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Analytical model for studying the role of ZnS-doped CdS on the performance of CZTSSe solar cells

H. A. Mohamed[a], Sh. S. Ali[a], M. R. Ahmed[a], W. S. Mohamed[b,c,*]
[a] *Physics department, Faculty of Science, Sohag University, 82524 Sohag, Egypt*
[b] *Physics Department, College of Science, Jouf University, Al-Jouf, Sakaka, P.O. Box 2014, Saudi Arabia*
[c] *Thin Films and Nanotechnology Lab, Physics Department, Faculty of Science, Sohag University, 82524, Sohag, Egypt*

This study focuses on thin-film structures made of ITO, CdS, ZnS, CZTSSe, and Mo (i.e., ITO/CdS:ZnS/CZTSSe/Mo) for solar cell applications. The effect of ZnS content on the performance of this cell has been theoretically investigated. The optical losses caused by reflection at various interfaces and absorption in ITO and CdS:ZnS layers have been calculated using the current structure's experimental data. The losses due to charge carrier recombination at the front and back surfaces of the CZTSSe absorber have been calculated using the absorber layer and depletion region parameters. It was discovered that increasing the ZnS content causes more photons to enter the absorber layer, causing the short-circuit current density to increase. Under consideration of optical and recombination losses, a maximum efficiency of about 13.75%, a fill factor of 81.6%, and an open-circuit voltage of 808 mV were obtained for ZnS-content = 0.5.

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1. Introduction

Thin-film solar cells have become competitive with first-generation solar cells because the amount of material utilized is lowered, resulting in cheaper production costs. The concept of using thin-film solar cells is based on the stacking of a number of thin layers, each of which plays an important part in the performance of the solar cells. These layers are composed of the substrate, transparent conducting oxides (TCO), window layer, absorber layer, and metal contact layer, respectively.

Copper indium gallium disulfoselenide (CIGSSe) [1, 2] and cadmium telluride (CdTe) [3, 4] have been extensively studied as a promising high efficient absorber material for second generation solar cells. However, the drawbacks of using elements like Cd, Se, In, and Ga as functional layer materials in solar devices, such as their toxicity, abundance, and high cost, have led to the search for alternative materials. These materials could be used in their alternative. [5-7]. Nowadays, $Cu_2ZnSn(S,Se)_4$ (CZTSSe) materials have been considered alternative to expensive and toxic CIGSSe and CdTe thin-film solar cells. These materials have unique properties such as, low cost, earth abundant, p-type electrical conductivity, suitable absorption coefficient (>10^4 cm^{-1}) and optimum optical energy gap between 1 and 1.5 eV [8, 9].

In solar cells, and with particular thin film heterojunction solar cells, which depend on their composition on chalcogen materials like Cd-(S,Se,Te), one finds that a CdS thin layer is utilized both as a window material and as a heterojunction partner such as CdTe [10], $CuInS_2$ [11], $CuInGaS_2$ [12], $Cu_2ZnSn(S,Se)_4$ [13] and PbS [14]. However, due to the fact that CdS has a relatively low energy gap (2.42 eV), photons with energies that are more than 2.42 eV are unable to penetrate the absorber layer. This issue can be resolved in one of three ways: the first is to

* Corresponding author: wsmahmed@ju.edu.sa
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decrease the thickness of the CdS layer; the second is to replace CdS with a semiconductor that has a large band gap; and the third is to enhance the energy gap of CdS using an annealing process. A direct rise in the short-circuit current density may be produced as a result of decreasing the thickness of the CdS layer. However, this typically results in a significant increase in the formation of pinholes and short circuit effects [15], in addition to a reduction in open circuit voltage and fill factor [16].

In this study, the findings of an examination into the efficiency of thin-film solar cells relying on CZTSSe by using CdS:ZnS as a window layer are presented. The properties of the cell, notably open circuit voltage, fill factor, and cell efficiency, have all been the subject of theoretical investigation. The values of these parameters have been determined on the basis of the findings that were obtained from the computation of the short-circuit current density. Both the optical and the recombination losses have been made sure to be taken into consideration as thoroughly as possible.

