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Effect of complex agent in the formation of copper selenide (CuSe) Thin Films on Optical and Structural Properties

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Abstract

Thin films solar cell (TFSC) has been considered as cohort to silicon solar cell due to their thin layers of materials required to absorb the electromagnetic radiations. TFSC needs at least two classes of semiconducting materials: a high optical bandgap employs as window layer and low optical band gap uses as absorber layer. Cadmium sulphide (CdS) grown by chemical bath deposition techniques commonly uses as a window layer but it contains a toxic element. In this study, CuSe thin films were deposited on soda lime glass substrates using chemical bath deposition method and the effect of complex agent was optimized at varying volume of TEA between 2.0 and 8.0 ml. The effect of variation of volume of TEA was investigated on optical and structural properties of the films. The optical properties revealed that the films have low absorbance, reflectance and high transmittance in the visible region of electromagnetic spectrum. The films have high optical band gap between 4.13 and 3.80 eV. The variation of the films has no effect on the structural property as the films are all amorphous as revealed by XRD. The findings show that the CuSe film can be used as a window and buffer layers in the heterojunction (TFSC).

Keywords: Cohort, Semiconducting, Window, Volume

1.0 Introduction

Thin films are the materials that created by random nucleation and condensing of atoms, ions, molecules on substrates. Recently, attentions have been given to copper selenide (CuSe) chalcogenide semiconductor by different researchers due to its numerous applications in solar cells, sensors, photoconductors and many other solid -state devices (Olusanmi, Akinsola, Aregbesola, Yusuf, & Alabi, D, 2025; Sadekar, 2018).The films of CuSe are binary semiconductors which exist in many phases and structural forms: different stoichiometry such as CuSe(klockmannite), Cu_2Sex, $CuSe_2$ (marcasite), α - Cu_2Se (bellidoite), Cu_3Se_2 (umagnite), Cu_5Se_4 (athabaskite), Cu_7Se_4 etc., as well with non-stoichiometric form such as $Cu_{2-x}Se$ (berzelianite),and can be constructed into several crystallographic forms (monoclinic, cubic, tetragonal, hexagonal, etc.(Petrovi et al., 2017).

Several techniques have been used to grow the films of CuSe. The techniques include thermal evaporation (Olasanmi, Akinsola, Yusuf, & Aregbesola, 2025; Petrovi et al., 2017; Rajesh, Chandrakanth, & Sunandana, 2013), sputtering (Kim, Lee, & Kim, 2023; Lee, Kim, Park, Oh, & Kang, 2021) , electrodeposition (Li, Gong, & Zhu, 2017; Olusola & Mukolu, 2024) , successive ionic layer adsorption and reaction (Güzeldir, Lam,

& Ate, 2012), chemical bath deposition (Ezenwa, Okereke, & Okoli, 2013; Khomane, A.S, 2012; Pejova & Grozdanov, 2001; Sadekar, 2018)

Chemical bath deposition is the mostly adopted method to deposit CuSe thin films because it is simple and inexpensive. Hence, the method is used in this study and effect of triethanolamine (TEA) as a complex agent in the optical and structural properties of CuSe thin films

2 Materials and Method

2.1 Materials

6.04 g of copper nitrate as source of copper ions, 25.00 g of anhydrous sodium sulphate and 9.00 g of selenium powder as source of selenium ions, distilled water, triethanolamine (TEA) as complex agent while hydrochloric acid (HCL) and sodium hydroxide (NaOH) serve as pH adjusters, soda lime glass (SLG) were used as the substrates.

2.2 Methods

2.2.1 Cleaning of the Substrates

Glass Slides were immersed in concentrated hydrochloric acid (HCl) for 24 hours, the HCl removes hydrocarbon-based contaminants, dissolves traces of metallic particles, and oxidizes surface residues that could interfere with nucleation during film growth on the substrates. The glass slides were carefully rinsed with distilled water. The rinsed slides were dried in an oven at approximately 100 °C for one hour.

