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Optical, Structural and Morphological Properties of Spin Coated Copper Zinc Tin Sulfide Thin Films

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Abstract: Thin films of Copper Zinc Tin Sulfide (CZTS) have been synthesized on glass substrates at various speeds by spin coating deposition. The optical, structural and morphological properties of the prepared samples have been studied via UV-Visible spectroscopy, X-Ray diffraction (XRD) and scanning electron microscopy (SEM), respectively. The optical band gap was found to be 1.50 eV and 1.45 eV for CZTS-1 and CZTS-2 thin films, respectively. The Urbach energy and the Steepness parameter for CZTS-1 thin film are 0.99 eV and 0.026 and that of for CZTS-2 are 0.95 eV and 0.027 respectively. The synthesized thin films were found to be polycrystalline. The XRD pattern showed that the dominating peak was at (112) corresponding to 2θ value of 28.437° for both the thin films. The SEM analysis revealed the texture structure for CZTS thin films.

Keywords: CZTS, Spin Coating deposition, UV-Visible spectroscopy, Band gap, texture structure.

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1 Introduction

The increasing demand for energy consumption accelerates the energy usage. The non-renewable energy sources have a barrier for continuous energy supply as the result of limited availability. To address the issues, the thin films technologies have been attracted much attention because of its unique size dependent properties and different potential applications, especially in the field of photovoltaic, i.e. solar cells [1, 2]. Solar cell based renewable energy is fruitful for long term energy supplying chain. The thin film solar cells are generally produced by using CdTe or CIGS based light absorber materials which are either toxic or expensive material. Replacement of those materials by earth abundant, non-toxic and strong light absorbing materials is urgently needed. The Cu_2ZnSnS_4 (CZTS) material is one of the promising candidates to fulfill the above requirements [3]. The availability of the elements Cu, Zn, Sn and S in the earth's crust is 50, 75, 2.2 and 260 ppm [4]. In recent years, a Cu_2ZnSnS_4 (CZTS) thin film is used as an absorber layer of solar cell and CZTS solar cells have been proposed and investigated. The CZTS is a kesterite quaternary semiconductor containing a suitable optical band gap ~ 1.50 eV and a sufficient absorption coefficient of 10^4 cm^{-1} for application of solar cells [5]. In the past few decades, different techniques, namely chemical bath deposition [6-8], atom beam sputtering [9-11], thermal evaporation [12, 13], RF magnetron sputtering [14, 15], co-evaporation, pulsed laser deposition [16, 17], ion beam sputtering precursor [18], photochemical deposition [19, 20], electrochemical deposition [21, 22], sol–gel [23-26], spray pyrolysis [27, 28], spin coating [29-31] and plasma polymerization [32] have been extensively used to synthesize the thin film coatings for photovoltaic applications. However, Spin coating is currently the predominant technique employed to produce uniform thin films of photosensitive organic/inorganic materials with thickness of the order of micrometers and nano meters. The physics of spin coating can be effectively modeled by dividing the whole process into four stages which are a) deposition b) spin-up, c) spin-off and d) evaporation of solvents. The first three are commonly

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sequential, but spin-off and evaporation usually overlap. Stage 3 (flow controlled) and stage 4 (evaporation controlled) are the two stages that have the most impact on final coating thickness. Spin coating has many advantages in coating operations with its biggest advantage being the absence of coupled process variables. Film thickness is easily changed by changing spin speed, or switching to a different viscosity photo resist. Another advantage of spin coating is the ability of the film to get progressively more uniform as it thins, and if the film ever becomes completely uniform during the coating process, it will remain so for the duration of the process. It is low cost and fast operating system [33]. Spin-coating deposition of PbS and CdS films from methanolic solution of M–TU complex precursors was studied by J. Patel *et al*. [30]. XRD confirmed the formation of cubic phase PbS and CdS layers. The PbS and CdS filmswere smooth and presented a band gap of 1.7 and 2.5 eV, respectively. Electrical properties revealed that PbS film is p-type with an electrical conductivity of around 0.8 S/cm. By using the developed PbS and CdS thin films, a thin film solar cell prototype of graphite/PbS/CdS/ITO/glass was developed. S. K. Swami *et al.* [29] fabricated Cu_2ZnSnS_4 thin film via spin coating technique. The results showed that a good quality kesterite polycrystalline structure had formed. The optical energy band-gap of the CZTS sample is about 1.5 eV, which is very close to the optimum value for a solar-cell. MoO_3 thin films have been successfully deposited by L. Chibane *et al.* [34] via spin coating technique. The results found that the transmittance of MoO_3 thin film annealed at 500°C is within the range 60-72 % in the visible range spectrum; while the reflectance is around 12%.The results obtained demonstrate the potential of the Spin coating method for preparation of MoO_3 thin films that may be useful for photovoltaic applications.

