eNergetics 2024
DOI: https://doi.org/10.5281/zenodo.14730922

Original Scientific Article

Numerical Simulation and Optimization of Stable $CH_3NH_3PbI_3$-based 2D/3D Mixed Dimensional Perovskite Solar Cell

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Abstract—**Over the recent decades, solar power has gained significant attention globally as a sustainable and cost-effective source of energy outpacing other renewable energy alternatives. Perovskite Solar Cells (PSCs) have been identified as a powerful photovoltaic technology due to their exceptional characteristics including improved efficiency and affordability. The perovskite material demonstrates superior properties such as higher absorption coefficients, adjustable band gaps and extended charge carrier lifetime. However, conventional 3D perovskite solar cells face instability issues upon exposure to elevated temperatures and moisture delaying commercialization. 2D perovskite materials demonstrate a higher stability level than their 3D counterparts. Therefore, 2D/3D mixed-dimensional PSCs have been developed to overcome these challenges and achieve balanced performance and long-term stability simultaneously. In this study, a 2D/3D mixed dimensional PSC was numerically simulated by incorporating 2D-$MAPbI_3$ capping layer on the top of 3D-$MAPbI_3$ using SCAPS-1D solar cell simulation software. The proposed device architecture is FTO/TiO_2/3D-$MAPbI_3$/2D-$MAPbI_3$/Cu_2O/Au. The device performance was optimized by varying the thickness and the defect density of 3D-$MAPbI_3$ layer. The optimized device demonstrates maximum power conversion efficiency (PCE) of 24.52% at 1µm thickness and defect density range of 10^{10}-10^{12} cm^{-3}. This analysis reveals that the 2D/3D mixed-dimensional PSC delivers enhanced efficiency while ensuring the prolonged operational stability of the solar cell.**

Keywords - perovskite solar cell, capping layer, stability enhancement, power-conversion efficiency, defects

I. INTRODUCTION

Global energy demand is rising day by day due to rapid industrialization and population growth. It is anticipated that global energy demand will triple by 2050, even the timely conservation measures are implemented. The availability of fossil fuel sources is limited making it challenging to fulfill the rising energy demand [1]. Moreover, the emission of greenhouse gases and air pollution caused by burning fossil fuels has become a pressing environmental issue emphasizing the need for energy alternatives. In recent decades, solar power has evolved as a clean and sustainable energy source providing solutions for both increasing energy demand and environmental pollution. Currently, Si-based solar cells dominate the solar cell market with enhanced power conversion efficiency [2]. However, their high manufacturing cost and associated environmental concerns limit their widespread adoption.

Perovskite Solar Cell is a promising solar cell technology due to its remarkable power conversion efficiency levels and simple and affordable manufacturing practices [3]. Organic-inorganic halide perovskites are rapidly gaining prominence in the solar cell industry due to their unique photovoltaic characteristics such as adjustable optical bandgap, greater absorption

ISBN: 978-86-82602-05-7

coefficient, improved carrier transportation and weak exciton binding energy [4]. Since the inception of PSCs, they have achieved 20% efficiency improvements in the past few decades due to technological advancements in the photovoltaic field. As per the efficiency chart published by the National Renewable Energy Laboratory 2023, the maximum certified PCE achieved by single-junction perovskite-based solar cells is 26.1% [5].

ABX3 is the standard chemical formula of perovskites. Here, A is denoted as monovalent cations such as methylammonium [MA+: CH3NH3+), formamidine [FA+], Cesium (Cs+), or their blend. B is divalent cations usually metals, such as Pb2+, Sn2+, Ge2+, or a blend of these. Monovalent anions are indicated by X, typically halides, including I−, Br−, Cl−, or a mixture of them [6]. 3D-MAPbI3 based PSCs are considered the most prominent PSCs in the market due to their higher PCE and low production cost. However, they are highly prone to degradation under external environmental factors such as heat, moisture and UV radiation resulting in deterioration of performance in long-term operation [7]. This is the main barrier behind the commercialization of MAPbI3 based solar cells.