2.2.2 Preparation of Precursor Solutions

2.2.2.1 Preparation of Copper Ion 0.1 M of Cu iron was prepared according to Eqn. 3.1. A mass of 6.04 g of copper nitrate was carefully weighed and transferred into a clean beaker. A small quantity of distilled water was added to the beaker, and the salt dissolved completely with gentle stirring. The resulting blue solution was then poured into a 250 ml conical flask and made up to the calibration mark with distilled water to ensure precise concentration and the conical flask was labelled Cu ion.

$$\boldsymbol{m} = \frac{DC \times Dv \times M}{100} \qquad \textbf{(1)}$$

Where:
m is the mass of the Cu $(NO_3)_2$
M is the molecular weight of the copper nitrate;
DC is the desired concentration which is 0.1 M
Dv is the volume of distilled water in litre
The solution was transferred into a 500 ml beaker and refluxed for one hour at rom temperature while stirring. The refluxing step ensured complete dissociation of the copper nitrate, enhanced homogeneity, and prevented localized precipitation. Constant stirring also maintained uniform ion distribution within the solution, making it suitable for deposition processes.

2.2.2.2 Preparation of selenium ion

Selenium ion was prepared by refluxing 9.00 g of selenium powder with an aqueous solution of sodium sulphite at constant temperature of 80°C and stirred for 3 hours to form sodium selenosulphate which served as anion and the solution was kept in a plastic bottle, labelled Se ion.

2.3 Deposition of the Films

Four precursor solutions of copper selenide thin films were formed by mixing 10 ml of copper ion with varying volume of TEA at 2.0, 4.0, 6.0 and 8.0 ml, the mixture was stirred for 10 minutes at room temperature after which the selenium ion of 20 ml was added and the solution was further stirred for another 10 minutes. The pH of the mixture was measured and adjusted by adding NaOH until pH of 10.42 was achieved. Each precursor solution at varying volume of TEA was poured into a 50 ml chromatographic tank in which the three pre cleaned glass substrates were inserted vertically and allowed to stayed for four hours at room temperature. Then, the substrates were removed from the precursor, rinsed with distilled water and annealed in an oven at 220°C for one hour. The thickness of the film was then determined according to Eqn,2. The composition of the films is presented in Table 1.

$$t = \frac{\Delta m}{A\rho}$$
(2)

Table 1: Deposition parameters of copper selenide thin film

SAMPLE NO.	COPPER ION (ml)	SELENIUM ION (ml)	VOL. OF DISTILLED WATER (ml)	VOL. OF TEA (ml)	TIME OF DEPOSITION (hours)
1	20	20	40	2	4
2	20	20	40	4	4
3	20	20	40	6	4
4	20	20	40	8	4

2.4 Characterisation of the Films

2.4.1 Optical Properties

The optical absorbance of the films was measured by double beam Cary 300 UV Vis spectrophotometer between 200 and 800 nm. Transmittance, reflectance and, absorbance coefficient, photon energy and optical band gap of the films were determined according to Eqns.3, 4, 5, 6 and 7, respectively.

$$T = 10^{-A} \qquad (3)$$

$$R = 1 - (A + T) \qquad (4)$$

$$\alpha = \frac{2{,}303A}{t} \qquad (5)$$

$$E = \frac{1243}{\lambda} \qquad (6)$$

$$(\alpha h\nu)^2 = A(h\nu - Eg) \qquad (7)$$

2.4.2 Structural Properties

X-ray diffraction (XRD) measurement was carried out with the aid of the Rigaku Miniflex machine The X-ray source is a copper anode ($\lambda C\ uK\alpha$ = 1.5418 Ȧ), I = 15mA (fixed). Measurement speed from 0.01 to 100°/min and the samples were scan between (2Θ) = 4° - 75°.

3.0 Results and Discussion

3.1 Optical Properties

The absorbance spectral as presented in Fig.1 reveals that the absorbance decreases gradually in the visible region of electromagnetic spectrum. The film coated at 0.6 ml of TEA has maximum absorbance of 13.2% at wavelength of about 401.0 nm and. Similarly, the film grew at 4.0 ml of TEA has maximum absorbance of about 8.9% at about 401.0 nm in wavelength and follows same pattern of the films grew at 6.0 ml at TEA. The minimum absorbance of the film was 0.99% for the film deposited at 2.0 ml. However, the absorbance of CuSe thin films deposited are very low in comparison with the films deposited by thermal evaporation where the effect of annealing temperature was studied (Ali et al., 2022).Similar lower absorbance .

