Optimization of Chalcogenide CdTe, CZTS and CZTSe Solar Cells PerformancesUsing $Cd_{1-x}Zn_xS$ Buffer Layer

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ABSTRACT: *Cadmium zinc sulfide ($Cd_{1-x}Zn_xS$) as a wide-bandgap material with x=0.7 was used in the present work as an alternative buffer material to CdS to improve the efficiency o fZnO/$Cd_{1-x}Zn_xS$/CdTe, ZnO/$Cd_{1-x}Zn_xS$/CZTS and ZnO/$Cd_{1-x}Zn_xS$/CZTSe thin film solar cells. The photovoltaic parameters such as efficiency, open circuit voltage (V_{oc}), short circuit current density (J_{sc}), and the Fill Factor (FF) have been computed using one-dimensional simulation programs such as Solar Cell Capacitance Simulator (SCAPS v3.3) and Analysis of Microelectronic and Photonic Structures (AMPS-1D). An improvement in conversion efficiency is noticed compared to the structure with the CdS buffer layer. It is found that the efficiencies of $Cd_{1-x}Zn_xS$/CZTSe and $Cd_{1-x}Zn_xS$/CdTeareincreased from 12.61% to 15.35% and from 17.53% to 18.83%, respectively. The simulations were performed for 1 µmthick absorber layers. It is also found that efficiency rises from 12.53% to 13.23% with $Cd_{1-x}Zn_xS$/CZTS structure for CZTS thickness of 2.5 µm. Moreover, the Quantum Efficiency (QE) characteristics display a maximum value of more than 80% in the visible range and the structures presented a slight improvement in the short wavelength. The present study shows that the suggested structures with a$Cd_{1-x}Zn_xS$ buffer layer may improve efficiency and reduce the amount of Cd, which is a toxic element.*

KEYWORDS: *$Cd_{1-x}Zn_xS$; Absorber Layer; Buffer Layer; AMPS-1D; SCAPS.*

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INTRODUCTION

Photovoltaic technology offers human beings unlimited-green energy sources by directly converting solar energy into electricity using certain semiconductor materials. In recent years, Thin Film Solar Cells (TFSCs) have been considered the second polarizing technology with a 5% share compared to the commercialized silicon technology, which dominates the global photovoltaic (PV) module production by 95% share [1]. Most TFSC devices are made using cadmium telluride(CdTe), Copper Indium Gallium Diselenide (CIGS), and Kestrite semiconductors due to their easy-to-fabricate and their low-cost constituents compared to the bulk silicon cells [2]. Among them, the CdTe has attracted the attention of researchers[3-7] due to its high absorption coefficient that exceeds $5\times10^5 cm^{-1}$ and an ideally very close-optimum direct band-gap of 1.45 which is suited for solar energy conversion [3,8,9]. Secondly, it can be prepared by a wide variety of physical, chemical, and electrochemical techniques, especially low-cost such as Chemical Vapor Deposition (CVD), Chemical Bath Deposition (CBD), Close-Space Sublimation (CSS) [10], and Metal-Organic Chemical Vapor Deposition (MOCVD) [11].In addition to the previous reasons, the conversion efficiency of CdTe TFSCs is also being improved at a very fast pace[12]. Where in 2012, A decent conversion efficiency of 13.4% was reported for the conventional CdS/CdTe heterojunction by *Chu et al.* [13]. A value of 15.8% was achieved by *Ferekides et al.* [14] for CdTe/CdS solar cell, where the CdTe absorber layer was prepared by CSS technique with a thickness of 4 ~ 8 µm. An efficiency of 16.9% was announced by *Matinet al.* [4] and the inserting of the inter-diffusion of $CdTe_{1-x}S_x$ improved the efficiency up to 19.6% with a CdTe absorber thickness of about 3.9 µm [15]. Recently, the study of kesterite compounds, such as Cu_2ZnSnS_4 (CZTS) and $Cu_2ZnSnSe_4$ (CZTSe) becomes one of the most attractive research in photovoltaic materials because they are non-toxic and naturally abundant, and because of their suitable band-gap energy of 0.95 and 1.5 eV for CZTSe and CZTS, respectively. Secondly, they have a high absorption coefficient of over 2×10^4 cm^{-1} for CZTSe and 5×10^5 cm^{-1} for CZTS [5, 6, 16-20]. Where in 2013, *Emrani et al.*synthesizedCu_2ZnSnS_4 thin film solar cells by sulphurization of sputtered Sn/Zn/Cu precursors, and they found a maximum efficiency of 5.75% [21].While, in the same year, *Jung et al.* found a maximum efficiency of 2.88%with depositingCZTSthin films on Mo-coated soda-lime-glass substrates by a single-stage co-evaporation process [22]. In 2015, the current record device of 4.83% is developed by *Engberg et al.*[23]by producing a CZTS absorber layer of high quality through a simple drop-casting technique.