2D perovskite materials show excellent moisture, heat and UV stability over their 3D counterparts. Ruddlesden–Popper (RP), Dion–Jacobson (DJ), and Alternation cation in the interlayer space perovskites (ACI) are the most prevailing 2D perovskite types [8]. The incorporation of 2D materials as a capping layer on the 3D perovskite has been proven as a successful way to boost the stability of the 3D perovskite materials. The resultant 2D/3D hybrid solar cell shows balanced efficiency and long-term stability compared to the PSCs [9-11]. For instance, [10] developed phenethylammonium based 2D with 3D perovskites. The stability of the system is enhanced, and it achieved a PCE of 19.98% with Voc of 1.17 V. Further, [11] incorporated thin 2D n-butylamine with 3D perovskite film to form a 2D/3D mixed dimensional solar cell which remained at 93% of the initial PCE after 55 days in the air with humidity of about 50% at room temperature.

The 2D form of MAPbI3 perovskite material is more environmentally stable than its 3D counterpart making them an excellent candidate for incorporation as a capping layer [12].

The layered structure facilitates reducing issues such as moisture sensitivity and thermal instability in 2D-MAPbI3. Furthermore, 2D structures can enhance charge transport properties resulting in greater carrier mobility and fewer recombination losses. Consequently, it offers better higher power conversion efficiencies in the PSCs. Nipuna L et al numerically simulated a solar cell inserting 3D-MAPbI3 between two 2D-MAPbI3 layers with the cell architecture of Glass/ITO/PEDOT: PSS/2D-MAPbI3/3D-MAPbI3/2D-MAPbI3/PC BM/Ag. Here, it has been investigated the effect of deep defect density of 3D-MAPbI3 layer on the performance of the PSC. The results show that the highest performance of the device is given when the deep defect density ranges from 1010-1012 cm-3 [13].

In this study, a 2D/3D hybrid solar cell was numerically simulated by applying 2D-MAPbI3 capping layer on the top of the 3D-MAPbI3 layer to reduce the degradation of the 3D-MAPbI3 upon exposure to heat and moisture. The simulated p-i-n device architecture was $FTO/TiO_2/3D\text{-}MAPbI_3/2D\text{-}MAPbI_3/Cu_2O/Au$. The effect of the thickness and defect density of the absorber layers were investigated to optimize the performance of the solar cell.

II. METHODOLOGY

The 2D/3D mixed dimensional PSC with the device architecture of FTO/TiO2/3D-MAPbI3/2D-MAPbI3/Cu2O/Au was simulated in this study. The front contact electrode was made of Fluorine-doped tin oxide (FTO) while Cu2O was utilized as the hole transport layer (HTL) which is a p-type hole-accepting material where the majority of carriers are holes. As the perovskite active absorber materials 3D-MAPbI3 was utilized and as a protective layer 2D-MAPbI3 was applied. In the proposed structure, TiO2 layer acts as the electron transport layer (ETL) and Gold (Au) acts as the back contact cathode material as depicted in Fig 1.

The Solar Cell Capacitance Simulator (SCAPS-1D) software was employed to simulate the proposed device. It is commonly used in the simulation of optoelectronic devices particularly for solar cells, due to its capability of solving the Continuity equation and Poisson equation, allowing accurately forecast the output of photovoltaic devices [14]. The presented cell structure was drawn in the software and simulation was performed using the drift-

diffusion model at 300 K temperature upon one sun illumination (AM1.5G, 100 mW/cm^{-2}).

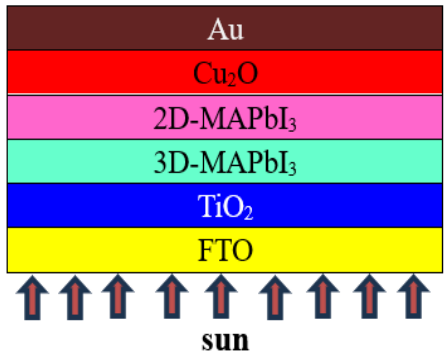

Figure 1. Proposed device architecture: Au/Cu_2O/2D-$MAPbI_3$/3D-$MAPbI_3$/TiO_2/FTO.

TABLE I. OPTICAL AND ELECTRONIC PARAMETERS USED IN SIMULATION FOR CHARGE TRANSPORT LAYERS AND FRONT ELECTRODE.

