

High-Performance MAPbI₃-Based Perovskite Solar Cell: Design, Simulation, and Analysis of Optoelectronic Properties and Efficiency Metrics Using SILVACO TCAD

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Abstract

In this work, a perovskite solar cell (PSC) based on MAPbI₃ has been designed, modeled, implemented, investigated and analyzed in Silvaco TCAD environment. A thin layer of MAPbI₃ serves as the photoactive absorber. A thinner layer of PEDOT: PSS based organic material acts as the hole transport layer to enhance the hole transport towards the ITO electrode and a thin layer of ZnO based inorganic material serves as the electron transport layer to assist electron transport to the aluminum electrode. Simulations have been carried out to obtain the energy band diagram and the electric field profile as well as the contour and 2D plots of photon absorption rate, the recombination rate and the photogeneration rate to gain a physical insight of electronic and optical behavior of the proposed solar cell. The current-voltage (JV) characteristics and the external quantum efficiency (EQE) of the device are also plotted. The deduced performance metrics of the proposed PSC demonstrates a short circuit current density (JSC) of 27.247 mA/cm², an open circuit voltage (VOC) of 0.978 V, a fill factor (FF) of 71.80% and a power conversion efficiency (PCE) of 21.25%, where the EQEs with and without considering the whole device absorption is more than 30% and 50% respectively over the visible and infra-red wavelength range (0.3 μm -1.0 μm).

Keywords: Perovskite solar cell, MAPbI₃, PEDOT: PSS, ZnO, ITO, Silvaco TCAD, PCE.

1. Introduction

The solar energy, an environmentally-harmless (Nayak *et al.*, 2019) resource of energy, can supply a huge energy- 4 million exajoule per year (Kabir *et al.*, 2017; Ho *et al.*, 2024). This ample energy can be harvested by the photovoltaic cells, commonly known as solar cells. Till 2021 solar panels covered 28% of total global capacity (Ho *et al.*, 2024; Nowsherwan *et al.*, 2023). Solar cells

also have the potential to deal with the climactic change enhanced by the global warming through carbon neutralization (Nowsherwan *et al.*, 2023). Theoretically, the maximum power conversion efficiency (PCE) that can be achieved by the solar cells is $\approx 33\%$ (known as Shockley-Quisser limit (Guillemoles *et al.*, 2019)). To achieve this limit with reduced material and fabrication cost seamless research endeavors have been conducted in the scientific community by utilizing different low-cost absorber materials with different structures which lead the evolution of different generations of solar cells (i.e., 1G, 2G, 3G). The 1G cells, the most primitive generation, involve costlier crystalline silicon (Si) and silicon-derived (i.e., SiGe) materials with p-n junction- and metal-insulator-semiconductor (MIS) junction-based structures (M. J. Islam *et al.*, 2016) and achieved a PCE of $\approx 25\%$. The PCE of p-n junction structured 1G solar cells can be further enhanced by developing a drift field in the absorber layer through the use of heavily and non-uniformly doping there in (Huqe *et al.*, 2013; S. K. Saha, Farhan, *et al.*, 2011; S. K. Saha, Ferdous, *et al.*, 2011; N. S. K. Saha, Ferdous, *et al.*, 2011; Huqe & Chowdhury, 2016; Huqe *et al.*, 2012). For MIS solar cells, use of low-cost materials (i.e., GaAs) have the promise of enhanced PCE with reduced cost. In 2G generation, the cell's higher PCE with significantly lower cost is ensured by employing a number of earth-abundant materials such as amorphous Si (M. I. Chowdhury *et al.*, 2020), CdTe (Haque *et al.*, 2014b; Biswas *et al.*, 2015; Rivon *et al.*, 2016), CIGS (Haque *et al.*, 2014a; Rahim *et al.*, 2015; M. J. Islam *et al.*, 2024), CZTS (Ayesha *et al.*, 2016; Nikita *et al.*, 2016) and CZTSSe (Hasan & Chowdhury, 2018). The fabrication cost of all these solar cells has been significantly reduced. The quantum well (Barnham & Duggan, 1990; M. J. Islam & Chowdhury, 2024; Nabiah *et al.*, 2024; Mostafa *et al.*, 2020; M. J. Islam *et al.*, 2020; Johir *et al.*, 2021; M. I. Chowdhury & Mostafa, 2020) and quantum dot (Aroutiounian *et al.*, 2001; Aissat *et al.*, 2016; Bi *et al.*, 2016; Nawaz *et al.*, 2023) based solar cells have been also reported with the promise of higher PCE with reduced cost. Low-cost, abundant materials such as organic semiconductors (W. H. Chowdhury *et al.*, 2021; Chakma *et al.*, 2024; M. R. H. Chowdhury *et al.*, 2024), dyes (O'Regan & Grätzel, 1991; Rahman *et al.*, 2023; Benespero *et al.*, 2018; M. N. Islam *et al.*, 2024) and perovskite semiconductors (Chen *et al.*, 2023; Pitaro *et al.*, 2021; Jeon *et al.*, 2018; Herz, 2017; T. A. Chowdhury *et al.*, 2023) are used as the absorber materials in 3G solar cells- all of which involves low-energy, roll-to-roll, solution-processible manufacturing process thereby enabling low-cost fabrication process (Ho *et al.*, 2024).