To solve the encountered problems during laboratory studies such as the previously mentioned research, a high concern has been devoted to the simulation studies of kesterite compounds based solar cells. Thus, a SCAPS-1D study was reported by *Simya et al.*and presented 12.03%, 13.16%, and15.77% efficiencies for CZTS,CZTSe, and CZTSSe devices, respectively, where the absorber layers were fixed to a thickness of about 2.50 µm[5, 6]. *Patel et al.* reported similar results in 2012 with a conversion efficiency of 13.41% [17]. Experimentally, it has been observed that using $Cd_{1-x}Zn_xS$ as a buffer layer in $Cd_{1-x}Zn_xS$/CdTe solar cell structure could achieve a high efficiency [24]. There are a lot of studies concluding that the optimal composition for the use of $Cd_{1-x}Zn_xS$ as a buffer layer ranges between$x = 0.5$ to$x = 0.7$. *Kartopu et al.* carefully fabricated solar devices using different $Cd_{1-x}Zn_xS$ compositions and then carried out an experimental evaluation of the devices' efficiencies. They concluded that $Cd_{1-x}Zn_xS$ optimal composition to be used as a buffer layer was around $x = 0.7$ [25]. A typical TFSC structure generally consists of Transparent Conductive Oxide (TCO) window, which also acts as an electron transport layer, a buffer layer, and an absorber, also acts as a hole transport layer, a back metal contact, and a substrate. Where the formed buffer/absorber junction works to separate the generated electron-hole pairs using the absorbed photons at the absorber layer.ZnO has attracted attention to use as a TCO layer material in photovoltaic devices due to its wide band gap (3.3 eV) and its large exciton binding energy (60 meV) [26].

In this study, cadmium zinc sulfide (Cd_{1-x} Zn_xS) material with x=0.7is introduced as a buffer layer replacing the conventional CDs buffer layer in ZnO/CdS/CdTe/Mo, ZnO/CdS/CZTS/Mo, ZnO/CdS/CZTSe/Mo solar cells. In the present study, three structures based on CdTe, CZTS, and CZTSe absorbers with a conventional buffer layer ofCdS are simulated to determine the optimum values of thicknesses and doping concentration that give better performances for conventional structures. Secondly, the replacement of the CdS

Table 1: Base parameters for CdTe , CZTS and CZTSe absorbers.

Parameters	n-ZnO-Al[3]	n-CdS[3 ,4,17]	n-Cd_{1-x} Zn_xS[24]	p-CdTe[3-4]	p-CZTS[5-6]	p-CZTSe[5-6]
E_g (eV)	3.3	2.4	3.05	1. 5	1.5	0.95
X_e (eV)	4.6	4.4	4.4	4.28	4.5	4.35
ε_r	9	10	8.3	9.4	10	10
N_C (cm^{-3})	2.2×10^{18}	2.2×10^{18}	1.5×10^{15}	7.5×10^{17}	2.2×10^{18}	2.2×10^{18}
N_V (cm^{-3})	1.8×10^{19}	1.8×10^{19}	1.8×10^{17}	1.8×10^{18}	1.8×10^{19}	1.8×10^{19}
μ_e(cm^2/Vs)	100	100	100	320	100	100
μ_p (cm^2/Vs)	25	25	25	40	25	25
CdTe solar cell						
n, p (cm^{-3})	1×10^{19}	1×10^{18}	1×10^{18}	1×10^{15}	_	_
CZTS and CZTSe solar cells						
n, p (cm^{-3})	1×10^{17}	1.6×10^{17}	1.6×10^{17}	_	2.5×10^{15}	1×10^{16}

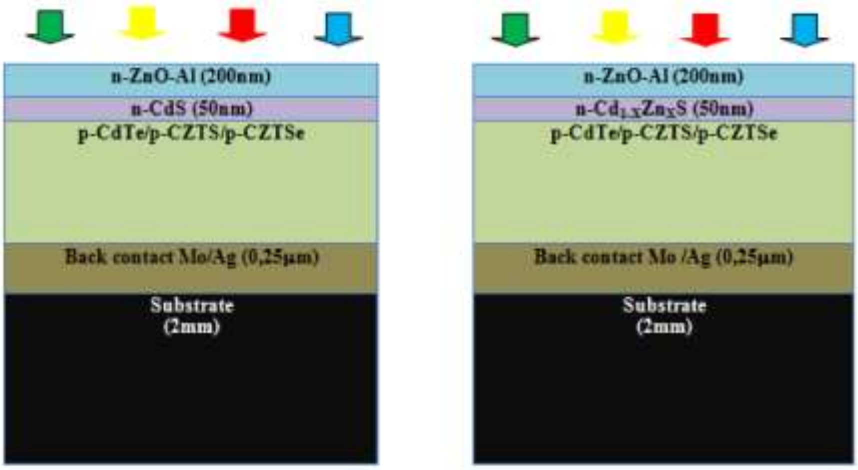

Fig. 1: Structures of CdTe, CZTS, and CZTSe solar cells: (left) Structure with the CdS buffer layer and (right) Structure with $Cd_{1-X}Zn_XS$ buffer layer.

with $Cd_{1-x}Zn_xS$ in three structures is investigated by numerical simulation through SCAPS and AMPS-1D simulators. Therefore, the results of the J-V characteristics; the QE, and the photovoltaic parameters are determined and discussed.

