Scientific Research Publishing

Advances in Materials Physics and Chemistry, 2022, 12, 207-219
https://www.scirp.org/journal/ampc
ISSN Online: 2162-5328
ISSN Print: 2162-531X

High Performance for Cu(In,Ga)Se$_2$ Quaternary System-Based Solar Cells with Alternative Buffer Layers

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How to cite this paper: Oubda, D., Kébré, M.B., Ouédraogo, S., Diasso, A., Zougmoré, F., Koalga, Z. and Ouattara, F. (2022) High Performance for Cu(In,Ga)Se$_2$ Quaternary System-Based Solar Cells with Alternative Buffer Layers. *Advances in Materials Physics and Chemistry*, **12**, 207-219.
https://doi.org/10.4236/ampc.2022.129015

Received: August 9, 2022
Accepted: September 26, 2022
Published: September 29, 2022

Abstract

In this study, the authors investigated the performance of different buffer layers through the electrical parameters such as J_{sc}, V_{oc}, *QE* and η of the quaternary system Cu(In,Ga)Se$_2$ solar cells. The performance of Cu(In,Ga)Se$_2$ solar cells has been modeled and numerically simulated by using the SCAPS-1D device simulation tool. The cells with a ZnSe, Zn(O,S) and (Zn,Mg)O buffer layers were compared with the reference CdS buffer layer. The investigation of ZnSe, Zn(O, S) and (Zn,Mg)O-based cells to substitute the traditional CdS in the future shows that the ZnSe-buffer layer is a potential material to replace CdS, which revealed the best efficiency of 20.76%, the other electrical parameters are: J_{SC} = 34.6 mA/cm^2, V_{OC} = 0.76 V and FF = 79.6%. The losses as a function of the temperature are estimated at 0.1%/K, among all kinds of buffer layers studied. We have also shown that the use of a high band-gap buffer layer is necessary to obtain a better short-circuit current density J_{SC}. From our results, we note that the chalcogenide solar cells with Zn-based alternative buffer layer have almost the same stability that the traditional CdS buffer layer solar cells have.

Keywords

Thin Film Solar Cells, CIGS Absorber, Alternative Buffer Layers, SCAPS-1D, Electrical Parameters

1. Introduction

Cu(In,Ga)Se$_2$(CIGS) quaternary system-based photovoltaic (PV) cells have demonstrated their potential to achieve the highest solar energy conversion efficiencies ever for thin film devices and they also exhibit excellent stability [1] [2]. Recently, $Cu(In,Ga)Se_2$ thin film solar cells with conversion efficiency of 23.35% have been reported in literature for laboratory cells [3], and commercial modules are now fabricated by large quantities [4]. Thin film solar cells with $Cu(In,Ga)Se_2$ chalcopyrite absorber layers have attracted significant research interest as an important light-to-electricity converter with widespread commercialization prospects.

Several factors are capable of improving the performance of $Cu(In,Ga)Se_2$ chalcopyrite solar cells among which we note, the thickness of the layer (absorber, buffer, windows), the graduation of the band gap, the affinity, the series, and the shunts resistances and the nature of the buffer layer. In addition, the conduction band offset and the valence band offset of CdS/CIGS layers are among the most important factors of the polycrystalline $Cu(In,Ga)Se_2$ solar cells with a CdS buffer layer leading to high performances [5]. Unfortunately, the cadmium present in the buffer layers of cadmium sulfide is pointed by many researchers to be toxic [4]. Therefore, these studies are moving towards the replacement of the CdS buffer layers. This is why we study the buffer layers in order to optimize the performances of CIGS solar cells and to fight against pollution due to the presence of toxic chemical material.

Several alternative materials have been investigated as buffer layers to replace the traditional CdS. We notice between other, ZnO, ZnS, ZnSe, (Zn,O)S, (Zn,Mg)O, SnO_2, $Sn(S,O)_2$ and ZrO_2 [6] [7] [8] [9] [10].

The aim of this work is to show the performances of certain Zn-based buffer layers capable to replace effectually the reference CdS buffer layer.

In this paper, we are interested in the study of the chalcogenide Mo/CIGS/buffer layer/ZnO structure with the following alternative buffer materials ZnSe, Zn(O,S), (Zn,Mg)O. We compare the results obtained with those reference CdS buffer layer.

In the numerical simulation, we investigate the current-voltage characteristic (J-V) and the quantum efficiency QE (λ) that better characterize the solar cell. We also analyse the energy band diagram.

2. Material and Method

2.1. Structure of CIGS Solar Cells

The structure of these solar cells (**Figure 1**) and the importance of each layer are described in [11]. Also, the so-called transportation electrical charge equations in the semiconductor, and regulating the functioning of these solar cells kind are presented [11].