Perovskite semiconductor-based solar cell, termed hereafter as PSC, is the fastest-growing 3G technology (Ho *et al.*, 2024), as the PCE of PSCs steeply rises from 3.8% (first invented in 2009 (Kojima *et al.*, 2009)) to more than 25% (reported recently) (Park *et al.*, 2023)). This steep rise in PCE can be attributed to the excellent structural (higher crystallinity, ionicity, and defect tolerance) (Ho *et al.*, 2024), electronic (higher relative permittivity, lower exciton binding energy, higher carrier mobility, and larger carrier diffusion length) (Chen *et al.*, 2023), and optical (higher absorption coefficient) properties (Pitaro *et al.*, 2021). PSCs are appropriate for indoor applications owing to their increased PCE under reduced illumination (Wu *et al.*, 2021). They can be used in portable and wearable electronics due to their ability of fabrication on flexible substrates (Tebbal & Hamida, 2023). They are superior choice in building-integrated photovoltaic (BIPV) applications (Bati *et al.*, 2023). In a typical planar regular PSC structure, the perovskite semiconductor is sandwiched between a hole transport layer (HTL) and an electron transport layer (ETL), where a transparent conductive oxide (i.e., ITO, FTO) next to HTL and a metal contact (i.e., aluminum, silver) next to ETL serve as the anode and the cathode respectively. The common HTLs are spiro-OMeTAD, PEDOT: PSS and NiO (Ho *et al.*, 2024), whereas, ZnO, tin oxide (SnO₂) and titanium oxide (TiO₂), are preferable ETLs (Chen *et al.*, 2023). Of various perovskite semiconductors reported till to date, Methylammonium lead iodide (MAPbI₃) is more preferable because of its excellent electrical and optical properties, low-temperature solution processability, long lifetime, and ferroelectricity (Husainat *et al.*, 2019).

This work aims to design and investigate a perovskite solar cell (PSC) in the Silvaco TCAD environment, as it is more versatile and has the ability to incorporate almost all the physics-based

models and hence, is a suitable tool to analyze complex structures (Hossain *et al.*, 2014). The intended solar cell uses MAPbI₃ as the perovskite absorber, PEDOT: PSS as the HTL (owing to its ability to enhance solar cell functioning (Murugesan *et al.*, 2015)), ZnO as the ETL (owing to its outstanding electronic and optical properties and affordable processing (Bati *et al.*, 2023)), ITO as the front electrode and aluminum as the back electrode.

2. Methodology

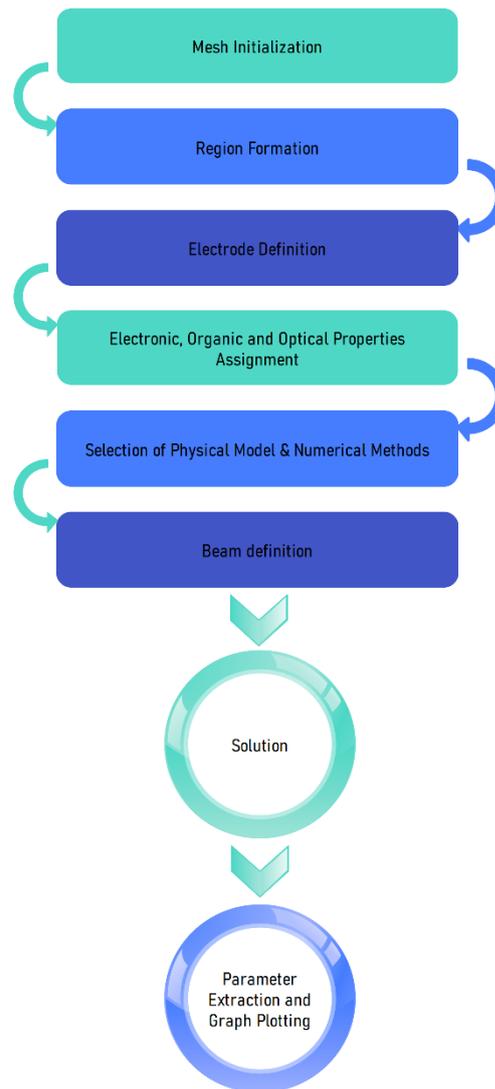


Figure 1 - Flowchart of Silvaco Atlas based solar cell modeling.

This work employs Atlas tool of Silvaco TCAD to model and simulate the proposed PSC. The first step is crucial to balance between the accuracy and the simulation time. Fine meshing has better accuracy with increased simulation time, whereas, course meshing reduces the simulation time at the cost of accuracy (M. R. H. Chowdhury *et al.*, 2024). The regions, electrodes and contacts are defined in the next two steps. Next, the electronic, organic and optical properties of each layer material are defined based on the literature. Electronic properties must include bandgap, permittivity, affinity, effective density of states in the conduction band (EC), effective density of states in the valence band (EV), electron mobility (μ_n) and hole mobility (μ_p), organic properties include relevant parameters for singlet dissociation and recombination as well as Langevin

recombination and optical properties include wavelength-dependent refractive index and extinction coefficient. In the following step, appropriate physical models (the most important one) and numerical methods are chosen. Specification of the solar irradiation profile in the next step completes the design of the PSC. Afterwards, the PSC has been simulated under illumination and bias conditions. Finally, simulation results are extracted to obtain the performance metrics and also, necessary graphs are plotted.