Device structure

Fig.1 illustrates the structure of the CdTe, CZTS, CZTSe solar cells. On the left side, we present the conventional structure with the CdS buffer layer, and on the right side, the structure with the proposed $Cd_{1-x}Zn_xS$ buffer layer is presented. The ZnO is introduced as an optical window layer. The argent (Ag) is used as a back metal contact for CdTe solar cell structure and the Molybdenum (Mo) is used also as a backmetalcontact for CZTS and CZTSe solar cell structures.

The used physical parameters in this simulation study are the band gap (E_g), electron affinity (X_e), dielectric constant (ε_r), the density of states at the conduction band (N_C), density of states at the valence band (N_V), electron mobility (μ_n), hole mobility (μ_p), and electron and hole concentration (n, p). The values of these physical parameters are given in Table 1. The given values are chosen on the basis of theoretical and experimental data [27-30]. The thicknesses are varied from 200 nm to 4000 nm for the CdTe absorber layer, from 200 nm to 2500 nm for the CZTS absorber layer, and from 200 nm to 1500 nm for the CZTSe layer. Furthermore, the thicknesses are fixed at 200 nm for the ZnO window layer and 50 nm for CdS and/or $Cd_{1-x}Zn_xS$ buffer layers.

RESULTS AND DISCUSSION

AMPS-1D and SCAPS-1D simulators are used to explore the conventional CdTe, CZTS, and CZTSe baseline cells and modified cells for achieving ultra-thin solar cells. Therefore, these programs have been used to realistically simulate the electrical parameters of the thin film hetero-junction solar cells [31,32].In our simulation, the temperature is kept at 300K, and the illumination of light isAM1.5 G(1000W/cm^2), which is used as a Standard Test Condition in stimulating research. The numerical simulations using the previously mentioned simulators are made by solving three basic equations related to semiconductors as the Poisson Equation (Eq.(1)) and the Continuity Equations (Eq. (2) and Eq. (3)) for free holes, and for free electrons[2,5,6,28]. Poisson's equation is solved by using formulas of the displacement vector, Gauss's Law of Electrostatics, and the relationship between electric field and flux. Drift, diffusion, and recombination-generation processes give rise to a change in the carrier concentrations in time. The combined effect of all types of carrier action within a semiconductor is described by the continuity equation [33].

$$Di(\nabla \varepsilon V) = -\rho = -q(N_d - N_a + p - n) \quad (1)$$

$$\frac{\partial n}{\partial t} = J_n \frac{1}{q}\nabla + \mu_n \quad (2)$$

$$\frac{\partial p}{\partial t} = -J_p \frac{1}{q}\nabla + \mu_n \quad (3)$$

In semiconductors, the carrier transport occurs by drift and diffusion only as mentioned above and these can be expressed by the below equations:

$$J_n = -\frac{\mu_n n}{q}\frac{\partial E_{Fn}}{\partial X} \quad (4)$$

$$J_p = +\frac{\mu_p p}{q}\frac{\partial E_{Fp}}{\partial X} \quad (5)$$

The first term is drift and the second one is diffusion. Whereas ε is the relative permittivity and V is the electrostatic potential, n and pare are the concentration of the electrons and holes, respectively, J_n is the electron current density, J_p is the hole current density, N_d is a donor

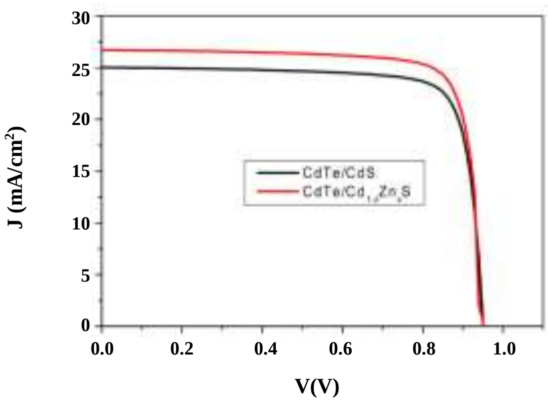

Fig. 2: J-V characteristics of CdTe/CdS and CdTe/$Cd_{1-x}Zn_xS$ solar cell structures.

concentration, N_a is the acceptor concentration, μ_n is the mobility of electrons,μ_p is the mobility of the hole, and q is the electric charge. Variable carrier concentrations for electron and hole are denoted by $E_{Fn}$and E_{Fp}.

The SCAPS and AMPS-1D simulators have different panels to enter thin film materials and their input settings and to show the results of J-V characteristics via a curve, which illustrates the power conversion efficiency (η), Fill Factor (FF), short-circuit current density (Jsc), and open-circuit voltage (Voc). Where the power conversion efficiency can be calculated by the below equation[31-32]:

$$\eta = \frac{FF \times V_{OC} \times J_{SC}}{P_{in}} \quad (6)$$

And, the input P_{in} is the emitted solar radiation on the solar cell device and it is assured as an input condition through panels of simulators.

