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Thickness optimization of Al_2O_3 tunneling layer in CBTS solar cells using SCAPS software

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Abstract: *The purpose of this paper is to explore the possibility for enhancing the performance of CBTS solar cells by incorporating Al_2O_3 insulating layer (Eg ~7 eV) between CdS buffer layer and CBTS absorber layer. initially, we performed a comparative analysis between Mo/MoS_2/CBTS/CdS/ZnO/AZO/Al and Mo/MoS_2/CBTS/ Al_2O_3/CdS/ZnO/AZO/Al using SCAPS-1D. Subsequently, we conducted an investigation into the impact of Al_2O_3 thickness on cell performance. Our findings indicate that photovoltaic parameters deteriorate with an increase in Al_2O_3 layer thickness, and a thickness of 3nm is sufficient to facilitate the electrons intra-band tunneling. The PCE of the reference cell (without Al_2O_3) is 6.75%. Upon inserting the alumina layer, the device exhibits a PCE of 11.89% with V_{OC}, J_{SC}, and FF equal to 1.08 V, 15.45 mA/cm² and 71.41 % respectively. Although cell efficiency is not fully optimized in this study, we've highlighted the significant utility of Al_2O_3 layer in advancing the CBTS solar cells development.*

Keywords: Al_2O_3; CBTS; Tunneling, Simulation, SCAPS-1D.

1. INTRODUCTION

Solar energy is one of the most widely used forms of renewable energy, which generates electricity using photovoltaic (PV) solar cells [1].
In recent years, thin films based on earth abundant elements such as Cu_2ZnSnS_4 (CZTS), $Cu_2ZnSnSe_4$ (CZTSe), $Cu_2ZnSn(S,Se)_4$ (CZTSSe), Cu_2BaSnS_4 (CBTS), Cu_2FeSnS_4 (CFTS), and Cu_2MgSnS_4 (CMgTS) were strongly investigated as PV absorbers [2].
The quaternary Cu_2BaSnS_4 (CBTS) is a p-type semiconductor with a direct bandgap and a high absorbance coefficient. Ge et al. (2017) realized the highest cell efficiency with a PCE of 2.03% at CBTS/CdS configuration [3]. Adding selenium to the absorber has brought an efficiency of over 5% [4].
The theoretically maximum efficiency of CBTS solar cell is around 25% [5]. However, an unwanted band alignment at the absorber / buffer heterojunction plays the prominent causes of low overall performance and limits V_{OC} and FF outputs [6] [7].
Several researchers found that using an interlayer at the buffer/absorber heterointerface could be beneficial for cell progress. Erkan et al. (2016) indicated the possibility of enhancing the Voc by the insertion of the Al_2O_3 layer in CZTS solar cells [8]. Ojeda-Durán et al. (2020) demonstrated enhancement in the V_{OC} and the FF using Al_2O_3 ultrathin layers for CZTS/CdS hetero-junctions [9]. The penetration of the carriers through is possible by tunnelling process.
Herein, we performed simulations of Mo/MoS_2/CBTS/Al_2O_3/CdS/ZnO/AZO/Al solar cell using SCAPS-1D software, then We studied the effect of the Al_2O_3 layer thickness on electrical performance, J-V characteristics and tunnelling current density.

2. DEVICE STRUCTURE AND SIMULATION

The schematics of the reference device structure (without Al_2O_3) and the proposed one (with Al_2O_3) are illustrated in Fig. 1. And Fig.2, respectively.
It consists of MoS_2 BSF layer, CBTS absorber layer, Al_2O_3 tunnelling layer, ZnO: Al Transparent Conducting Oxide, and i-ZnO window layer. The Mo back contact and Al front contact.

