Moroccan Journal of Chemistry
ISSN: 2351-812X
http://revues.imist.ma/?journal=morjchem&page=login
Olawale & al. / Mor. J. Chem. 8 N°3 (2020) 673-682

PERFORMANCE ANALYSIS OF COPPER ZINC TIN SULFIDE, Cu_2ZnSnS_4 (CZTS) WITH VARIOUS BUFFER LAYERS BY USING SCAPS IN SOLAR CELLS

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Received 02 Jan 2020,
Revised 13 May 2020,
Accepted 28 Jun 2020

Abstract

Numerical modeling has become an essential tool for scientists and engineers as it enhances the ability to understand certain device properties and several processes that occur in solar cell operation. The performance of CZTS thin film based solar cell was investigated with the aid of a simulation program called Solar Cell Capacitance Simulator (SCAPS 1-D) while varying the thickness of the absorber layer. CZTS semiconductors serves as the absorber layer in the cell structure, Al:ZnO/ZnO was utilized as the front contact and CdS/In_2S_3/ZnSe/SnS_2/ as the window layer. Observation shows that ZnSe produced the most efficient cell of 13.79% (with Voc = 0.7289V, Jsc = 22.77mA/cm^2, FF= 83.12%). Also, Temperature, carrier concentration and thickness of layers all play an important role in performance of the cell.

Keywords: *Cu_2ZnSnS_4, Carrier Concentration, Solar Cell, SCAPS*

1.0 INTRODUCTION

The interest of the quaternary kësterite Cu_2ZnSnS_4 (CZTS) for solar cells have greatly increased over the last decade.[1] Its preference owns to the non-toxic nature of the elements and the abundance of the constituent elements. It is also worthy of note that band gap and the absorption coefficient make it an interesting alternative compared to the conventional CIGS solar cell. In improving the performance of a photovoltaic solar cell, it is expedient to optimize the absorber/buffer interface involving hetero-junction structure. Due to the complex nature of heterostructure like CZTS, numerical simulation serves as an efficient approach to predict the effect of changes of different electrical & optical properties on the output performance.[2] In this work, SCAPS (solar cell capacitance software) is utilized as our simulation software.

2.0 Material and Methods

The relationship between potential and space charges can be described using Poisson's equation.

$$\frac{\partial^2}{\partial x^2}\varphi(x) = \frac{q}{\varepsilon}$$

where φ is the potential, q is the elementary charge, ε is the permittivity, n is the density of free electron, p is the density of free hole, N_D^+ is the ionized donor-like doping density, N_A^- is the ionised acceptor-like doping density, p_t is the trapped hole density, n_t is the trapped electron density.

These sets of the equation below is known as continuity equations and it defines the transportation of carriers

$$\begin{cases} q\frac{\partial n}{\partial t} = \frac{\partial J_n}{\partial t} + qG - qR \\ q\frac{\partial p}{\partial t} = \frac{-\partial J_p}{\partial x} + qG - qR \end{cases} \qquad \begin{cases} J_n = qn\mu_n\frac{\partial \varphi}{\partial x} + qD_n\frac{\partial n}{\partial x} \\ J_p = -qp\mu_p\frac{\partial \varphi}{\partial x} + qD_p\frac{\partial p}{\partial x} \end{cases}$$

where G is the optical generation rate, R is the recombination rate, D_n is the electron diffusion coefficient, D_p is the hole diffusion coefficient, μ_n is the electron mobility, and μ_p is the hole mobility.

2.1 Device structure & simulation:

SCAPS was developed at University of Gent, Belgium. It is a one-dimensional solar cell simulation program. Marc Burgelman, Alex Niemegeers, Koen Decock, Johan Verschraegen, Stefaan Degrave have contributed to the development of this software.[3] The program was designed to simulate comprehensively several characteristics of thin-film heterojunction solar cells. CdTe thin-film and CIGS family have had initial test using the software. It has been observed that there are good agreements between the experimental results and the simulated results using SCAPS, it thus motivated us to use the simulation tool in this work.[6] The software has an interface where we input different physical parameters for each layer in order to analyze the overall cell performance. In this study the following configurations were considered: [ZnO:Al/iZnO/CdS/CZTS],[ZnO:Al/iZnO/SnS2/CZTS],[ZnO:Al/iZnO/In_2S_3/CZTS] and [ZnO:Al/i-ZnO/ZnSe/CZTS] for a comparison study of Kesterite based thin film photovoltaic devices. The configuration, Al:ZnO/ZnO/CdS/CZTS/back contact, is considered for the simulation of single junction solar cells with CZTS as the absorber layer material. Al:ZnO serves as the front electrical contact and simultaneously serves as invisible or transparent to the incident solar radiation. ZnO layer serves as the window layer. This layer acts as a passivation layer and n-type CdS layer acts as the buffer layer, making a heterostructure p-n junction with p-type CZTS absorber layer. In this simulation, at each interface, carrier transport via tunnelling mechanism is considered coupled with the effective mass. The material parameters that are required for the present simulation studies are either gotten from literature, cited wherever used or assumed with rationale for better

