

Optimum Defect Density of Organic–in Organic for High Efficiency Perovskites Solar Cells

Murtadha J. Edam^{1,*}, Hawraa M. Khadier², Samir M. AbdulMohsin¹, Baraa H. Auad³, Dhuha E. Tarek⁴ and Mushtaq O. Oleiw¹

¹ Department of Physics, College of Education for Pure Sciences, University of Thi-Qar, Thi-Qar, 64001, Iraq

² Department of Physics, College of Sciences, University of Thi-Qar, Thi-Qar, 64001, Iraq

³ Dhi-Qar Health Department, Ministry of health, Thi-Qar, Iraq

⁴ Department of Biomedical Engineering, College of Engineering, University of Thi-Qar, Thi-Qar, 64001, Iraq

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Abstract: In this paper, device modeling of lead-free perovskite solar cells was done using the perovskite absorber layer $Cs_2AgBi_{0.75}Sb_{0.25}Br_6$ with a big bandgap of 1.8 eV, a PCBM layer for the electron transport layer, which is renowned for its anti-hysteresis effect, and a NiO layer for the hole transport layer, which increases stability and boosts the cell's efficiency. The SCAPS-1D, which is highly suited for analyzing photovoltaic design, was used to examine new architecture. Using this software technique, we conducted an analysis the thickness, defect density, operating temperature and work function of the model through modeling under different circumstances. With the optimize the thickness to be (0.3 μm) corresponding best efficiency among several perovskite thicknesses, the defect density of absorber layer (10^{13} cm^{-3}) and maximum power conversion efficiency PCE rose to 35.47%, which is a promising result, the short-circuit current density J_{sc} is 33.498903 (mA/cm^2), and fill factor FF is 78.00% and open-circuit voltage V_{oc} is 1.3584 (V). while, the effect of the working temperature was evaluated, and the outcomes demonstrate that lead-free perovskite cells work quite well increase with increase in temperature then start decrease at 400 K. Finally, study effects for change of the work function on the major parameters of perovskite solar cells.

Keywords: Perovskites, SCAPS-1D, high efficiency, Solar Cells, thickness, work-function.

1. Introduction

In the past few decades, solar PV technology has grown quickly thanks to technological advancements, the search for new, inexpensive materials, and a rise in commercial manufacturing [1]. The efficiency of solar cells must be increased as much as feasible in order to commercialize this technology. One of the limitations of single-junction photovoltaic devices is when the photon energy is uneven with the frequency gap energy because this results in loss of power. Energy loss caused by the difference between the photon energy and the material's bandgap energy (E_g) is one of the variables that restrict the performance of single-junction photovoltaic (PV) devices. When the photon energy is equal to the frequency gap energy, the photon energy can be converted into electric energy with high efficiency, but if the photon energy is less than the frequency gap energy, cannot be absorbed, and when the photon energy is greater than the frequency gap energy it does not contribute to thermal conductivity because it loses the additional energy through the pregnant thermal coding [2]. Through this study we were able to analyze and examine the effects of the thickness change of the absorbed layer, HTM and ETL on system output, the SCAPS program was manufactured to

simulate the one-dimensional solar cell then this program was developed in Belgium by the University of Gent Department of Electronics and Information Systems. Many researchers were involved in the development SCAPS: Mark Burgelman, Queen Decock, Alex Nemigers, Stefan DeGraff, Johan Verschriegen [3-8].

2. Simulation and Modeling Computer

The use of simulation is essential for gaining a thorough understanding of physical activity, assessing the validity of proposed physical interpretations, and determining the impact of physical alterations on the performance of solar cell devices. In order to simulate solar cells there are a lot of simulation models like (SCAPS, AMPS, SCAP, etc.), SCAPS is a program consisting of 7 input layers for semiconductors [9]. It is impractical and a waste of effort and money to construct a solar cell without stimulation. It can analyze layers and their functions and also increase the ability of solar cells in addition, it reduces risks, shortens time and saves money. Key input parameters must be carefully selected to act as a real counterpart for computer simulation [10-11]. Simulation phases using SCAPS are shown step by step in the flow chart below:

