

Optical and Electrical Simulation of $\text{CH}_3\text{NH}_3\text{PbI}_3$ -Based Perovskite Solar Cells

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ABSTRACT— Recently, organo–metal halide perovskites have attracted much attention of the scientific community relating to their successful application in the absorber layer of low-cost solar cells. However, enough is known about the material and device properties, to realize that much remains to be learned.

In this paper, the electrical and optical properties of perovskite solar cells are investigated using the COMSOL Multiphysics simulation program. It is a study of the influences of carrier diffusion length (L), dielectric constant (ϵ_r), the valence band offset (VBO) of absorber/hole transport materials (HTM) and illumination intensity on fill factor (FF), short-circuit current density(J_{SC}), performance (PCE), and open-circuit voltage(V_{OC}). Also, J-V characteristics are calculated for different ϵ_r values. The simulation results point to the great dependence of efficiency on the carrier diffusion length of absorber layers. It is shown that, to obtain a high rate of efficiency, the relative permittivity should not be higher than 45.

KEYWORDS: Perovskite solar cells, Fill Factor, Power Conversion Efficiency, Simulation.

I. INTRODUCTION

Mixed organic–inorganic halide perovskites solar cells are encouraging as future-generation photovoltaic tools due to their benefits such as easy processing, high cost-effectiveness and efficiency [1, 2]. Principally, perovskite solar cells can serve as planar-junction devices or mesoscopic-junction devices. A performance rate of over 15% has been obtained for the two categories [3-5].

Proposing that the mesoporous is not the pre requisite for high performance [6].

The highest performance of 19.3% is attained by optimizing the expansion atmosphere and the band positions [7-9]. The dielectric and optical features of perovskites are vital to optimize the structure of perovskite solar cells [10,11]. It was obtained that the dielectric constant of perovskite displayed a frequency dependency and temperature [12-14].

In 1994, a broadly cited optical frequency dielectric constant of 6.5 was reported for perovskites. It was computed from diffuse powder reflection, and currently declined to 5.06 inferred from detailed analysis of transmittance and reflectance of films [15-17]. A great dielectric constant has been also reported for the iodide recently, in which another extra factor of 1000 is made under the illumination of one sun when measured at very low frequencies (<1 Hz) [18-20].

Detailed studies of the dark IV features of perovskite solar cells have suggested that the low-frequency relative permittivity of perovskites is like that of HTL [21]. However, the impact of dielectric constant on light characteristics is not known yet. Highly efficient perovskite solar cells generally benefit from organic hole transport materials (HTMs) which are often costly and have bottom low hole mobility [22]. Perovskite solar cells based on inorganic p-type semiconductors are expanded, but their performance is lower than that of cells based

on organic materials.¹⁴ This shows the valence band offset ($\text{VBO} = E_{\text{V,perovskite}} - E_{\text{V,HTM}}$) of the absorber/HTM layers should be significant.

The literature on the topic is growing, both in fully simulating and empirical forms. Simulation studies provide deep insights into the function of cells and the restrictions to their efficiency. Those studies are generally appropriate for monitoring, fast characterization and prediction of panel efficiency [23, 24].

A simulation program is very beneficial for clarifying the key roles of different parameters in final operations. In this paper, to determine the action mechanism of perovskite devices, theoretical analyses are performed of the influence of carrier diffusion length, VBO and dielectric constant on the photovoltaic parameters of perovskite solar cells. The COMSOL Multiphysics simulation software is used for both electrical and optical responses [25].

II. DEVICE SIMULATION PARAMETERS

COMSOL is a window application program, developed at the University of Harford with which the user can set the parameters or in which the results are calculated [25]. COMSOL analyses the physics of the model and it explains the recombination profiles, electric field distribution, carrier transport mechanism and individual current densities. The continuity equations of electrons and holes are:

$$\frac{1}{q} \frac{dJ_n}{dx} = R_n(x) - G(x) \quad (1)$$

$$\frac{1}{q} \frac{dJ_p}{dx} = G(x) - R_p(x) \quad (2)$$

where, J_n is Electron Current Density, J_p is Hole Current Density and R is generation rate.

