Optical, structural, and electrical investigations of Spin Coated Cu_2ZnSnS_4 (CZTS) kesterite Thin Films for solar cell applications

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Abstract

In recent decades, there has been an increase in the demand for energy due to the fast economic and population growth which has led to a high reliance on non-renewable energy sources and has adverse environmental impact. Because of its limitless supplies and lack of environmental harming pollutants, solar energy has become a viable alternative for renewable energy. This work intends to propose the advancement of new technique of spin coated pure Cu_2ZnSnS_4 solar cell by taking into account the CZTS kesterite structure, secondary phases, bandgap and absorbance properties. In order to produce the high-performance solution-processed CZTS solar cells, this work required to take into account the atomic ratio of Cu-poor, Zn-rich, and the film consistency during the spin coating process. The impact of the deposition processes on the variation of the CZTS films' structural, optical, optoelectrical, and electrical properties has been studied. According to the XRD patterns, CZTS thin films are polycrystalline and have a kesterite crystal structure. The evaluated films showed a direct energy gap transition that found to be 1.48 eV. A prominent increase in the nonlinear parameters has been noticed with the increase in the deposition time. This study can assist in determining the possibility of using CZTS in nonlinear devices. As the film thickness has increased, the activation energy values were reduced which confirming an improvement in film uniformity. The improved CZTS's lowest dielectric loss, which has attained with less inherent defects, proved the materials' viability. Therefore, the paper provides a simple and suitable approach for fabricating CZTS without post-treatment to make low-cost solar cells.

***Keywords:* kesterite, CZTS solar cells, polycrystalline, atomic ratio**

1 Introduction

Copper-Zinc-Tin-Sulphide (CZTS) made exclusively from abundant materials, has drew a great interest due to its potential applications in sustainable thin-film solar cell devices. Although stable power conversion efficiencies of over 9% have been achieved in the commercial production of chalcogenide-based solar cells like Cu (In, G) (S, Se), (CIGS) and cadmium telluride (CdTe) also their widespread use has been constrained by their dependence on pollutants (Cd) or elements with very limited quantity (In and Te) [1, 2].

Therefore, one of the more promising thin-film photovoltaic semiconducting materials for next-generation applications is relatively abundant and non-toxic kesterite, such as copper zinc tin sulphide, which offers low production costs, scalable manufacturing, a suitable optical band gap of ∼1.5 eV, and a significant absorption coefficient of over 104 cm^{-1} [3-5]. These materials also have a considerably wider range of applications because it is possible to alter their chemical makeup to enhance their functionality [6]. In other words, a thin coating of this material can serve as an absorber layer for solar cells by being able to capture more photons from solar radiation.

CZTS was deposited using a number of different deposition methods. These include, but are not limited to, thermal and electron-beam evaporation [7], co-evaporation [8], DC and RF magnetron sputtering [9 - 10], hybrid sputtering [11], sol-gel spin-coated deposition, and electro deposition (co-electroplating) [12 - 13], chemical bath deposition [14], the one temperature method [15], and SILAR [16]. The majority of the CZTS thin film depositions that have been reported so far have film thicknesses of at least 1000 nm. On the other hand, annealing thin films is known to enhance their characteristics [17–20].

Only one study that looked at the spin coating of CZTS films [21–26] showed a solar-to-electrical conversion efficiency that was high enough (11.1%) to be regarded as being at the level of marketable efficiency [16]. However, these methods have demonstrated that both tin (Sn) and sulphur (S) are lost in the procedure. Given that CZTS is a quaternary compound, it is important to comprehend how compositional variation from stoichiometry may affect its structural, crystallographic, and electrical properties [27- 30]. In order to achieve this, this work examines how the structural, electrical, and optical properties of CZTS thin-film absorbers for PV applications are affected by the compositions of the deposition precursors and the degree of crystallisation.