2. The model

In this model, thin film solar cells, namely those with a multilayer structure that includes absorber (CZTSSe), window (CdS:ZnS), and charge-collecting contacts (TCO) layers, have been investigated. Indium tin oxide was used as the charge-collecting contacts in this investigation (ITO). Before reaching the absorber layer where electron-hole pairs are generated, the light flow must travel via the solar cell's ITO and window layers. Figure 1 depicts a simplified view of substrate thin-film solar cell. Thus, the optical losses are due to air/ITO reflection, ITO/CdS:ZnS reflection, CdS:ZnS/CZTSSe reflection, and light absorption in the ITO and CdS:ZnS layers. Optical losses can be estimated quantitatively by using the refractive index, extinction coefficient, and energy gap of the materials in use.

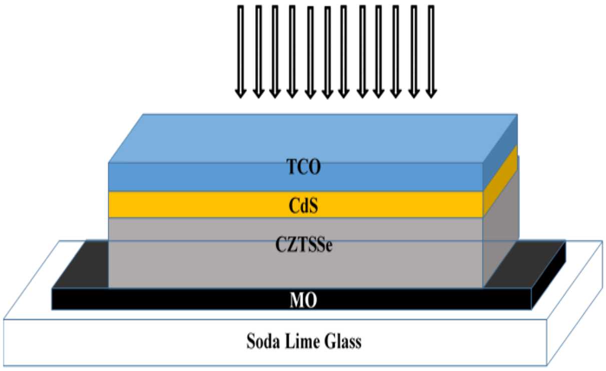

Fig. 1. Thin-film solar cell schematic structures based on CZTSSe.

Besides, the photogenerated carriers in depletion region of CdS:ZnS/CZTSSe p-n heterostructure and those carriers that generated outside this region may recombine before reaching the front and back electrode and, therefore, do not contribute to the formation of the photocurrent. This model enables us to calculate the quantum yield efficiency and estimation of the recombination losses either at the front surface or at the rear surface of the absorber layer. The continuity equation is used in order to calculate the recombination losses that need to be taken into account. The drift and diffusion components of the photocurrent are both accounted for by this equation. Quantifying the recombination losses requires knowledge of the absorber layer's physical qualities like its absorption coefficient, energy gap, thickness, carrier lifetime, and mobility, as well as the junction's properties like its barrier height and the breadth of its space-charge area. The physical properties of the absorber layer are taken into consideration in order to arrive at this conclusion. In addition, the spectrum was calibrated to the international standard of Am 1.5, and the temperature of operation was held constant at 300 K. Table 1 is a listing of the parameter values that were utilized for this research project.

Table 1. Values of the parameters that were utilized in the model. Thickness of ITO (d_{ITO}) , thickness of CZTSSe (d_{CZTSSe}) ,width of space charge region (W) , the barrier height(φ_0) ,the elementary charge (q), the velocity of recombination at the front surface of CZTSSe (S_f) , the recombination velocity at the back surface of CZTSSe (S_b) , the mobility of charge ($\mu_{n,p}$) ,and the lifetime of charge ($\tau_{n,p}$).

parameter	d_{ITO}	d_{CZTSSe}	W	φ_0-qv	$S_{f,b}$	μ_n	μ_p	$\tau_{n,p}$
value	100 nm	4 µm	0.5 µm	1 eV	10^6 cm/s	10^3 cm^2/(V s)	80 cm^2/(V s)	4*10^{-8} s

The well-known Fresnel equations [2] are used to calculate the reflectivity, denoted by the letter "R," at two different adjusting layers, numbered "1" and "2."

$$R_{12}(\lambda) = \frac{|n_1^* - n_2^*|^2}{|n_2^* + n_2^*|^2} = \frac{(n_1 - n_2)^2 + (k_1 - k_2)^2}{(n_1 + n_2)^2 + (k_1 + k_2)^2} \tag{1}$$

where n represents the refractive index, and k stands for the extinction coefficient.