The Drift and Diffusion Equations are:

$$J_n = \frac{I_n}{A} = -nq\mu_n E + qD_n \nabla n \quad (3)$$

$$J_p = \frac{I_p}{A} = -pq\mu_p E - qD_p \nabla p \quad (4)$$

where, D_n is electron diffusion coefficient, μ_n is electron mobility and μ_p is Hole mobility.

Figure 1 explains the simulation process using COMSOL.

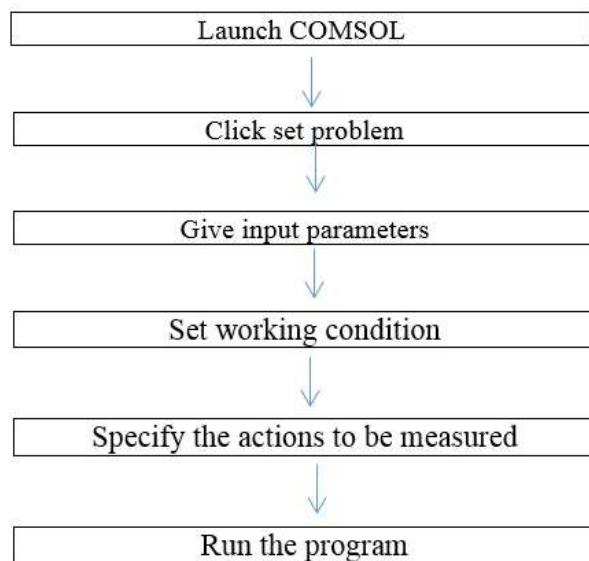
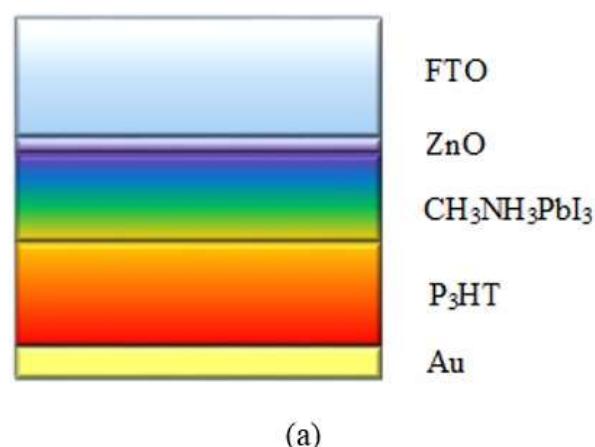


Fig. 1. COMSOL working procedure.

A simulation structure was devised for n-i-p perovskite solar cells. As displayed in Fig. 2, the specifications included FTO (500 nm)/ZnO (ETM layer, 50 nm)/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ (absorber layer, 400 nm)/ P_3HT (HTM layer, 350 nm)/Au (100 nm). The standard AM1.5G spectrum was presented as the accident light source, and the k , n values of the stack as well as FTO, ZnO, $\text{CH}_3\text{NH}_3\text{PbI}_3$, P_3HT , and Au values were derived from the literature [26].



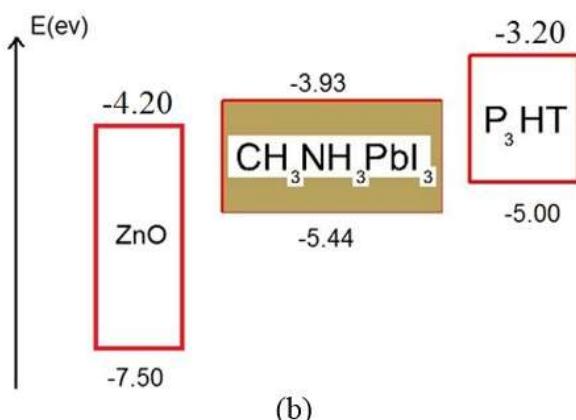


Fig. 2. (a) Schematic diagram of the perovskite solar cells, (b) Energy levels of different layers of the perovskite solar cells

The photo-generation rate obtained through the optical computation was used to determine the electrical characteristics, where only the three layers of n-type ZnO, CH₃NH₃PbI₃ and p-type P₃HT were taken into consideration. The simulation parameters are presented in Table 1 [27-30].