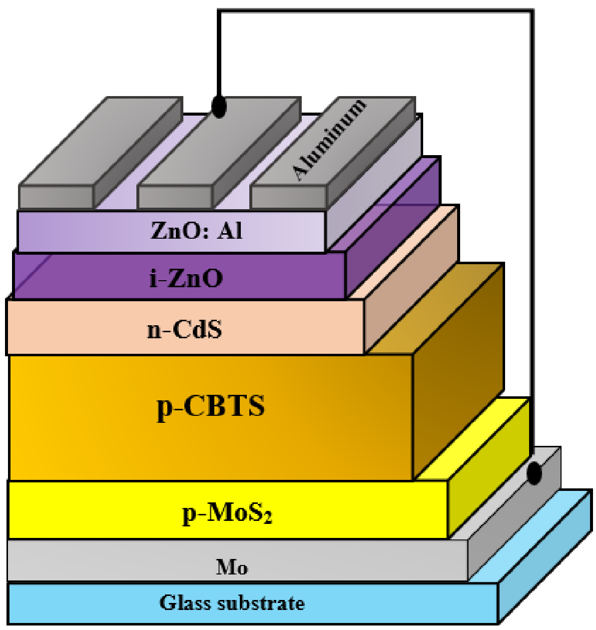

Fig. 1. The schematic representation the reference device.

Table 1. Layer properties [8, 10-25]

	MoS_2	CBTS	Al_2O_3	CdS	ZnO	AZO
W (µm)	0.1	2	0.003	0.02	0.05	0.1
Eg (eV)	1.7	1.9	7	2.42	3.37	3.6
χ (eV)	4.1	3.6	2.5	4.1	4.5	4.6
ε_r	13.6	5.4	9.8	9	9	9
N_C (cm^{-3})	7.5×10^{17}	2.2×10^{18}	1.0×10^{12}	1.8×10^{18}	2.2×10^{18}	2.2×10^{18}
N_V (cm^{-3})	1.8×10^{18}	1.8×10^{19}	1.0×10^{12}	2.4×10^{19}	1.8×10^{19}	1.8×10^{19}
μ_e ($cm^2\ V^{-1}s^{-1}$)	100	30	247	160	150	150
μ_h ($cm^2\ V^{-1}s^{-1}$)	150	10	247	50	25	25
N_D (cm^{-3})	0	0	0	5.0×10^{18}	1.0×10^{17}	1.0×10^{20}
N_A (cm^{-3})	1.0×10^{16}	5.0×10^{15}	0	0	0	0
N_T (cm^{-3})	1×10^{14}	1×10^{15}	2.10^{15}	1×10^{17}	1×10^{17}	1×10^{17}

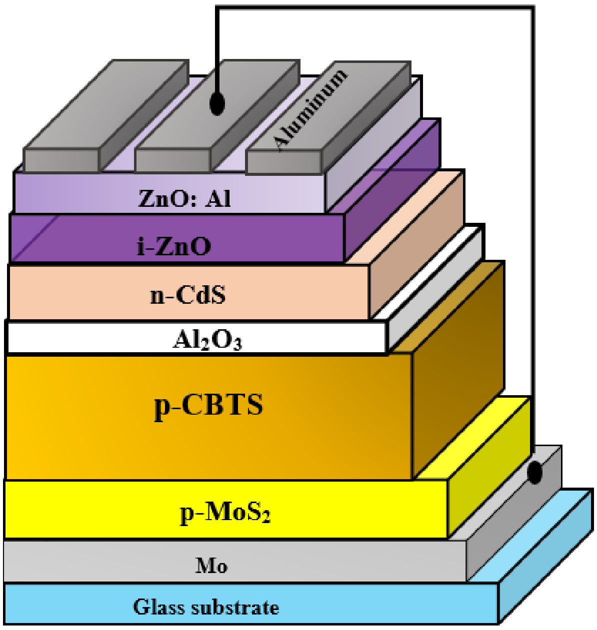

Fig. 2. The schematic of the proposed device.

