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Numerical investigation of a new approach based on perovskite CH3NH3PbI3 absorber layer for high-efficiency solar cells

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Abstract

Recently, metal halide perovskite materials for solar cells have gained a wonderful attention from researchers around the world. In this paper, a novel perovskite layer in combination with other layers is numerically investigated to enhance the performance of perovskite-based solar cells (PSCs). General-purpose photovoltaic device model (GPVDM) is used to simulate the proposed structure with Methylammonium Lead Iodide (CH3NH3PbI3), sandwiched between a bi- and tri-layer. The results show enhanced short circuit current density ($J_{sc}$) and power conversion efficiency (PCE) for PSCs. The proposed PSCs show a $J_{sc}$ of 39.6 mA cm$^{-2}$ and PCE 31.4% under standard AM 1.5 G. The bi- and tri-layer sandwiched CH3NH3PbI3 structure is an elegant solution for the realization of high efficiency PSCs.

1. Introduction

Due to the increasing demand of energy and lack of conventional resources, solar technology is considered an ultimate choice for electric power generation. The core reason behind solar technology adaptation is, its potential to cater all the demands due to abundance, eco-friendly, sustainable and reliable nature [1]. The Photovoltaic (PV) cells are predicted to deliver one-third of global power by end of this decade [2]. Reports [3, 4] show that 90% of the PV products throughout the global markets are fabricated using crystalline i.e. (monocrystalline and polycrystalline) Silicon (Si) wafers with power conversion efficiency (PCE) of around 26%, commonly referred as 1st generation PV. However, these PV cells are costly, due to lower conversion efficiency, high processing expenditure and raw material of Si [3, 4]. Moreover, technological and material-wise challenges are a barrier to further improvements in the PCE of Si-based solar cells [5]. The PV cells, fabricated using Copper Indium Gallium Selenide (CIGS), Amorphous Si and Cadmium Telluride (CdTe) could not gain success over crystalline Si PV cells due to technological complications and stability problems [6, 7]. The Multi-junction solar cells, i.e. a combination of III–V materials with silicon solar cells are an attractive solution to enhance the power conversion efficiency [8, 9]. However, these solar cells performance is hindered by poor crystalline quality of the III–V materials [10].

To provoke these hurdles, researchers are investigating novel materials for high efficiency PV cells. With the advent of novel discoveries, perovskite material for fabrication of solar cells is the most potential candidate due to its inherent material properties for PV applications [5], and hence, it has gained much attention as a potential replacement of Si PV cell market, as well as usage in the tandem based solar cells [4, 5]. Within the past decade, an incredible improvement has been observed in its PCE from 3.8% to 25.5%, which has attracted the attention of solar community towards the use of perovskite-based solar cells [11]. Furthermore, comparatively low costing of perovskites over Si solar cells serves as the potential advantage for future commercialization of perovskite based PV technologies [12]. Hence, PSCs have been considered as auspicious PV cell option for the eco-friendly energy industry, in the adjacent few years.
The Methylammonium Lead Iodide (CH$_3$NH$_3$PbI$_3$) perovskite is among the potential candidates for its utilization as an active layer in the PSCs with a tunable energy bandgap from 1.5eV to 2.3eV [13–15]. CH$_3$NH$_3$PbI$_3$ has been reported as a better light harvester than N$_719$ ruthenium sensitizer used in dye-sensitized solar cells (DSSC) [16]. It occurs in cubic crystal structure at temperature 330 K, the cubic state converts into the tetragonal state at a temperature of 236 K. The tetragonal crystal converts into the orthorhombic phase at a temperature of 177 K [17]. This shows a strong dependence of the crystalline structure of CH$_3$NH$_3$PbI$_3$ on a range of temperatures, ultimately affecting the electronic properties of the fabricated device, which can severely alter the PCE. Liu et al [18] reported that once the film thickness significantly exceeded the carrier diffusion lengths in CH$_3$NH$_3$PbI$_3$, the PCE is sharply declined.