Table 1. Semiconductor parameters used for the simulation

	ZnO [27-28]	CH ₃ NH ₃ PbI ₃ [29]	P ₃ HT [30]
Thickness (nm)	50	400	350
E _g (eV)	3.3	1.5	1.8
X (eV)	4.6	3.9	3.9
ε _r	9	6.5	3
N _C (cm ⁻³)	2.2×10 ¹⁸	2.5×10 ²⁰	10 ²⁰
N _V (cm ⁻³)	1.8×10 ¹⁹	2.5×10 ²⁰	10 ²⁰
μ _e (cm ² /Vs)	100	50	10 ⁻⁴
c (cm ² /V _s)	25	50	10 ⁻⁴
N _D (cm ⁻³)	10 ¹⁸		
N _A (cm ⁻³)	--	10 ¹³	10 ¹⁶

Here, ε_r is the relative permittivity, N_D and N_A denote the donor and acceptor densities, X refers to the electron affinity, N_V and N_C are the effective densities of the states of valence and conduction bands respectively, E_g is the band gap energy, and μ_e and μ_h are the mobility rates of electrons and holes respectively.

Ideal Ohmic and Schottky contacts with the surface recombination rate of 10⁷ cm/s were used in the case of back and front contacts.

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III. RESULTS AND DISCUSSION

The spectral dependence of the absorptance of perovskite solar cells on the thickness of their absorber layers is shown in Fig. 3.

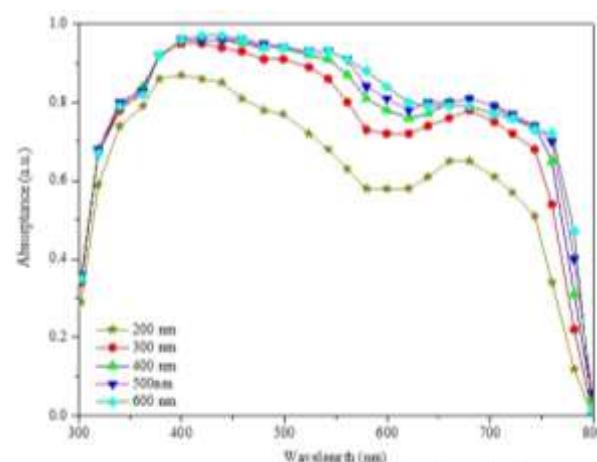


Fig. 3. Spectral affiliation of the light attracted by absorber layers with different thicknesses

The absorptance is enhanced quickly with the increase of the absorber thickness, and it reaches saturation for a thickness of more than 400. The integrated absorptance of absorber layers with different thicknesses between 200 and 600 nm is displayed in Fig. 4(a). It increases with the increase of the absorber layer thickness, and a value of upper 0.75 is obtained at 600 nm. Also, the generation current, where the photo-generated carriers are prepared without recombination, i.e. the theoretical maximum current, is enhanced with an increase in the absorber layer thickness and reaches around 21 mA/cm² at 600 nm, as shown in Fig. 4(b).

Then illumination intensity was investigated as displayed in Fig. 5. To design the J-V curves of the cells, different illumination intensities were tried. The results show that, with the

increase of illumination intensity, J_{SC} would increase but V_{OC} would remain almost unchanged.

