Design and Simulation of High Efficiency Tin Halide Perovskite Solar Cell

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Abstract-: Perovskite solar cells are becoming dominant alternative for the traditional solar cells reaching an efficiency of 22.1% in a short span of eight years (2008-2016). In this work, we designed a tin-based perovskite simulated model with the novel architecture of Glass/ZnO: Al/TiO_2/ $CH_3NH_3SnI_3$/CuI/Au and analyzed using the solar cell capacitance simulator (SCAPS-1D), which is well adapted to study the photovoltaic architectures. Using this software tool, we studied the effect of absorber layer parameters on the photovoltaic parameters of the designed model. We studied the thickness, defect density, band gap of the absorber layer and operating temperature of the model by simulating at various conditions. With the predicted parameters such as thickness (0.6 μm), defect density of absorber layer (10^{14} cm^{-3}), band gap(1.3 eV) the encouraging result of maximum power conversion efficiency(PCE) 24.82%, the short-circuit current density(J_{sc}) is 25.67 mA/cm^2, and fill factor(FF) is78.14% and open circuit voltage(V_{oc}) is 1.0413V are predicted. The results are indicating that the lead-free $CH_3NH_3SnI_3$ is having the great potential to be an absorber layer with suitable inorganic hole transport material like CuI to achieve high efficiency. This simulation model will become a good guide for the fabrication of high-efficiency tin-based perovskite solar cell.

Keywords: Perovskite, Design, Solar cell, SCAPS-1D, Simulation, Absorber layer

1.Introduction

From past few years, good efforts have been made to develop the perovskite-based solar cells[1-3]. In 2009, the maximum efficiency of perovskite solar cell was 3.8%[4]. By the improved fabrication methods, power conversion efficiency was reached to 22.1% by 2016[5]. Organic-In organic perovskite material has shown great potential to use as an absorber material in the thin film, solar devices which include efficient light harvesting characteristics[6], low processing cost, low temperature and solution processing[7-8].

Lead perovskite is the most used hybrid material but the toxicity of lead causing health and ecological challenge[9]. To overcome this problem, lead-based perovskite material is replaced with tin-based perovskite material[10]. From the literature, it was proved that tin-based perovskite material is a best alternative light harvester to the $CH_3NH_3PbI_3$[11]. Most of the experimental and theoretical analysis was done using lead perovskite material and very few studies were done using tin perovskite material. Few researchers observed that tin perovskite material is giving low *Voc* than the lead perovskite solar cell[12]. The maximum power conversion efficiency(PCE) of a reported tin halide perovskite solar cell was 6.4%[13]. The basic fundamentals regarding the architecture and materials can be get from the literature[14-21].

Tin based perovskite solar cells are suitable to the planar heterojunction architecture[22-23]. Tin perovskite solar cells are unstable in the ambient atmosphere; those are easily affected by the oxidation process. Due to the oxidation Sn^{2+} into Sn^{4+}, the device efficiency is reducing drastically. In few experiments by adding SnF_2 in the architecture; the device stability is greatly improved. This simulation work will give useful information on the material parameters and their effect on the performance of a device. Besides we used an inorganic material i.e. CuI as

a hole transporting material(HTM) and TiO_2 as an electron transport material(ETM) and $CH_3NH_3SnI_3$, is used as an absorber material. With the predicted parameters, the efficiency of the simulated device was reached to the 24.34%.

2. Methodology:

SCAPS is a computer based window application program written in C code. SCAPS was developed at the Department of Electronics and Information Systems of Gent University, Belgium[24]. SCAPS will work on using the basic semiconductor equations such as Poisson equation, continuity equation of electrons and holes. SCAPS calculates various profiles of the defined architectures those are steady state band diagram, carrier transport, and recombination and bulk defects. SCAPS is well adapted for modelling of various micro and polycrystalline thin films devices and photonic structures[25-27]. Thus, SCAPS-1D can be used to simulate the perovskite solar cell architectures. From the literature, experimental results are coinciding with the simulated results of the SCAPS-1D, to design and develop high efficiency Tin Halide Perovskite solar cell.

2.1.The architecture of device:

Tin based perovskite solar cell is having inverted planar heterojunction architecture. In this type of architectures, three layers are sandwiched between the two electrode materials. $CH_3NH_3SnI_3$ is used as an absorber layer, CuI as HTM, TiO_2 as an electron transport material (ETM), ZnO: Al acts as a front contact and 'Au' as a back contact as shown in Fig-1.