In this scenario, research-based on experimental investigations has been done to increase PCE by improving the electrical performances of PSCs. Meanwhile, efforts on simulations [19–23] are also conducted in parallel to improve PCE by varying the different physical parameters, such as bandgap, doping concentration, and layer thickness etc for minimizing, the experimental expenditure before the actual fabrication of PSCs. Recently, numerous researchers have reported PSCs based simulations using different software, such as SILVACO ATLAS, wxAMPS and SCAPS, which are based on a single physical model: the drift–diffusion transport model [19–23]. To achieve a better simulated model, that can represent experimental results, such can be demonstrated using General-purpose Photovoltaic Device Model (GPDVM) device software [24], to evaluate PV equations in both forms i.e. in steady-state and in the time domain. Generally, the model can be applied on both electron and hole drift–diffusion, and carrier continuity equations in position space, to trace the apparent movement of charges within the device. The internal electrostatic potential can be calculated by applying the Poisson equation. Moreover, GPDVM can also solve Shockley–Read–Hall (SRH) formalism for recombination and carrier trapping phenomena.

In this work, numerical investigation of a new approach for high-efficiency PSCs is presented. For the first time, a bi- and tri-layer are simultaneously proposed and simulated to study their effect on PCE of PSCs. The proposed structure of device with, Methylammonium Lead Iodide (CH$_3$NH$_3$PbI$_3$), sandwiched between a bi- and tri-layer shows a tremendous improvement in short circuit current density ($J_{sc}$) and PCE of PSCs. Furthermore, this research calculates, numerically the effect of thickness of active layer versus PCE, the thickness
of all layers versus PCE, and temperature versus PCE of CH3NH3PbI3 absorber based PSCs, using GPVDM simulations software.

2. Proposed model

The proposed PSC architecture is shown in figure 1 as upside down. The device area is taken as 400 μm × 400 μm under AM 1.5G illumination. It includes a glass layer, which is used to converge visible light followed by anode, which is made from Fluorine-doped Tin Oxide (FTO). This is than followed by a tri-layer which is also known as Electron Transport Layer (ETL) that plays a critical role in the PCE of PSCs, this layer is composed of three layers i.e. Titanium dioxide (TiO2), Zinc oxide (ZnO) and Buckyballs (C60). A highly productive ETL should be of high quality to ensure maximum electrical conductivity and marginal conduction band. Furthermore, they must have a point-to-point correspondence with the active layer otherwise it badly affects the PCE of PSC [21]. In the tri-layer each layer plays a critical role, in improving or degrading the PCE of the PSCs. The functional detail of each layer is such as, appropriate energy adjustment; TiO2, C60, and ZnO are indispensable for improving the open-circuit voltage (Voc), whereas the C60 improves the carrier extraction mechanism from the CH3NH3PbI3xClx layer. The charge recombination process is eliminated through the FTO surface, using the TiO2, by assuring the electron collection procedure indispensable for improving the open-circuit voltage.

Further, the V2O5 material is proposed with its numerous characteristics being reported in the literature, such as a light-harvesting layer. With efficient light-absorbing property, it has now been considered as a favorite light-absorbing element. The next important layer is the bi-layer, which is also called a hole transport material (HTL). HTL is composed of two-layers, which are Vanadium Pentoxide (V2O5) and Poly (3, 4-Ethylene Dioxythiophene): Poly (Styrene Sulfonate) (PEDOT: PSS). PEDOT: PSS has been extensively used as HTL in most recognized architectures. Due to high optical transparency, low redox potential, PEDOT: PSS is highly hygroscopic and absorbs moisture, which could cause decomposition of the perovskite absorber layer. So for the proposed PSC, V2O5 has been incorporated to improve the HTL mechanism and to reduce the decomposition. Further, the V2O5 material is proposed with its numerous characteristics being reported in the literature, such as an auspicious HTL nominee for PSCs due to its superior transparency to visual light and high hole mobility, and suitable energy bands for hole transport in PSCs [26, 26]. The energy levels of V2O5 and PEDOT are already reported in the literature [27–29]. The valence band (VB) of V2O5 is approximately 4.7 eV [27, 28], nearer to the Occupied Molecular Orbital level of PEDOT: PSS, which is around 5.0–5.2eV [27–29]. Collectively, they create an electrical barrier, which is free cascading contact for hole transport across the interface. Further, such modified PSCs structures have shown improved conversion efficiency of around 20% compared to PEDOT only devices [30, 31]. The last layer is the silver (Ag), while Ag and HTL combined act as an anode. The structure used in this work is as, anode/bi-layer (HTL)/CH3NH3PbI3/tri-layer (ETL)/cathode/glass as shown in figure 1.

