constant that indicates the degree of polarization of a solid material in response to an electric field. For most optical material, the X_c can be express in terms of the n as follows (Emegha *et al.*, 2019b):

$$\left(X_c\right) = n^2 - 1 \qquad (13)$$

The X_c value at λ = 900 nm was 2.80. Since the calculated value is greater than 1, the deposited material could be polarizing easily when a more intense light is incident on it, thus reducing the stored energy (Thirumavalavan *et al.*, 2015).

Electrical Properties: The I-V characterization of CuCdSTFs was carried out using a 4-point probe method at room temperature. The variation of average current and voltage plot is indicated in Figure 4. It was evident from the plot that the film has a non-linear I-V curve, suggesting that the conduction mechanism of the deposited film is non-ohmic. The sheet resistance (R) was estimated using the relation (Emegha *et al.*,2022);

$$R = G\left(\frac{V}{I}\right) = 4.532\left(\frac{V}{I}\right) \qquad (14)$$

Where I and V are the average current and voltage respectively, G is a constant that depends on the geometry and configuration of the 4-point probe technique. In this instance, the electrical resistivity is calculated in terms of the average sheet resistance (R) and thickness as follows (Emegha *et al.*, 2019a):

$$\rho = R \times d \qquad (15)$$

Here, d is the thickness of the film. Knowing the resistivity, the electrical conductivity (σ) was taken as the inverse of the resistivity. The estimated values of the electrical sheet resistance, resistivity and conductivity were 6.87 x 10^7 Ω/Sq, 2.5 Ω/cm and 4.0 x 10^{-1} (Ω/cm)$^{-1}$ respectively. According to the values, the electrical conductivity was found to be within the range (10^{-13} to 10^2) reported for conductive TFs, indicating that the deposited material is a semiconductor (Emegha *et al.*, 2019a). Correspondingly, two reasons could have led to the conductive nature of the deposited material; (i) the substitution of cadmium ions into copper sulphide ions sites and (ii) cadmium ions interstitial incorporation into the copper sulphide matrix.

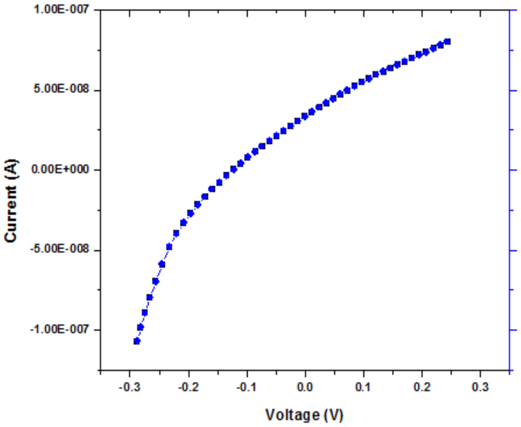

Fig 4: The current-voltage plot of CuCdS TFs

Conclusion: The O&E characterizations of chemical bath deposited CuCdS TFs have been discussed. The BG was determined to be 2.36 eV using the ASF method. The optical properties of *n*, *k*, ε_r and ε_i constants, tan ϕ, X_c were evaluated to analyse the optical relationship within the material. The electrical study indicates that the I-V plot of the film was non-linear, suggesting a non-ohmic conduction mechanism of the deposited film. The determined properties confirmed that CuCdS TFs are a potential ternary material for various optoelectronic applications.

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