To design an efficient solar cell, we need to understand the process which was undertaken in between the layers of a solar cell. Following steps were observed, while converting electromagnetic energy into electrical energy in a solar cell.

a. When the photons are incident upon the absorber, then the photon interacts with the absorber and the excitons are produced i.e. the absorber jumps to the excited state.

b. The excitons separate into the electron and hole and moves towards the respective electrodes.

c. The separated carriers will be flown to an external circuit via contacts, electron losses its energy by passing through the load and comes back to initial position, the absorber comes to the ground state and it is ready to generate another electron-hole pair.

This process is said to be photovoltaic action. For the smooth flow of electrons, the absorber properties and other materials nature is very important.

2.2.Simulated parameters:

The parameters set for simulation are carefully selected from the literature and previous experimental work[28-40]. The Table.1 summarizes all the parameters used in the simulation process.The thermal velocities of electron and hole are taken in the order of 10^7 cm/sec. From the literature, the acceptor density value is taken as $3.2x10^{15}$ $1/cm^3$,the radiative recombination coefficient value is taken as $3x10^{-11}$ cm^3/sec, auger electron capture coefficient is $1x10^{-29}$ cm^6/sec and auger hole capture coefficient is $1x10^{-29}$ cm^6/sec[31].

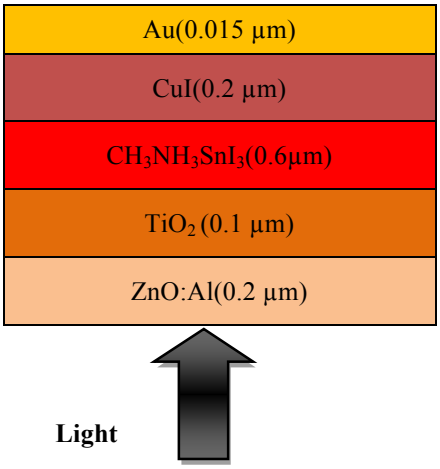

Fig-1: Architecture of Simulated model

Parameter	CuI	$CH_3NH_3SnI_3$	TiO_2	ZnO: Al
Thickness (µm)	0.200	0.700	0.100 [39]	0.200
Band gap(eV)	3.100 [32]	1.30 [37]	3.260 [39]	3.300 [40]
Electron affinity(eV)	2.100 [33]	4.170 [37]	4.200 [39]	4.600 [40]
Dielectric Permittivity	6.5 [34]	6.50 [30]	10.0 [39]	9.000 [40]
CB effective density of states ($1/cm^3$)	2.200E+19[35]	1.000 E+18 [37]	2.200E +18 [38]	2.200E+18[40]
VB effective density of states ($1/cm^3$)	1.800E+19[35]	1.000 E+19 [37]	1.800E +19 [39]	1.800E+19[40]
Electron thermal velocity(cm/S)	1.00E+7	1.00E +6	1.00E+7 [39]	1.00E+7 [40]
Hole thermal velocity(cm/S)	1.00E+7	1.00E +6	1.00E+7 [39]	1.00E+7 [40]
Electron mobility(cm^2/VS)	1.00E+2 [35]	1.60E-+0[37]	1.00E+2 [39]	1.00E+2 [40]
Hole mobility(cm^2/VS)	4.390E+1[36]	1.600 E+0 [37]	2.500E +1 [39]	2.500E+1[40]
Shallow uniform donor density ND ($1/cm^3$)	0	0	1.00E+19	1.00E+18[40]
Shallow uniform acceptor density NA($1/cm^3$)	1.00E+18	3.200 E+15 [37]	0	0

Table 1. The parameters set for $CH_3NH_3SnI_3$ based solar cell at 300K and at A.M. 1.5G.

3.The output of Simulation Studies:

3.1.Influence of thickness of absorber layer:

Absorber layer thickness plays a major role in determining the efficiency of the device. Absorber layer thickness variation affects the diffusion length of carriers. If the absorber layer thickness is very less, then absorption rate decreases, ultimately efficiency also decreases. If the absorber layer thickness is too high, then the charge carriers may not travel up to the charge collecting layers. So estimation of absorber layer thickness is necessary to achieve good efficiency.