To decrease the toxicity of cadmium (Cd) in the Cadmium sulfide CdS buffer layer to 20% with the replacement of the CdS layer by $Cd_{0.8}Zn_{0.2}S$ layer for the best performance is also the aim of this study.

Optimum thicknesses of CdTe, CZTS, and CZTSe layers

Case of CdTe solar cells structure

The numerical simulation is carried out using the AMPS-1D simulator to get of thinner CdTe absorber. It is expected to obtain stable solar cells with high efficiency using the CdTe absorber and $Cd_{1-x}Zn_xS$ buffer layer. The thicknesses are fixed at about 1 µm for the CdTe absorber layer and about 50 nm of CdS and $Cd_{1-x}Zn_xS$ buffer layers. Fig. 2 presents the J-V characteristics

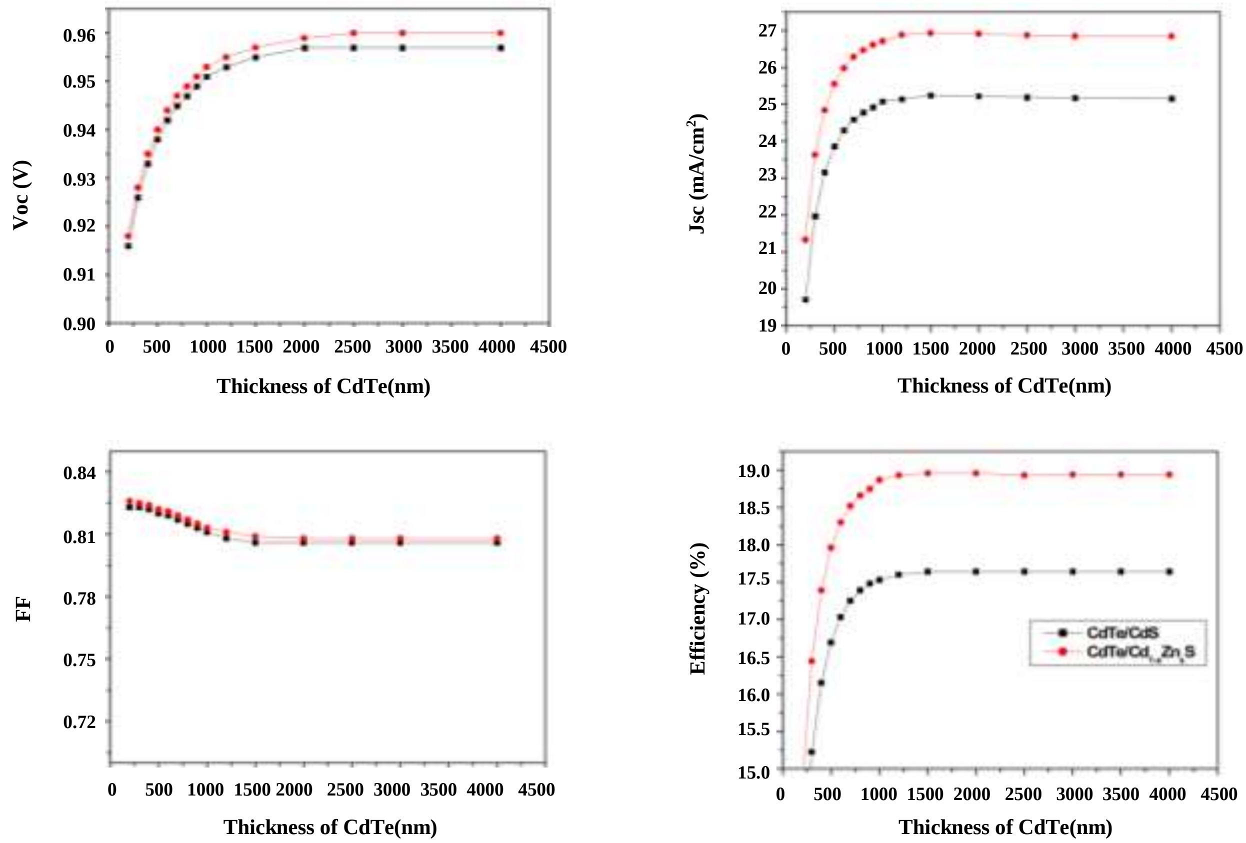

Fig. 3: Variation of photovoltaic parameters as a function of CdTe absorber thickness.

of CdTe solar cells with CdS and $Cd_{1-x}Zn_xS$ buffer layers. The curves show a better J_{sc} for CdTe/$Cd_{1-x}Zn_xS$ solar cell of about 26.71 mA/cm^2. The cell with a CdS buffer layer presents only 25.02 mA/cm^2 J_{sc}. The V_{oc} is almost unchanged for both cells at about 0.95 Volt. It is known that the efficiency is improved whenever the increase appears either in the current or in the voltage or the two of them.