Similarly, the films transmittance as observed in Fig. 2 shows opposite pattern to the absorbance spectral. The sample coated at 2.0 ml of TEA has minimum transmittance of about 97.7 at wavelength 401nm while the maximum transmittance of 99.7% occurred at the wavelength 517 nm. The transmittances of the film coated at o.4 ml of TEA is between 81.3 and 84.6 % measured between wavelength 401 nm and 581 nm. For the film deposited at 0.6 ml has its transmittance ranges between 73.8% and 78.8% at the wavelength between 403 nm and 595 nm. The films of CuSe deposited at 0.8 ml of TEA has its minimum transmittance of 88.5% occurred at the wavelength 401 nm while the maximum transmittance of radiation is 90.8% at point 535 nm in the wavelength axis. It is observed that the samples have high transmittance in the visible region of electromagnetic spectrum this may attributed to lower dipping time which might have enhance its thickness. It has been established that dipping time has a direct relationship with film's thickness, hence thinner films have higher transmittance (Olusanmi et al., 2025).High transmittance and low absorbance suggest that the films are suitable as buffer layers in thin film solar cells.

Moreover, the reflectance in Fig.3 shows that the films reflected little of incoming radiation. The maximum reflectance of 1.3 % was recorded for the sample coated at 2.0ml of TEA. This shows the films have low reflectance and can be used for antireflection coating (Ezenwa et al., 2013).

Furthermore, the optical band gap as extrapolated from Fig. 4.4(a), 4.4(b), 4.4(c), and 4.4(d) are 4.13 eV, 3. 88 eV, 3.72eV and 3.80 eV, respectively. These results show that the optical band gaps are high and deviated from the theoretical bandgap varying between 2.1 and 2.3 eV. However, the similar higher bandgap between 3.61 and 3.99 eV were recorded in the films of CuSe deposited by thermal evaporation (Olasanmi, Akinsola, Yusuf and Aregbesola, 2025). The higher band gap can be attributed to increase in Cu^{2+} and low refluxing time of Se ions during preparation of precursor solutions. However, higher band gap is used in design and fabrication of window layer in TFSC.(Babatunde, Odunaike, Talabi, & Adeleke, 2020)