It is possible to make an estimate of the transmittance by utilizing the optical calculation that takes into account the refractive indices and extinction coefficients of the materials. In this particular instance, the transmission is provided by:

$$T(\lambda) = (1 - R_{12})(1 - R_{23})(1 - R_{34}) \tag{2}$$

where R_{12}, R_{23} and R_{34} are the reflectivity at the interfaces between air/ITO, ITO/CdS:ZnS and CdS:ZnS/CZTSSe, respectively.
whenever the process of absorption that happens in TCO and CdS:ZnS. The term "optical losses" refers to the losses that occur as a result of reflection and absorption. In this scenario, Eq.2 is written as follows:

$$T(\lambda) = (1 - R_{12})(1 - R_{23})(1 - R_{34})e^{-\alpha_1 d_1}e^{-\alpha_2 d_2} \tag{3}$$

where, α_1 and α_2 are the absorption coefficient of ITO and CdS:ZnS and d_1 and d_2 their thickness, respectively. The values of n and k of ITO and CZTSSe is taken from [2, 17].

In a general sense, the absorption coefficient α(λ can be computed by making use of the data of the extinction coefficient k(λ), and doing so in accordance with the following equation:

$$\alpha = \frac{4\pi}{\lambda}k \tag{4}$$

Through the process of calculating the short-circuit current density Jsc, one is able to gain a quantitative analysis of the optical losses and the effect that this has on the performance of the solar cell. The formula for determining the spectrum distribution of the photons is $\boldsymbol{\Phi_i/h\nu}$, where $\boldsymbol{\Phi_i}$ is the spectral power density and $\boldsymbol{h\nu}$ is the photon energy. The current density in a short circuit, abbreviated as J_{SC}, can be determined with the use of the following equation [18]:

$$J_{SC} = q\sum_i \frac{\Phi_i(\lambda_i)}{h\nu_i}T(\lambda_i)\Delta\lambda_i \tag{5}$$

where Δλ is the distance between two adjacent values of the wavelength.

Reflection and absorption optical losses can be calculated with Eq. (5) and the following formula.

$$Losses\ (\%) = \left(1 - \frac{J_{SC}}{J_{SC(\max)}}\right) \times 100 \tag{6}$$

where $J_{SC(\max)}$ is the greatest value of short-circuit current density that may be obtained when $T(\lambda)=1$. It is also important to keep in mind that the summing in Equation (5) should be performed spanning the spectral range from λ=300 nm to $\lambda=\lambda_g=hc/E_g$, where E_g is the optical band gap of the absorber material. This should be done with the following caveat in mind.

Lavagna et al [19] solved the continuity equation using the boundary conditions and they obtained an expression of the quantum efficiency. Internal quantum efficiency was simplified by Kosyachenko et al. to the following formulation [20].

$$\eta_{int} = \frac{1+(S_f/D_p)[\alpha+(2/W)(\varphi_o-qV)/kT]^{-1}}{1+(S_f/D_p)[(2/W)(\varphi_o-qV)/kT]^{-1}} - \frac{\exp(-\alpha W)}{1+\alpha L_n} \tag{7}$$

where S_f is the front surface recombination velocity, D_p is the hole diffusion coefficient related to the mobility μ_p by the Einstein relation $qD_p/kT= \mu_p$, α is the absorber's absorption coefficient, $\boldsymbol{W}$ is the space-charge region's width, $\boldsymbol{V}$ is the voltage, φ_o is the barrier height, L_n $(=(\tau_n D_n)^{1/2})$ is the diffusion length of minority carriers, τ_n is the electron's lifetime, and D_n is the diffusion coefficient of the electrons related to the mobility μ_n by the Einstein relation $qD_n/kT= \mu_n$. One thing to keep in mind is that Eq. (7) doesn't account for is recombination happening on the absorber layer's rear surface.

The drift part of the photoelectric quantum yield can be expressed using Eq. (7). In the space-charge area, photoelectric quantum yield is proportional to the absorptivity of this layer, or 1 – exp(–α*W*). We may derive the expression for the diffusion component of the photoelectric quantum yield by subtracting the phrase 1 – exp(–α*W*) from the right side of Eq. (7).