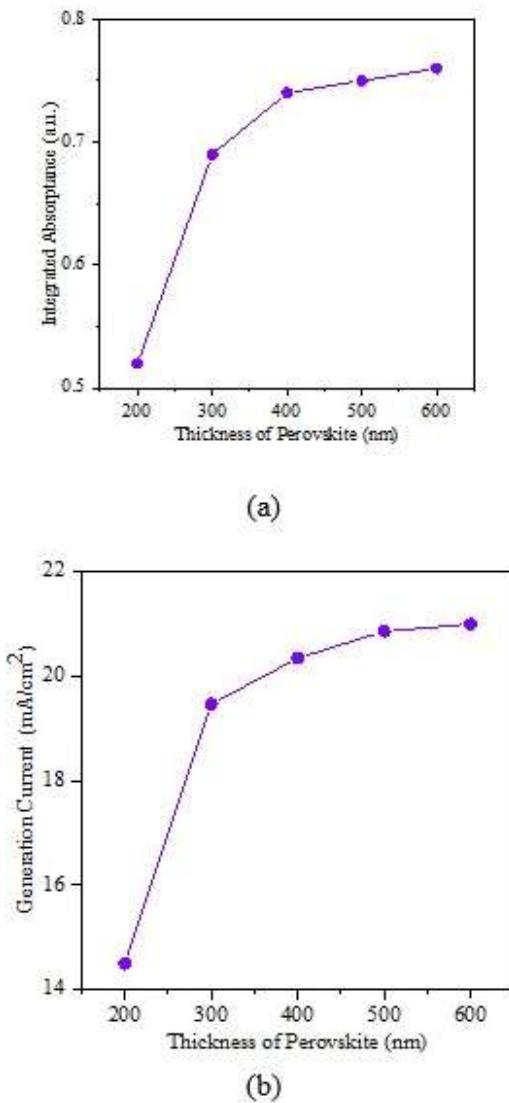


Fig. 4. Effect of absorber thickness on (a) integrated absorptance and (b) generation current

The absorber quality, namely carrier diffusion length for hole (L_p) and electron (L_n), is the main parameter of the efficiency of perovskite solar cells. The L_n of 100 nm estimated from the single-step solution-processed perovskite thin films was the minimum value, but it increased by one order (i.e., $>1 \text{ lm}$) with Cl doping [19]. Furthermore, high crystalline films were entrusted by vapor deposition methods and the sequential, propose that the L_n as in the scale 100 nm.

In addition, high crystalline films were deposited by the sequentia and vapor

deposition methods, suggesting that the L_n was in the scale of hundreds of nanometers.

In addition, an efficiency rate of higher than 12% was gained in perovskite solar cells with a thickness about 300 nm, denoting that maximizing the quality of absorber films can lead to L_n and $L_p > 300 \text{ nm}$ [30-33].

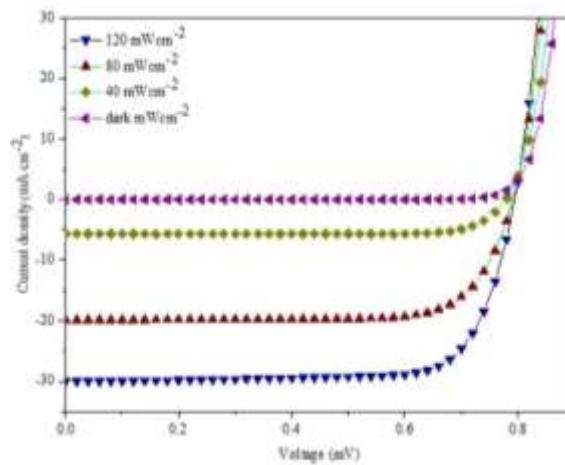


Fig. 5. J-V curves of the perovskite solar cells simulated with different illumination intensity values

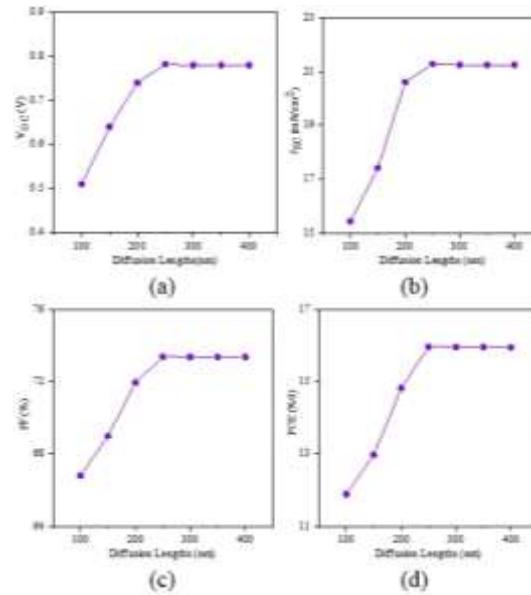


Fig. 6. Effect of the diffusion lengths (L) of perovskite on (a) V_{OC} and (b) J_{SC} and (c) FF and (d) PCE.