To calculate the optimum absorber thickness, CdTe absorber layer thickness is varied from 200 nm to 4000 nm using the CdS buffer layer. In the modified structure, the $Cd_{1-x}Zn_xS$ alternative buffer layer has been replaced, and the solar cell structure consists of ZnO-Al/$Cd_{1-x}Zn_xS$/CdTe/Ag. Ag back-contact material is used with a barrier height (Φ_{bL}) of 1.35 eV for both baseline and modified cells [4]. Fig. 3 demonstrates the variation of the CdTe absorber thickness and their influence on the photovoltaic parameters. As comprehended in Fig. 3, the three photovoltaic parameters: efficiency, $V_{oc,}$ and J_{sc} are increasing with the increase of CdTe absorber layer thickness up to 1 µm. After 1 µm, the output parameters remain unaffected with increasing in CdTe thickness. However, the *FF* shows a little decrease before 1.5 µm due to the reduction of bulk resistance of CdTe material. This indicates that the selection of 1 µm of CdTe absorber thickness with CdS and Cd_{1-x} Zn_xS layers is the best. On the other hand, the use of the $Cd_{1-x}Zn_xS$ buffer layer improves all the solar cell output parameters, where a significant improvement of 6.33% was forJ_{sc} and hence an improvement of 7.86% in conversion efficiency. Note that at 1 µm CdTe layer-thick and with $Cd_{1-x}Zn_xS$ buffer layer, the conversion efficiency of 18.83% is obtained with V_{oc} = 0.95 Volt, J_{sc} = 26.71 mA/cm^2 and *FF* = 0.81, however with CdS buffer layer only a efficiency of 17.53% is obtained with V_{oc} = 0.95 Volt, J_{sc} = 25.02 mA/cm^2, *FF* = 0.81.

The Quantum Efficiency (QE) measurement of a solar cell is an indicator of the converting sunlight to electricity effectiveness and then is computed by the ratio of the number of carriers collected by the solar cell structure to the number of photons of a given wavelength on the solar cell structure [34]. Therefore, Fig. 4 depicts the variation

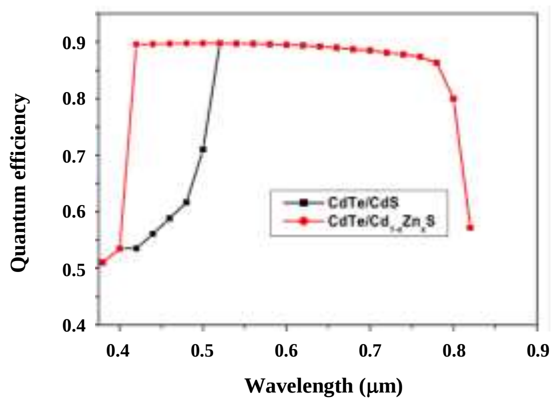

Fig. 4: Comparison of the quantum efficiency of the cell with CdS and $Cd_{1-x}Zn_xS$ buffer layers.

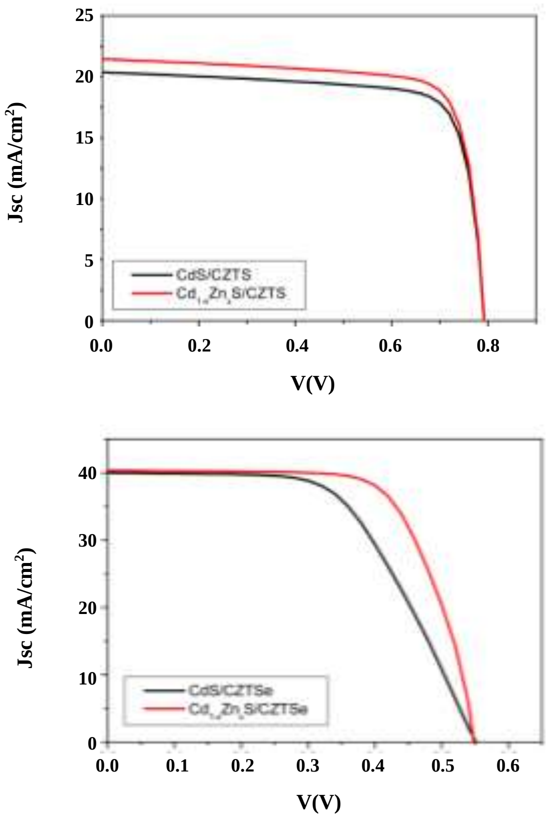

Fig. 5: J-V characteristics of CZTS and CZTSe solar cell structures.

of the QE asa function of wavelength with 1µm of CdTe absorber thickness. The cell with $Cd_{1-x}Zn_x$Sbuffer layer shows a better QE up to a wavelength of 520 nm. After this value, the quantum efficiency remains the same for the proposed and conventional structures. This indicates that $Cd_{1-x}Zn_xS$ material has a good absorbance than the CdS material in the wavelength region from 400 to 520 nm.