The equation for the drift component of the photoelectric quantum yield is given by Eq. (7). Within the space-charge area, the photoelectric quantum yield is proportional to the absorptivity of this layer, or 1 – exp(–α*W*). The diffusion component of the photoelectric quantum yield can be calculated by subtracting the term 1 – exp(–α*W*) from the right side of Eq. (7)

$$\eta_{dif} = \exp(-\alpha W)\frac{\alpha L_n}{1+\alpha L_n} \tag{8}$$

The back-surface recombination of the absorber layer is neglected in this equation. When we account for surface recombination at the front interface, we can calculate the drift component of the photoelectric quantum yield by subtracting the right side of Eq. (8) from the right side of Eq. (7).

$$\eta_{drift} = \frac{1+(S_f/D_p)[\alpha+(2/W)(\varphi_o-qV)/kT]^{-1}}{1+(S_f/D_p)[(2/W)(\varphi_o-qV)/kT]^{-1}} - \exp(-\alpha W) \tag{9}$$

From the solution of the continuity equation, we also obtain the diffusion component of the internal quantum efficiency (η_{dif}). The continuity equation has been reduced to the form [21], which is sufficiently accurate.

$$\eta_{dif} = \frac{\alpha L_n}{\alpha^2 L_n^2-1}\exp(-\alpha W) \times \left\{\alpha L_n - \frac{\left(\frac{S_b L_n}{D_n}\right)[\cosh((d-W)/L_n)-\exp(-\alpha(d-W))]+\sinh((d-W)/L_n)+\alpha L_n\exp(-\alpha(d-W))}{\left(\frac{S_b L_n}{D_n}\right)sinh[(d-W)/L_n]+cosh[(d-W)/L_n]}\right\} \tag{10}$$

in which S_b is the recombination velocity at the absorber layer's rear surface and d is the absorber's thickness. When recombination losses at the front and rear surfaces of the absorber layer are included, the total internal quantum efficiency is calculated using Eqs.(9) and(10).

$$\eta_{int} = \eta_{drift} + \eta_{dif} \tag{11}$$

To get an approximation of the recombination losses, Eq. (5) must be rewritten as follows:

$$J_{SC} = q \sum_i \frac{\Phi_i(\lambda_i)}{h\nu_i} \eta_{int}(\lambda_i) \Delta\lambda_i \tag{12}$$

Also Eq.(6) is used to calculate the recombination losses. In the present case, the maximum short-circuit current density $J_{SC(\max)}$ is obtained at $S_f = 0$ and $S_b = 0$.

It is possible to calculate the external quantum efficiency of solar cells by taking into account the optical losses that are caused by reflection at various interfaces as well as absorption in the TCO and window layers.

$$\eta_{ext} = T(\lambda)\, \eta_{int} \tag{13}$$

where $T(\lambda)$ is provided by Eq.3, which accounts for optical losses.

The short-circuit current density can be written as follows to account for the impact of optical and recombination losses:

$$J_{SC} = q \sum_i T(\lambda) \frac{\Phi_i(\lambda_i)}{h\nu_i} \eta_{int}(\lambda_i) \Delta\lambda_i \tag{14}$$

And the total losses are calculated using Eq.6.

The p-n junction current is calculated using the conventional diode equation, which states:

$$J = J_0 \left[exp\left(\frac{qv}{AkT}\right) - 1 \right] - J_L \tag{15}$$

The elemental charge q, the Boltzmann constant k, the absolute temperature **T**, and the ideality factor **A**; J_0 is the reverse saturation current; J_L is the photogenerated current; Using J_o and **A** from [22] as a starting points a solar cell's performance can be quantified as:

$$\eta = \frac{FF \times J_{SC} \times V_{0C}}{P_{in}} \tag{16}$$

using the formula: where V_{0C} is the open circuit voltage, P_{in} is the density of the total solar radiation power at AM 1.5, and FF is the fill factor:

$$FF = \frac{J_m \times V_m}{J_{SC} \times V_{0C}} \tag{17}$$

The allowable maximum current density and voltage are denoted by J_m and V_m, respectively.