3. Results and discussion

3.1. Simulations model

General-purpose Photovoltaic Device Model (GPVDM) software [24] has been used for simulating the proposed PSCs model. The temperature, individual layer thicknesses and combined layer thicknesses effects on the PCE were investigated under standard AM 1.5 G spectrum. The GPVDM solves the Poisson equation (equation (1)), bipolar drift-diffusion equation (equations (2) and (3)) and continuity equation (equations (4) and (5)), given as below;

$$\frac{d}{dx} \varepsilon \varepsilon_r \varepsilon_v = \frac{\partial \varphi}{\partial x} = q(n - p)$$  \hspace{1cm} (1)

$$J_n = q \mu_n \frac{\partial E_v}{\partial x} + qD_n \frac{\partial n}{\partial x}$$  \hspace{1cm} (2)

$$J_p = q \mu_p \frac{\partial E_v}{\partial x} - qD_p \frac{\partial p}{\partial x}$$  \hspace{1cm} (3)

$$\frac{\partial J_n}{\partial x} = q \left( R_n - G + \frac{\partial p}{\partial x} \right)$$  \hspace{1cm} (4)

$$\frac{\partial J_p}{\partial x} = -q \left( R_p - G + \frac{\partial n}{\partial x} \right)$$  \hspace{1cm} (5)

During first run of simulations, the optimum layer thicknesses such as ETL, HTL and active layer were investigated. Furthermore, the simulations have also been performed to study the impact of temperature on PCE, in dual proposed layer of the PSCs. Meanwhile, different optimal thickness values were obtained by
keeping the temperature constant at 300 K, later on through the simulation procedure, the obtained optimum critical layer thickness were utilized and then, the temperature was changed from 250 K to 370 K, observing the effect on PCE of the proposed PSCs. Table 1. Parameters used in simulation shows the different parameters used during the simulations of this study [31]. During simulations model, no light losses at the surface/interfaces has been considered.

3.2. Active layer thickness (perovskite layer) impact on proposed PSC performance
The active layer thickness plays a crucial role in performance of the PSCs. This layer affects all the critical electrical parameters of the PSCs, such as \(I_{sc}\), \(V_{oc}\), Fill Factor (FF) and PCE.
For this part of the PSCs simulations, thicknesses of all layers except absorber were kept constant, to observe the impact of variation of active layer thickness on PCE of the PSCs. To achieve this, the constant layers thicknesses were kept as, cathode 80 nm, for ETL 0.1 mm, 0.1 mm and 1 pm respectively and for HTL 8 nm and 1 nm respectively.