Case ofCZTS and CZTSe thin film solar cells

The SCAPS simulation program has been employed to study the current density-voltage (*J-V*) characteristics and related output parameters of conventional ZnO/CdS/CZTS/Mo and ZnO/CdS/CZTSe/Mo solar cells with the proposed $Cd_{1-x}Zn_xS$ buffer layer. An evident improvement is seen in the simulated *J-V* characteristics and shown in Fig. 5. This improvement is drawn mainly from J_{sc} in the case of the CZTS solar cell and *FF* in the case of the CZTSe solar cell. The improvement in the performance of the solar cell is obtained using the $Cd_{1-x}Zn_xS$ buffer layer. To get more in-depth, the thickness of the absorber layer is varied between 0.2 µm to 2.5 µm for the CZTS absorber layer and from 0.2 µm to 1.5 µm for the CZTSe absorber layer. The results obtained from the SCAPS simulation are shown in Fig. 6 and Fig 7.

For CZTS solar cells, it is clearly evident from Fig. 6 that the efficiency, J_{sc}, *FF*, and V_{oc} increase with increasing CZTS layer thickness. This overall enhancement may be due to the large absorber thickness when more electron-hole pairs are generated and consequently improves the conversion efficiency of the structure [4]. The cell using the $Cd_{1-x}Zn_xS$ buffer layer presents an efficiency of 13.23%, while the cell with the CdS layer presents only an efficiency of 12.53% with 2.5 µm of CZTS absorber thickness. This decent enhancement is estimated at a 5.29%, and may be due to the good absorbance of $Cd_{1-x}Zn_xS$ buffer layer than the CdS buffer layer in the wavelength region from 400 to 520 nm, as depicted by the QE in Fig. 5. Where this means that more photons are absorbed and hence more electron-hole pairs are generated and collected which consequently improves the conversion efficiency of the structure. Alternatively, this 5.29% enhancement may also be due to the reduction in the carrier recombination in the $Cd_{1-x}Zn_xS$/CZTS interface region.

For CZTSe solar cells, the conversion efficiency increases from 12.61% to 15.35% using only 1 µm of CZTSe absorber thickness and with the insertion of $Cd_{1-x}Zn_xS$ buffer layer (Fig. 7). Therefore, an optimal enhancement in efficiency is estimated at a19.22%whichmay be due to the good absorbance of $Cd_{1-x}Zn_xS$ buffer layer than the CdS buffer layer in the wavelength region from 400 to 520 nm as mentioned before. In addition,

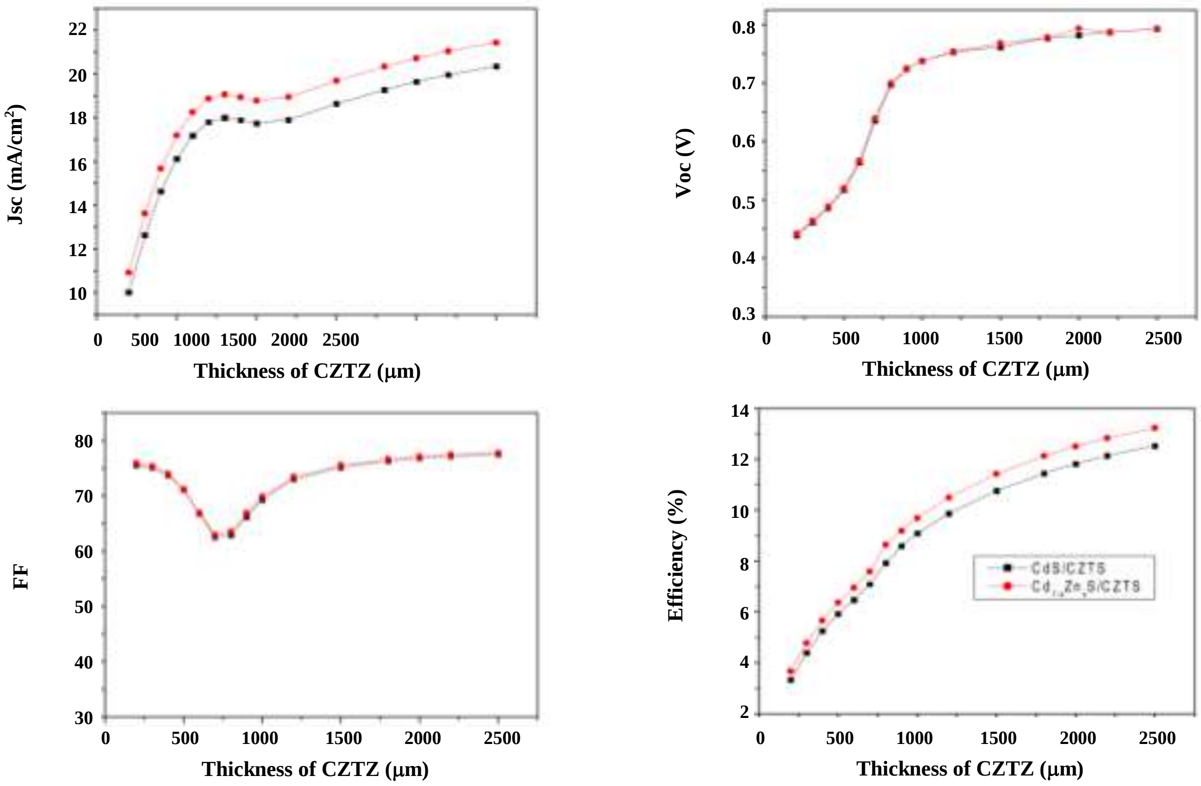

Fig. 6: Variation of photovoltaic parameters as a function of CZTS absorber thickness.