3. Results and discussion

A trial has been carried out [23] where the thermal evaporation technique was employed to obtain thin films of $(CdS)_{1-x}(ZnS)_x$ (x=0, 0.1, 0.2, 0.3, 0.4 and 0.5) and the optical and the structural properties of these films were studied. The obtained energy gap as a function of x-ratio was estimated and listed in Fig. 2-a. Figure 2-b represents the spectral dependence of transmission coefficient $T(\lambda)$ on the ZnS content (x-ratio) of the system ITO/CdS:ZnS. The results are carried out using Eq.(3), which takes into consideration the reflection at various interfaces and the absorption in ITO and window layer.

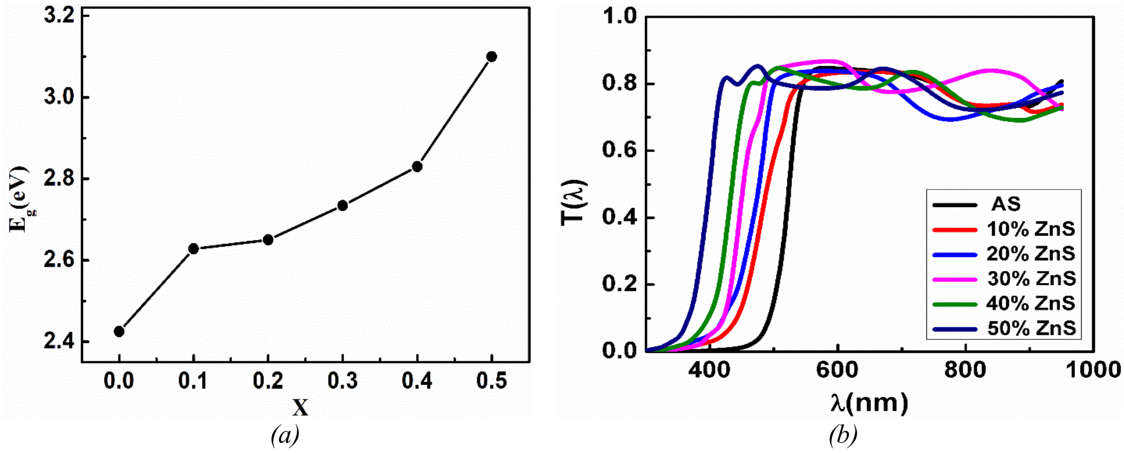

Fig. 2. The optical band gap of (CdS)1-x:(ZnS)x as determined by [23]. (a) and the calculated optical transmittance of the system ITO/CdS:ZnS (b) at various values of ZnS content (x-ratio).

$T(\lambda)$ grows as the concentration of ZnS in the alloy $(CdS)_{1-x}:(ZnS)_x$ increases. The low values of $T(\lambda)$ at short wavelengths can be explained by the fact that both the ITO and CdS:ZnS layers are able to absorb some of the incident photons.

The short circuit current density dependence on the concentration of ZnS contents is shown in Fig.3. It is clear that J_{SC} increases from 18.6 mA/cm^2 for x=0 to 23.5 mA/cm^2 for x=0.5. This refers that when the ratio of addition of ZnS reaches 50% of the window layer (buffer layer), the value of J_{SC} increase by a ratio of 26%. On the other hand, the reflection losses at different interfaces (air/ITO, ITO/CdS:ZnS and CdS:ZnS/CSTSSe) and the losses due to the absorption in ITO and CdS:ZnS are shown in Fig.3.

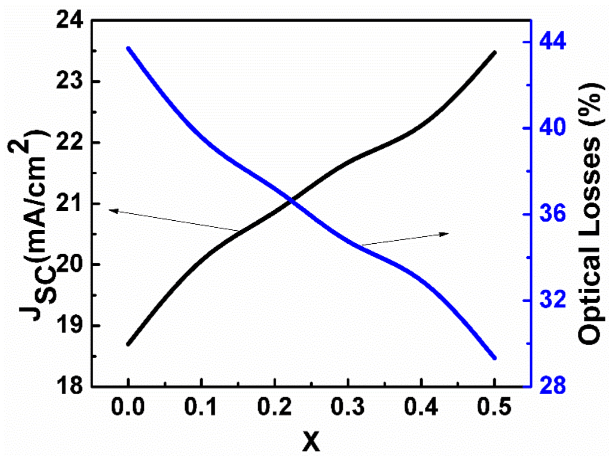

Fig. 3. Optical losses and short-circuit current density in a (CdS):(ZnS)/CdTe/CdSe solar cell for varying concentrations of ZnS (x=0, 0.1, (0.2, 0.3, 0.4, and 0.5).