3. Structure and Simulation

In this work, a reference structure implemented in SCAPS 1D and reported in Husainat *et al.* (2019) has been modified to enhance the PCE. The reference PSC structure is based on FTO/TiO₂/MAPbI₃/Spiro-OMETAD/Au with a PCE of 20.34%, where MAPbI₃ is used as the absorber layer (thickness is 300 nm), TiO₂ as the ETL, Spiro-OMETAD as the HTL, FTO is the front electrode and Au (gold) as the back electrode. Severe recombination has been observed between the perovskite films and the FTO electrode in the presence of pinholes in TiO₂ layers leading to poor performance (Ho *et al.*, 2024). Therefore, ZnO is proposed as ETL in this work to improve performance. Spiro-OMeTAD has inherent stability problem, Bati *et al.* (2023), whereas, PEDOT: PSS enhances solar cell functioning (Murugesan *et al.*, 2015). Hence, PEDOT: PSS is better alternative as HTL than Spiro-OMeTAD for improved cell stability. Au is a costlier metal than aluminum (Al) and hence, use of Al can reduce the cost. Therefore, the proposed PSC in this work has been modified to a structure of ITO/PEDOT: PSS/ MAPbI₃/ZnO/Aluminum to enhance the PCE, reduce the cost and improve the cell stability.

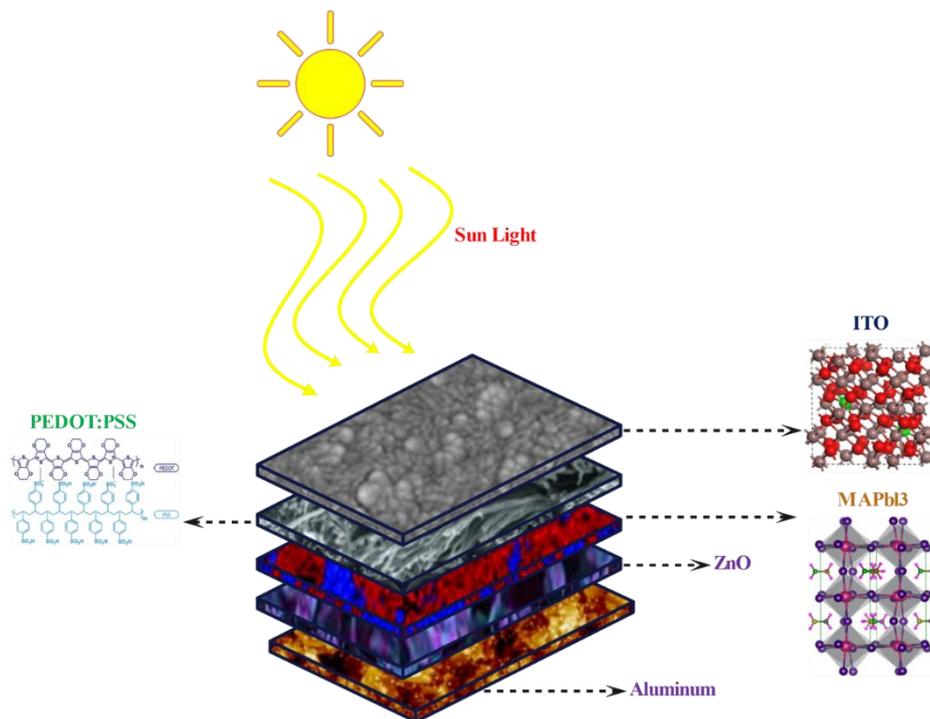


Figure 2 - The proposed structure of MAPbI₃ based PSC.

Figure 2 shows the schematic of the proposed PSC structure investigated in this work. The photoactive absorber layer MAPbI₃ is sandwiched between the top PEDOT: PSS HTL and the bottom ZnO ETL, ITO and aluminum are used as the front and back electrodes. The dimensions of all these materials are shown in Table 1. The electronic properties of MAPbI₃, PEDOT: PSS and ZnO are listed in Table 2. Work function values of ITO and aluminum are chosen as 4.8 eV and 4.1 eV respectively. The optical properties i.e., the refractive index and the extinction coefficient have been collected from literature (RefractiveIndex.INFO - Refractive Index Database, n.d.). As incident

solar irradiation, the terrestrial solar spectral irradiance specified by the American Society for Testing and Materials (ASTM) G-173 (Reference Air Mass 1.5 Spectra, n.d.) has been used.

Table 1 –Layer, Material, and thickness for proposed PSC structure.

Layer	Material	Thickness
Front Electrode	ITO	120 nm
HTL	PEDOT: PSS	30 nm
Absorber	MAPbI3	300 nm
ETL	ZnO	200 nm
Back Electrode	Aluminum	100 nm

Table 2: Electronic properties of different layer materials used in the proposed PSC.