2 Methodology

2.1 Thin Film Preparation

The CZTS precursor solutions for spin coating were prepared from copper (II) chloride dihydrate, zinc (II) chloride, tin (II) chloride dihydrate and thiourea a in 2-methoxylethanol solvent, to which monoethlyamine (MEA) was added as a dispersant to prevent the formation of precipitates. This was accomplished by first dissolving CuCl2.2H2O and SnCl2.2H2O in 50 ml of 2-methoxylethanol while vigorously magnetic swirling the mixture until a bluish-green solution was formed. A light-greenish-yellow solution was produced after ZnCl2 was added and stirred until fully dissolved. This shows that ZnCl2 aids in a redox reaction between Cu2+ and Sn2+ ions. The addition of thiourea was the final step to stop the development of secondary phases and the loss of sulphur.

A stoichiometric set with a Cu:Zn:Sn:S ratio of 2:1:1:4 and a non-stoichiometric set with a Cu/(Zn+Sn) of 0.88 or 0.92 were both prepared. The two sets of solutions had varying stoichiometry. To deposit CZTS thin films onto meticulously cleaned substrates of soda lime glass slides measuring 37x 26x 1 mm^3, a spin-coating kit was utilised. The solvent in the pre-cursor solutions was removed by heating for 80 to 120 minutes at 110 ^{0}C before spin coating.

Thereafter, for roughly 60 seconds, a high spinning speed of 3200 rpm is employed to make sure that the precursor fluid was evenly distributed across the substrate by centrifugal force. Up until the necessary film thickness was reached, this process was repeated. Films were moved to a rapid thermal processing boiler for additional annealing in a N_2 environment after the sample had been pre-annealed on a hot plate.

An wafer with a high thermal conductivity was placed on the sliding quartz flange onto which the sample holder was inserted in order to maintain a constant temperature throughout the sample. The quartz tube was emptied and filled with N_2 gas at a rate of 50 ml/minute when the boiler was turned on. The boiler was then heated using a halogen tube (10 kW) at a maximum rate of 120 ^{0}C/s for 10 minutes, reaching a working temperature of 400 ^{0}C. The boiler was then turned off, and the chamber was left to cool naturally for an additional 120 minutes until it reached room temperature.

2.2Characterization Analysis

XRD analysis is executed using XPER – PRO diffractrometer with the wavelength of Cu – Kα (λ = 1.54060 Å) and Cu – Kβ (λ = 1.39225 Å). The data analysis is done from 2θ = 10.0231° to 2θ = 80.9231° at 25°C. The morphological analysis of the sample is done using field-emission Scanning Electron Microscope (FESEM - VEGA3 SBH setup) with 5.0 kV; range of magnification (1000 – 2000 x); spot size and WD at 107.67 nm and 9.7868 mm. The transmission T(λ) and reflection R(λ) spectra of the films have been recorded in the wavelength range 400 nm – 2500 nm at normal light incidence using a UV - Vis spectrophotometer. The J-V curves of CZTS/Si heterojunction solar cells were acquired based on thickness and V_{oc}, FF, J_{sc}, η parameters of the heterojunction solar cells.

3. Results and Discussions

a) Structural Analysis

X-ray diffraction (XRD) pattern of the spin coated CZTS thin film on glass plate is illustrated in figure 1. The diffraction peaks of the thin film are matched with the kesterite CZTS (JCPDS No. 26-0575). The extended diffraction indicates a nanocrystalline nature of the obtained thin films. The three main diffraction peaks correspond to (112), (220) and (312) crystal planes of CZTS structure at 28.5 ◦, 47.3 ◦ and 56.1 ◦, respectively. The intensity of the diffraction peaks on (112) plane is the highest. Since the X-ray diffraction peaks are close to those of ZnS (JCPDS, Card, no. 65-0309) and Cu2SnS3 (JCPDS, Card, no. 27-0198). It can also be seen from figure 1 that the film has better crystalline structure.