It can be seen that at pure CdS, the optical losses (reflection and absorption) represent the maximum value of 44%. And with increasing the ZnS content, this losses decrease and record the minimum value of 29% at x=0.5.

In order to investigate the manner in which the ZnS concentration influences the behavior of the quantum efficiency, we plotted the external quantum efficiency (η_{ext}) vs the wavelength for a variety of ZnS concentrations in Fig.4. The ZnS ratio of 50% (x=0.5) exhibits the highest value of the external quantum efficiency particularly at wavelength of 450-550 nm. Where at this ratio, the smallest value of absorption can be taken place in window layer and most incident light passes to the absorber layer.

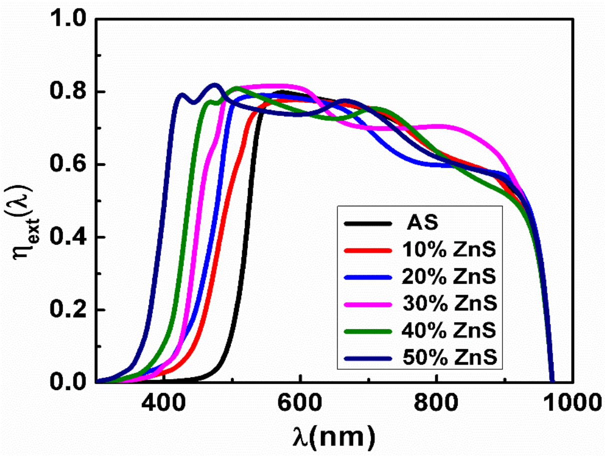

Fig. 4. Spectral external quantum efficiency (η_{ext}) at different concentrations of ZnS.

CZTSSe solar cell performance has been accurately portrayed by accounting for both optical and recombination losses. Optical and recombination losses as percentages, as well as the short-circuit current density of CZTSSe devices with varying ZnS contents, are shown in Fig. 5.

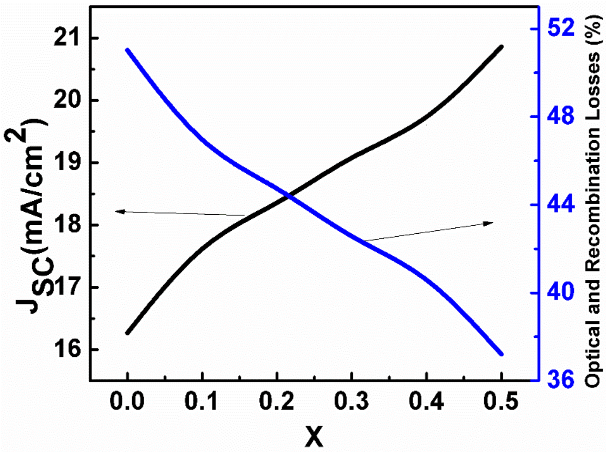

Fig. 5. (CdS):(ZnS) /CZTSSe solar cell with various concentrations of ZnS (x=0, 0.1, 0.2, 0.3, 0.4, and 0.5), showing the short-circuit current density and the related optical and recombination losses for each concentration.

It can be noted that J_{SC} records 16.2 mA/cm^2 for x=0 and about 21 mA/cm^2 for x=0.5. These results are considered smaller compared to the case of optical losses (see Fig. 3). Also, the lowest optical and recombination losses of 37% are observed at x=0.5. It is clear that recombination losses have not significant effect on J_{SC} comparing with the effect of optical losses.