Electronic Property	MAPbI3	PEDOT: PSS	ZnO
Bandgap (eV)	1.5	1.6	3.37
Permittivity	3.9	3.0	9.0
Affinity (eV)	30	3.5	4.54
Nc (cm-3)	2.5×10^{20}	5.0×10^{19}	2.2×10^{18}
Nv (cm-3)	2.5×10^{20}	5.0×10^{19}	1.8×10^{19}
μ_n (cm ² /V-sec)	50	0.1	100
μ_p (cm ² /V-sec)	50	0.08	25
Work function (eV)	-	-	-

The proposed structure has been implemented in the Silvaco TCAD environment. The modeling includes Fermi statistics, and SRH, Auger and optical recombination mechanisms. Since MAPbI3 and PEDOT: PSS are not defined in Silvaco TCAD, the user-defined material option of TCAD is used; MAPbI3 is defined as ‘polysilicon’ and PEDOT: PSS is defined as ‘organic’. The 2D view of the proposed PSC structure, implemented in Silvaco TCAD, is shown in Figure 3.

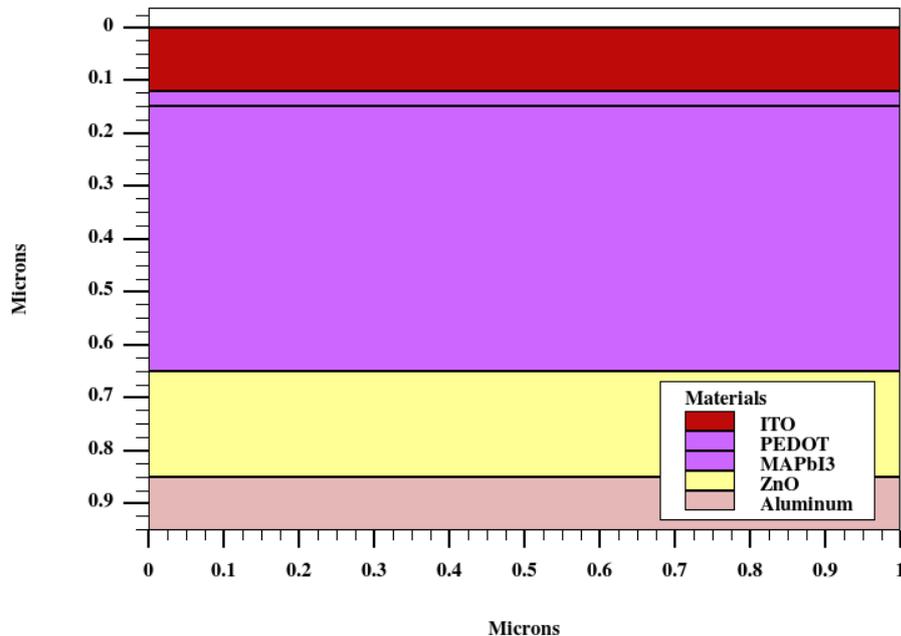


Figure 3 - Silvaco TCAD-implemented structure of the proposed PSC.

4. Results and Discussion

This section presents, analyses and discusses the simulation results obtained from TCAD simulation of the proposed PSC structure. These results include the plots of energy band diagram, electric field, photon absorption rate, recombination rate and photogeneration rate. Internal and external quantum efficiencies (IQE and EQE) and current-voltage (JV) characteristics of the proposed PSC are also presented. Finally, various performance metrics which include short circuit current density (JSC), open circuit voltage (VOC), fill factor (FF) and PCE of the proposed PSC are presented and compared with the reference structure reported in Husainat *et al.* (2019). Figs. 4(a) and 4(b) present the energy band diagram and the electric field profile respectively of the proposed PSC under zero-bias and no light condition. The vertical lines separate the layer materials, where ITO, PEDOT: PSS, MAPbI₃, ZnO and Al are presented from left to right. The linearity of the conduction and valence band energies observed in MAPbI₃ layer represents its undoped nature, for which electric field in this layer becomes constant, as seen from Figure 4(b). The discontinuity in the energy bands observed at the interfaces is due to the work function difference between the materials involved at each interface. This discontinuity results in a depletion region near each interface, for which energy bands are bent [Figure 4(a)] and nonuniform electric field has been developed [Figure 4(b)] near these interfaces. The observed higher level of electric field in the absorber layer confirms the efficient electron-hole separation of the excitons photogenerated therein. The even higher field observed at the interfaces of ITO/ PEDOT: PSS and ZnO/Al helps improved charge carrier extraction towards the electrodes.