*Corresponding author E-mail: murtadhaj11@utq.edu.iq

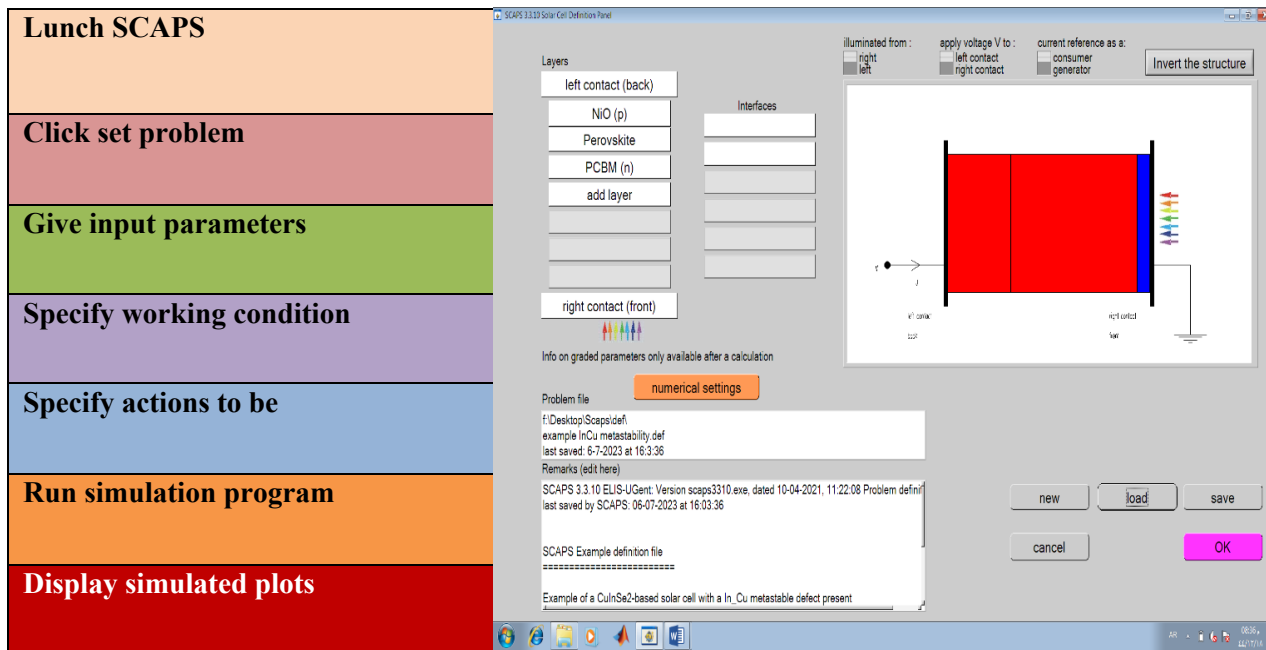


Fig. 1: Explains the simulation process and panel showing the NiO/C_{s2}AgBi_{0.75}Sb_{0.25}Br₆/PCBM heterojunction solar cells definition using SCAPS.

HTM has three main functions: acting as a hole-selective contact, extracting photo-generated holes, and carrying the holes to the metal contact. The ETL reduces charge recombination and enhances total PCE by blocking direct contact between the counter electrode and the perovskite/photoactive layer. Fig 2 shows a comparison of the energy levels of between HTM layer, absorb layer and ETL layer.

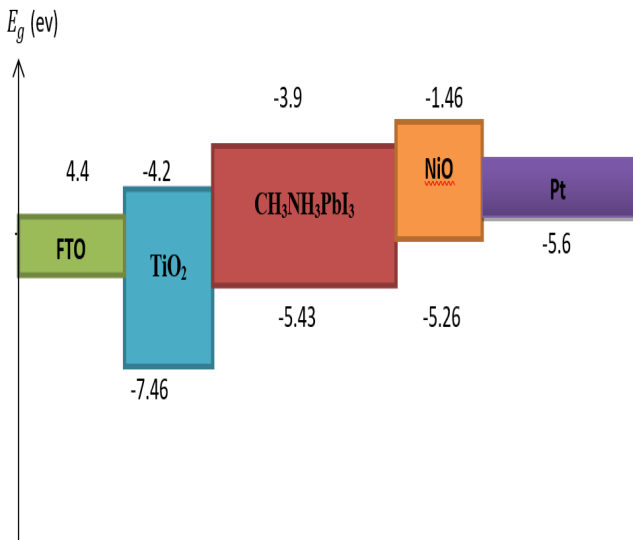


Fig. 2: Energy band alignment.

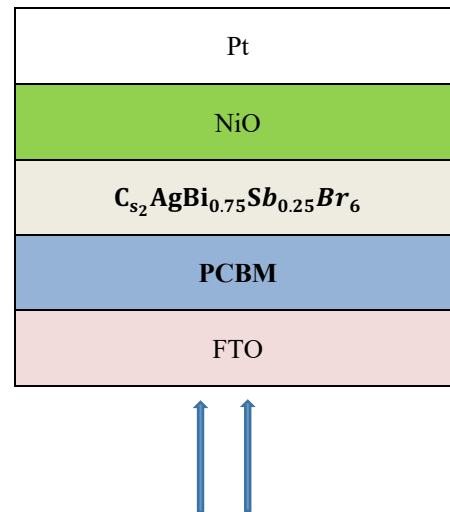


Fig. 3: Schematic representation device architecture (ITO/NiO / C_{s2}AgBi_{0.75}Sb_{0.25}Br₆/ PCBM/Pt).

3. Modeling and simulation

A model of perovskite solar cell involving C_{s2}AgBi_{0.75}Sb_{0.25}Br₆ is proposed. Device simulations for the ITO/NiO/C_{s2}AgBi_{0.75}Sb_{0.25}Br₆/PCBM/Pt structure have been performed using SCAPS1D. From I-V characteristics of structure, it was evident that the highest efficiency can be namely, Poisson's equation (1), continuity equation for holes (2) and continuity equation for electrons (3) as follows:

$$\frac{d}{dx} (-\epsilon(x) \frac{d\psi}{dx}) = q[p(x) - n(x) + N_d^+(x) - N_a^-(x) + p_t(x) - n_t(x)] \quad (1)$$

$$\frac{dp_n}{dx} = G_p - \frac{p_n - p_{n0}}{\tau_p} - p_n \mu_p \frac{d\xi}{dx} - \mu_p \xi \frac{dp_n}{dx} + D_p \frac{d^2 p_n}{dx^2}, \quad (2)$$

$$\frac{dn_p}{dx} = G_n - \frac{n_p - n_{p0}}{\tau_p} + n_p \mu_n \frac{d\xi}{dx} + \mu_n \xi \frac{dn_p}{dx} + D_n \frac{d^2 n_p}{dx^2} \quad (3)$$

Where, D is the diffusion factor, ψ represent the electrostatic voltage. The value of q represents the amount of the electron charge while G represents the rate of generation and ξ is permittivity. n represents free electrons either p free hole, n_t represents trapped electrons and p_t represents trapped holes. N_a^- refers to ionized acceptor-like doping concentration while N_d^+ stands for ionized donor-like doping concentration [12].