Figure 6 displays the J-V curves of a CZTSSe solar cell under full sunlight. CZTSSe has excellent carrier transport capabilities, as evidenced by the nearly rectangular form of the J-V curve. It is clear that with increasing the ZnS content more photons reach the absorber layer and thus more photogenerated carriers have been carried out. This can be seen in downward shifts in *J-V* curves.

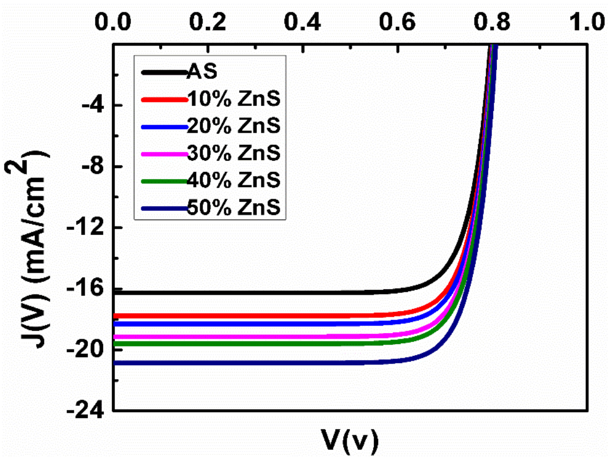

Fig. 6. J-V characteristics of CZTSSe solar cells at various ratios of ZnS.

In Fig. 7, we see a trend in the photovoltaic parameters that are in line with the illuminated J-V characteristics. As the percentage of ZnS in the sample rises, so does the FF and V_{OC} levels. Where, *FF* varies from 0.808 to 0.816 and V_{OC} varies from 0.793 V to 0.808 V for ZnS-content = 0 and 0.5, respectively. However, the efficiency increases dramatically from 10.42% to 13.75% with increasing the ZnS content from 0 to 0.5. Accordingly, we can achieve the maximum efficiency of about 13.75% with *FF* = 81.6% and V_{OC}= 0.808 V at ZnS content of 50%.

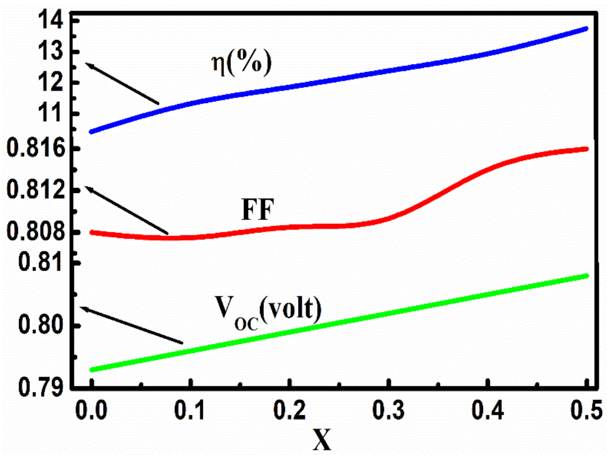

Fig. 7. Dependence of CZTSSe cell parameters on ZnS content.

This enhancement in efficiency by a ratio of 32% is mainly due to the increase of short-circuit current density, which increases from 16.26 mA/cm^2 for x=0 to 20.86 mA/cm^2 for x=0.5. Hence, according to these results, we can say that ZnS plays a significant role in improvement the efficiency of CZTSSe solar cell through allowing more photons to reach the absorber layer and effectively contributing in photogenerated carries.

4. Conclusions

In this work a theoretical model was implied to study thin film solar cells based on CZTSSe with new window layer consists of CdS:ZnS. Such solar cells have been considered alternative to expensive and toxic CIGSSe and CdTe thin-film solar cells due to their unique physical properties. The main parameters used in this model were taken from experimental literatures. Analysis of the impact of ZnS content on short-circuit current density and on cell efficiency metrics was performed while taking optical losses and recombination losses into account.

The short-circuit current density grew noticeably when the open-circuit voltage and the fill factor were both increased by a small amount. The optical losses from interface reflection and absorption in front charge-collecting contacts and the window layer were more significant than recombination losses. The maximum efficiency of about 13.75% with FF = 81.6% and V_{OC}= 0.808 V were achieved at ZnS-content of 50%.

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