The plot of the PV parameters versus the carrier diffusion length of the perovskite layers is shown in Fig.6. The L changed from 100 nm to 400 nm. In this study, L_p and L_n are identical. It was observed that, with the

increase of the absorber diffusion length, i.e. with the improvement of the film quality, J_{SC} , V_{OC} , FF and PCE increased. This increase was due to the suppression of recombination. And then more $L=250$ nm they are constant. It means that, at $L=250$ nm, the values $PCE = 15.9\%$, $J_{SC} = 21.2\text{mA}/\text{cm}^2$, $V_{OC} = 0.78\text{ V}$ and $FF = 74.2\%$ could be obtained.

If the absorber thickness is too low, full light absorption does not occur and the cell performance is low. When the absorber thickness is increased, the PCE is improved gradually. Once the absorber layer is too thick, it is not possible to collect the photo-generated carriers effectively because, before they are recombined, they must move through the absorber to reach the layers that collect them. The role of the charge diffusion length is critical in designing the thickness and structure of the perovskite layer. The effect of the absorber layer thickness on the performance of perovskite solar cells with different charge carrier diffusion lengths is displayed in Fig. 7.

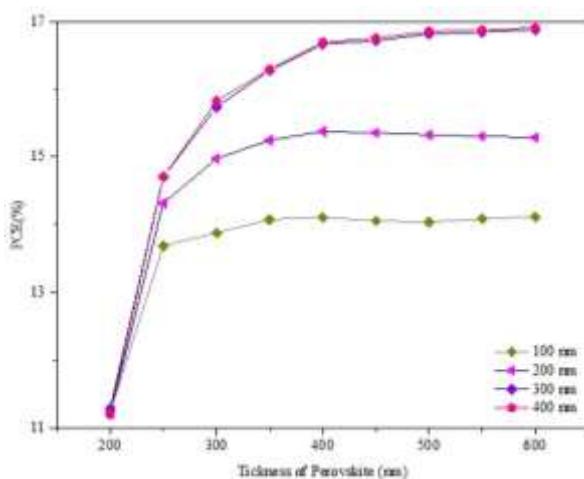


Fig. 7. Effect of the absorber thickness on PCE with different diffusion lengths.

As it can be seen, the performance is enhanced with the increase of the absorber thickness, but, when the perovskite thickness is above 400 nm, the performance is almost constant.

The influence of the dielectric constant on the performance of the perovskite solar cells was evaluated by the change from 5 a.u. to 135 a.u. versus the PV parameters. The results are given in Fig. 8.

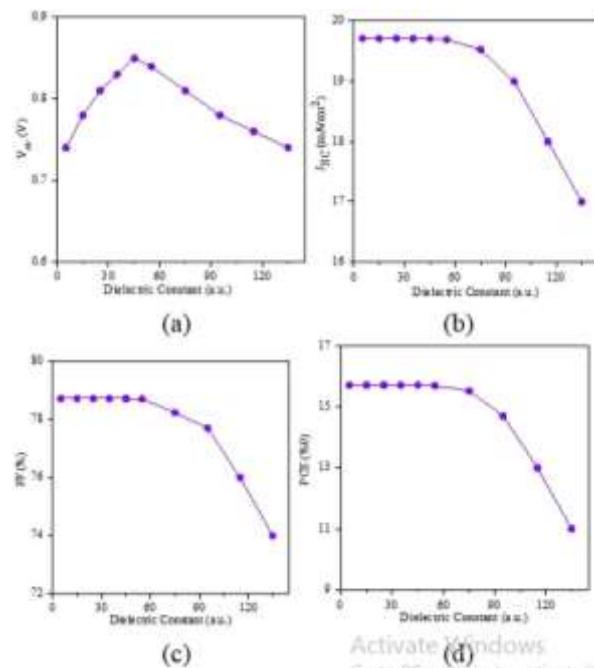


Fig. 8. Effect of the dielectric constant (ϵ_r) of perovskite on (a) V_{OC} and (b) J_{SC} and (c) FF and (d) PCE.