The simulated parameters such as PCE, FF, Jsc, Voc of the CH3NH3SnI3 solar cells, with varying perovskite thickness is as shown in Fig-2. e maximum PCE of 30.49%, with Jsc=30.58mA/cm2,FF=87.84%,Voc=1.14V is achieved when the thickness reaches 0.6µm and in this case, the absorber layer is considered as defect free. When the thickness is increasing from 0.1µm to 0.8µm, the PCE and Jsc also increases up to 0.6µm, representing the increase in the generation of the electron, hole pairs in the absorber layer. From 0.6µm to higher thickness the PCE and Jsc both undergo saturation representing the recombination of charges inside the absorber layer before reaching to the contacts. For the large thickness of perovskite solar cell 'Voc' decreases; because of 'Voc' is a function of Jsc and Jo(as given in equation (1) which depends upon the perovskite layer thickness[40].

$$V_{OC} = \frac{KT}{e} l_n \left[\frac{J_{SC}}{J_0} + 1 \right] \quad \text{---------(1)}$$

Where,

K= Boltzmann constant

T= Operating temperature

Jsc= Short circuit current density

Jo =Reverse saturation current

e = Electron charge

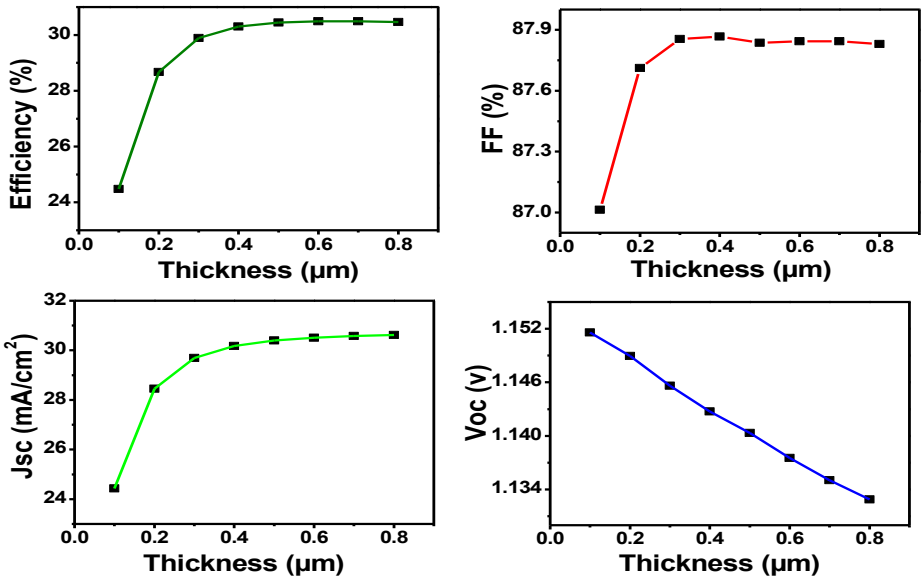

Fig-2 Representing the variation of solar cell parameters with the thickness of absorber layer.

From Fig-2, the open circuit voltage decreases slightly with the increase of thickness, it represents that recombination, generation, a collection of charge carriers can be controlled by adjusting the

thickness of absorber layer. For further studies on efficiency, the desired thickness of absorber layer
is taken as 0.6µm.

3.2. Influence of Defect Density of Absorber:

Generation, recombination, transportation process occurs inside the absorber layer, so the absorber layer quality and defect parameters greatly effect the device performance[41]. To explain the defect energy levels of perovskite layer, Gaussian distribution model is used[42-43].

The corresponding equations of Gaussian distribution model are:

$$g_D(E) = G_{Md} \exp\left[-(E - E_{pkd})^2 / 2\sigma_d^2\right] \quad -(2)$$

$$g_A(E) = G_{Ma} \exp\left[-(E - E_{pka})^2 / 2\sigma_a^2\right] \quad -(3)$$

The variation of solar cell parameters, with the defect density, is as shown in Fig-3. In the simulation model, the defect density is varied from 1013 cm-3 to 1018 cm-3. It was observed that, if the defect density absorber layer is increasing from 1013 cm-3, to 1018 cm-3, the photovoltaic parameters randomly decreases and at 1018 cm-3 the PCE reaches to the 2.12% and fill factor is 29.16%,Jsc=13.12 mA/cm2, Voc=0.55V. An abrupt decrease in the PV parameters with the increase in defect density indicating that the increased defects act as a recombination center which decreases the lifetime of carriers. The minimum defect density of the absorber layer is predicted as 1014 cm-3, at this condition the maximum attainable PV parameters are efficiency is 25.03%, fill factor is 78.34%,Jsc is 29.69mA/cm2,Voc=1.08V.