Table 1: Basic parameters used in modeling [13-20].

parameters	NiO	C _{s2} AgBi _{0.75} Sb _{0.25} Br ₆	PCBM
Band gap (ev)	3.600	1.800	2.000
Electron affinity (ev)	1.800	3.580	3.900
Dielectric permittivity	11.700	6.500	4.000
CB effective density of states (1/cm ²)	2.500E+20	2.200E+18	1.000E+21
VB effective density of states (1/cm ²)	2.500E+20	1.800E+19	2.000E+20
Electron mobility (cm ² /v.s)	2.800E+0	2.000E+0	1.000E-2
Hole mobility (cm ² /v.s)	2.800E+0	2.000E+0	1.000E-2
Donor Concentration N _D /cm ⁻³	0.000E+0	1.000E+13	5.000E+14
Acceptor Concentration N _A /cm ⁻³	3.000E+8	1.000E+17	0.000E+0

Table 2: Parameters used to simulate SCAPS.

Left contact electrical properties(Pt)	
Thermionic emission /surface recombination velocity electron and hole (cm/s)	10 ⁷
Metal (Pt) work function (ev)	5.6
Right contact electrical properties	
Thermionic emission /surface recombination velocity electron and hole (cm/s)	10 ⁷
work function of ITO (ev)	4.4

4. Result and Discussion

Effect of layer thickness, Defects, Temperatures and Work function change on the NiO/ C_{s2}AgBi_{0.75}Sb_{0.25}Br₆/PCBM solar cells devices

4.1. Effect of the NiO (HTM) layer thickness change on solar cells

The thickness of NiO as the (HTM) must be carefully controlled, to thoroughly a full cover the perovskite layer, absorb the most photons possible, and create electron-hole pairs. Layer thickness NiO has been range from (0.1 to 0.7) μm . A thin HTM layer does not completely cover the absorber layer, as a result, at thickness 0.1 μm , the efficiency is low. At thickness of the absorber layer C_{s2}AgBi_{0.75}Sb_{0.25}Br₆ increases from 0.2 to 0.5 μm , chances of recombination are rises because an increase in the path length of charge carrier and the electric resistance of the device, which boosts efficiency. Figure 4 sketches the change in the PV parameters of perovskite solar cell with thickness NiO as the HTM, the graph show an efficiency, voltage, and current density decreases with increasing

thickness, while the filling factor stays constant. when the thickness is increased from 0.2 μm to 0.5 μm , we note efficiency stay constant 0.57 then decrease at thickness 0.6 μm , while voltage is constant at the thickness 0.1 μm and 0.2 μm then increase at thickness 0.3 μm Then it starts to decrease in thickness 0.5 μm as illustrated in Figure (4). As a result, the best efficiency is obtained at a thickness of 0.2 μm , where the efficiency hits 0.57%. The resistance rise with the layer thickness increases from 0.6 to 0.7 μm , further lowering the cell's performance. The drawing data is shown in table number three.

Table 3: Variation of Thickness for NiO with device parameters.

Thickness(μm)	V _{oc} (V)	J _{sc} (mA /cm ²)	F.F (%)	η (%)
0.1	1.2295	1.488065	30.88	0.56
0.2	1.2295	1.488097	30.88	0.57
0.3	1.2296	1.488135	30.88	0.57
0.4	1.2296	1.488134	30.88	0.57
0.5	1.2295	1.488056	30.88	0.57
0.6	1.2294	1.487893	30.88	0.56
0.7	1.2292	1.487655	30.88	0.56