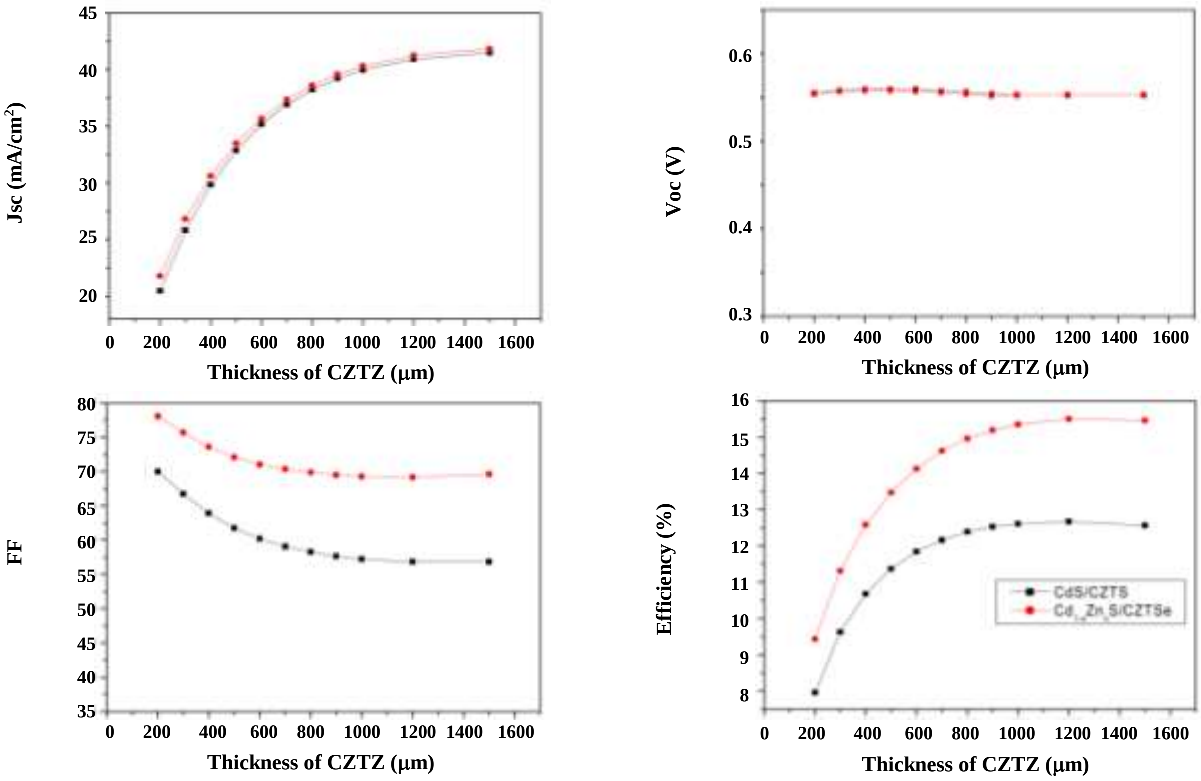

Fig. 7: Variation of photovoltaic parameters as a function of CZTSe absorber thickness.

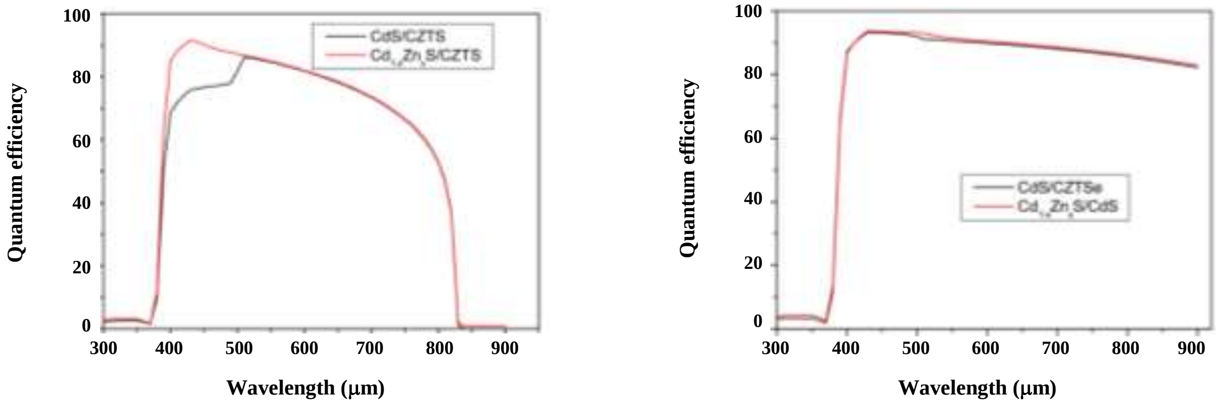

Fig. 8: Spectral response of CZTS and CZTSe solar cells.