This study primarily aimed to develop CZTS thin films through an easy and fast spin-coating route. After an extensive characterization of the developed films for their structural, morphological and optical properties, these films were used to develop thin film solar cell devices.

2 Experimental Details

2.1 Sample Preparation

In this research work, soda lime glass (SLG) was used as substrates. The SLG substrates were cleaned in an ultrasonic acetone bath for 20 min, and then rinsed with deionized water. CZTS precursors were prepared by using the mixture of copper (II) chloride dihydrate, zinc (II) chloride, tin (II) chloride dihydrate and thiourea of 1.9, 1.17, 1.0 and 3.0 mol/L respectively and dissolved in de-ionized water containing 40 vol% ethanol. The solutions were stirred at 58°C for 25 min and then spinning coated on the SLG substrate at a speed of 2500 rpm (CZTS-1) and 3000 rpm (CZTS-2) for 2 minutes. After the spin coated deposition, the films were annealed at 280°C for 30 min. The spin-coated and synthesizing processes were repeated 3 times to obtain a suitable thickness of CZTS.

2.2 Characterization techniques

The optical properties of the CZTS thin film was studied with the help of Ultraviolet-Visible Spectroscopy (UV-1601, UV-Vis spectrophotometer (Shimadzu Corporation, Japan) at Pilot Plant and Process Development Center (PP & PDC), BCSIR, Dhaka. The structural properties of thin films were studied using X-ray Diffractometer (BRUKER D8 XRD system with Cu-K_{α} radiation sing the wavelength of 1.5406Å, operated at a 40 kV and 40 mA, at PP & PDC, BCSIR, Dhaka. The surface morphological study of the formed film was performed by using Scanning Electron Microscopy (Analytical Scanning Electron Microscope, Model: JEOL JSM-6490LA, at IPD, BCSIR, Dhaka).

3 Results and Discussion

3.1 Optical properties of Spin coated CZTS thin films

3.1.1 Absorbance and Transmittance

The UV-Vis absorption and the transmission spectra for CZTS-1 and CZTS-2 thin films of different thicknesses have been recorded at room temperature as shown in Fig. 1 and Fig. 2.

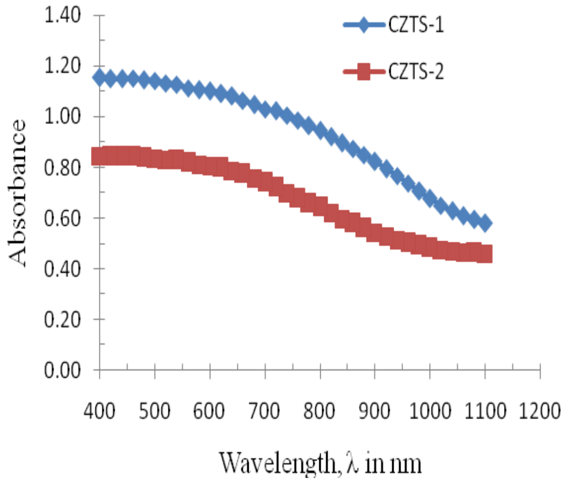

Figure 1: Absorbance Vs Wavelength Graph of CZTS Thin Films.