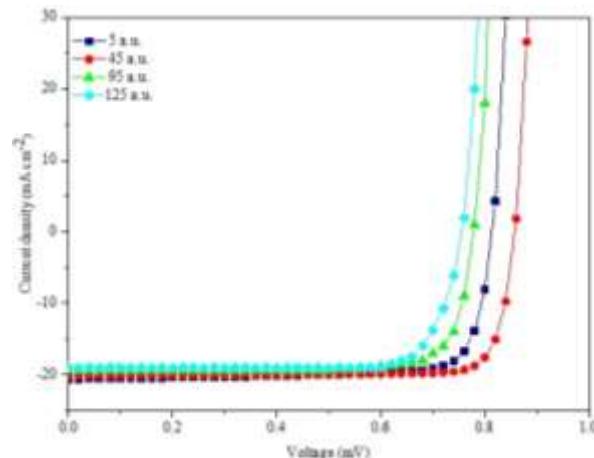


Fig. 9. J-V curves of the perovskite solar cells simulated with different dielectric constant values.

According to the figure, with the increase of ϵ_r to 45 a.u., J_{SC} , FF and PCE values remain unchanged. Then, with further increase of ϵ_r , those values decrease. As for V_{OC} , however, it first increases with the increase of ϵ_r to 45 a.u. and then decreases with higher ϵ_r . Also, as presented in Fig. 9, the influence of different dielectric constants on the J-V characteristics of the solar cells was examined. The same result was reached in this examination; with an increase of ϵ_r up to 45 u.a., V_{OC} increases and then decreases with more raise in ϵ_r .

The effect of the band offset values of the absorber/HTM interface on the performance of the perovskite solar cells was calculated according to the PV parameters. The results are displayed in Fig. 10. As the figure shows, for $\text{VBO} < 0$, with an increase of VBO, the values of V_{OC} , FF, PCE increase, while the short-circuit current density remains almost unchanged.

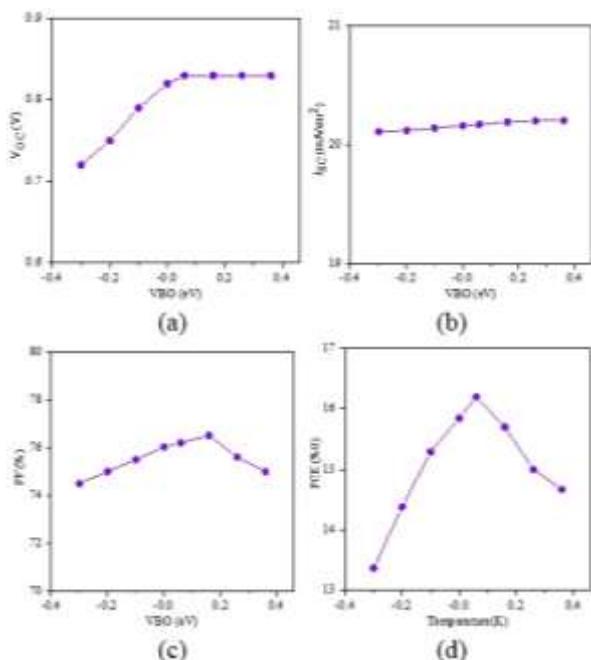


Fig. 10. Effect of valence band offset (VBO) on (a) V_{OC} and (b) J_{SC} and (c) FF and (d) PCE

In the case of $\text{VBO} = 0.13$, Fig. 10 suggests a high performance. With further enhancement of VBO, a double-diode like a curvature is presented, resulting in low FF but similar V_{OC} . However, VBO is negative for most HTMs, such as CuSCN (-0.13 eV) and CuI (-0.23 eV) [34-36]

IV. CONCLUSION

In summary, planar PSCs have been simulated, which calculates both the optical and electronic responses of a solar cell structure. The effect of carrier diffusion length, dielectric constant, the valence band offset of absorber/HTM and illumination intensity on the PV parameter of perovskite solar cells as well as the spectral affiliation of the light attracted by absorber layers with different thicknesses. Also, the effect of absorber

thickness on the integrated absorptance and generation current was calculated. According to the simulation results, the optimal carrier diffusion length, absorber layer thickness, VBO and dielectric constant were 250 nm, 400nm, 0.13 and 45 a.u. respectively. Also, the optimal values and the calculated photovoltaic parameters were $\text{VOC} = 0.81 \text{ V}$, $\text{J}_{\text{SC}} = 20.04 \text{ mA/cm}^2$, $\text{PCE} = 15.36\%$ and $\text{FF} = 75.03\%$. Based on this study, some insight is provided into the working mechanism of perovskite devices and the properties of perovskite films. This can be of benefit for designing better perovskite solar cells.

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