by comparing the obtained efficiency of 15.35% for 1 µm of CZTSe absorber thickness with the obtained efficiency of 13.23% for 2.5 µm of CZTS absorber thickness (Fig. 6) with the insertion of $Cd_{1-x}Zn_xS$ buffer layer, it is better to use the CZTSe absorber layer with $Cd_{1-x}Zn_xS$ layer instead of CdS layer for profit in conversion efficiency and profit in the amount of material used in solar cell structure. On the other hand, the decrease of J_{sc} and *FF* with increasing the absorbent layer thickness is due to the reduction of bulk resistance for CZTS material [9]. As a comparison, the $Cd_{1-x}Zn_xS$/CZTS cell structure presents 0.79 Volt and 21.44 mA/cm^2 of V_{oc} and J_{sc}, respectively, with 2.5 µm CZTS absorber-thick, while $Cd_{1-x}Zn_xS$/CZTSe cell structure presents 0.55 Volt and 40.32 mA/cm^2 of V_{oc} and J_{sc}, respectively, with only 1 µm CZTSe absorber-thick.

The Quantum Efficiencies (QE) of the cells are obtained in the wavelength range of 300 nm to 900 nm for the above-mentioned structures and shown in Fig. 8, indicating a significant improvement of QE in the case of $Cd_{1-x}Zn_xS$/CZTS structure in the short wavelengths range, while no improvements are observed in the case of $Cd_{1-x}Zn_xS$/CZTSe structure.

As an indicator of the stability of solar cell devices, the researchers deliberately checked the dependence of V_{oc} and hence efficiency to the operating temperature [35]. Thus, to testify to this vital dependence relation on the studied hetero-structures, we varied the operating temperature from 300 to 400 K and we have shown results of three structures in Fig. 9. From the shown curves, it found that the conversion efficiency decreases linearly when the operating temperature increased from 300K to

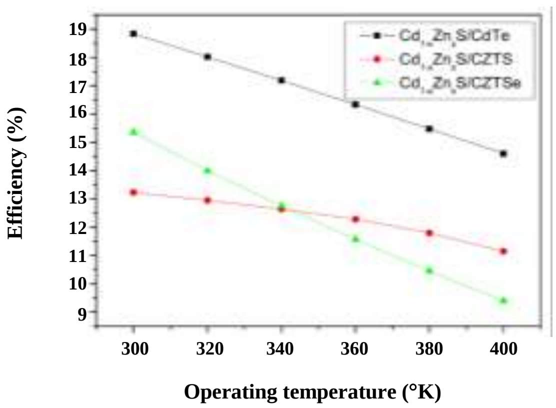

Fig. 9: Effect of operating temperature on the performances of the cells.

for three structures, while in terms of stability the $Cd_{1-x}Zn_xS$/CZTS have shown better stability compared to the other cells [18].

In the present numerical study, it has demonstrated that decent conversion efficiency of 15.35% and 18.83% can be obtained by applying the $Cd_{1-x}Zn_xS$ material as a buffer layer in CZTSe and CdTe TFSCs respectively, which will support the researchers in the experimental field to make further development. For example, *Clayton et al.* reported an efficiency of about 8.8% for Cd1-xZnxS/CdTe TFSC heter-structure produced by MOCVD method [36]. Consequently, all of the results in this study support the scientific knowledge that the gap energy of semiconductors such as a wide band-gap $Cd_{1-x}Zn_xS$ semiconductor represents fundamental physical aspects that characterize their optical and electronic properties. The photovoltaic properties such as efficiency, V_{oc}, J_{sc}

and *FF* also aids in the performance assessment of band gap engineering structures for continuous and optimal absorption of broad-band spectral sources.

CONCLUSIONS

A comparative study of solar cells performances based on CdTe and Kesterite (CZTS, CZTSe) compounds, by inserting a novel buffer layer $Cd_{1-x}Zn_xS$ has been carried out using AMPS-1D and SCAPS-1D simulation software. Optimum thicknesses of CdTe, CZTSe, and CZTS layers have been obtained at 1 µm, 1 µm, and 2.5 µm, respectively. By using the novel buffer layer, $Cd_{1-x}Zn_xS$ enhances the conversion efficiency of about 18.83%, 15.35%, 13.23% for CdTe, CZTSe, and CZTS solar cells, respectively, instead of 17.53%, 12.53%, and 12.61% by using a conventional buffer layer, CdS, respectively. These values were obtained by using 50 nm of $Cd_{1-x}Zn_xS$, 200 nm of ZnO, 1 µm of CdTe, CZTSe, and 2.5 µm of CZTS. These results provide valuable insight, guidance, paths, and windows of opportunity in setting up future PV designs for cleaner and sustainable energy production.

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