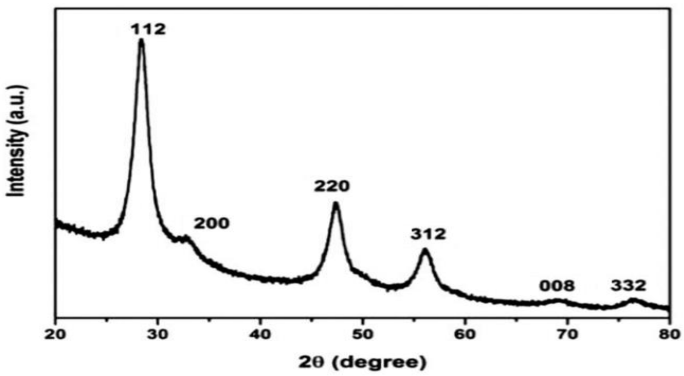

Figure 1. XRD Analysis of Spin Coated CZTS Thin Film (JCPDS No. 26-0575)

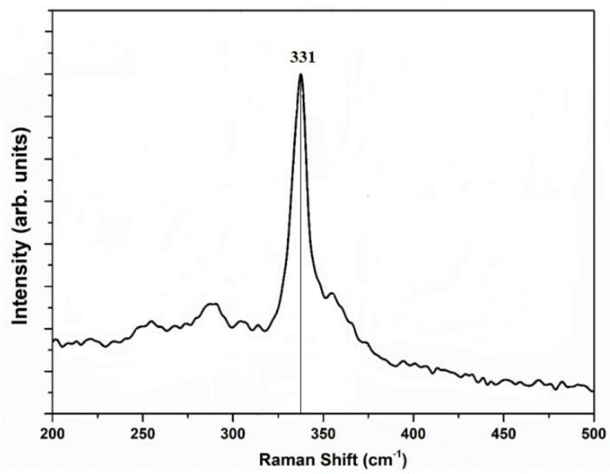

Figure 2. Raman Shift of CZTS Thin Film

Raman spectrum is characterized to verify the presence of CZTS structure, as shown in figure 2. The spectra show several peaks characteristics of vibrational modes of CZTS and some of its secondary phases. The scattering at 331 cm -1 is identical to the literature values of CZTS. The absence of peaks at 278 cm $^{-1}$, 351 cm $^{-1}$ and 295–303 cm $^{-1}$, 355 cm $^{-1}$ suggests the absence of ZnS and Cu_2SnS_3 in the sample. The XRD and Raman results indicate that the main phase of sample is CZTS.

The average crystal size of 3.8 nm estimated from XRD patterns therefore indicates a compact polycrystalline nature of the thin films. Holes with diameters of 165–285 nm are observed on the smooth surface of the thin films. These holes might be formed due to the evaporation of the solvents when the thin films were annealed in argon. This is an interesting result because generally porous structures or cracks were resulted upon decomposition of extra sulfur precursors (TAA or thiourea) and solvents. A higher surface area in porous structures was proposed to lead to an enhanced photoelectrochemical solar energy conversion efficiency due to a higher density of surface reaction centres.

The EDX spectrums of CZTS is illustrated in figure 3 and the atomic ratios of the elements of CZTS thin film is given in Table 1. CZTS thin films are Cu-poor, Zn-rich, and Sn-rich according to the table. As a result, the rate of Cu element deposition may continue to be low. For the Sn element to evaporate, the 350 ^{0}C temperature used to anneal CZTS thin films is a low value. In contrast to other element atoms, Sn atoms in thin films at 350 ^{0}C tend to diffuse more towards the surface.