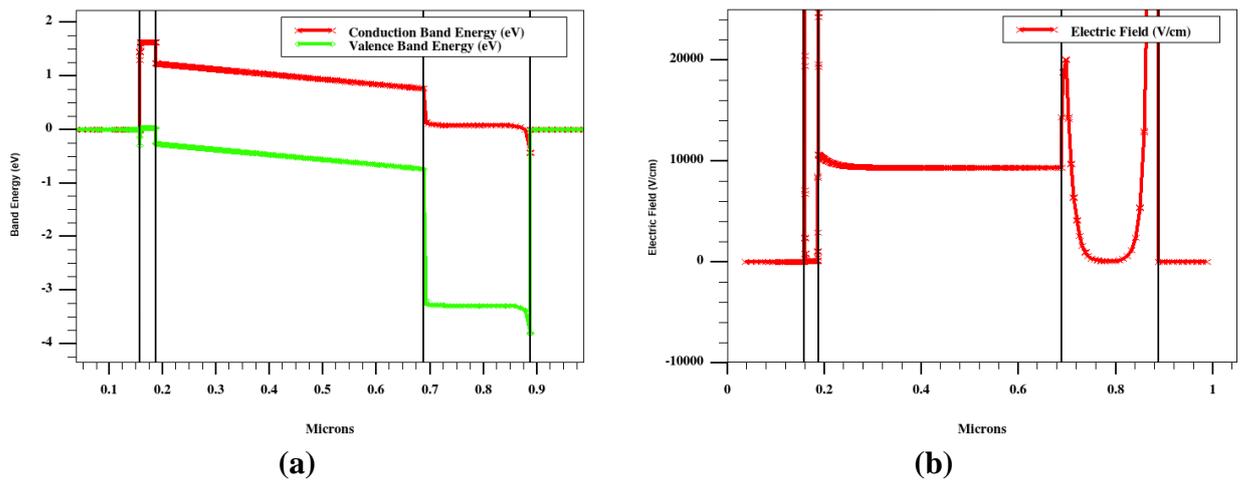


Figure 4 - (a) Energy band diagram and (b) Electric field profile of the proposed PSC.

The contour and 2D views of the photon absorption rate under no bias condition are presented in Figs. 5(a, b). These plots reveal that the photons are mostly absorbed in the MAPbI₃ layer. This absorption is almost exponential, which peaks at the interface with the PEDOT: PSS layer and exponentially decays as moves toward the ZnO layer. Figs. 5(a, b) also shows a small amount of absorption in the PEDOT: PSS and ZnO layers.

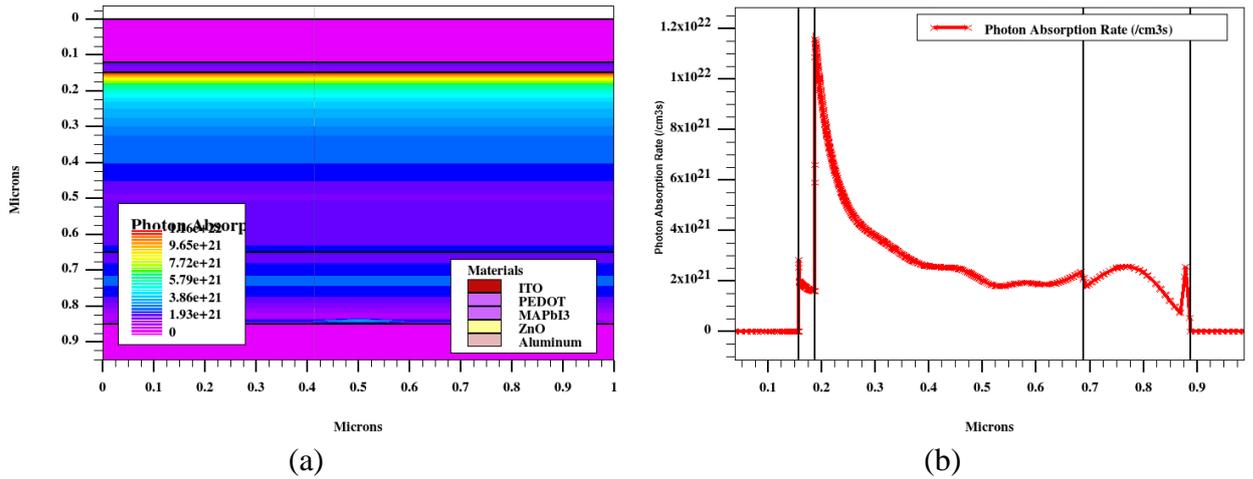


Figure 5 - (a) Contour and (b) 2D plot of the photon absorption rate in the proposed PSC.

The contour and 2D plots in Figure 6 present the recombination rate in the different layers of the proposed PSC. These plots show that the recombination of carriers happens in all three layers of absorber, HTL and ETL. The highest recombination happens in the ETL, lowest in the HTL, and in between in the absorber layer. This means that the mismatch between the absorber and ETL is the highest and that between the absorber and HTL is the lowest, whereas, almost constant recombination in the absorber reveals its structural uniformity.