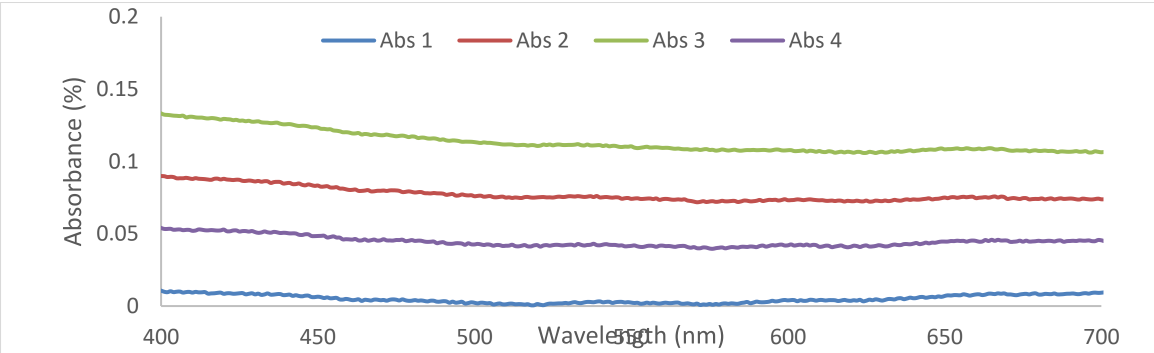

Fig 1: Absorbance against wavelength

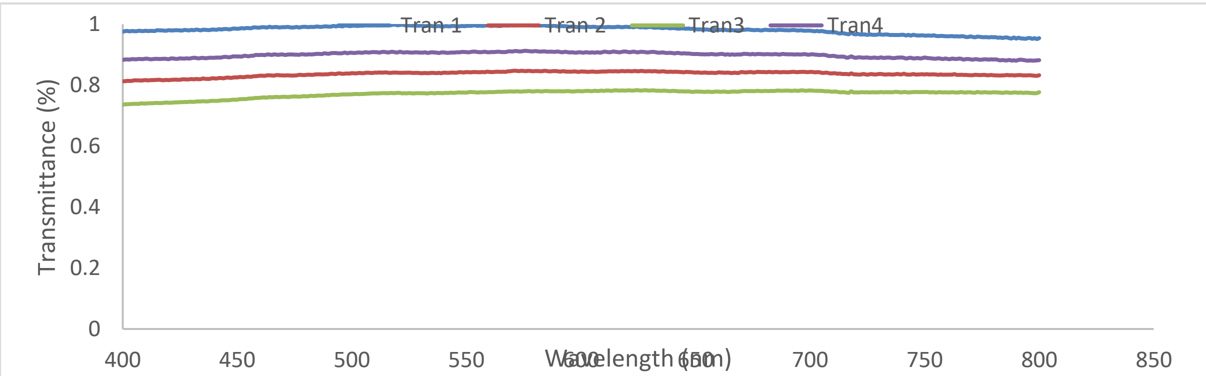

Fig 2: Transmittance against wavelength

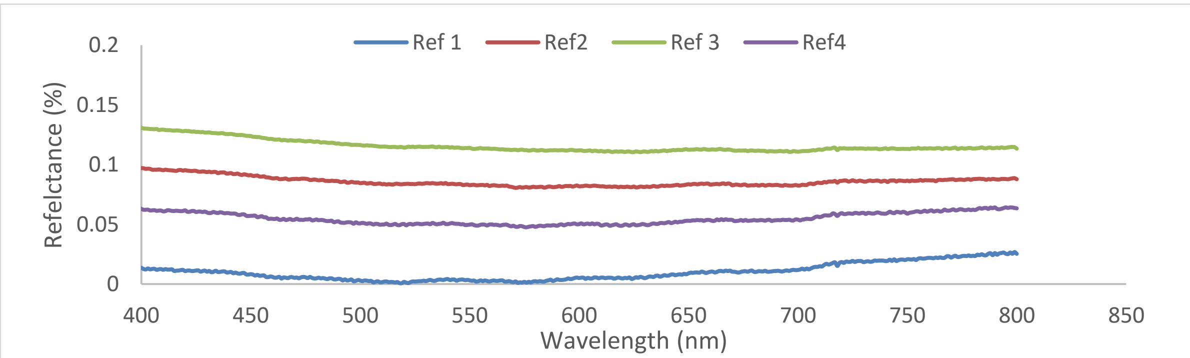

Fig 3: Reflectance against wavelength

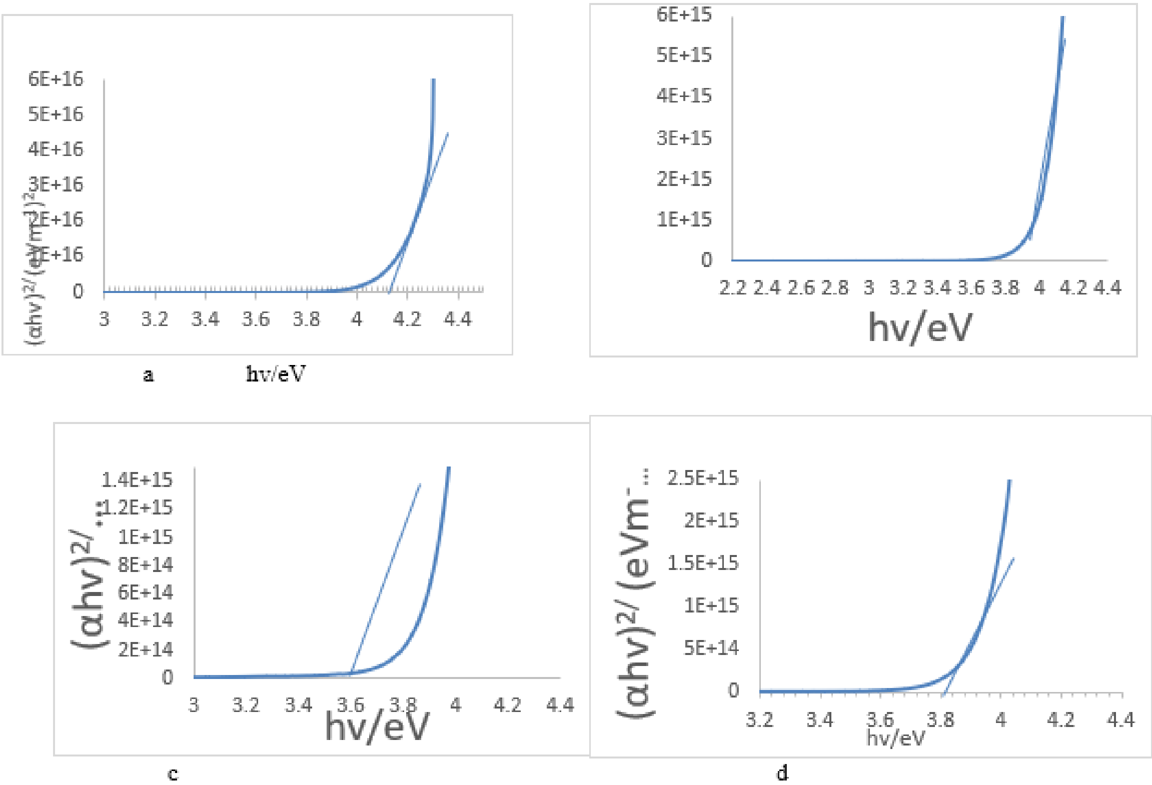

Fig 4 Plot of $(\alpha h\nu)^2$ against hν for CuSe films (a) 2.0 (b) 4.0,(c) 6.0 and (d) 8.0ml of TEA

3.2 Structural Properties

The structural pattern of all the films is the same with no defined peak as revealed by the XRD shows in Fig 5 and that shows the thin film is amorphous unlike CuSe films deposited by thermal method which has polycrystalline structure (Ali et al., 2022). The deviation from the polycrystalline to the amorphous may be attributed to low refluxing time when preparing selenium ions. The similar pattern has been reported in the structural properties of CuSe films grown CBD method (Sadekar, 2018)