Here, the simulation is done by the SCAPS-1D. the SCAPS-1D works on three fundamental semiconductor equations which are:

The Poisson's equation:

$$\frac{\partial}{\partial x}\left(\varepsilon_0\varepsilon_r\frac{\partial\Psi}{\partial x}\right) = -q\left(p - n + N_D^+ - N_A^- + \frac{\rho_{def}}{q}\right) \quad (1)$$

The continuity equations

$$\frac{dn(t)}{dt} = G_n - R_n - \frac{1}{q}\frac{\partial J_n}{\partial x} \quad (2)$$

$$\frac{dp(t)}{dt} = G_p - R_p - \frac{1}{q}\frac{\partial J_p}{\partial x} \quad (3)$$

The drift-diffusion equations

$$J_n = nq\mu_n E + qD_n\frac{dn}{dx} \quad (4)$$

$$J_p = pq\mu_p E - qD_p\frac{dp}{dx} \quad (5)$$

where Ψ is the electric potential, ε_0 and ε_r are the permittivity of the free space and relative, respectively. ρ_{def} is the defect density, N_D^+ and N_A^- are the densities of ionized donors and acceptors, R and G are the recombination and generation rates of carriers, respectively. Jn and Jp are the current densities of electrons and holes. D and µ is the diffusion coefficient and mobility of carriers, respectively.
The parameters of the materials used to simulate the CBTS solar cell are summarized in Table 1.
The interface and Al_2O_3 layer defects densities are listed in table 2.
The measurements were performed at zero bias, under 300 K, 1.5 AM and light of 1 KW/m^2.

3. RESULTS AND DISCUSSION

We first simulate the cell structure with and without Al_2O_3 insulator layer, to show how the presence of Al_2O_3 layer can affect the device performance. The obtained results are summarized in Table 3.

Table 3. Functional parameters of the studied cell with and without Al_2O_3 layer.

cell	V_{OC} (V)	J_{SC} (mA/cm²)	FF (%)	η (%)
Without Al_2O_3 layer	0.69	15.09	64.84	6.75
With Al_2O_3 layer	1.08	15.45	71.41	11.89

Table 2. added defects values

properties	CBTS/CdS [12]	Al_2O_3 [8]	CBTS/Al_2O_3	CdS/Al_2O_3
N_T (cm^{-3})	/	2.10^{15}	/	/
N_{it} (cm^{-2})	1.10^{15}	/	1.10^{15}	1.10^{11} [8]
δ_e (cm²)	2.10^{-15}	1.10^{-15}	2.10^{-15}	2.10^{-15}
δ_h (cm²)	2.10^{-15}	1.10^{-13}	2.10^{-15}	2.10^{-15}

According to Table 3, the cell with Al_2O_3 layer shows highest photovoltaic parameters (V_{OC}, J_{SC}, FF, and η).
To explain these results, Fig. 3 has been drawn. the band alignment at the CBTS/CdS heterojunction (without Al_2O_3 layer) is a cliff.
A cliff-like offset triggers the accumulation of electrons in CdS and increases the recombination process at CBTS/CdS interface between the majority carriers [26], leading to the V_{OC} degradation.
By inserting Al_2O_3 dielectric layer, the built-in electric field at CBTS/Al_2O_3, MoS_2/CBTS and Al_2O_3/CdS interfaces increases, indicating higher values of V_{OC}.