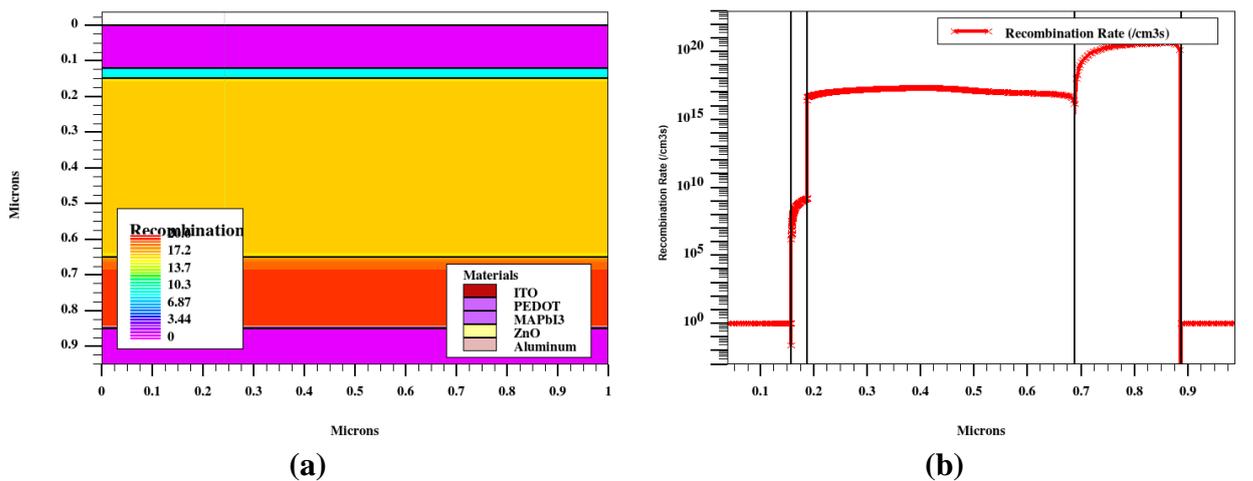


Figure 6 - (a) Contour and (b) 2D plot of the recombination rate in the proposed PSC.

The contour and 2D plots of the photogeneration rate in the different layers of the proposed PSC are presented in Figure 7. Both these plots reveal that the highest photogeneration occurs in the MAPbI3 absorber layer, which almost exponential decays from left to right. A comparatively smaller photogeneration also occurs in the ZnO layer. However, no photogeneration is happened in the PEDOT: PSS layer.

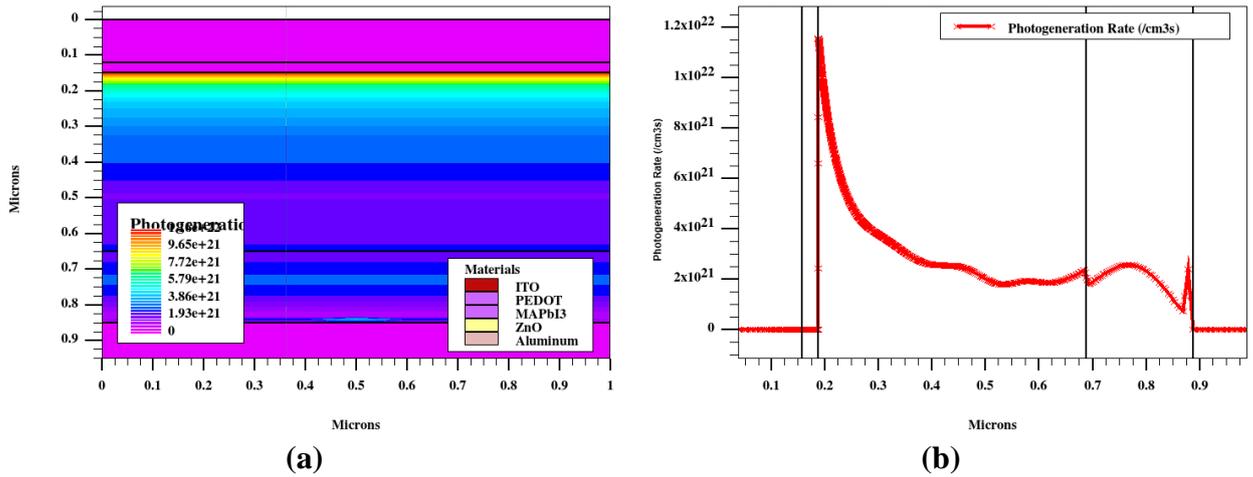


Figure 7 - (a) Contour and (b) 2D plot of the photogeneration rate in the proposed PSC.

The reflection, transmission and absorption spectra of the proposed PSC over a wavelength range 0.3 μm -1.0 μm are presented in the plots of Figs. 8(a-c). The reflection is $< 30\%$ for wavelengths up to 0.75 μm , then starts to rise and reaches $\approx 50\%$ at a wavelength of 0.98 μm , as seen from Figure 8(a). On the other hand, Figure 8(b) shows that the transmission is zero for wavelengths up to 0.52 μm and then increases, however, it remains $< 0.24\%$. As seen from Figure 8(c), the absorption is $> 50\%$ over all the wavelength span and it is $> 70\%$ for wavelengths up to 0.76 μm . The lower reflection and transmission and the higher absorption over most of the wavelength span shows higher photon absorption capability of the proposed PSC.

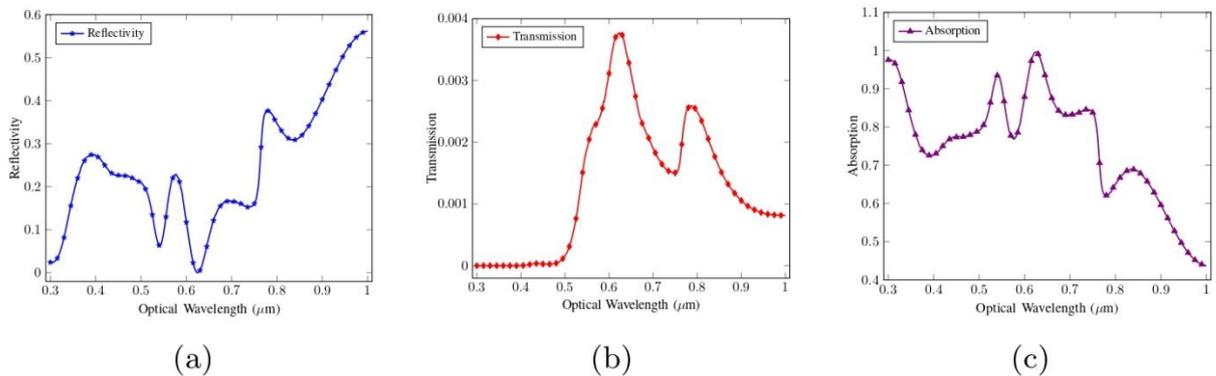


Figure 8 - (a) Reflection, (b) transmission and (c) absorption profile of the proposed PSC.