G_{Md} G_{Ma} are effective defect densities

σ_d *and* σ_a are the standard energy deviations of the Gaussian donor and acceptor levels.

E_{pkd}, E_{pka} are donor peak energy position measured positive from Ec and the acceptor peak energy position measured positive from Ev.

The recombination centers at the deep energy levels are known as Schokley-Red Hall non-radiative(SRH) recombination center[44]. SRH model clearly explains the defect density of a perovskite layer.

SRH model equation is as follows

$$R = \frac{np - n_i^2}{\tau_p\left(n + N_C e^{\frac{(E_g - E_t)}{KT}}\right) + \tau_n\left(p + N_V e^{\frac{E_t}{KT}}\right)} \quad (4)$$

Here ‘ R’ is the rate of recombination
‘n’ and ‘p’ are the concentration of electrons and holes.

If qV>KT; then n_i^2 will be neglected.

τ_n, τ_p are the lifetime of electron and hole.

Same as the simulation of lead perovskite, when the defect density increases, the cell performance dropped significantly. However, tin-based perovskite displays excellent charge transport in real devices[45-46].

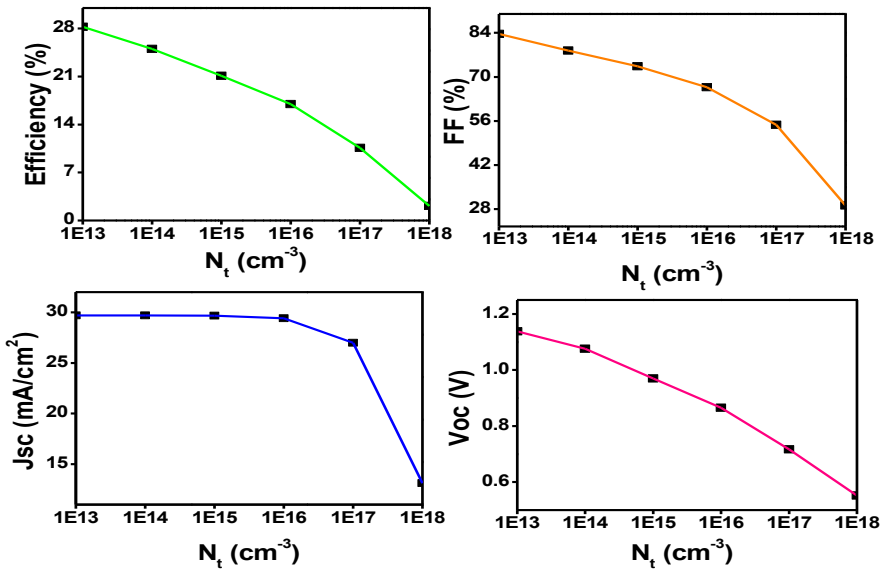

Fig-3: Representing the variation of solar cell parameters with the defect density

3.3.Influence of absorber bandgap:

Previous experiments showed that tin halide perovskite material band gap can be tunable in a range between 1.3 eV to 2.15 eV. In an article by Albrecht.et.al., did simulations of C-Si/tandem perovskite material and finally suggested the optimized band gap, which yields the highest performance of the device[47]. In a perovskite solar cell band gap of the absorber layer plays a major role in optical absorption. The perfect band matching of HTM, an absorber layer, ETM benefit for the improvement of the device performance.

Below fig-4 represents the variation in the PV parameters by changing the band gap of the absorber material from 1.3 eV to 2.15 eV, by considering the former suggested thickness(0.6 µm) and the defect density of absorber layer as(10^{14} cm^{-3}). When the band gap increases from 1.3 eV to 2.15 eV, the PV parameters such as efficiency, fill factor, short circuit current density decreases, but the open circuit voltage slightly increases. The reason behind the decrease in the *Jsc* is decreased in the optical absorption of the photons having the less energy than the band gap of the perovskite material. The open circuit voltage is a function of the band gap, as the higher band gap leads to the higher *Voc* and low radiative recombination. The reduction in the fill factor by the increase of band gap is due to the mismatch between the HTM and absorber layer. Above all reasons finally, leads to the poor efficiency of the model at higher band gap of the absorber layer.