S/ No	Description of the parameter	FTO [15]	TiO_2 [14,16]	Cu_2O [14,16]
1	Thickness (μm)	0.5	0.1	0.4
2	Band gap (eV)	3.5	3.26	2.22
3	Electron affinity (eV)	4	4.2	3.40
4	Dielectric permeability relative	9	38	7.11
5	VB effective density of state (states/cm^3/eV)	2.2x 10^{18}	2.2 x 10^{17}	2.02x 10^{17}
	CB effective density of state (states/cm^3/eV)	1.8x 10^{19}	1.8x 10^{18}	3.0x 10^{20}
6	Hole thermal velocity (cms^{-2})	1x10^7	1x10^7	1x10^7
	Electron thermal velocity (cms^{-2})	1x10^7	1x10^7	1x10^7
7	Hole mobility (cm²/Vs)	20	2x10^4	30
	Electron mobility (cm²/Vs)	10	1x10^4	30
8	Shallow uniform donor density (cm^{-3})	10^{15}	6x10^{19}	0
	Shallow uniform donor density (cm^{-3})	0	0	1x10^{18}
9	ElectronCapture cross section (cm^2)	1x 10^{-15}	1x10^{-15}	1x10^{-15}
10	Hole Capture cross section (cm^2)	1x 10^{-15}	1x10^{-15}	1x10^{-15}
11	Defect density	1x10^{12}	1x10^{12}	1x10^{12}

The thickness of the 3D-$MAPbI_3$ varied between 0.2 μm-1.5 μm as reported in the literature. The 2D-$MAPbI_3$ should be a thin layer so that its thickness is kept at 0.03 μm as reported in previous studies [13]. In this theoretical study, the defect density of 3D layers varied from 10^{10}-10^{17} cm^{-3} to explore the impact of defect densities on the overall performance of the device keeping the thickness of the layers at the optimum values. The parameters used in SCAPS-1D simulations for the different absorber materials, transport layers and electrodes were adopted from previously published reports as depicted in Tables I and II.

TABLE II. OPTICAL AND ELECTRONIC PARAMETERS USED IN THE SIMULATION FOR ABSORBER LAYERS.

S/No	Description of the parameter	3D-$MAPbI_3$ [14,17]	2D-$MAPbI_3$ [13]
1	Thickness (μm)	varied	0.03
2	Band gap (eV)	1.55	1.63
3	Electron affinity (eV)	6.90	3.9
4	Dielectric permeability relative	6.50	6.5
5	VB effective density of state (states/cm^3/eV)	2.75x 10^{18}	1.94×10^{20}
	CB effective density of state (states/cm^3/eV)	3.9 x 10^{18}	1.94×10^{20}
6	Hole thermal velocity (cms^{-2})	1x10^7	1x10^7
	Electron thermal velocity (cms^{-2})	1x10^7	1x10^7
7	Hole mobility (cm²/Vs)	11.8	414
	Electron mobility (cm²/Vs)	11.8	1184
8	Shallow uniform donor density (cm^{-3})	2.2x10^{18}	1x10^{16}
	Shallow uniform donor density (cm^{-3})	9x10^{20}	1x10^{16}
9	ElectronCapture cross section (cm^2)	1x10^{-15}	1x10^{-15}
10	Hole Capture cross section (cm^2)	1x10^{-15}	1x10^{-15}
11	Defect density	varied	2.5x10^{12}