Figure 9 plots the variation of three photo-generated currents, namely source (I_S), available (I_A) and load (I_L) against over the wavelength span of the irradiated solar spectrum. These currents are calculated based on the consideration of optical and electrical losses- I_S does not consider any one, I_A considers only the optical one and I_L considers both Johir *et al.* (2021), where reflection and transmission losses from the optical loss and recombination of charge carriers form electrical loss. As seen from Figure 9, the optical loss is more severe than the electrical losses, as I_A is significantly reduced from I_S , whereas, I_A and I_L are very close throughout the wavelength range. This means that the reflection is higher and the recombination is lower in the proposed PSC.

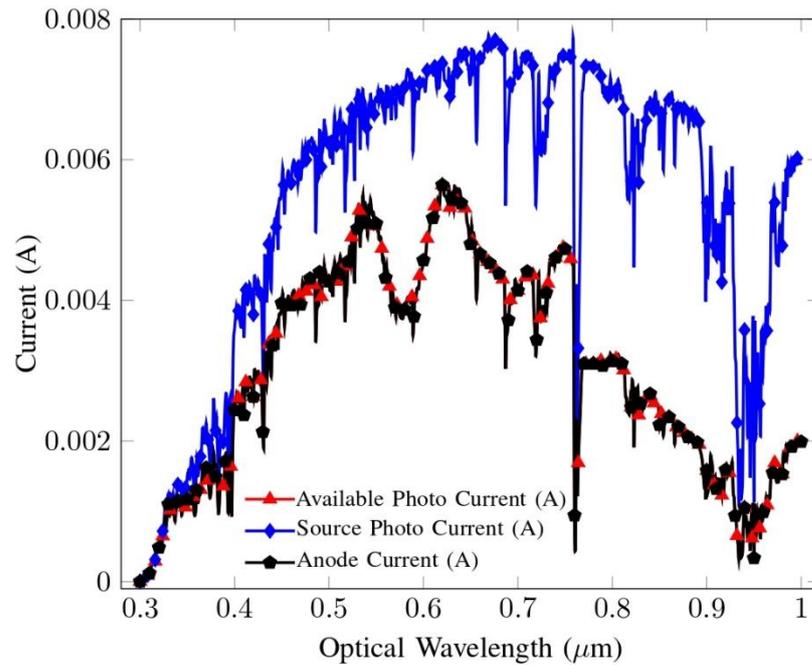


Figure 9 - The current profiles Vs. wavelength of the proposed PSC.

The IQE (η_{int}) and EQE (η_{ext1} and η_{ext2}) plots of the proposed PSC are presented in Figure 10, where these quantities can be defined in terms of I_S , I_A , I_L and absorption (A) as Johir *et al.* (2021):

$$\eta_{int} = \frac{I_L}{I_A}$$

$$\eta_{ext1} = \frac{I_L}{I_S}$$

$$\eta_{ext2} = \frac{I_L}{I_S A}$$

The IQE plot observed in Figure 10 approaches to 100% over the full wavelength of interest, as I_L and I_A are very close as observed in Figure 9 due to reduced recombination. On the other hand, the EQE plots have been observed severely degraded in the most part of the wavelength spans of interest. Since the absorption spectra is $> 50\%$ up to the wavelength of $0.76 \mu\text{m}$, the EQEs are observed higher ($>60\%$) in this span. However, EQE2 which considers absorption of the whole device becomes lower than EQE1 at the wavelengths $> 0.55 \mu\text{m}$ and $> 0.78 \mu\text{m}$, meaning that parasitic absorption in the HTL and ETL increases at these wavelengths.

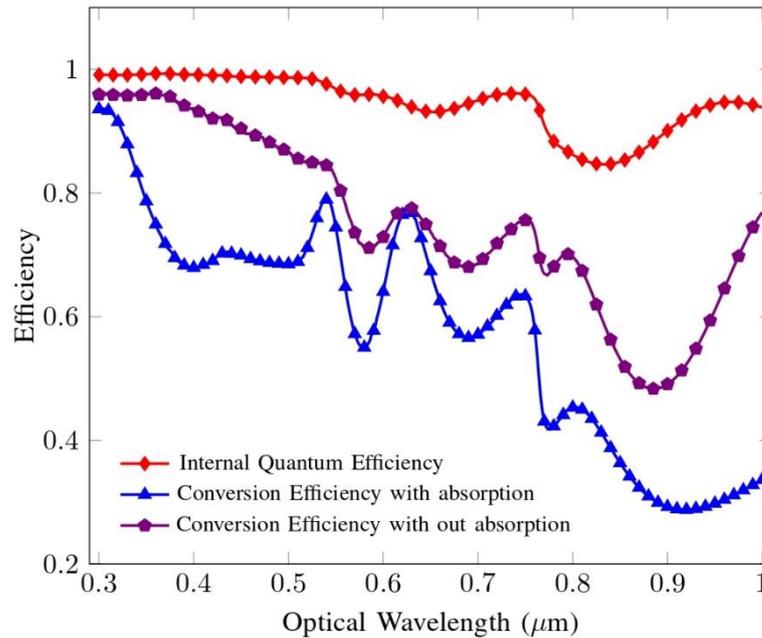


Figure 10 - IQE and EQE characteristics of the proposed PSC.