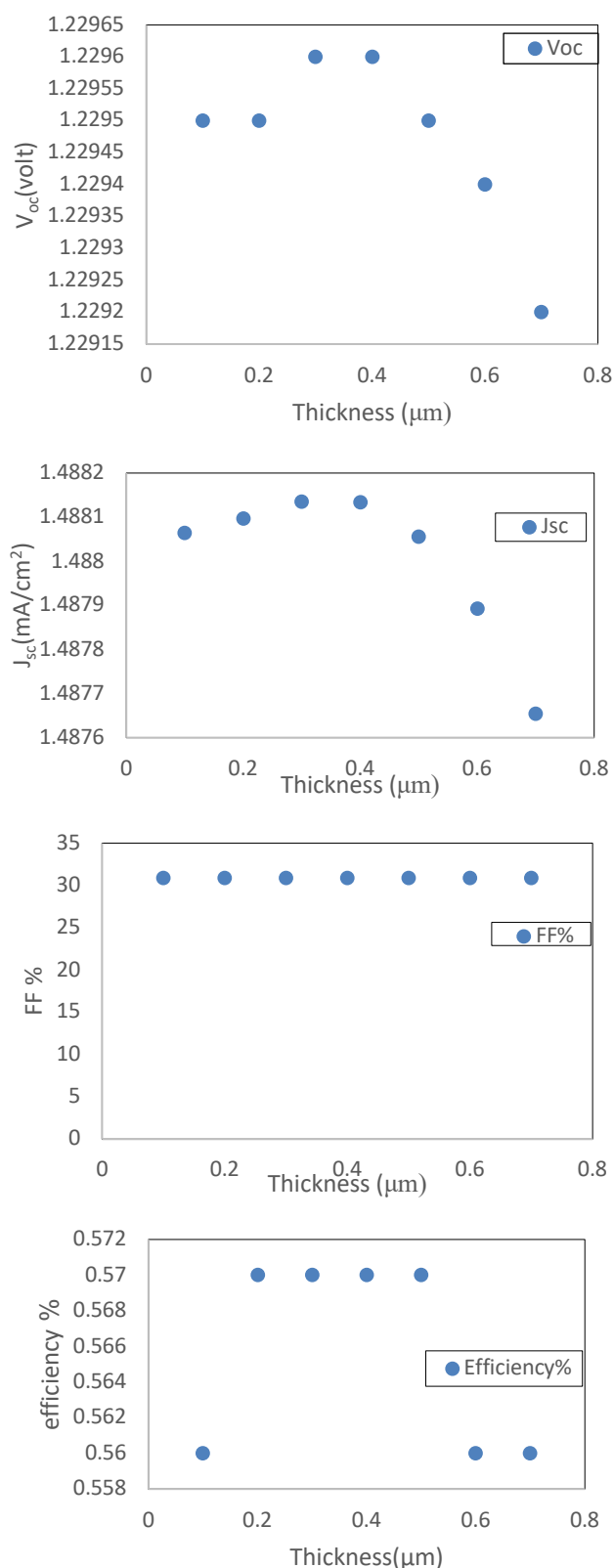


Fig. 4: Photoelectric parameters change by NiO thickness change.

4.2. Effect of the $\text{C}_{\text{s}_2}\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ layer thickness change on solar cells

The thickness of the absorber layer significantly affects the efficiency of the solar cell. The absorber layer thickness of $\text{C}_{\text{s}_2}\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ has varied between 0.1 μm and 1.6 μm . Fig. 5 Reflects the change of the photovoltaic characteristics with perovskite where is value the efficiency 0.56 at thickness 0.1 μm and 0.2 μm then thickness the efficiency of the solar cell increase at thickness 0.3 μm to value 0.57 then stay constant same the value to thickness 1.6 μm , 0.3 μm is considered to be the ideal thickness for the solar cell, where the value of efficiency 0.57%. By increasing the thickness of the absorber layer $\text{C}_{\text{s}_2}\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$, the depletion layer becomes extremely similar to the back contact, and more electrons are gathered for recombination Through the back contact. by looking at the (F.F/ thickness), (V_{oc} /thickness) and (J_{sc} /thickness) graphs, we can be noted that FF value increase at thickness 0.2 mm then stay constant to thickness 1.6 mm, while V_{oc} stay constant, finally the value of J_{sc} increases and then decrease with increasing thickness. The drawing data are shown in table number four.

Table 4: Photoelectric parameters change by $\text{C}_{\text{s}_2}\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ thickness change.

Thickness (μm)	V_{oc} (V)	J_{sc} (mA/cm^2)	F.F (%)	η (%)
0.1	1.2295	1.488562	30.86	0.56
0.2	1.2295	1.488002	30.88	0.56
0.3	1.2295	1.488079	30.88	0.57
0.4	1.2295	1.488097	30.88	0.57
0.5	1.2295	1.488105	30.88	0.57
0.6	1.2295	1.488127	30.88	0.57
0.7	1.2295	1.488184	30.88	0.57
0.8	1.2295	1.488118	30.88	0.57
0.9	1.2295	1.488202	30.88	0.57
1	1.2295	1.488163	30.88	0.57
1.1	1.2295	1.488255	30.88	0.57
1.2	1.2295	1.488211	30.88	0.57
1.3	1.2295	1.488171	30.88	0.57
1.4	1.2295	1.488142	30.88	0.57
1.5	1.2295	1.488309	30.88	0.57
1.6	1.2295	1.488233	30.88	0.57