The absorbance increases with increasing thickness but decreases with increase in wavelength in the visible region. The maximum absorbance in the visible region (400-700 nm) of the spectrum is 1.1542 and 0.8451 for CZTS-1 and CZTS-2 samples whereas maximum absorptance is 0.9299 and 0.8571 for the same samples respectively. This means that CZTS-1 thin film absorbs maximum 92.99% and CZTS-2 thin film absorbs maximum 85.71% light of the incident radiation in the visible region of the spectrum. Comparing among these two samples it is found that the absorbance as well as absorptance of CZTS-1 sample is higher than CZTS-2 in the visible regions.

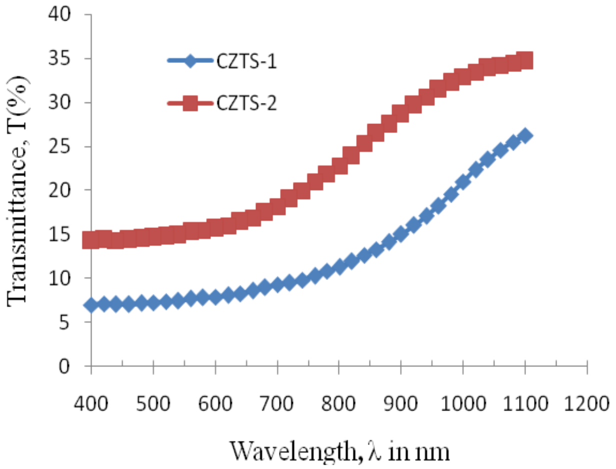

Figure 2: Transmittance Vs Wavelength of of CZTS Thin Films.

It is also found that the absorbance decreases with rpm. Since CZTS thin film is used as an absorber layer in thin film solar cell, so high absorption is required and in this respect 2500 rpm spin coated CZTS-1 thin film is much suitable for solar cell applications [35]. It is observed from Fig. 2 that the transmittance for CZTS-1 is lower than that for CZTS-2 thin film and increases with increase in wavelength as well as with increase of rpm. In case of CZTS thin film absorber layer low transmittance is desired. So CZTS-1 thin film may be a good absorber layer due to its low transmittance [35].

3.1.2 Absorption Coefficient and optical band gap

The value of absorption coefficient can be measured either using transmittance (T) or absorbance (A). In the present work, the absorption coefficient, α, was calculated from the absorption data for samples of different coating speed using the relation [36],$\alpha = \frac{2.303\ A}{d}$, where A is the absorbance and d is the thickness of the films. One of the most significant optical parameters, which is related to the electronic structure, is the optical band gap. The optical band gap E_{opt} can be calculated by Tauc relation, $\alpha h\upsilon = B(h\upsilon - E_{opt})^n$ where hυ is the incident energy, n is the parameter connected with distribution of the density of states and B is the proportionality factor, called Tauc parameter [37]. The value of B is a measure of the steepness of band tail density of states (Urbach region). The index n equals 1/2 and 2 for allowed direct transition and indirect transition energy gaps respectively. Thus, the allowed direct and indirect energy gaps of insulators and/or dielectrics can be determined from the straight-line plots of $(\alpha h\upsilon)^2$ versus hυ and $(\alpha h\upsilon)^{1/2}$ versus hυ respectively [32].

Fig. 3 represents the absorption coefficient, α as a function of photon energy, hυ, for all CZTS. The graph indicates that the absorption coefficient of CZTS-1 thin film is greater than the CZTS-2 thin film and this absorption coefficient increases with increase in photon energy. The absorption coefficient of CZTS-1 thin film is 0.2373×10^4 cm^{-1} maximum. Since CZTS thin film is used as an absorber layer in solar cell, so its absorption coefficient should be high and due to this reason CZTS-1 may be a good choice to be used as an absorber layer for solar cell applications [35].