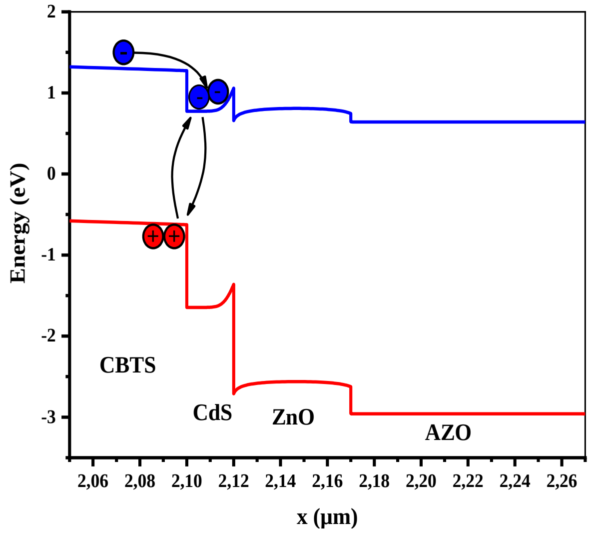

Fig. 3. The energy band diagram of structure without Al_2O_3 layer

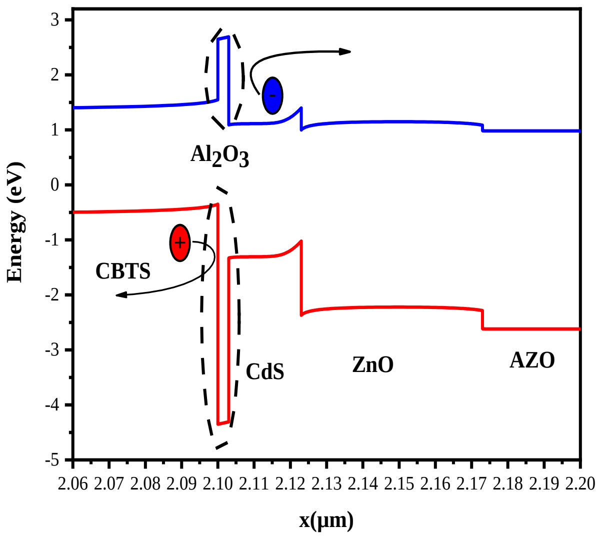

Fig.4 The energy band diagram of device with Al_2O_3 layer.

3-1 Effect of Al_2O_3 thickness on PV parameters

In this section, we aim to optimize the Al_2O_3 layer thickness, the thickness was changed from 1 nm to 15 nm [9, 27]. The impact of Al_2O_3 thickness on J-V curve and the PV parameters is shown in Fig.6 and Fig.7,

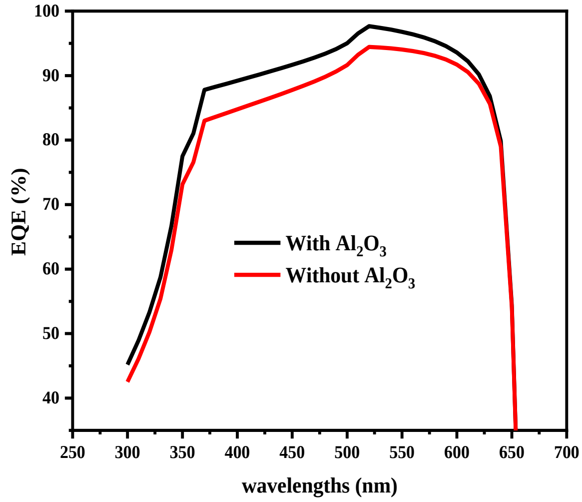

Fig. 5. The EQE vs. Wavelength of both structures (with and without the Al_2O_3 layer).

The band diagram of the Mo/ MoS2/ CBTS/ Al_2O_3/ CdS/ ZnO/AZO/Al structure is shown in Fig.4.
By adding the Al_2O_3 layer, the structure is characterized by a barrier of 4 eV at valence band at CBTS/ Al_2O_3 interface and a desired spike-like CBO at CdS/ Al_2O_3 interface.
The presence of insulator layer is beneficial to block the electrons at front side of the cell and the holes at back side so they cannot recombine with each other, resulting in high carrier collection and this explains why the presence of Al_2O_3 improve the EQE values (Fig.5).