understanding of the device performance under realistic situations. Auger recombination is relatively neglected for this study since it is significant only at higher carrier concentrations.

Table 1: Material Properties used for simulation

Parameter	CZTS	CdS	ZnO	ZnS	In_2S_3	ZnSe	i:ZnO	ZnO:Al
Thickness(µm)	2.00	0.050	0.80	0.050	0.50	0.50	0.80	0.020
Band gap(ev)	1.50	2.42	3.37	3.50	2.80	2.90	3.37	3.37
Electron affinity(eV)	4.3	4.50	4.6	4.5	4.70	4.10	4.6	4.6
Dielectric Permittivity	6.950	9.00	9.00	10.00	13.50	10.00	9.00	9.00
CB effective density of states(cm^{-3})	2.200E+18	1.800E+19	2.200E+18	1.800E+19	2.2 E+17	1.800E+18	2.200E+18	2.200E+18
VB effective density of states (cm^{-3})	1.800E+19	2.40E+18	1.800E+19	2.400E+18	1.800E+19	1.800E+19	1.800E+19	1.800E+19
Electron thermal velocity (cms^{-1})	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7
Hole thermal velocity (cms^{-1})	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7
Electronmobility(cm^2/V_s)	1.00E+2	1.600E+2	1.50E+2	1.600E+2	1.600E+2	1.600E+2	1.50E+2	1.50E+2
Hole mobility(cm^2/V_s)	3.500E+1	5.00E+1	2.50E+1	5.00E+1	5.00E+1	5.00E+1	2.50E+1	2.50E+2
Shallow uniform donor density, N_D (cm^{-3})	0.00	1.00E+17	1.00E+17	1.00E+17	1.00E+14	5.00E+18	1.00E+17	1.00E20
Shallow uniform donor density, N_A(cm^{-3})	5.00E+16	0.00	0.00	0.00	0.0	0.00	0.00	0.00
Defect type at bulk/interface	Donor/Neutral							
Radiative recombination coefficient[cm /s]	1.04 x 10^{-10}	1.04 x 10^{-10}	1.04 x 10^{-10}	1.04 x 10^{-10}	1.04 x 10^{-10}	1.04 x 10^{-10}	1.04 x 10^{-10}	1.04 x 10^{-10}
Effective mass electron	0.18[8]	0.25[7]	0.275[7]	0.25	0.25	0.25	0.275[7]	0.275[8]
Effective mass hole	0.71[8]	0.7[7]	0.59[7]	0.7	0.7	0.7	0.59[7]	0.59[8]
Hole capture cross section (cm 2)	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15} [8]
Electron capture cross section (cm 2)	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}	1x 10^{-15}
Interface recombination speed (cm/s)	10^4 [9]							
Absorption coefficient [cm eV]	file[6]	SCAPS	SCAPS	1.060E+4	1.00E+5		SCAPS	

In simplifying the calculation, capture cross section and thermal velocity for electron and holes are standardized to 10^{-15} cm ^{2} and 10^7 cm/s respectively. The minority carrier life time at the bulk of absorber layer and the interface recombination speed at absorber/buffer layer interface are taken 10 ns, 5 ns, and 10^4 cm s^{-1} ,10^3 cms^{-1} for CZTS [9].