2.2. SCAPS-1D

SCAPS-1D is a software application on Windows, it is the work of the team of Marc Burgelman of the department of electronics and information systems (ELIS), of the University of Gent in Belgium.

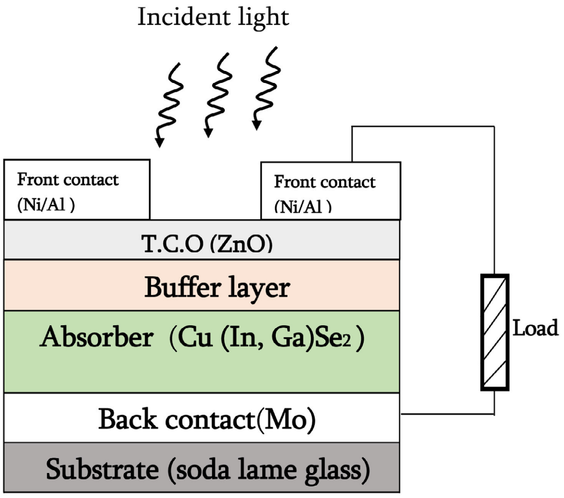

Figure 1. CIGS solar cells structure.

SCAPS-1D is one-dimensional numerical simulation software that was developed to obtain the electrical characteristics of heterojunction and thin-film solar cells. Its development is inspired by work on solar cells based on CdTe and Cu(In,Ga)Se_2 [12] [13]. The simulated results are in excellent agreement with the measured results [14] [15], but also with other previously developed software such as AMPS-1D [16].

SCAPS-1D is interested in the most sensitive characteristics of the solar cells namely: J-V, C-V, C-f, and QE (λ) under illumination and in darkness [12].

When the desired simulation is complete, the energy band diagram, the generation-recombination profile and the curves of the current-voltage density, capacitance-voltage, capacitance-frequency, and quantum efficiency characteristics of the solar cell can be viewed on the screen [15]. The numerical data of its electrical quantities can also be extracted.

Available free of charge for research into thin-film photovoltaic solar cells in general and CdTe and CIS/CIGS-based solar cells in particular, SCAPS-1D is a very attractive software for optimizing the performance of PV solar cells. Thus, we retained this software for the simulation of our solar cell model.

2.3. Alternative Buffer Layers

In this part, we present the physico-chemical and optoelectrical properties of different alternative buffer layers. Presently, in the field of the chalcogenide thin-film, the best solar cells in the point of view of conversion efficiency and stability are elaborate with the traditional CdS buffer layers. Unfortunately, the gap of 2.4 eV of the CdS layer seems to be weak and stops a good part of the photons of short wavelength to reach the absorber layer. In addition to the toxicity of cadmium which represents a serious problem [4], many studies are oriented toward alternative buffer layers not toxic [17], these layers equally present physico-chemical and optoelectrical properties very interesting. Many varieties of zinc compounds could be used with success like of alternative buffer layers [7].

ZnS, ZnSe, ZnO and certain of their compounds ternary are among the materials which can be exhibiting the best electrical performance [18] [19].

Zinc-based buffer layers have most often an adjustable wide gap [20], a zinc-based buffer layer presents in addition, a certain quality of interface with the CIGS layer which permits the control of defect diffusion towards this interface [21]. It has been found that the diffusion of the zinc element in the absorber presents the same advantages as cadmium [22]. Moreover, the abundance of the main raw material, which is zinc, is one of the most important particularities of the zinc-based compound. The presence of a buffer layer is beneficial for the solar cell, the first aim is to form the heterogeneous pn junction with the absorber [23], it must optimize in addition, the band alignment between the transparent conductive oxide (T.C.O) layer and the absorber. The following functions are attributed to the buffer layer [22] [24] [25] [26]:

- From a physical point of view, the effects of the buffer layer affect the structure of band energy. Especially, it affects the band offset;
- From an optical point of view, it must have good transparency to the solar spectrum and a well-adapted refraction index.

2.4. Thin Film Solar Cells

In the field of the thin film, it is necessary to notice that the variation of nanometer order of the thickness of one of the layers, in particular the buffer layer (CdS, ZnSe, Zn(O,S), (Zn,Mg)O) or the absorber layer $Cu(In,Ga)Se_2$, has considerable effects on the functioning of the solar cells. In addition, comparing the characteristic J-V of solar cells with an alternative buffer layer to reference CdS buffer, must be done with many precautions. In the field of thin film, it is hard to know if the weak performance of cells results from unexpected problems that occur during one of the phases of the process, or if the evolution observed is due to a change on the buffer layer. It is therefore difficult to interpret a change in electrical behavior without considering all the modifications made. Despite this difficulty, it is nevertheless possible to carry out this comparison by interpreting the graphs relating to the quantum efficiency QE (λ) and to the current-voltage density J-V.