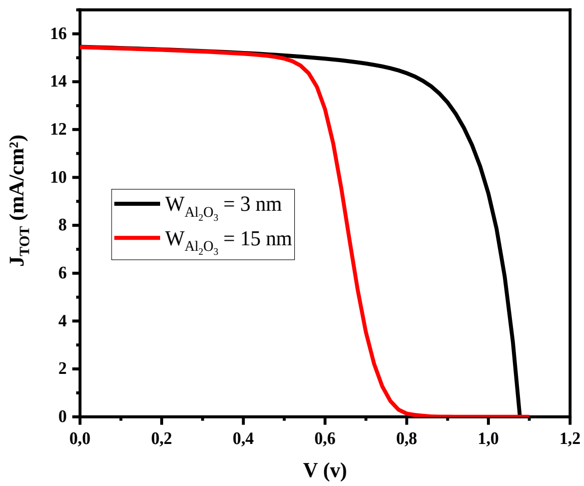

Fig.6 Effect of thickness of Al2O3 on J-V curve.

respectively. Fig. 7(a) illustrates that the open circuit voltage (V_{OC}) is not affected by the Al_2O_3 thickness variation and its value remains around 1.08 V.
As shown in Fig. 5(b), the J_{SC} decreases with the increase of the Al_2O_3 layer thickness.

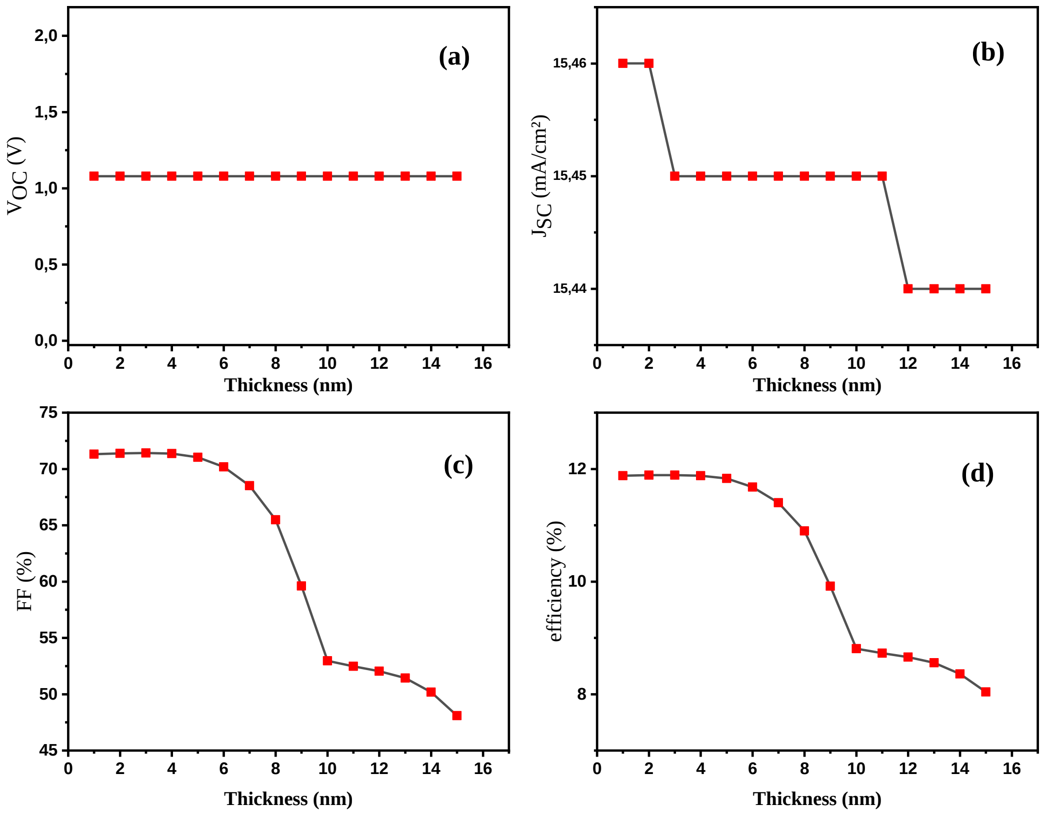

Fig.7 Effect of Al_2O_3 thickness variation on PV parameters.