Band to band recombination is considered as well for the bulk materials and also considered for radiative recombination with recombination coefficient equal to 1.04 x10 $^{-10}$ cm^3 s^{-1} for all the layers in the simulation[10]. In this study, illumination was set to 1000 W/m^2, temperature to 300k and air mass of 1.5 global spectrum were considered for simulation. The device and material parameters used in the simulation are all cited from experimental study, literature values or in some cases reasonable estimation

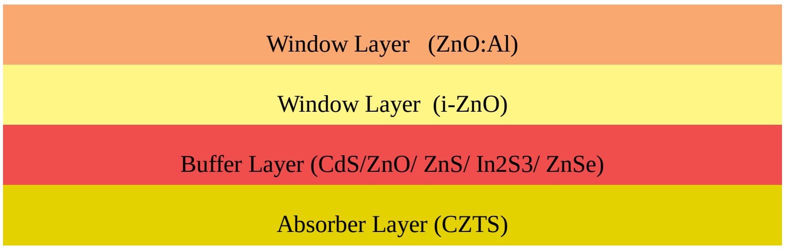

Figure 1: Device structure of CZTS solar cell

Effect of Absorber layer thickness against Efficiency of the cell

The high absorption rate of CZTS makes it a good material for solar cell application. CZTS thickness varies from 500nm to 3000nm. It is observed that as there is increase in thickness, the efficiency increases along with Open circuit voltage (V_{oc}), Current density (J_{sc}) and Fill factor (FF). 2000nm is considered good for the thickness of this layer. The incoming solar radiation will be absorbed conveniently due to the high absorption coefficient of CZTS and its direct band gap. The thicker absorber layer will absorb more photons with longer wavelength, which will in turn make a contribution to the generation of electron-hole pairs. This account for the increased efficiency resultant from increased thickness.

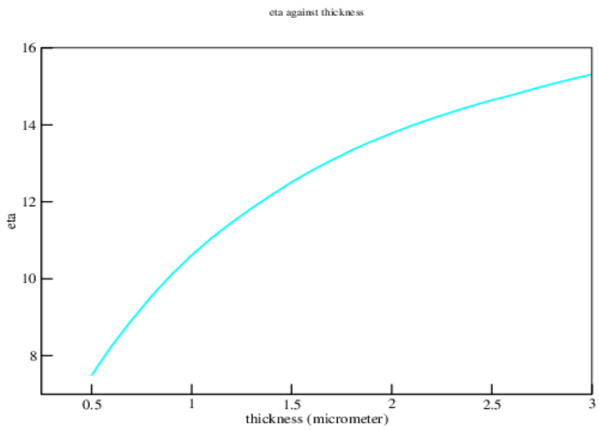

Figure 2: Plot of efficiency against thickness

Effect of Temperature against efficiency of the cell

Temperature was varied and the resultant effect on the cell has been studied. It was observed that increase in temperature had a negative effect on the efficiency of the cell. Increased temperature resulted to low efficiency. This may be due to the fact that certain spectrum of the incoming solar radiation is required by the cell, so exposing it to more than required solar radiation may only heat up the cell without causing any more carriers to be generated.

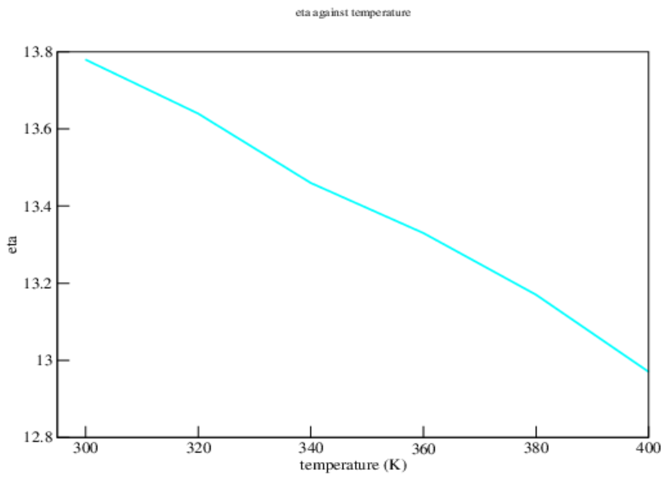

Figure 3: Plot of efficiency against temperature

Effect of buffer layers against efficiency

In this study, some selected buffer layers were considered namely CdS, In_2S_3, ZnSe and SnS_2.. The thickness was varied from 0.010 µm to 0.100µm. The performance of a solar cell depends on the band alignment at the heterointerface.[1] A thin buffer layer may result to leakage in current and too thick layer may also result to low carrier separation rate.

(a) CdS as Buffer Layer

CdS is a commonly used buffer layer. The toxicity of cadmium poses a disadvantage to the use of Cadmium Sulphide as a buffer layer, alternative element is sourced for to be used in the buffer layer.