Figure 11 represents the JV characteristics curve of the proposed PSC. This curve closely resembles to typical square-like shape observed in inorganic solar cells, as reported in the literature (M. I. Chowdhury *et al.*, 2020; Haque *et al.*, 2014b; Biswas *et al.*, 2015; Rivon *et al.*, 2016; Haque *et al.*, 2014a; Rahim *et al.*, 2015; M. J. Islam *et al.*, 2024; Ayesha *et al.*, 2016; Nikita *et al.*, 2016; Hasan & Chowdhury, 2018) and follows the two-diode model (A. Islam & Chowdhury, 2014) of inorganic solar cells. This typical inorganic JV behavior results in higher fill factor (FF) (> 70%) of the proposed PSC, which is also reflected in Table 4.

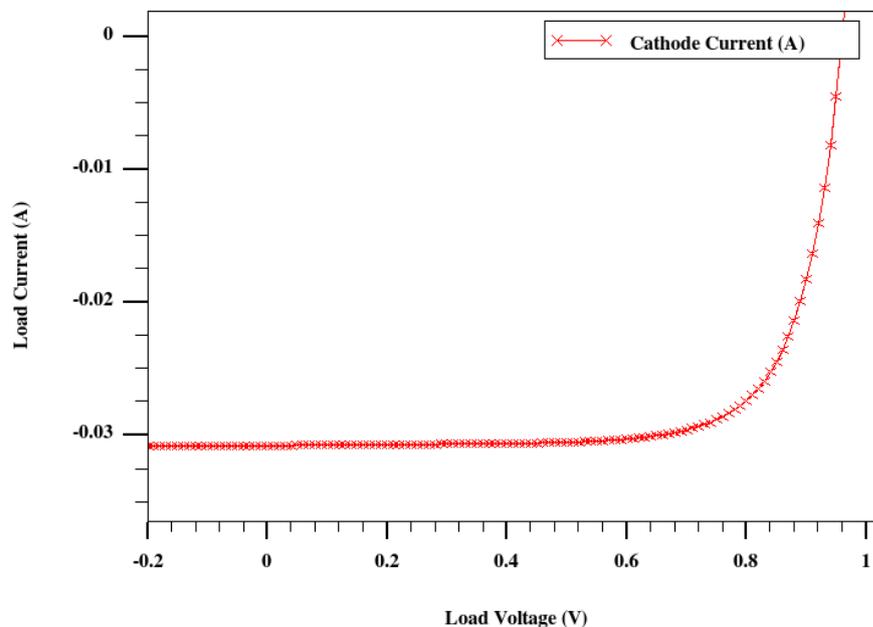


Figure 11 - JV characteristics of the proposed PSC.

The performance metrics comparison of the proposed PSC with the reference structure reported in (Husainat *et al.*, 2019) has been presented in Table 4. The proposed PSC has much higher short circuit current density (JSC) than the reference cell, which reflects its better charge carrier collection. However, owing to the increased recombination in ZnO ETL, the open circuit voltage (VOC) and the FF of the proposed PSC becomes lower than the reference cell. However, the significant increase in JSC outperforms the decreased VOC and FF, resulting in the higher PCE of the proposed PSC.

Table 4: Performance metrics comparison of the proposed PSC with the reference structure (Husainat *et al.*, 2019).

Performance Metrics	FTO/TiO ₂ /MAPbI ₃ /SiproOMeTAD/Au (Husainat <i>et al.</i> , 2019)	ITO/PEDOT: PSS/MAPbI ₃ /ZnO/Al (This work)
Jsc (mA/cm ²)	15.158	27.247
Voc (V)	1.476	0.978
FF (%)	91	71.80
PCE (%)	20.34	21.25

5. Conclusion

In this work, an MAPbI₃-based perovskite solar cell has been designed, modeled, and investigated for improved performance. The MAPbI₃-based absorber layer is sandwiched between two electrodes (ITO and aluminum). The PEDOT: PSS placed atop of this absorber layer serves as the hole transport layer and enhances the hole transport to the ITO electrode. Instead of TiO₂, ZnO is used as an electron transport layer to overcome the recombination problem associated with the presence of pinholes in TiO₂. ITO and aluminum are used two electrodes. The proposed solar cell has been implemented and simulated in the Silvaco TCAD environment. Compared to the reported values in the literature, the proposed structure shows an increased PCE of 21.25% with significantly enhanced short-circuit current density (JSC) of 27.247 mA/cm². This significant improvement in JSC counterbalances the less values of open circuit voltage and fill factor as compared to the reported ones in the literature. The analysis regarding the degraded external quantum efficiency of the proposed PSC suggests that the continued research endeavors should be conducted to reduce the optical losses and hence, to enhance the PCE further.

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