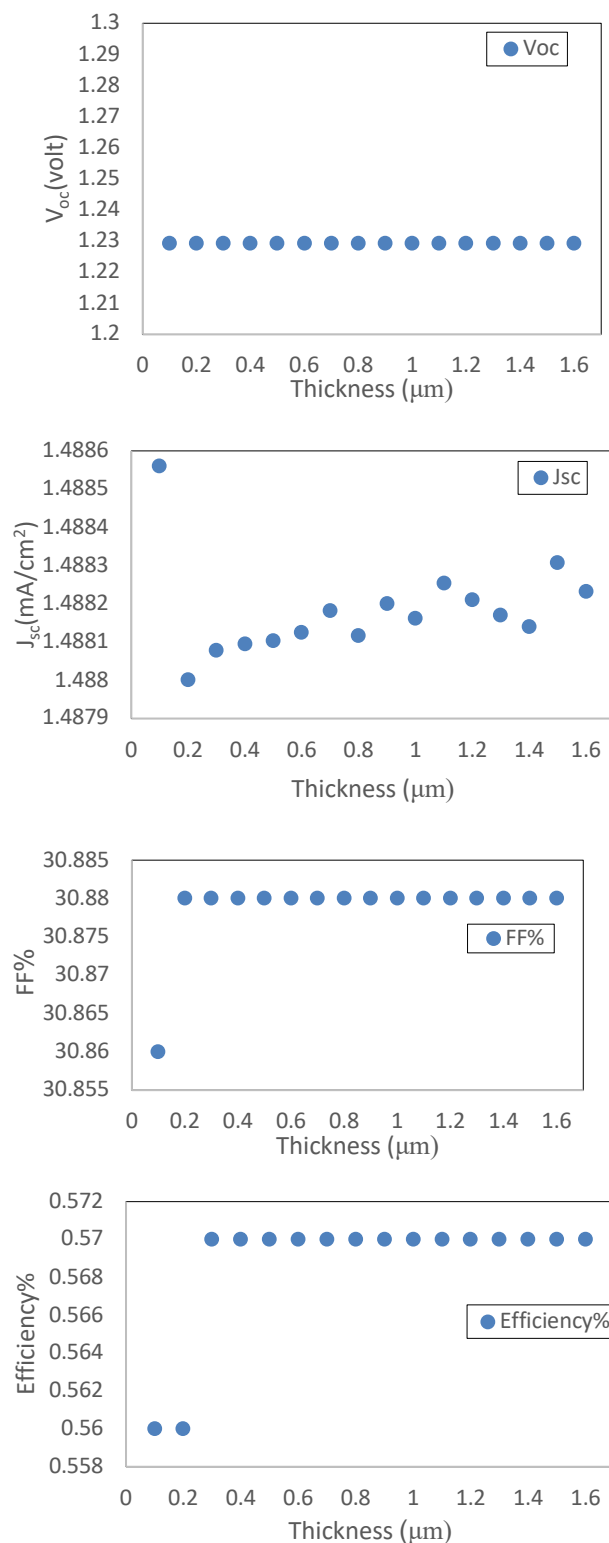


Fig. 5: Photoelectric parameters change by PCBM thickness change.

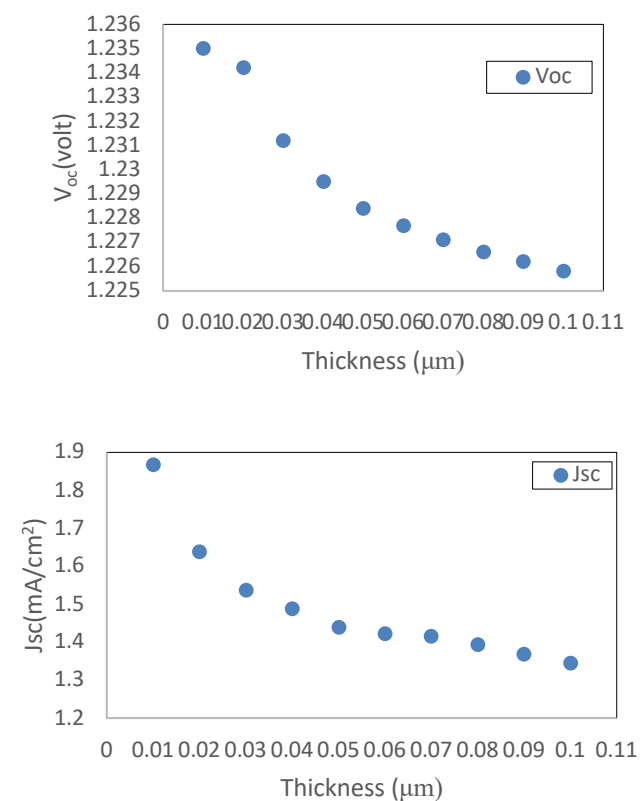
4.3. Effect of the PCBM (ETL) layer thickness change on solar cells

The value of the thickness of the (PCBM) layer has been changed from (0.01 to 0.1) µm. When the thickness of the

(PCBM) layer increases from 0.01 µm to 0.05µm, the PCE exhibit a steady state behavior till the thickness 0.05 where (PCE) reached to 0.57 then declined efficiency gradually to 0.53 at thickness 0.1 µm, while the J_{sc} decrease gradually with increase the thickness due to increase the resistivity with thickness, where highest value of J_{sc} at thickness 0.01µm reached to 1.868658 (mA/cm²), as well V_{oc} decreases gradually with increase the thickness, afterwards, at increases thickness from 0.06 µm to 0.1µm, decreases PCE, J_{sc}, and V_{oc}, while the FF increase from (24.74) to (32.36) in all interval from thickness (0.01 to 0.1)µm, which can be noticed in Fig 6. Additionally, increasing the PCBM thickness results in the creation of bigger pinholes and an uneven surface, which lowers the J_{sc} and V_{oc} and also decreases the PSC's overall effectiveness. The drawing data are shown in Table five.

Table 5: Change of Thickness for PCBM with device parameters.