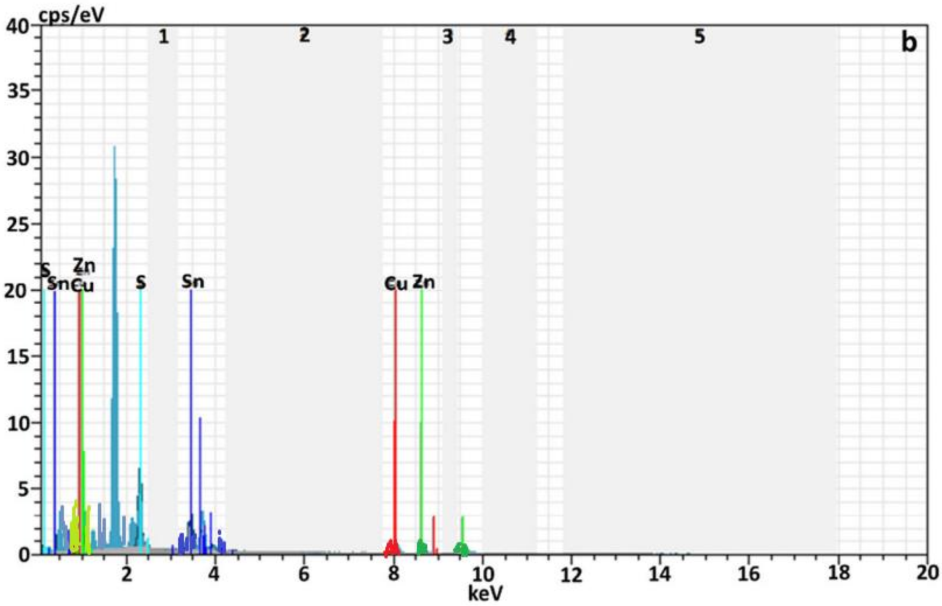

Figure 3. EDX spectrums of CZTS thin film

In addition, the pressure may be higher during annealing process under vacuum conditions, which may increase the amount of Sn atoms in CZTS thin films. In CZTS thin films, SnZn donor defects, Vcu (Cu vacancy) acceptor defects, and ZnCu (Zn occupied Vcu) acceptor defects can develop. CZTS ultrathin films' p-type properties are improved by VCu and ZnCu defects, however their p-type properties are negatively impacted by SnZn defects. The ratio of Cu/Sn in CZTS thin films can also be used to modify the lifetime of minority charge carriers in these thin films. Even though the heterojunction solar cells in this work were constructed from CZTS thin films, this circumstance causes the Jsc and values of these CZTS heterojunction solar cells to be greater.

b) Morphology analysis

FESEM have been utilized to analyse the morphology of the Cu_2ZnSnS_4 films. The FESEM micrographs of the CZTS thin films is magnified at 1000x, 2000x and 5000x that shown in figure 4 (a), (b) & (c). The results showed that the films are uniform and homogeneous. The grains in the films are shaped like circles, and as the deposition period increases, so do their sizes. The grains' diameters and spherically shaped nanoparticles show a homogeneous distribution of direct contact between Cu and Zn. The spherical nanoparticles of the deposited sample were changed by the annealing temperature of 350 ^{0}C into granular particles that covered the whole surface of the substrate.