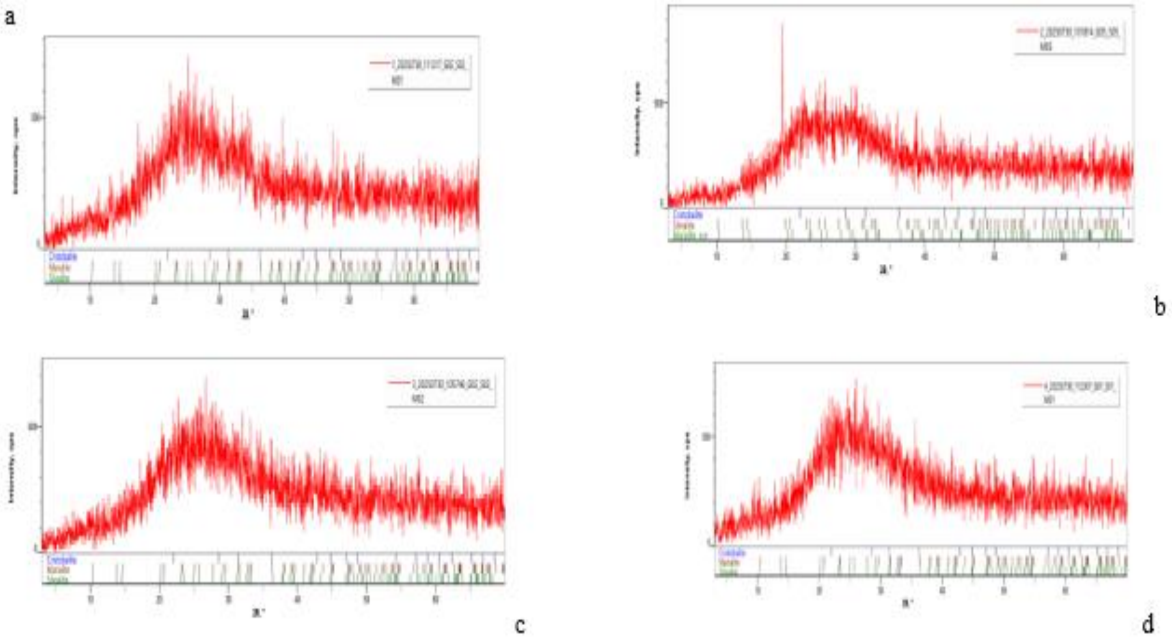

Fig 5 Structural pattern of CuSe films (a) 2.0 (b) 4.0,(c) 6.0 and (d) 8.0 ml of TEA

4. Conclusion

The study CuSe thin films were deposited by chemical bath deposition (CBD) by varying the volume of TEA as complex agent and keep other parameters such as time of deposition, concentration of Cu and Se ion and deposition time. The results show that the thin film coated at 2.0 ml of TEA has maximum transmittance and high optical band gap while the film coated at 6.0 ml has the least absorbance and lowest band gap in the visible region of electromagnetic spectrum. However, the variation of TEA volume has no significant impact on the structural properties of the films because the XRD revealed that all the thin film are amorphous at varying volume of TEA in the formation of CuSe thin films

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