Thickness (µm)	V _{oc} (V)	J _{sc} (mA /cm ²)	F.F (%)	η (%)
0.01	1.2350	1.868658	24.74	0.57
0.02	1.2342	1.638597	28.39	0.57
0.03	1.2312	1.537761	29.86	0.57
0.04	1.2295	1.488097	30.88	0.57
0.05	1.2284	1.440060	32.10	0.57
0.06	1.2277	1.423482	32.20	0.56
0.07	1.2271	1.415616	31.95	0.56
0.08	1.2266	1.393949	32.02	0.55
0.09	1.2262	1.369400	32.18	0.54
0.1	1.2258	1.346208	32.36	0.53



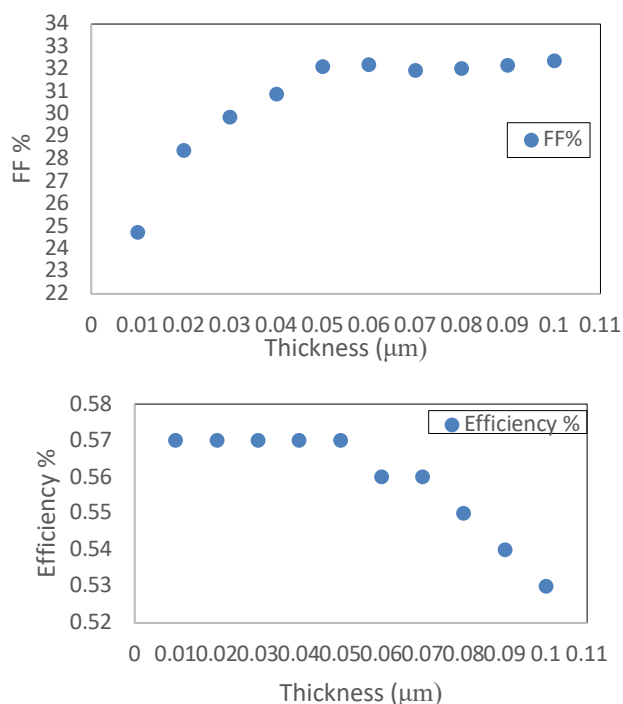


Fig. 6: Photoelectric parameters change by PCBM thickness change.

4.4. Effects of the Defect State of the Interface Defect Layer NiO/C_{s2}AgBi_{0.75}Sb_{0.25}Br₆/PCBM

Recombination and generation take place in the absorber layer of the proposed C_{s2}AgBi_{0.75}Sb_{0.25}Br₆, where the parameters of the defects have a substantial impact on the device's effectiveness. According to Fig 7, the difference of parameters for solar cell, versus the defect density. The proposed defect density values in this simulation range from 10¹³ cm⁻³ to 10²¹ cm⁻³, where we note that the defect density value for the absorber layer when increases from 10¹³ cm⁻³ to 10²¹ cm⁻³, the efficiency decrease significantly from (35.47 to 0.10) %, as for filling factor also decrease from (74.83 to 27.94)%, while current density of the photovoltaic parameters and open circuit voltage decrease at increasing defect, which indicates that the increasing defects serve as a recombination hub that shorten of carrier lifetime. We get it optimal defect efficiency through results obtained at 10¹³ cm⁻³, where is the efficiency value 35.47 %, F.F =74.84%, J_{sc} =32.810481 (mA/cm²) and V_{oc}=1.4444 (Volt).

Table 6: Change of defect for NiO/C_{s2}AgBi_{0.75}Sb_{0.25}Br₆/PCBM with device parameters.

Defect Nt(1/cm ³)	V _{oc} (V)	J _{sc} (mA/cm ²)	F.F (%)	η (%)
1*10 ¹³	1.4444	32.810481	74.83	35.47
1*10 ¹⁴	1.3794	16.181453	59.89	13.37
1*10 ¹⁵	1.3141	4.668733	48.31	2.96
1*10 ¹⁶	1.2729	3.044798	37.52	1.45

1*10 ¹⁷	1.2295	1.488097	30.88	0.57
1*10 ¹⁸	1.1699	0.615897992	27.56	0.20
1*10 ¹⁹	1.0944	0.428778448	26.12	0.12
1*10 ²⁰	1.0161	0.389453702	26.81	0.11
1*10 ²¹	0.9606	0.379571926	27.94	0.10