Table 1 below summarizes the parameters of the different buffer layers studied [1] [3] [11] [20] [25] [27] [28].

The ZnSe material has a large gap of 2.67 eV, the Zn(S,O) and (Zn,Mg)O materials are ternary materials, and the composition of these materials depends on the concentration of sulfur atoms and magnesium. The sulfur or magnesium content has a strong impact on the position of the conduction band minimum and therefore on the discontinuity of the conduction band. There is therefore a sulfur or magnesium content in these ternary compounds allowing good band alignment at the buffer/absorber layer interface. The sulfur or magnesium content ranges from 0 to 1, the gap of pure materials, ZnO is 3.3 eV, ZnS is 3.6 eV, and MgO is 3.9 eV (Table). Indeed, the graduation of the gap of Zn(O,S) or

Table 1. Bases parameters of CIGS cell properties. W—thickness, ε—dielectric constant, E_g—band gap energy, ΔE_C—conduction band offset, σ_e, σ_h—capture cross section electrons and holes, χ_e—electron affinity, υ—thermal velocity, N_a, N_d—shallow uniform acceptor and donor density.

Parameters	CdS	ZnSe	Zn (S,O)	(Zn, Mg)O
W (nm)	30	30	30	30
E_g (eV)	2.4	2.67	3.3 - 3.6	3.3 - 3.9
χ_e (eV)	4.15	4.44	4.38	4.34
$\varepsilon/\varepsilon_0$	10	9.1	9	9
υ_e (cm/s)	10^7	10^6	10^6	10^6
υ_h (cm/s)	10^7	10^6	10^6	10^6
N_a (cm^{-3})	7×10^{15}	3×10^{15}	3×10^{15}	3×10^{15}
N_d (cm^{-3})	2×10^{17}	10^{17}	10^{17}	10^{17}
ΔE_C (eV)	−0.3	−0.3	−0.3	−0.3
σ_e (cm^2)	10^{-17}	10^{-12}	10^{-12}	10^{-12}
σ_h (cm^2)	10^{-13}	10^{-12}	10^{-12}	10^{-12}

generally $ZnO_{1-y}S_y$ and $Zn_{1-x}Mg_xO$ vary as a function to the composition x and y, according to the Equations (1) and (2) [29] [30] in a parabolic profile case.

$$E_g\left(\mathrm{ZnO}_{1-y}\mathrm{S}_y\right) = yE_g\left(\mathrm{ZnS}\right) + \left(1-y\right)E_g\left(\mathrm{ZnO}\right) - by\left(1-y\right) \qquad (1)$$

$$E_g\left(\mathrm{Zn}_{1-x}\mathrm{Mg}_x\mathrm{O}\right) = xE_g\left(\mathrm{MgO}\right) + \left(1-x\right)E_g\left(\mathrm{ZnO}\right) - bx\left(1-x\right) \qquad (2)$$

where y represents the concentration of sulfur in ZnO, *i.e.* the ratio S/(O + S), E_g(ZnS) and E_g(ZnO) are the gap of the pure compounds ZnS and ZnO respectively and b is a constant fixed at 3 eV [29]. x represents the concentration of magnesium in ZnO, *i.e.* the ratio Mg/(Zn + Mg) and E_g(MgO) the gap of the pure compound MgO respectively [29] [30]. For our study, the content of sulfur is 0.26 and that of magnesium is 0.28, so we obtain a gap of 2.8 eV for Zn (O,S) and 2.86 for (Zn,Mg)O.

3. Results and Discussions

3.1. Buffer Layer of Cadmium Sulfide (CdS)

In our paper intituled numerical simulation of Cu (In,Ga)Se_2 solar cells performances [11], the detailed study of the solar cells based on the CICS compound as a function of the variation of the CdS buffer layer thickness for a temperature of 300 K, permits us to extract the electrical parameters through the exploitation of J-V characteristic. These results obtained show that the values of every electrical parameter are more important for low thickness of the CdS buffer layer as confirmed in **Figure 2**.

The analysis of the results allows us to understand that the recombination inside the CdS buffer layer and at the CdS/CIGS layer interface becomes more and more important when the thickness of the CdS increases.