We observe a similar shape of efficiency and fill factor (FF) curves, implying that the decrease in efficiency is mainly related to the deterioration in FF.
The drop in FF with the increase in insulator layer Al_2O_3 thickness is due to the increase of tunnelling resistance.

In other words, when the Al_2O_3 thickness is increased, the width of the barrier reaches an extent that the photo-generated carriers do not have the energy and time sufficient to cross the barrier.
In Fig.8, we show the influence of the thickness of Al_2O_3 on J_{Tunnel}. It is obvious that the smaller the thickness of Al_2O_3, the higher tunnelling current density delivered (J_{Tunnel}).
The optimum thickness of Al_2O_3 was observed to be about 3 nm and the PCE is significantly deteriorated from 11.89% to 8.04 % when the thickness increases from 3 to 15 nm.
We found good agreement of the optimum thickness of Al_2O_3 with the experimental values that had previously published (Table 4).

4. CONCLUSIONS

In this paper, we have described the effects of Al_2O_3 insulator layer on cell performance of CBTS solar cells. To be more precise, we have intended to explain how the insertion of Al_2O_3 insulator layer can greatly improve the cell's performance. Furthermore, we have found that a thickness of only 3 nm is enough to allow the transport of photogenerated carriers by tunnelling effect.
Despite the fact that the presence of an Al2O3 layer enhances device performance, an increase in the thickness of this layer by a few nanometers leads to a significant reduction in FF. our results indicate that an optimized Al2O3 has positive impact on the VOC and the FF and PCE.

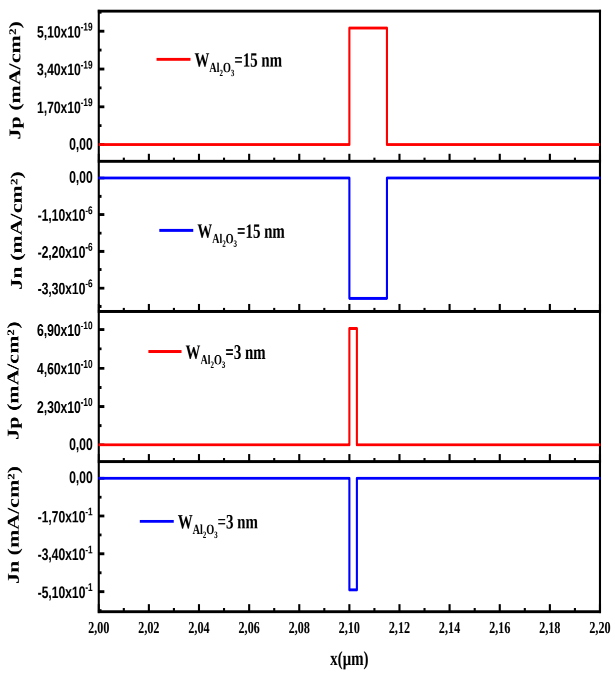

Fig. 8 Effect of Al_2O_3 thickness variation on tunnel current density.

Table 4. comparison of PV parameters of this paper with other reported works.

Structure	V_{OC}(V)	J_{SC} (mA/cm²)	FF (%)	η (%)	Optimal Al_2O_3 thickness	Ref
Mo/CZTSSe/Al_2O_3/CdS/i-ZnO/AZO/Ni-Al	0.336	13.8	29.14	1.35	1 nm	[8]
Mo /CZTS/ CdS/Al_2O_3/ /ITO	0.515	32.1	69.2	11.5	1 nm	[28]
Mo/ Al_2O_3/ CZTSSe /CdS/AZO/Ni-Al	0.364	35.35	55.46	7.13	5 nm	[29]
Mo/MoS_2/CZTS/Al_2O_3/CdS/ZnO/ITO	0.669	15.7	58.0	6.1	3 nm	[9]
Mo/MoS_2/CBTS/Al_2O_3/CdS/ZnO/AZO/ Al	1.08	15.45	71.41	11.89	3 nm	Our work [30]

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