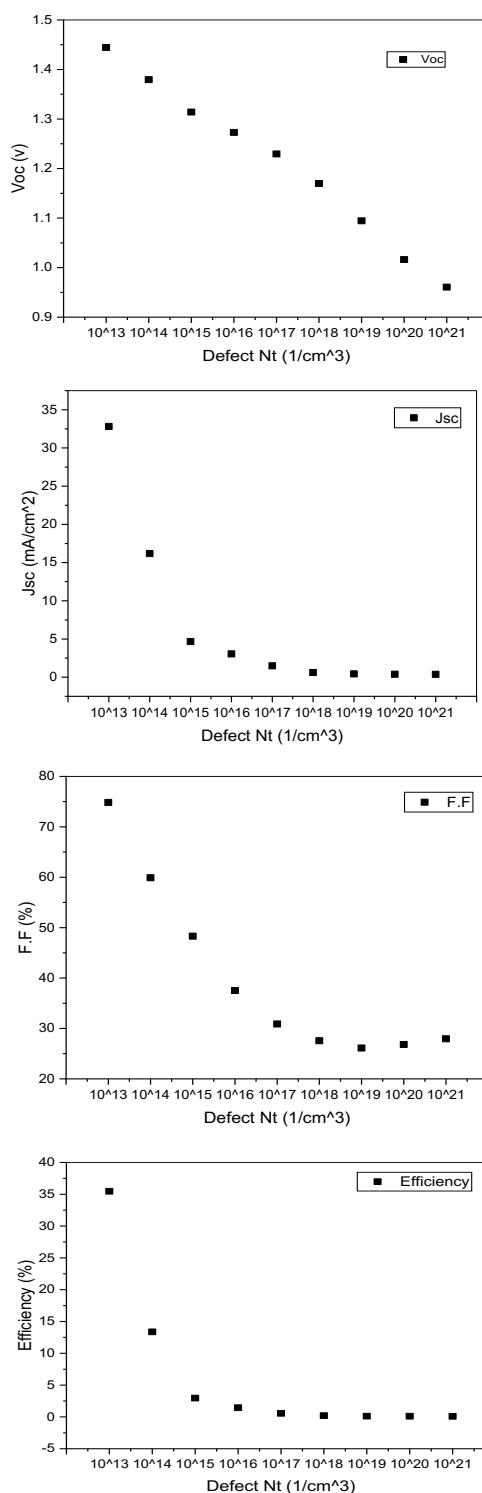


Fig 7: Defect density of the HTM/C_{s2}AgBi_{0.75}Sb_{0.25}Br₆/ETL layer.

4.5. Effect of annealing Temperatures for $Cs_2AgBi_{0.75}Sb_{0.25}Br_6$

In this model we note when the temperature achieved to 300 K, get greatest value of efficiency is 35.47% with $J_{sc} = 32.810481$ (mA/cm²), FF = 74.83% and $V_{oc} = 1.4449$, therefore the best result at the standard temperature. The temperature of the surrounding environment has a significant impact on productivity. For example, when the temperature rises from 100 K to 350 K, the PCE, FF, and J_{sc} increase due to an increase in the generation of electron-hole pairs in the perovskite materials, while the V_{oc} value decreases from 1.4545 to 1.3584 as shown in Fig 8, then decrease PCE, V_{oc} , FF at increase temperature from 400 k to 500 k. As shown in Fig 8, the open-circuit voltage reduces gradually generally with rising temperatures, and the device performance can be changed with temperature changed because the ability to of controlling recombination, create, and a collecting charge carriers, so the appropriateness temperature for the use of perovskites solar cells with $Cs_2AgBi_{0.75}Sb_{0.25}Br_6$ as PVSC is 300 k.

Table 7: The parameter of the for NiO/ $Cs_2AgBi_{0.75}Sb_{0.25}Br_6$ /PCBM heterojunction solar cells.

Temperature (K)	V_{oc} (V)	J_{sc} (mA/cm ²)	F.F (%)	η (%)
100	1.4545	29.145484	65.78	27.89
150	1.4545	29.145484	65.78	27.89
200	1.4709	30.778628	68.01	30.79
250	1.4836	31.952901	70.26	33.30
300	1.4449	32.810481	74.83	35.47
350	1.3584	33.498903	78.00	35.49
400	1.2359	34.013773	76.84	32.30
450	1.0863	34.362837	76.41	28.52
500	0.9506	34.634793	75.26	24.78

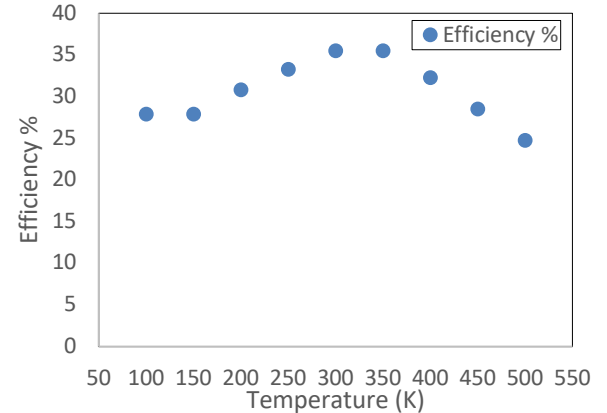
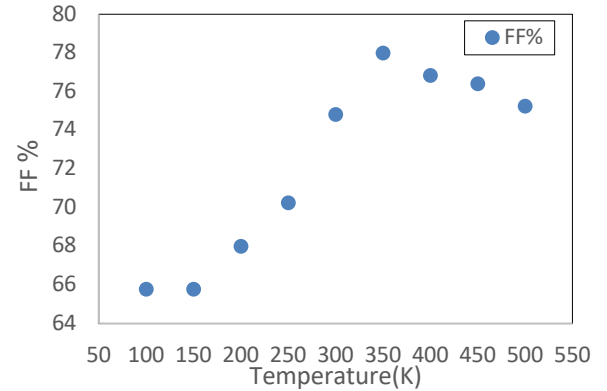
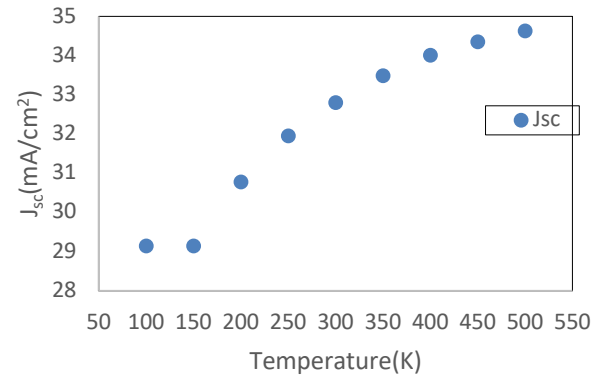
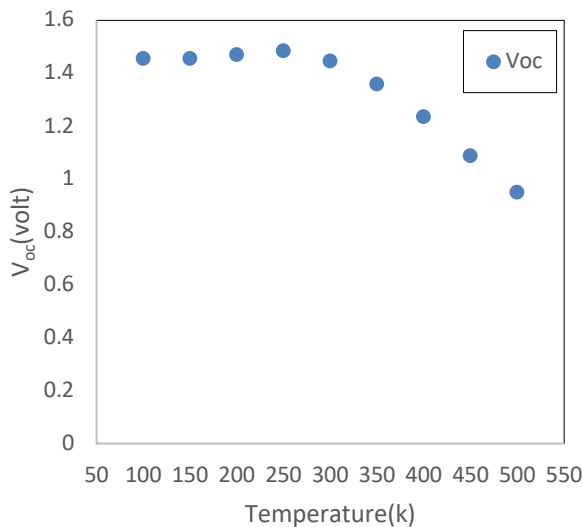