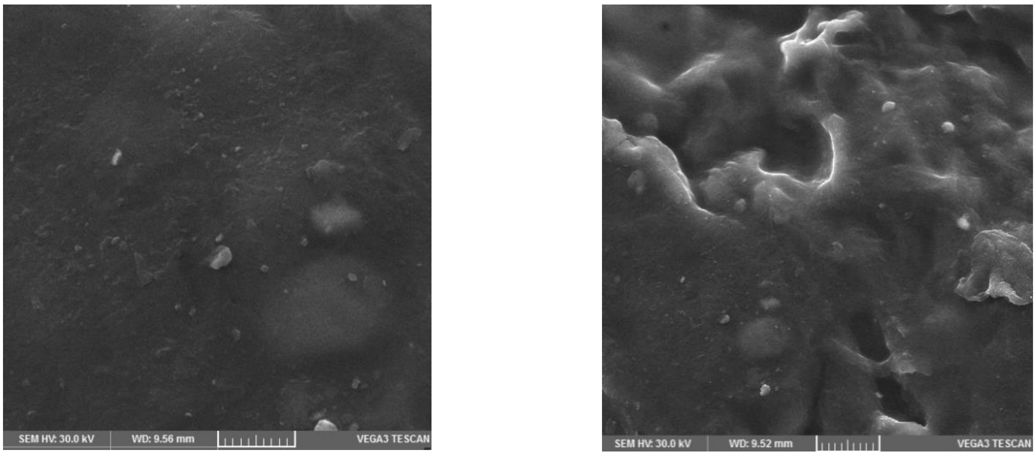

Figure 4 (a) & (b) FESEM images of CZTS Thin Film for 1000x and 2000x

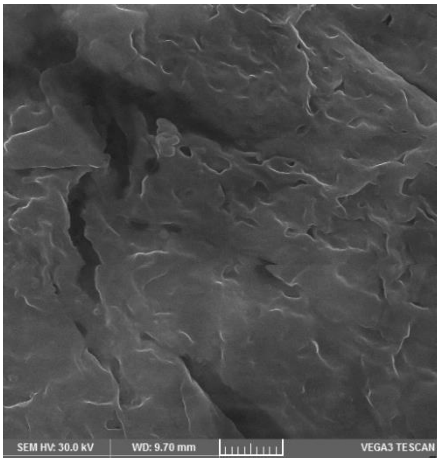

Figure 4 (c) FESEM images of CZTS Thin Film for 5000x

The average grain size demonstrated the presence of a nanostructure in the Cu_2ZnSnS_4 thin films. EDX has examined the elemental make-up of Cu_2ZnSnS_4 thin films. The pattern proves that copper, zinc, tin, and sulphur exist. In the as-deposited Cu_2ZnSnS_4 thin films, the atomic ratio of Cu, Zn, Sn, and S is close to 2:1:1:4, which is close to the stoichiometric composition. The rings pattern in the chosen area of the electron diffraction demonstrates the nanocrystalline nature of the films, which is revealed by the electron diffraction. Also, the distinct spots in the ring pattern prove that the film is made up of nanoscale grains.

From this, the nonstoichiometric films can be seen to have a smoother and more highly dense surface with smaller grains (< 1 µm) when compared to the stoichiometric films. Furthermore, the degree of crystallization is lower for the nonstoichiometric films, but increases with increasing spinning speed. This is quite significant, as the efficiency of a solar cell generally increases with increasing grain size in the absorber layer

c) Optical characterizations

The application of CZTS thin films in solar cells requires the detailed study of the optical properties. By acquiring the transmission and reflection spectra, we have used UV-Visible-NIR spectroscopy to explore how light interacts with the material in this study. The spectrophotometric measurements of T(λ) and R(λ) have been utilized in determining the optical parameters such as; the absorption coefficient (∝), extinction coefficient (k) and the refractive index (n) of CZTS thin films.

Figures 5(a) and 5(b) show the variation in optical reflectance and transmittance for deposited Cu2ZnSnS4 thin films as a function of wavelength (b). These figures show that whereas reflectance has been found to rise with increased deposition time, transmittance has been found to decrease with increased deposition time. The absorption coefficients, α, of the CZTS thin films deposited has been determined from the transmittance T (λ) and reflectance, R (λ) and film thickness (d).

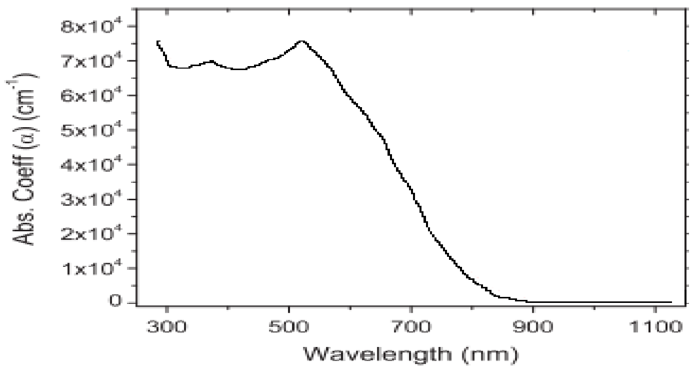

Figure 5 (a) Absorption Coefficient Vs Wavelength of CZTS Thin Films

Figure 5(a) explains the spectral dependence of the absorption coefficient on the wavelength for the Cu_2ZnSnS_4 thin films. The absorbance coefficient of Cu_2ZnSnS_4 thin films has been reported to rise with wavelength as well as with an increase in deposition time. Moreover, films show a high absorption coefficient in the visible light range exceeding 128 cm^{-1}, which is consistent with the literature [2, 4]. This is evidence of the superiority of the developed films that will be used as an absorber film in solar cells. Several optical parameters are determined by the extinction coefficient (k), as those are related to the absorption of light waves and the dielectric constants. The extinction coefficient (k) of the Cu_2ZnSnS_4 thin films deposited has been evaluated.