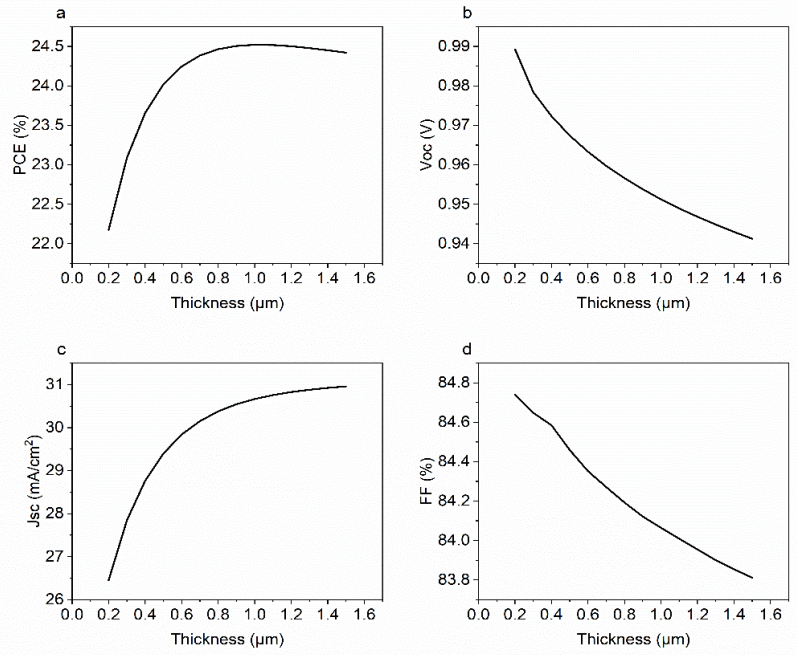

Figure 2. (a) efficiency, (b) Voc, (c) Jsc, (d) FF as a function of 3D-$MAPbI_3$ layer thickness.

III. Results and Discussion

A. *Effect of 3D-$MAPbI_3$ Absorber Layer Thickness on the Performance of the PSC*

As illustrated in Fig. 2a, the PCE of the device increases with the thickness of the 3D-$MAPbI_3$ layer until a threshold thickness of 1μm and then starts to decline further increasing the thickness. The recorded maximum PCE is 24.52%. The main reason behind this behavior can be related to the thickness-dependent absorption characteristics of the active absorber layer. In the relatively lower thicknesses, the absorption in the longer wavelength is limited leading to the creation of limited electron-hole pairs. Increasing the perovskite layer thickness promotes the absorption of longer wavelengths resulting in improved electron-hole pair generation [14]. However, when further enhancing the thickness beyond a critical value, even a greater percentage of the solar spectrum can be absorbed, the generated charge carriers are required to travel long distances to reach the electrodes which increases the likelihood of carrier recombination [18]. Subsequently, the PCE of the device declines after the thickness surpasses a critical threshold. As per Fig. 2b and Fig. 2d the V_{oc} and FF are decreasing when the absorber layer thickness rises. This is mainly caused by the increased carrier recombination and series resistance due to thickness enhancement. The thicker absorber layer introduces more series resistance leading to voltage drop under the current flow and reducing the effectiveness of charge collection, thereby reducing the V_{oc}. The decrement in the V_{oc} drops the maximum output power and thereby reduces the FF [19]. At the optimum thickness value, the device shows V_{oc} of 0.95 V, J_{sc} of 30.66 mA/cm^{2,} and FF of 84.07%.

B. *Effect of 3D-$MAPbI_3$ Absorber Layer Defect Density on the Performance of the PSC*

The defect density of the perovskite absorber layer greatly affects the photovoltaic characteristics of the PSCs. If the defect density is high, it declines the efficiency and the overall performance of the device as it increases the recombination losses. The behavior of photovoltaic parameters with defect density of the 3D-$MAPbI_3$ layer is shown in Fig. 3. As per Fig. 3a, when defect density increases up to 10^{12} the PCE remains unchanged at 24.52%, however, when further increases the defect density, PCE starts to decline. Similar behavior can be observed in V_{oc}, J_{sc} and *FF* as well. To determine the impact of the defect density of the perovskite layer, the Shockley – Read – Hall (SRH) model can be used. This model portrays

the way defects within the bandgap of a semiconductor can behave as recombination centers for charge carrier pairs influencing carrier dynamics and device performance as mentioned in Eqs. (1) and (2) [18].

$$R_{SRH} = \frac{np - n_i^2}{\tau_p\left(n + n_i\right) + \tau_n\left(p + p_i\right)}, \qquad (1)$$

$$\tau_{n,p} = \frac{1}{\sigma v_{th,n,p} N_t}. \qquad (2)$$

Here R_{SRH} is denoted by the rate of the Shockley–Read–Hall recombination, n and p are the concentration of electron and hole respectively. τ_p, τ_n are the lifetime of electron and hole. $\upsilon_{th,n,p}$ are the thermal velocities of electrons and holes, respectively. With relatively low defect density levels, the recombination rate (R_{SRH}) is small. Therefore, the majority of the photo-generated carriers can be effectively collected by the electrodes before recombination [14]. Hence, up to a certain defect density value, the device performance remains unaffected. However, at moderate and higher defect density levels the trap states become significant, and the performance starts to decline. As per equation 2, defect density (N_t) is inversely proportional to the lifetime of the electron and hole. When increasing the defect density, the non-radiative recombination enhances and thereby reduces the overall performance of the device [13].