Fig. 8: Solar cell parameters change with temperature used in simulation changes.

4.6. Effect of different back contact material

Fig 9 shows the effect of change back contact on efficiency. Increase work function lead to increase in open-circuit voltage from 0.7280 (volt) to 1.4449 (volt) and also lead to increase in current density from 9.343896 (mA/cm²) to 32.810481(mA/cm²), And we note also, perovskite solar cell efficiency increasing with increase the work function where it reaches the value 35.47 % at Pt=5.6 eV, while filling factor increases with increasing the work function then decreases at Ir=5.25 eV. Pt is one of the prospective rear contact materials that can develop perovskite solar cell performance, according to the simulation results.

Table 8: The effect of metal contact change on the efficiency of the solar cell used in this simulation.

Efficiency %	F.F (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	Work Function (ev)	Metals
4.62	67.96	9.343896	0.7280	4.26	Al
5.95	71.17	9.422077	0.8880	4.42	Sn
7.47	73.63	9.495905	1.0680	4.6	Cu
8.32	74.71	9.533903	1.1680	4.7	Ag
9.18	75.61	9.571443	1.2679	4.8	Fe
14.43	76.81	13.230065	1.4197	5.1	Au
17.94	76.43	16.464692	1.4257	5.15	Ni
28.48	75.36	26.264640	1.4388	5.25	Ir
35.47	74.83	32.810481	1.4449	5.6	Pt

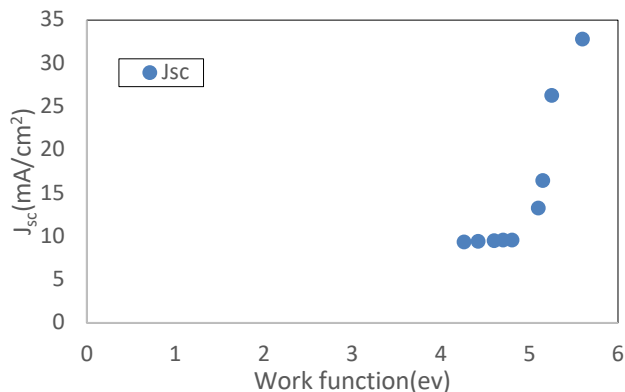
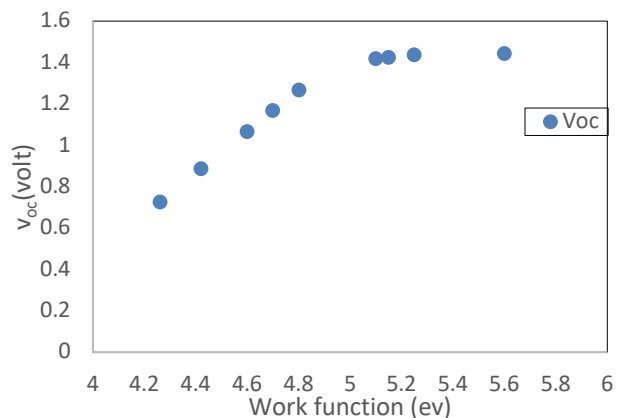


Fig. 9: Chang photovoltaic parameters V_{oc} , J_{sc} , FF and $\eta\%$ with metal function change.

5. Conclusions

In this work, has been analyzed perovskite solar cells with SCAPS-1D simulation software. The thickness was changed from $0.1\mu\text{m}$ to $0.7\mu\text{m}$ for NiO as the ETL, from $0.01\mu\text{m}$ to $0.1\mu\text{m}$ for PCBM and thickness for $\text{C}_{s_2}\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ from $0.1\mu\text{m}$ to $1.6\mu\text{m}$, a device's performance is good at $0.3\mu\text{m}$ thickness, therefore the ideal efficiency at $0.03\mu\text{m}$ thickness is 0.57% . Controlling in the defects lead for high efficiency of solar cells, the minimum defect density for the good efficiency was $1 \times 10^{13} \text{ N}_i(\text{1}/\text{cm}^3)$ where top efficiency of the structure NiO/ $\text{C}_{s_2}\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ /PCBM device which reached to 35.47% . After that the temperature change was studied from 100K 500K to observe the changes in the photovoltaic parameters (efficiency, voltage, filling factor and current density) of the device. Then, we changed work functions, which improved efficiency by 35.47% at 5.6eV . The simulation shows that the $\text{C}_{s_2}\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$, attain a PCE of 35.47% .

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