Hence, the necessary band gap of tin perovskite absorber layer is 1.3 eV, attaining the efficiency is 24.82%, open circuit voltage (V_{oc})is 1.04 V, short-circuit current density (J_{sc}) is 30.50 mA/cm^2, Fill factor is 78.14%.

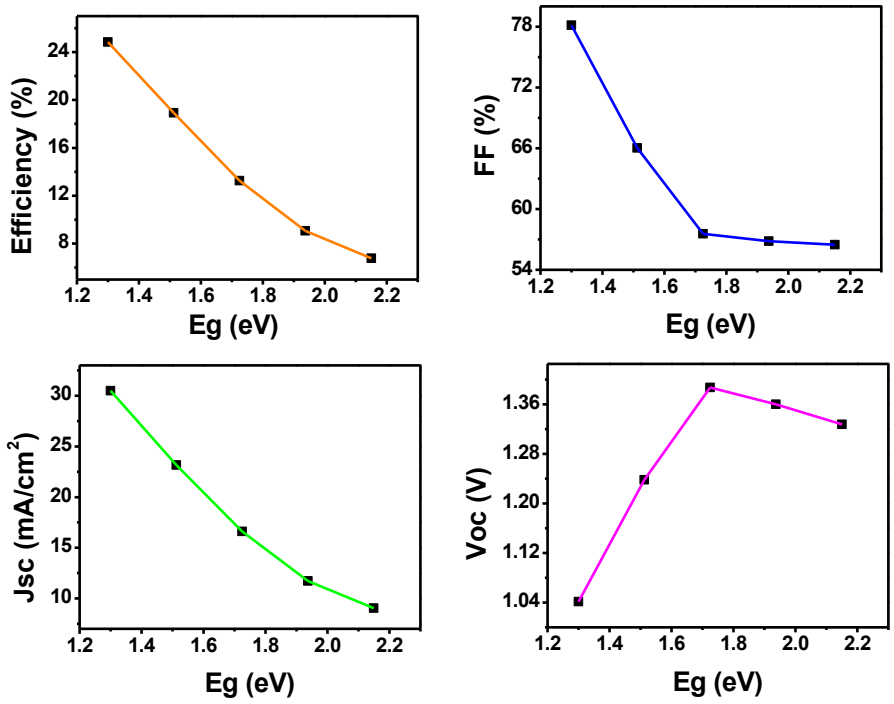

Fig-4: Representing the variation of solar cell parameters with the band gap(eV) of absorber layer

3.4.Influence of operating temperature:

Working temperature plays a major role in the performance of a device. Generally, the testing temperature of a solar cell device is at 300^0 K, but at the installed conditions, the working temperature is more than 300^0K. In this case, the absorber is considered with the defect density of 10^{14} cm^{-3}. In this simulation model Glass/ZnO: Al/TiO_2/$CH_3NH_3SnI_3$/CuI/Au the operating temperature varied from 300^0K-450^0K, then the changes in the characteristics are observed and given in Fig-5. As the temperature increases from 300^0 K to 450^0 K, then the efficiency drops to18.60% and fill factor is 71.1%,J_{sc}=28.66 mA/cm^2,V_{oc}=0.93V. Increased temperature may lead to the more stress and deformation resulting in increased interfacial defects and poor interconnectivity between the layers. The increase in defect density leads to increased recombination and reduction in diffusion length. Decrease in diffusion length increases of series resistance, by this fill factor and efficiency will be decreased[48]. By the increase of temperature, parameters like electron and hole concentrations, carrier mobility's, band gaps of the materials are affected because of change in the resistance and finally device efficiency drops. To achieve the best performance the operating temperature of the model is considered as 300^0K. At this temperature the maximum achievable efficiency of the model is 25.03%, fill factor is 78.34%, J_{sc}=28.66 mA/$cm^{2,}$ and V_{oc}=1.08V