Figure 2(a) shows the dependence of J_{sc} on thickness of the perovskite absorber layer. The J_{sc} increases rapidly with the thickness of the absorber from 101 A cm^{-2} and reaches to a maximum of 396.6 A m^{-2} with the cast of layer thickness 600 nm. This increase in J_{sc} with absorber thickness is expected as more electron-hole pairs are generated in the active layer and collected at the output circuit. J_{sc} relies majorly upon the production of charge carriers. The increase in thickness of the CH_{3}NH_{3}PbI_{3} absorber layer leads to a higher absorption percentage of the exposed light, consequently leading to an increase J_{sc}. However, the J_{sc} decreases with further thickness of the absorber because of limited penetration of the incident light AM 1.5 G. Figure 2(b) shows dependence of V_{oc} on thickness of the perovskite absorber layer. A maximum value of 0.98 V is obtained at an
absorber layer thickness of 0.4 mm. The Voc decreases as thickness of the CH$_3$NH$_3$PbI$_3$ absorber increases. This is due to the increased recombination in absorber, because of the short lifetime of charge carrier’s. Within the thick absorber layer of PSCs, charge carriers generated near the center of CH$_3$NH$_3$PbI$_3$ layer recombines when the thickness surpasses the diffusion length $[32]$. Furthermore, the increase in thickness of the absorber layer leads to reduction of the effective bandgap, which consequently results in a reduction of the Voc $[33, 34]$. Figure 2(c) shows the dependence of FF on thickness of absorber layer. The FF starts to decrease from a maximum of 86.03% with increase in thickness of the absorber layer. FF is mainly associated with the shape of J-V curve and parasitic resistances $[35]$. Since resistance increases with increase in thickness of the absorber layer, resulting in decrease of FF $[36]$. From figure 2(d), it is evident that the PCE reaches to a maximum of 31.4% than it decreases with further increase in the thickness of the absorber layer, which is attributed to the reduction of $J_{sc}$, $V_{oc}$ and FF with increase in the thickness of CH$_3$NH$_3$PbI$_3$ absorber layer.

3.3. Impact of other layers thicknesses on the PCE

The other layers except the active layers have also a strong impact on PCE of the PSCs. Figure 3(a) shows the variation of PCE with thickness of the C$_{60}$ layer. The PCE remains stable around 31.4% up to 0.02 nm of C$_{60}$ thickness than decreases for increasing the thickness. During the variation of C$_{60}$ layer, the other layers were kept constant and thus an optimum value of 31.2% using 0.02 nm was obtained. The insertion of ultra thin C$_{60}$ layer reduces the electron accumulation at the CH$_3$NH$_3$PbI$_3$ absorber interface. Further, improves the efficient electron extraction from the CH$_3$NH$_3$PbI$_3$ perovskite absorber, resulting in a high current. A previously published fluorescence microscopy measurements showed a much stronger photoluminescence, indicating a larger electron accumulation for the devices without C$_{60}$ than with it $[31]$. Similarly Golubev et al $[36]$ reported that inserting an ultra thin C$_{60}$ layer prevent electrons from moving back to the perovskite, consequently reduces the loss of free carriers due to carrier recombination within the pervoskite layer. The decrease in PCE of the PSCs with thickness of C$_{60}$ supports the results of Liu et al $[35]$ and Golubev et al $[36]$ that an ultra thin C$_{60}$ is needed to enhance carrier extraction. Figure 3(b) shows variation of PCE with thickness of the ZnO layer. The downfall of efficiency with increase in thickness of the ZnO layer is mainly due to the increase of trap density at the interface layers. It has been reported $[37]$ that the defect density at CH$_3$NH$_3$PbI$_3$/ZnO interface should be controlled.

![Figure 4. Effect of Temperature on (a) short circuit current density, (b) open circuit voltage, (c) fill factor, and (d) power conversion efficiency.](image)
under \( \approx 10^{17} \text{ cm}^3 \), to achieve high PV performance. Figure 3(c) shows the effect of TiO\(_2\) layer thickness on PCE. The PCE remains around 31.4\% until 600 nm, then drops down with further increase in thickness. This is consistent to the previous studies and attributed to higher probability for charge trapping and subsequent recombination as well as increased light blocking \([38, 39]\). Figure 3(d) shows variation of PCE with the thickness of V\(_2\)O\(_5\) layer. The PCE shows a decrease with increase in thickness of the layer. Similarly, figure 3(e) shows that increase in thickness of PEDOT:PSS shows a slight decrease in the PCE. This HTL is reported \([33]\) to be thin enough to reduce recombination and light absorption while thick enough to enhance carrier collection from the active layer.