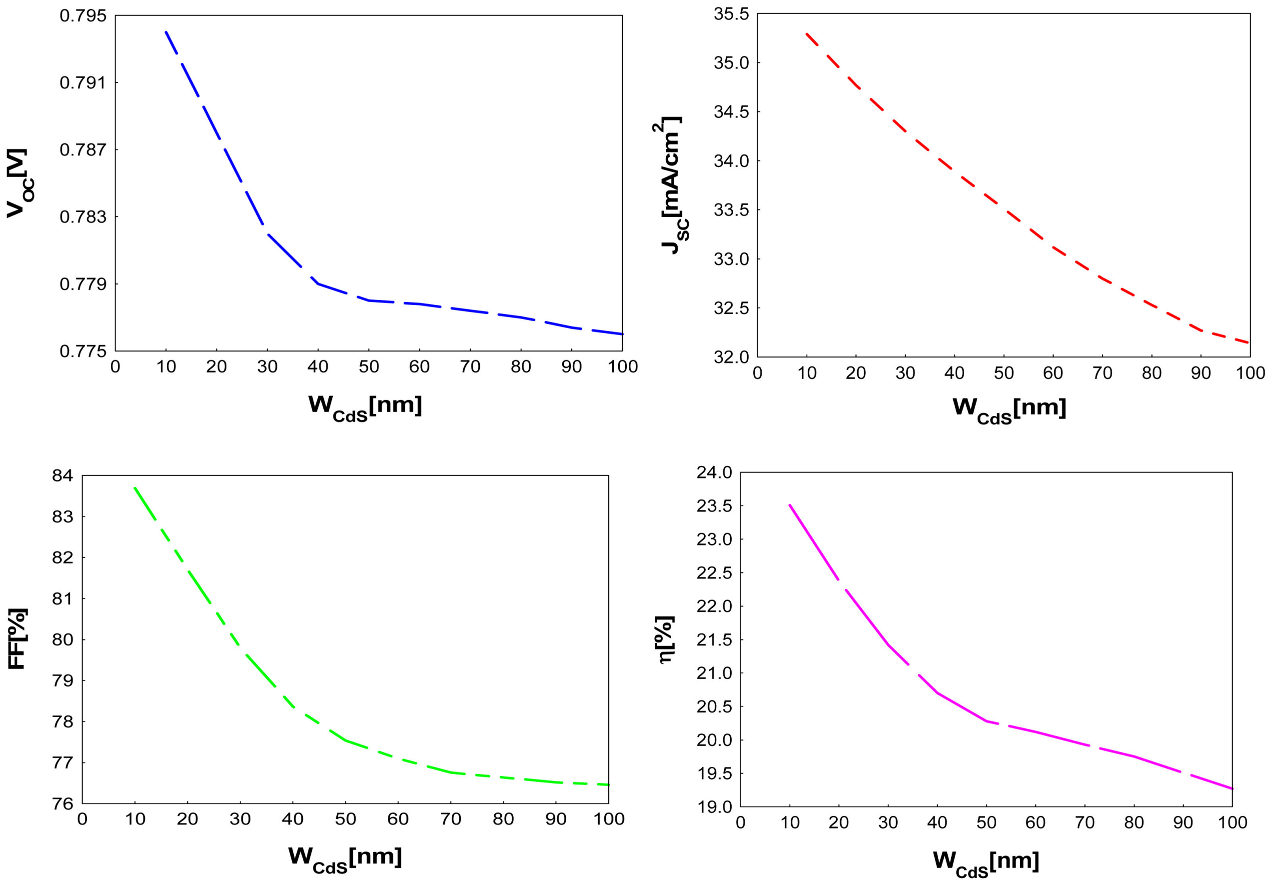

Figure 2. Electrical parameters as a function of CdS layer thickness.

Theoretically (**Figure 2**), we note that a very low thickness of the CdS layer is a source of good performance, which is not the case in practice because this low thickness of the CdS buffer layer does not effectively protect the CIGS absorber layer during of the energetic deposition of the transparent conductive oxide layer (T.C.O), this is in a good agreement with Zongo *et al.* [2]. In the rest of our work, we therefore, retain a 30 nm thickness of the CdS for good stability and better optimization of the performance of the CIGS solar cells. These results will serve as support in order to be able to carry out a comparative study with the different alternative buffer layers.

3.2. Study of Alternative Buffer Layers

In this part, we make a comparative study of the performance of the CIGS solar cells with an alternative buffer layer to the performance of the CdS buffer layer cell. In agreement to the results that we obtained on the CdS buffer layer and the absorber layer [31], we consider a thickness of 2000 nm and a gap of 1.2 eV for the CIGS absorber layer according to Oubda and Zongo *et al.* [2] [31]. The series resistance is zero and the shunt resistance is infinite in our model. The buffer layers we study are zinc selenide (ZnSe), sulfurous zinc oxide (Zn(O,S)), and zinc and magnesium oxide ((Zn,Mg)O).

3.2.1. Electrical Characteristics and Quantum Efficiency

Figure 3(a) and **Figure 3(b)** present the J-V and QE characteristics for the different alternative buffer layers studied, so that in CdS for comparison.