CdS with wide bandgap of ~2.42 eV allows the maximum photo-absorption in the absorber layer. Thickness of a buffer layer is determined basically by its conductivity. The higher the conductivity of buffer layer, the more the penetration of depletion region towards the absorber side and thus the higher device efficiency. In this study, it was observed that increase in the thickness of the layer resulted into increase in efficiency of the cell. At 0.050, the increase is not high appreciably so it recommended that the thickness of the layer is 0.050µm.

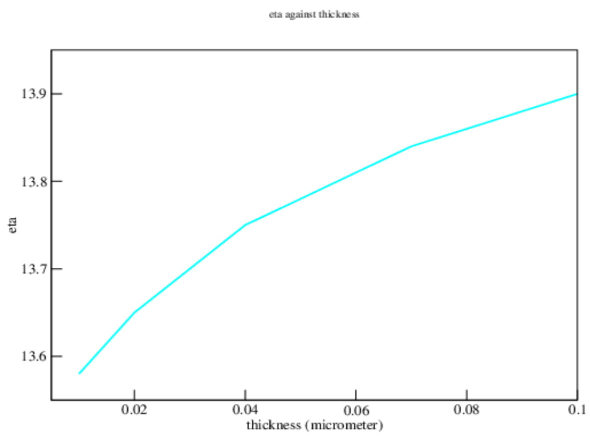

Figure 4: Plot of efficiency against thickness

Table 2: Result of variation of thickness of CdS as buffer layer

Thickness (µm)	Voc	Jsc	FF	Eta
0.010	0.7290	22.589598	82.46	13.58
0.020	0.7291	22.650930	82.65	13.65
0.030	0.7291	22.708123	82.76	13.70
0.040	0.7292	22.762437	82.81	13.75
0.050	0.7293	22.812829	82.83	13.78
0.060	0.7293	22.856950	82.85	13.81
0.070	0.7294	22.894524	82.87	13.84
0.080	0.7294	22.926736	82.88	13.86
0.090	0.7295	22.954688	82.90	13.88
0.100	13.88	22.978974	82.91	13.90

(b) SnS_2 as Buffer Layer

An alternative material considered for the buffer layer is SnS2. It has a bandgap of ~1.559eV. It forms a p-n junction with the absorber layer. It has been observed that as the thickness increases efficiency, J_{sc}, FF declines.

It is best to use a thickness of less than 0.10 µm.

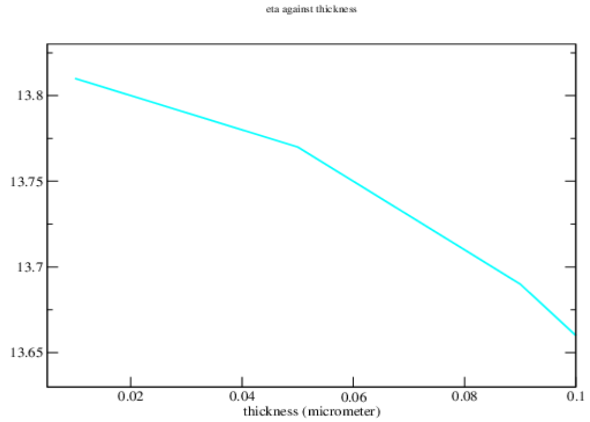

Figure 5: Plot of efficiency agaainst thickness

Table 3: Result of variation of thickness of SnS_2 as buffer layer

Thickness (µm)	V_{oc}	J_{sc}	FF	Eta
0.010	0.7286	22.761793	83.27	13.81
0.020	0.7286	22.748234	83.27	13.80
0.030	0.7286	22.733978	83.27	13.79
0.040	0.7286	22.715988	83.28	13.78
0.050	0.7286	22.693557	83.28	13.77
0.060	0.7286	22.666432	83.28	13.75
0.070	0.7286	22.634552	83.28	13.73
0.080	0.7286	22.597964	83.28	13.71
0.090	0.7286	22.556791	83.28	13.69
0.100	0.7286	22.511211	83.28	13.66

(c) In_2S_3 as Buffer Layer

An alternative material considered for the buffer layer is In_2S_3. It has a bandgap of ~0.835eV. It forms a p-n junction with the absorber layer. It is observed that as the thickness increases, efficiency, Jsc, FF declines.

It is best to use a thickness of less than 0.10 µm.