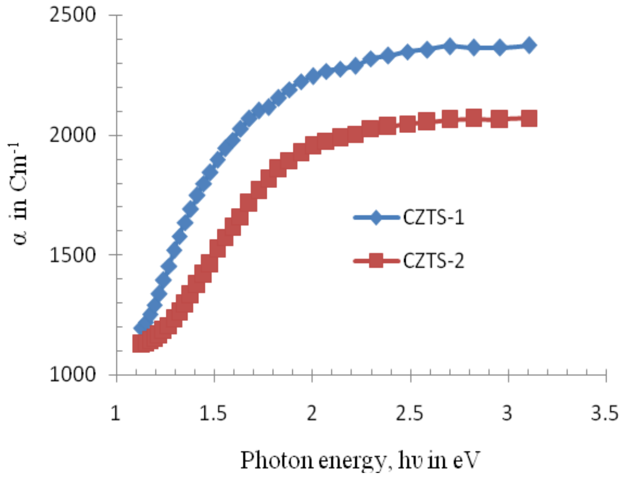

Figure 3: Absorption Coefficient Vs Photon Energy Graph of CZTS Thin Films.

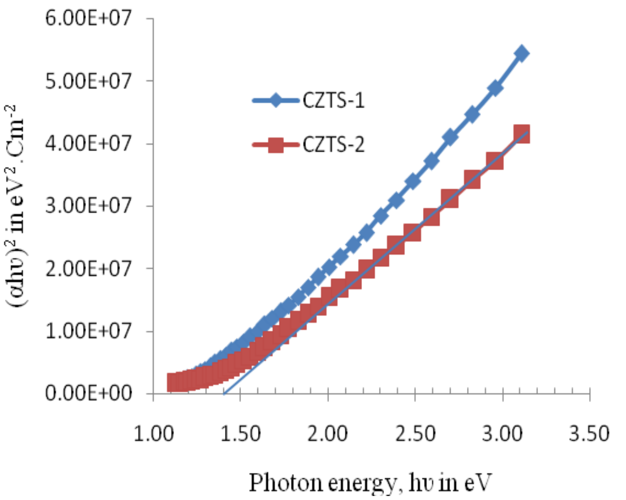

Figure 4: $(\alpha h\upsilon)^2$ Vs Photon Energy, hυ Graph of CZTS Thin Films

The band gap energy E_g for CZTS thin films were determined by plotting $(\boldsymbol{\alpha} h\upsilon)^2$ versus photon energy (hυ) for the corresponding wavelength (λ), where α is the coefficient of absorption. Fig. 4 shows the variation of $(\alpha h\upsilon)^2$ with photon Energy, hυ of the CZTS thin films. Extrapolating the straight line portion of the curve in energy axis gives the values of band gap energy E_g as shown in Fig. 4. The estimated values of band gap for different samples are listed in Table-1. From Table-1, it is seen that the band gap energy of CZTS-1 thin film is 1.50 eV which is comparable to the light energy and is nearly equal to the theoretical value (~ 1.50 eV) of band gap and that's why it will absorb more light. So we can interpret that CZTS-1 thin film sample is better than CZTS-2 with respect to band gap energy. It is also observed that the band gap decreases with decrease in film thickness, i.e., the band gap is thickness dependent.

3.1.3. Urbach Energy, Steepness Parameter and Extinction Coefficient

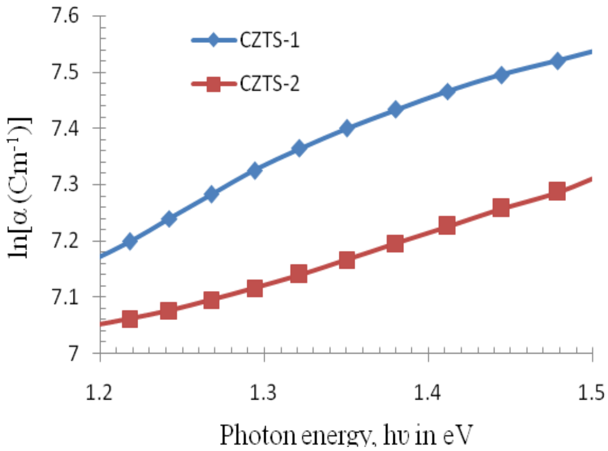

Figure 5: Lnα Vs photon energy, hυ Graph of CZTS Thin Films.