We note that the short-circuit current density (J_{SC}) of the alternative buffer layers increases (**Figure 3(a)** and **Figure 4(b)**) and on the other hand their open-circuit voltage decreases (**Figure 3(a)** and **Figure 4(a)**) comparatively to those of the CdS buffer layer: the cell with the ZnSe, Zn(O,S) or (Zn,Mg)O buffer has a higher J_{sc}, but a lower V_{oc}. The increase in the short-circuit current density is explained by a contribution of the charge carriers resulting from the absorption of very energetic short wavelength photons $465 \le \lambda \le 520$ nm for ZnSe and $376 \le \lambda \le 520$ nm for the Zn(S,O) and (Zn,Mg)O in the CIGS absorber (**Figure 3(b)**).

Figure 4 shows, in addition to the open-circuit voltage (**Figure 4(a)**), that the CdS buffer layer displays the best fill factor (**Figure 4(c)**) and the best efficiency conversion (**Figure 4(d)**).

The analysis of these results therefore, shows that the recombination at the interface of the layers and inside the space charge region (SCR) of the photogenerated charge carriers is less important for the CdS buffer layer and very important for the alternatives buffer layers.

In addition, we notice high stability of the CdS buffer layer due to its very interesting physico-chemical and electro-optical properties, which favor obtaining a very large fill factor (FF) and a very high conversion efficiency (η) (**Figure 4**).

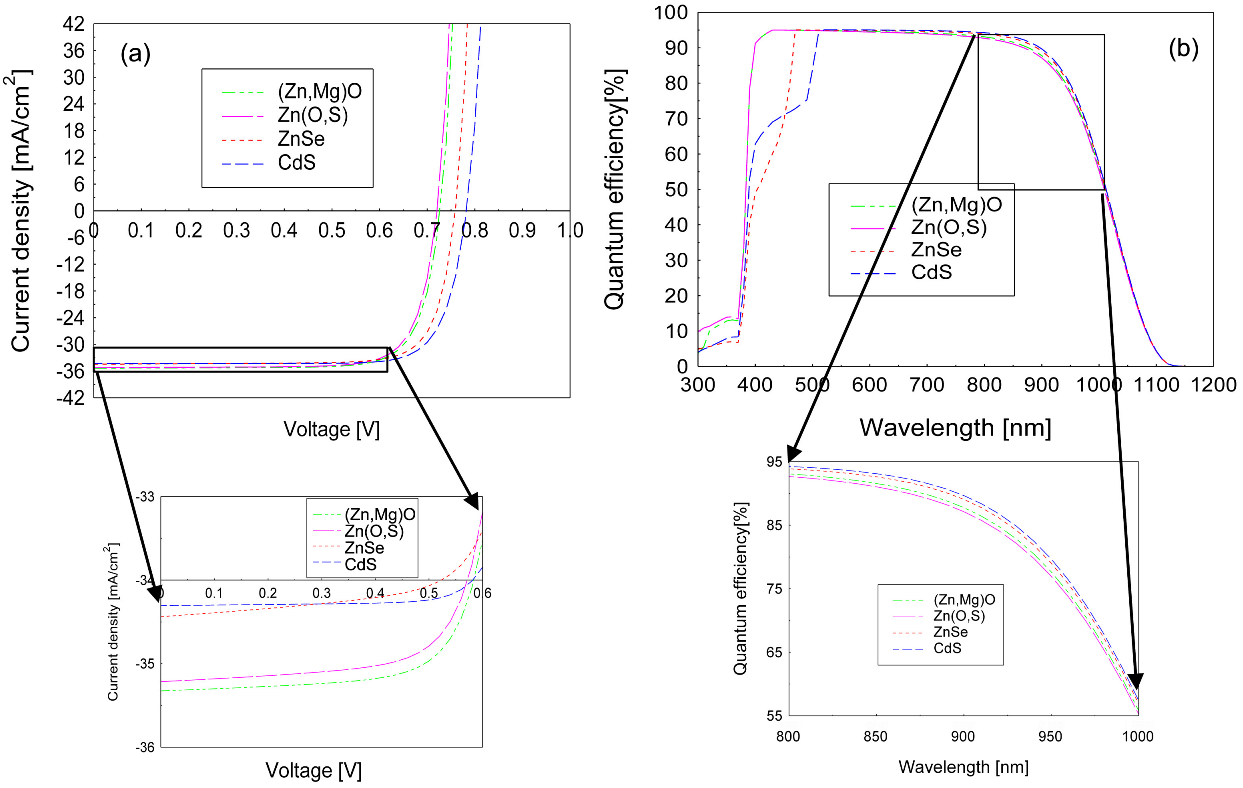

Figure 3. (a) Current density characteristic and (b) quantum efficiency as a function of the buffer layer.