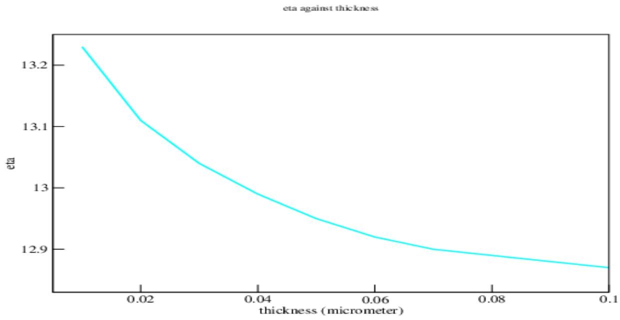

Figure 6: Plot of efficiency against thickness

Table 4: Result of variation of thickness of In_2S_3 as buffer layer

Thickness (µm)	Voc	Jsc	FF	Eta
0.010	0.7295	22.498320	80.59	13.23
0.020	0.7297	22.475363	79.96	13.11
0.030	0.7298	22.453397	79.57	13.04
0.040	0.7298	22.432561	79.32	12.99
0.050	0.7299	22.413104	79.16	12.95
0.060	0.7299	22.394389	79.06	12.92
0.070	0.7299	22.376656	79.00	12.90
0.080	0.7300	22.359949	78.96	12.89
0.090	0.7300	22.344171	78.95	12.88
0.100	0.7300	22.329466	78.95	12.87

(d) ZnSe as Buffer Layer

An alternative material considered for the buffer layer is ZnSe. It has a bandgap of ~1.166eV. It forms a p-n junction with the absorber layer. It is observed that the as the thickness increases, the efficiency, Jsc, FF decline.

It is best to use a thickness of less than 0.10 µm.

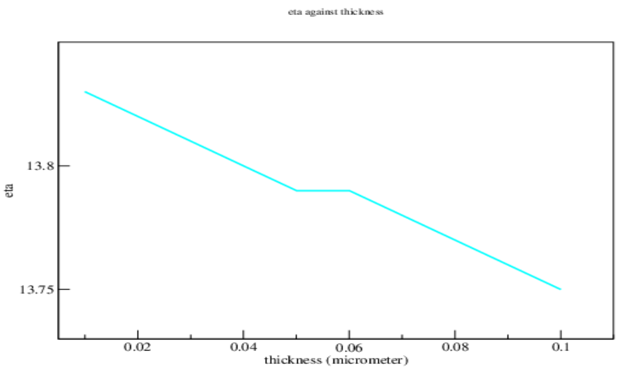

Figure 7: Plot of efficiency against thickness

Table 4: Result of variation of thickness of ZnSe as buffer layer

Thickness (µm)	Voc	Jsc	FF	Eta
0.010	0.7290	22.842662	83.03	13.83
0.020	0.7289	22.817505	83.12	13.82
0.030	0.7289	22.799193	83.12	13.81
0.040	0.7289	22.784401	83.12	13.80
0.050	0.7289	22.770419	83.12	13.79
0.060	0.7289	22.756485	83.12	13.79
0.070	0.7288	22.742248	83.12	13.78
0.080	0.7288	22.727513	83.12	13.77
0.090	0.7288	22.712179	83.12	13.76
0.100	0.7288	22.696165	83.12	13.75

Impact of absorber layer acceptor concentration

CZTS is an intrinsic p-type photovoltaic absorber material, where carrier concentration relies on the non-stoichiometry and defects present in the synthesized material. The carrier concentration is varied from 5 x10^{12} cm^{-3} to 5 x10^{18} cm^{-3} for the absorber layer. There's a steady increase in the open circuit voltage, V_{oc}, with increasing carrier concentration. J_{sc} decreases as the carrier concentration increase.

Effect of absorber layer band gap on efficiency of the cell

CZTS has a reported efficiency between (1.4 – 1.6) eV. In this study, we tried to vary the band gap from 1.48 – 1.60 all with thickness of 0.050µm. It is observed that with increase in band gap, there is a reduction in efficiency and J_{sc} of the cell. An optimized value of 1.48 is recommended.

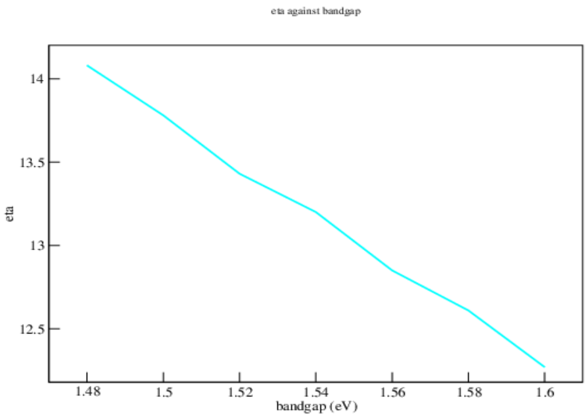

Figure 8: Plot of efficiency against bandgap

J-V Characteristics

Finally, based on the best possible optimization, the J-V characteristics for different solar cells have been simulated.