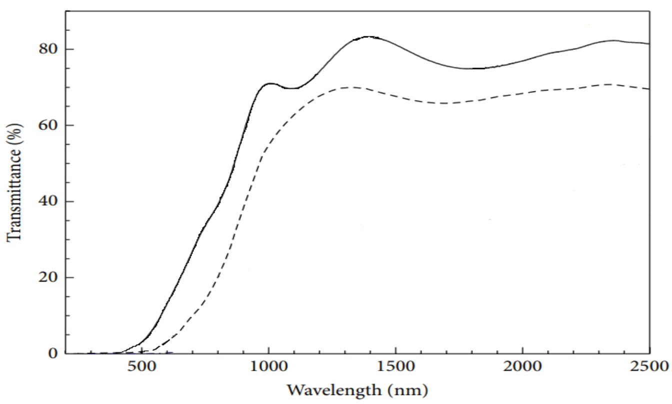

Figure 5 (b) Transmittance Vs Wavelength of CZTS Thin Films

The transmittance of the extinction coefficient on the wavelength of CZTS thin films is shown in Figure 5(b). For the materials used to make optical switches, optical filters, and other devices, the calculation of refractive index is essential for this present study. The spectrum modulation of the reflective index of CZTS thin films as a function of wavelength has been used to derive the refractive index (n) of the Cu_2ZnSnS_4 thin films from the extinction coefficient (k) and reflectance (R). The refractive index of Cu_2ZnSnS_4 thin films has been shown to have anomalous dispersion for wavelengths below λ<675 nm and normal dispersion for wavelengths above λ<675 nm. During longer deposition times, a rise in the refractive index has been studied.

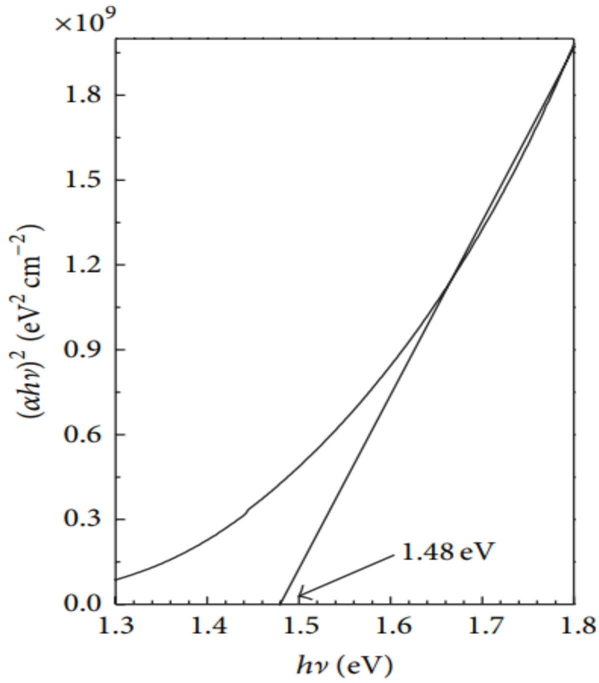

Figure 6 Tauc curves of CZTS Thin Films

Electrons move from the lower energy band (valence band) to the higher energy band as a result of light's interaction with the materials (conduction band). Tauc's relation has been used to comprehend the type of transition and the energy needed (optical band gap) for the transformation. The optical band gap (E_g) of CZTS thin films can be obtained by employing Tauc's relation is shown in figure 6[27, 28]: The values of (E_g) have been determined to be 1.48 eV, which is extremely close to the ideal band gap value of "1.5 eV" for semiconductors employed in photovoltaic conversion.