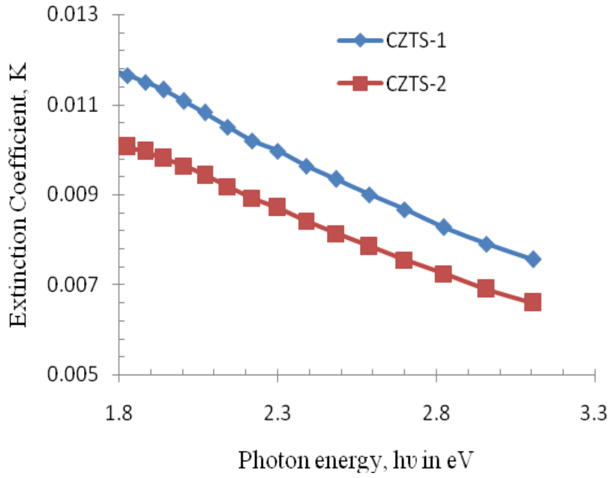

Figure 6: Extinction Coefficient Vs Photon Energy Graph of CZTS Thin Films.

The spectral dependence of absorption coefficient**,** α was studied at photon energies less than the energy gap of the films, i.e. in the region of the so called Urbach spectral tail, which characterizes the slope of the exponential edge and is expressed as, $\alpha=\alpha_0 \exp(E/E_U)$ where, α_0 is a constant, E is the photon energy hυ and E_U is the Urbach energy. Thus, the plots of lnα vs hν should be linear whose slope gives Urbach energy (E_U), interpreted as the width of the tails of localized states in the band gap. The lnα vs hν plots for the CZTS thin films of different thicknesses are shown in Fig. 5 and the corresponding values of E_U are also listed in Table 1.

It is noticed that the values of E_U, which is the band width of the localized states, decreases as the thickness decreases. This behavior may be due to the decrease in the degree of disorder and decrease in density of defect states [14].

The steepness parameter, σ which characterizes the broadening of the optical absorption edge due to electron-phonon or exciton-phonon interactions [14], can be calculated by the equation, $\sigma = kT / E_U$ where, k is the Boltzmann constant, T is the absolute temperature and E_U is the Urbach energy. The values of σ were calculated by taking T as room temperature 298 K. The steepness parameter, σ for the CZTS thin films is listed in Table 1. It is seen that the magnitude of Steepness parameter slightly increases with decrease in film thickness.

The extinction coefficient, K, can be calculated from α using the relation, $\alpha=4\pi K/\lambda$, where, λ is the wavelength and α is the absorption coefficient. It is seen from Fig. 6 that the extinction coefficient for CZTS-1 thin film is higher than the other. The extinction coefficient decreases with increase in photon energy.

3.2 Structural Properties of Spin coated CZTS thin films

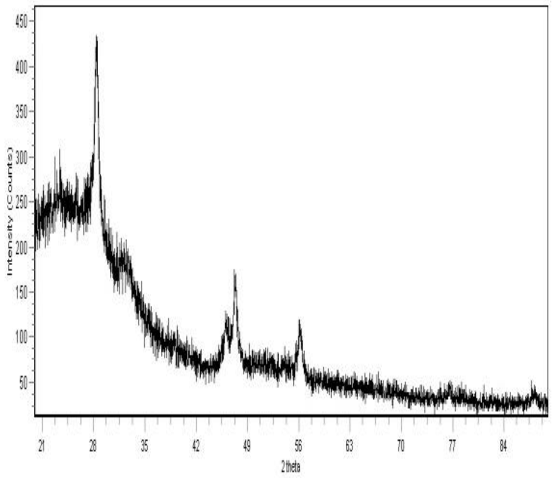

Figure 7: X-Ray Diffraction Pattern of CZTS-1 Thin Film.

Fig. 7 and 8 show the XRD patterns of CZTS thin films. The spin coated CZTS thin films were found to be polycrystalline in nature. The X-ray diffraction peaks of CZTS compounds are expected to share a large number of reflections. The 2500 rpm spin coated CZTS-1 thin film shows the (112), (220) and (312) planes corresponding to 2θ values of about 28.437°, 47.302° and 56.094° respectively .The 3000 rpm spin coated CZTS-2 thin film presents the (112), (220) and (215) planes demonstrating 2θ values of 28.437°, 47.302° and 56.817° respectively. So it is said that the crystalline nature of both the CZTS samples is almost same. The intensity of the peaks of CZTS thin films slightly decreases with increasing rpm (in case of spin coating).This effect indicates that the crystalline quality is slightly good for CZTS-1 thin film than CZTS-2 thin film.