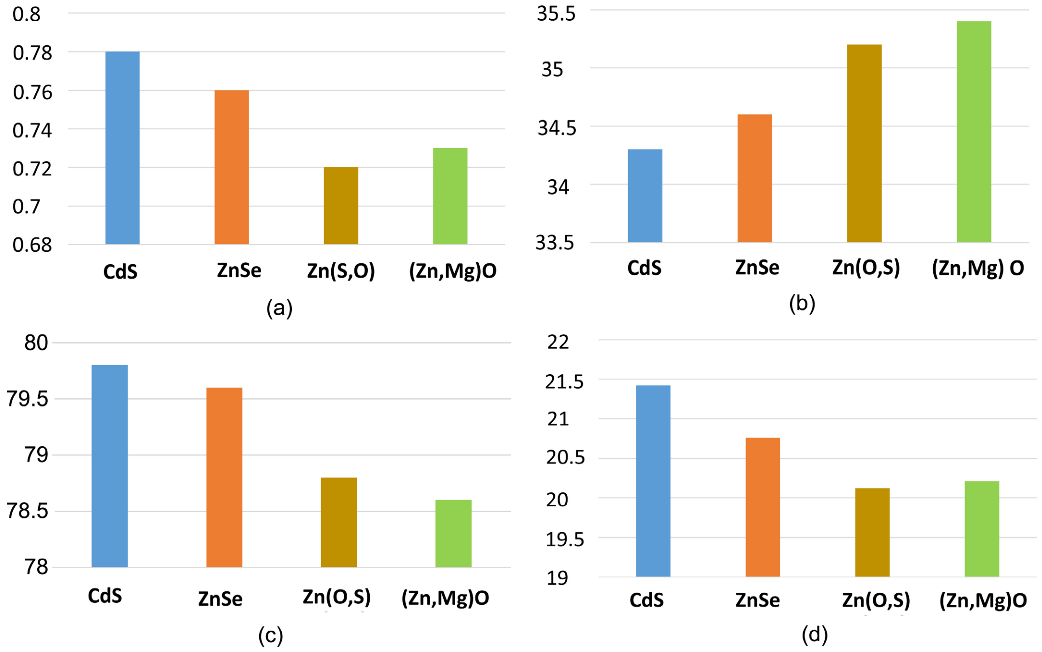

Figure 4. Evolution of electrical parameters as a function of the nature of the buffer layer. (a) V_{OC} (V); (b) Jsc (mA/cm^2); (c) FF (%); (d) η (%).

In this part of our paper, we can affirm that the use of a buffer layer with a wider gap than that of CdS reduces the losses related to the absorption of high energy photons of low wavelength in the CdS buffer layer.

3.2.2. Effect of Temperature on the Electrical Parameters

The operating temperature plays a vital role in the performance of solar cells. **Figure 5** presents the evolution of the electrical parameters as a function of the temperature.

The temperature is a limiting factor for the proper functioning of the solar cells. This work effectively shows that the electrical parameters are affected by temperature (**Figure 5**).

However, we note that the parameters of the different buffer layers have a similar behavior under the effect of temperature. The results obtained show a significant decrease in the values of the V_{OC} of the FF and of the η with the increase in temperature and a slight increase in the J_{SC}. At high temperatures, the gap of the materials decreases, which justifies the small increase in the values of the J_{SC}.

In addition, when the temperature increases, the mobility of the electron and hole decreases and leads to significant recombination of the charge carriers. Likewise, the concentration of charge carriers decreases, which greatly decreases the performance of the solar cell.

Among our buffer layers, the ZnSe layer holds our attention, because it is getting closer and closer to the reference buffer layer (CdS) by the evolution of these curves as well as the values of these electrical parameters.