The production of defects may be to account for the reduction in (E_g) values, which in turn influences the materials' optical properties, where the optical band gap is dependent on composition. Other factors, like as changes in film morphology, could be the source of band gap fluctuation. The existence of unstructured defects may be viewed as the cause of the drop in (E_g) in our investigation, which ultimately resulted in a rise in the grain size of the film.

d) Photovoltaic properties of CZTS Thin Films

The photovoltaic properties have been studied by analyzing the current-voltage (I-V) measurements in dark and under light illuminations for the fabricated hetero-junction. Figure 7 illustrates the I-V characteristics of n-Si/P-Cu_2ZnSnS_4 heterojunction when exposed to light and dark conditions It is clear that the n-Si/P-CZTS heterojunction's current value under light exposure is higher than it is under dark circumstances. This demonstrates that the formation of electron-hole pairs as a result of the absorbed light results in photocurrent that contributes to the carrier. The power conversion efficiency of the manufactured hetero-junction can be used to assess its performance.

The key parameters that describe the power conversion energy are V_{oc} (the open circuit voltage), J_{sc} (the short circuit current density), and FF (the fill factor). The V_{oc} of the solar cell can be prescribed as the voltage that balances the current flow through the external circuit. The J_{sc} can be described as the current during the external circuit in the absence of applied voltage. To estimate the efficiency of our solar cell, an illuminated cell has been brought together to a load resistance from zero to infinity.

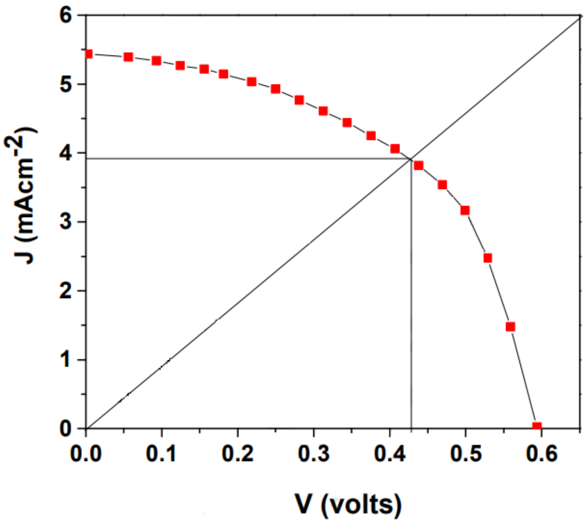

Figure 7 current-voltage (I-V) measurements of CZTS Thin Films

The efficiency (η) of a solar cell is a metric used to compare the performance of one solar cell to that of another. Moreover, it is equivalent to the proportion of solar cell output energy to solar input energy. The following equation [45] can be used to calculate the solar cell efficiency (η): The ratio of the highest power point to the product of the open circuit voltage and short circuit current density is known as the fill factor (FF) of the manufactured cell (J_{sc}). The solar cell has the following estimated device parameters as V_{oc} = 0.598 V, J_{sc} =5.56 mAcm^{-2} , FF = 0.535 and efficiency η = 3.65 %. The good value of the FF could be attained the good interface between Cu_2ZnSnS_4 film and electrodes surface.

Conclusions

The present work intends to propose the advancement of new technique of spin coated pure Cu_2ZnSnS_4 solar cell by taking into account the CZTS kesterite structure, secondary phases, bandgap and absorbance properties. In order to produce the high-performance solution-processed CZTS solar cells, this work required to take into account the atomic ratio of Cu-poor, Zn-rich, and the film consistency during the spin coating process. The impact of the deposition processes on the variation of the CZTS films' structural, optical, optoelectrical, and electrical properties has been studied. According to the XRD patterns, CZTS thin films are polycrystalline and have a kesterite crystal structure. The evaluated films showed a direct energy gap transition that found to be 1.48 eV. The ratio of the highest power point to the product of the open circuit voltage and short circuit current density is known as the fill factor (FF) of the manufactured cell (J_{sc}). The solar cell has the following estimated device parameters as V_{oc} = 0.598 V, J_{sc} =5.56 mAcm^{-2} , FF = 0.535 and efficiency η = 3.65 %. Therefore, the paper provides a simple and suitable approach for fabricating CZTS without post-treatment to make low-cost solar cells.

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