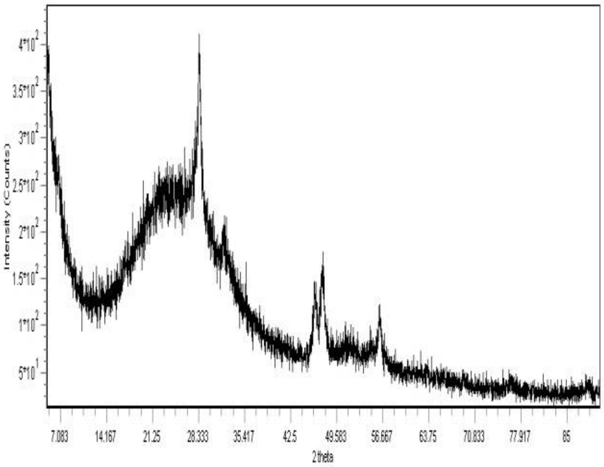

Figure 8: X-Ray Diffraction Pattern of CZTS-2 Thin Film

3.3 Surface Morphology of Spin coated CZTS thin films

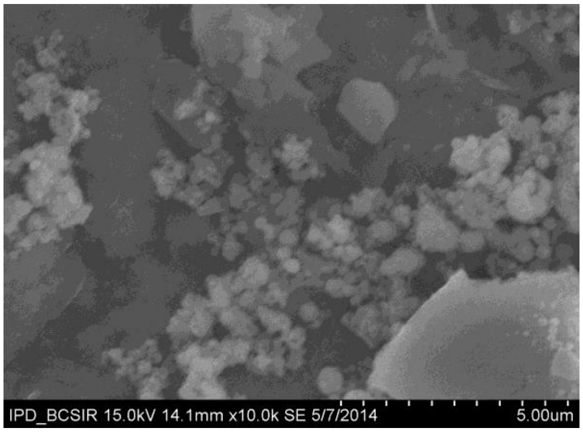

Figure 9: SEM Images of CZTS-1 Thin Films

Fig. 9 and 10 show the surface morphology of the two CZTS thin film samples. From the SEM images it is seen that the texture structures are formed for the CZTS thin films.

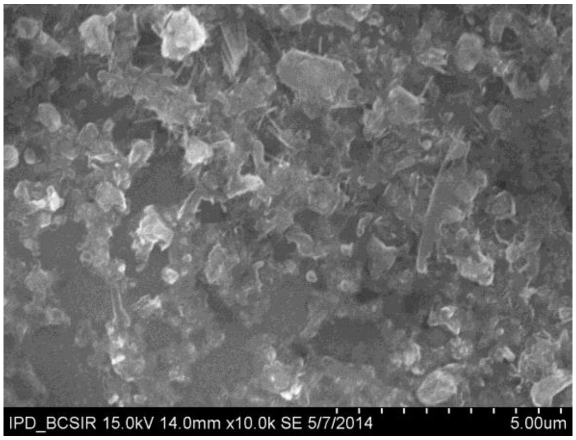

Figure 10: SEM Images of CZTS-2 Thin Films

4 Conclusions

The 2500 rpm spin coated CZTS-1 thin film shows maximum absorbance than that of 3000 rpm CZTS-2 thin film in the visible and infrared regions. Since CZTS is used as an absorber layer in solar cell so its absorbance needs to be high and due to this CZTS-1 may be a preferable choice as an absorber layer for solar cell applications. The band gap of CZTS thin film depends on the film thickness. The band gap of the absorber layer should be comparable with the photon energy to absorb light. Since CZTS-1 with band gap 1.50 eV is comparable with the light spectrum, so it may be a good choice to be used as an absorber layer in solar cell.

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