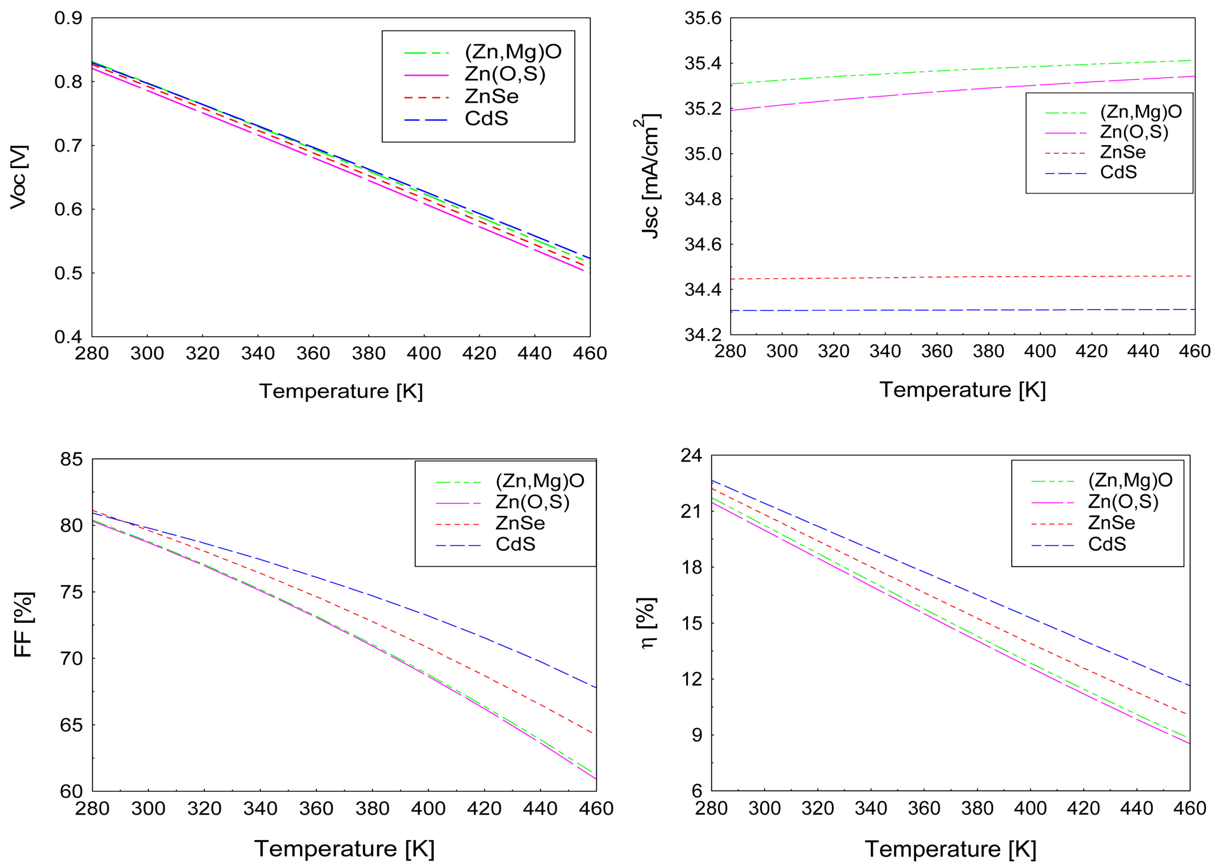

Figure 5. Electrical parameters as a function of the temperature.

3.2.3. Energy Band Diagram

Conduction and valence band offsets are among the most important factors of the CdS layer that allow obtaining very good performances. It is important for us to analyze the energy band diagram of our alternative buffer layers in order to be able to evaluate their efficiencies and their stabilities.

The results obtained are presented in **Figure 6**. We actually see a good alignment of the discontinuities of the valence and conduction bands (E_V and E_C), which is very important for the good performance of the solar cell. Similarly, we appreciate the good curvature of the Fermi levels for the electrons and for the holes (E_{F_n} and E_{F_p}) (**Figure 6**) of the different buffer layers, which is a sign of good stability

The good alignment of the ΔE_V and ΔE_C as well as the good curvature of the E_{F_n} and E_{F_p} can also be explained by the fact that the materials have different gap, in addition, the gap of each one is direct.

This actually shows that each of the alternative buffer layers has very interesting Physico-chemical and electro-optical properties to be used as a buffer layer for solar PV devices.

We also note the stability of the alternative buffer layers close to the stability of the CdS reference buffer layer, which encourages their use in photovoltaic solar cells based on CIGS.