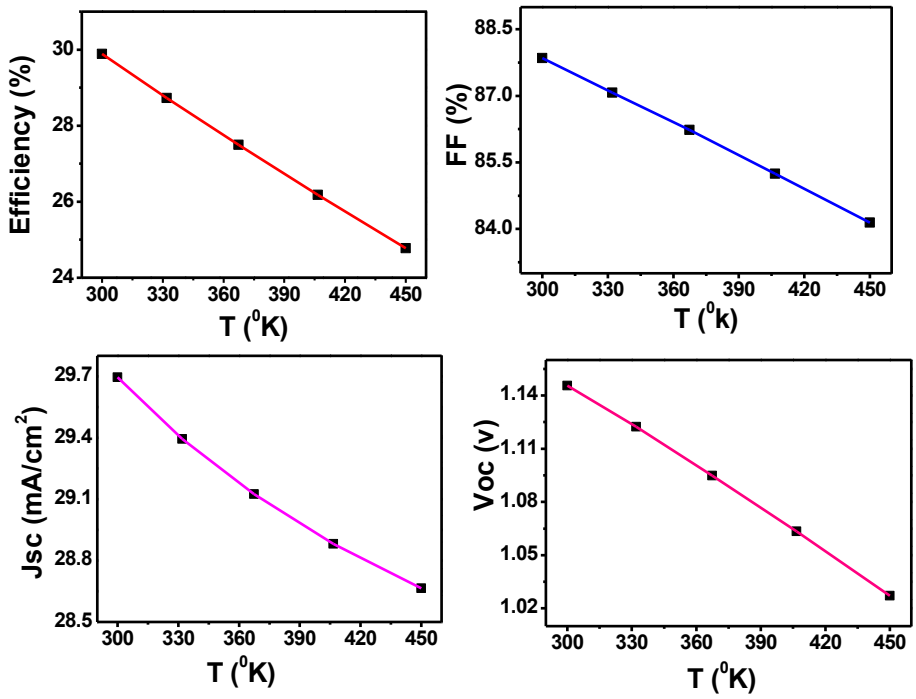

Fig-5: Variation of solar cell parameters with the Operating temperature

3.5. Simulated J-V curve:

With all the predicted parameters of the model those are thickness of absorber layer is 0.6 µm and the total defect density is 10^{14} cm-3, the perovskite solar cell model is simulated, and the final J-V curve is as shown in the fig-6, Represents the characteristic of perovskite solar cells giving the open circuit voltage (V_{oc})is 1.04 V, short-circuit current density (J_{sc}) is 30.50 mA/cm^2, Fill factor is 78.14%, and PCE is 24.82 %.

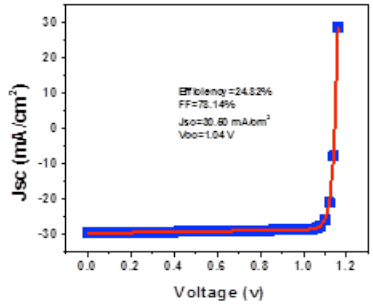

Fig-6:J-V curve of Glass/ZnO:Al/TiO_2/ $CH_3NH_3SnI_3$/CuI/Au

4.Conclusion:

Tin based architecture for perovskite solar cells are simulated using the SCAPS-1D. The thickness of absorber layer was changed from 0.1µm to 0.8µm, the good performance of a device is achieved at the thickness of 0.6 µm hence the thickness of the absorber layer is set at 0.6µm. Decreasing the defects in the absorber layer and improving the material quality, the device performance will be increased and the minimum defect density for the good efficiency was10^{14}cm^{-3}. The suitable band gap for the good absorption and to reach higher performance is predicted as 1.3eV. The operating temperature was changed from 300K to 450K to observe the changes in the PV parameters of the device. The simulation demonstrates that the $CH_3NH_3SnI_3$, attain a PCE of 24.82%.

The architecture of tin-based perovskite solar cell is having the remarkable characteristics like non-toxicity, easy fabrication, and low cost. By simulating the model, the material parameters influences on the device performance are analyzed in detail. Results are showing that CuI is a best alternative organic hole transport material for high-efficiency devices. Astoundingly, a maximum PCE of 24.82% and J_{sc}=25.67mA/cm^2, FF=75.24%,V_{oc}=1.26V are attained.

These simulation results illustrating that the material(absorber) parameters will largely affect the device performance.

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