Figure 9(a) CdS

Figure 9(b) In_2S_3

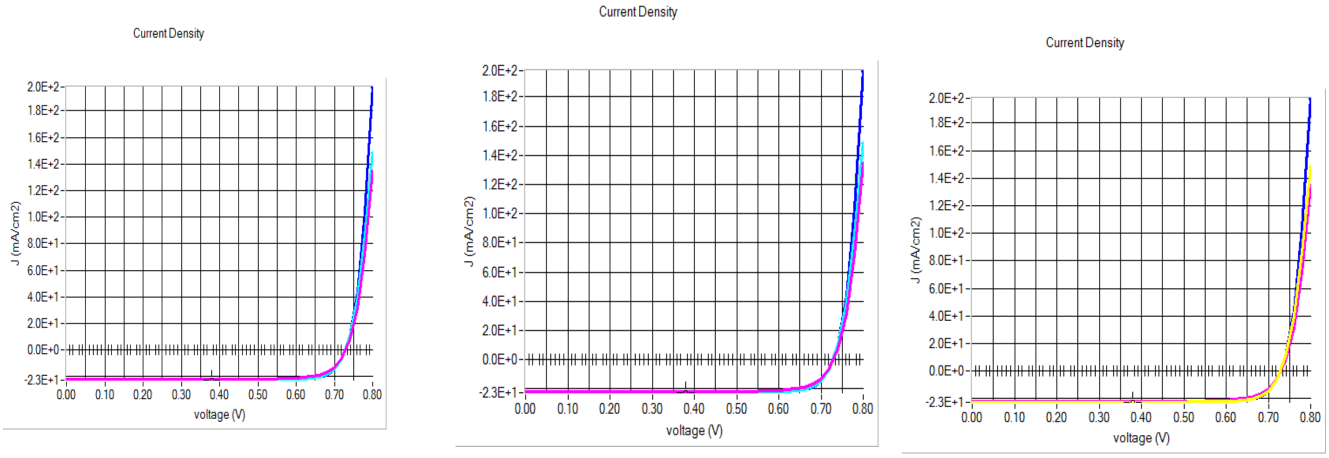

Figure 9(c) ZnSe

Table 5: CZTS absorber based photovoltaic cell performance parameters

Thin Film Solar Cell	Efficiency(%)	Fill Factor (%)	J_{sc} (mA/cm)	V_{oc}(V)
ZnO:Al/i-ZnO/CdS/CZTS	13.78	82.83	22.812829	0.7293
ZnO:Al/i-ZnO/In_2S_3/CZTS	12.95	79.16	22.413104	0.7299
ZnO:Al/i-ZnO/ZnSe/CZTS	13.79	83.12	22.770419	0.7289
ZnO:Al/i-ZnO/SnS_2/CZTS	13.77	83.28	22.693557	0.7286

In obtaining these results, the optimal values considered in this paper are, absorber layer thickness of 2 µm, buffer layer thickness of 0.050 µm and absorber carrier density of 5×10^{17} cm^3. Simulating all the parameters it has been found that the solar cell with ZnS buffer layer has the highest efficiency (efficiency= 13.79%, FF= 83.12%, J sc = 22.770419 mA/cm 2 and V oc = 0.7289V) among all other considered buffer layers.

Conclusion

Keskerite CZTS solar cell is investigated using one dimensional SCAPS 1D solar cell simulator. The numerical simulation has been done via varying the absorber layer thickness, buffer layer thickness, doping concentration of absorber layer, varying temperature and varying band gap. In obtaining a better performance, optimization of the parameters was done and considered for J-V characteristics. Solar cell with ZnSe as the absorber layer shows the best performance (eficiency= 13.79%, FF= 83.12%, Jsc = 22.770419 mA/cm^2 and Voc = 0.7289V).

The above configuration can lead to developing higher efficiency CZTS thin film solar cells.

Acknowledgement

The authors acknowledge Dr. Marc Burgelman at the University of Gent. We would like to thank him for providing SCAPS.

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