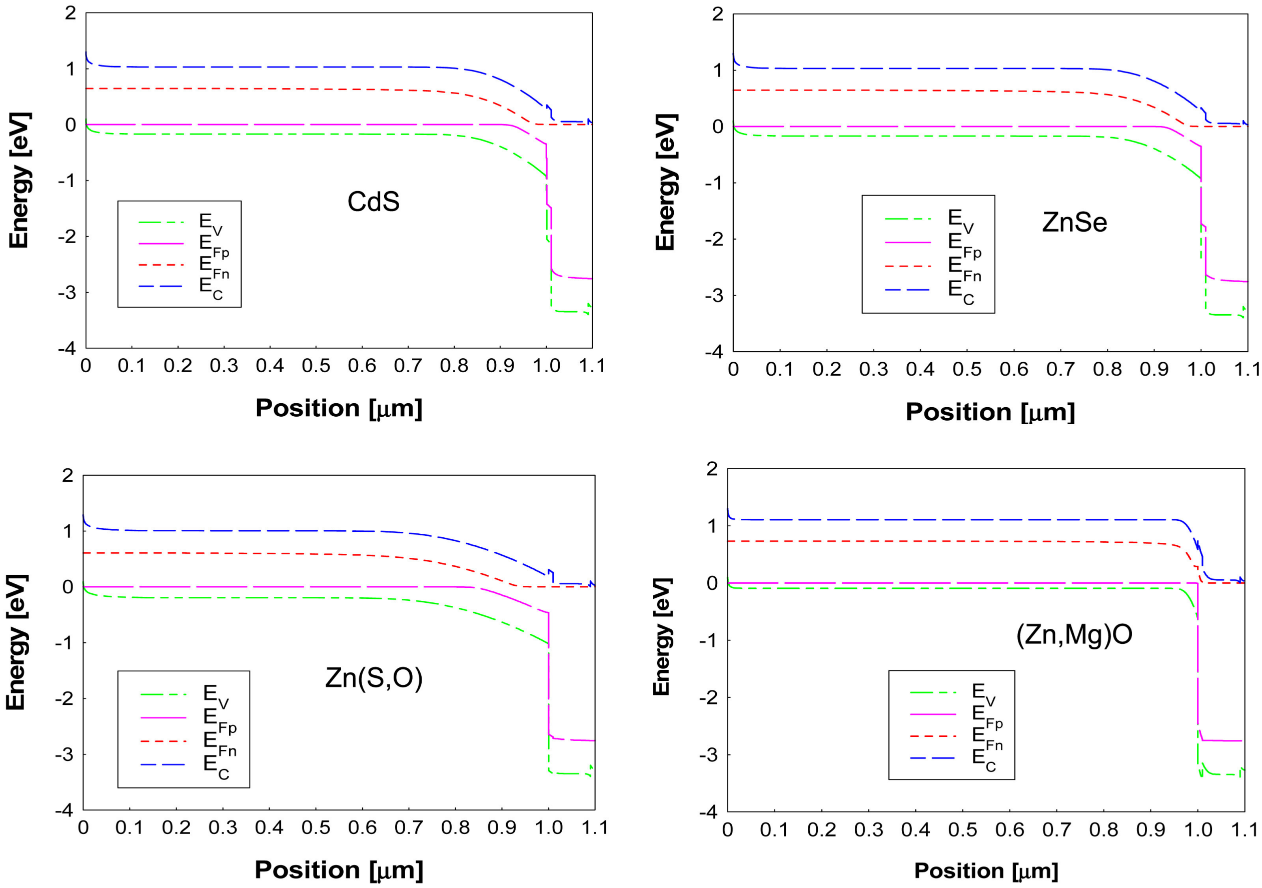

Figure 6. Band offset diagram and Fermi level alignment.

4. Conclusions

In this paper, the study of the polycrystalline thin film GIGS solar cells has been carried out. Various buffer layers such as ZnSe, Zn(O,S), and (Zn,Mg)O for possible replacement of CdS have been investigated in terms of overall solar cell performance for different buffer layer thickness and at various operating temperatures. For an optimum thickness and band gap of about 2000 nm respectively 1.2 eV of high-efficiency CIGS absorber, the optimum thickness of buffer layers is 30 nm.

The simulation results show that the performance of solar cells is affected by the increase in the thickness of the buffer layer and the operating temperature, but also by the nature of the buffer layers. The absorption of CIGS chalcogenide in the buffer layer being a one source of loss in Cu (In,Ga)Se$_2$ thin-film solar cells, this work allowed us to show that the use of a buffer material with energy of band gap higher allows to obtain a higher short-circuit current density J_{SC}.

The investigation of cells based on ZnSe, Zn(O,S) and (Zn,Mg)O with the aim to replace traditional CdS in the future shows that all buffer layers are promising materials with higher efficiency than 20%. Of these alternative materials which have a band gap larger than CdS, therefore the absorption losses in the buffer layer are minimized. The losses as a function of temperature are estimated at

0.1%/K, for all the types of buffer layers studied. The cell with ZnSe buffer layer has promising potential to replace CdS, it revealed the best efficiency of 20.76%, the other electrical parameters are important, we note: J_{SC} = 34.6 mA/cm², V_{OC} = 0.76 V and FF = 79.6%.

In view of the results obtained, if the use of a buffer layer seems to be a good way to obtain very high performance, it is clear that the use of an alternative buffer in addition to the good performance must make it possible to avoid environmental pollution thanks to non-polluting materials such as zinc and its compounds.

Acknowledgements

The author gratefully acknowledges the use of SCAPS-1D developed by Marc Burgelman and colleagues at the University of Gent.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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