3.4. Temperature effect on various electrical parameters

Thermal stability is a significant parameter for the measurement of PSCs performance and is measured by operating temperature of the cell. Bandgap of the materials, carrier concentration, electrons and holes mobility and density of states performance parameters are influenced incredibly at elevated working temperature. The overall energy output differs from what is expected compared to the standard test condition measurements.

For this part of the PSCs simulations, optimized thicknesses of all layers were used, to observe the impact of temperature variation on PCE of the PSCs. To achieve this, the thickness of optimized layers was kept as, ETL (FTO: 80 nm, TiO\(_2\): 100 nm, ZnO: 100 nm, C\(_6\)O: 1 pm), Perovskite absorber (CH\(_3\)NH\(_3\)PbI\(_3\): 400 nm) and HTL (V\(_2\)O\(_5\): 1 nm, PEDOT:PSS: 8 nm, Ag: 80 nm).

Figures 4 (a)–(d), shows the effect of temperature on \( J_{sc}, V_{oc}, FF \) and PCE respectively. The increase in \( J_{sc} \) with temperature was expected because \( J_{sc} \) is proportional to the incident light. The dependence of FF is shown in figure 4(b). It decreases with increase in temperature, because of the increase in series resistances at high temperature. There exists a linear relationship between FF and series resistance

\[
FF = FF_0 X \left( 1 - \frac{R_s}{R_{ch}} \right) = FF_0 X \left( 1 - \frac{R_s J_{sc}}{V_{oc}} \right)
\]

The \( V_{oc} \) is strongly temperature dependent as shown in figure 4(c). It decreases as temperature increases due to the change in entropy, which causes to minimize the electron–hole pair, electrochemical energy in the conduction and valence bands, this leads to widening of the Fermi–Dirac distribution. Meanwhile, impels the quasi-Fermi levels; so it leads to apart from the band edges. Further, at high temperature, it may be assumed that ions create thermal imbalance, consequently leading to the cause of observed degradation phenomena.

Figure 4(d) shows the temperature dependence of PCE. It is evident that PCE decreases with increase in temperature because of the decrease of \( V_{oc} \) and FF.

Figure 5 presents the J-V characteristic of the proposed PSC with comparison to earlier reported simulated models. The high \( J_{sc} \) is attributed to the layers optimization as well as insertion of the ultra thin C\(_6\)O layer in ETL of the device \([35, 36]\). The \( V_{oc} \) is comparable to the literature; however, it is lower than the \( V_{oc} \) reported by...
This is attributed to the HTL doping concentration and increased rate of recombination at the interface between the perovskite absorber and the HTL layer.

Furthermore, table 2. Comparison of Proposed Model with Literature demonstrates the electrical parameter results of the proposed PSC model and their comparison with reported simulation results.

Figure 6 shows the corresponding energy band level diagram for the optimized proposed structure with the J-V characteristics shown in figure 5. For the given incident light (i.e. standard AM 1.5G), the electron-hole pair generated at the absorber will move towards the ETL and HTL in the device extracted at the output circuit.

4. Conclusion

In this work, a novel perovskite CH3NH3PbI3 absorber layer, sandwiched between a bi- and tri-layer, was investigated to enhance the performance of PSCs using GPVDM simulations software. The Thicknesses of ETL (C60, ZnO, TiO2) and HTL (V2O5, PEDOT:PSS) were individually varied to investigate their impact on Jsc, Voc, FF and PCE of the PSCs. The simulation results show that thickness of each layer has a significant contribution to the electrical performance of PSCs. Further, there is a trade-off in selection of optimized layer thickness and PCE. The proposed PSCs show a Jsc of 39.6 mA cm$^{-2}$ with PCE of around 31.4% under standard AM 1.5 G. The bi- and tri-layer sandwiched CH3NH3PbI3 structure is an elegant solution for the realization of high efficiency PSCs. The numerical analysis discussed in this paper, evidently shows that in future experimental demonstration of the proposed structure is crucial to investigate the real world challenges that exist in material synthesis, device fabrication and structural and optical characterization.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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