The I-V characteristics of the proposed device after optimizing the thickness and the defect density of 3D-$MAPbI_3$ absorber layer is depicted in Fig. 4. In the same graph, IV curves $MAPbI_3$ based solar cells without the capping layer for the same device architecture is also included. The two curves are identical. In this study, a thin capping layer was incorporated so that it does not have a substantial impact on bulk properties of solar cells such as charge transport and light absorption. Therefore, incorporating 2D capping layer has a limited effect on overall performance.

The effect of the other parameters such as interface defect densities, recombination rates, series, shunt resistance and light intensities can

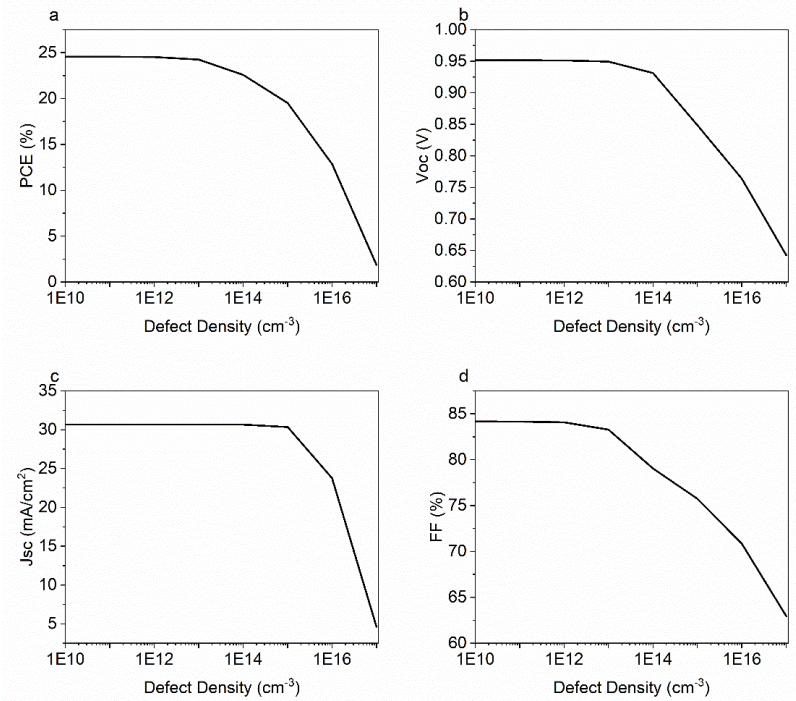

Figure 3. (a) efficiency, (b) Voc, (c) Jsc, (d) FF as a function of 3D-$MAPbI_3$ layer defect density.

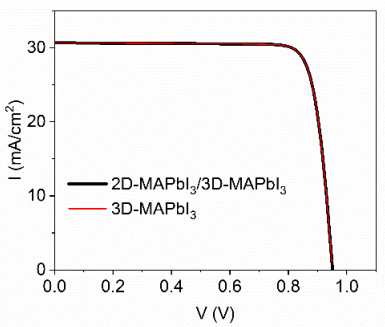

Figure 4. I-V characteristics of devices with and without the capping layer.

be comprehensively analyzed to further optimization for the device. In addition to that, by changing the number of layers in 2D-$MAPbI_3$, the overall efficiency of the hybrid device can be enhanced as increasing the number of layers results in lower bandgap and enhanced carrier mobilities [12]. Further, the literature suggests that performance and stability enhancement of these 2D/3D mixed dimensional PSCs can be achieved by changing the composition by substituting different organic spacer cations [20,21].

The previous research studies have shown that 2D perovskite exhibits enhanced thermal and moisture stability compared to their 3D counterparts. Furthermore, several studies have demonstrated that incorporating 2D capping layers on 3D perovskite materials can enhance the stability of the 3D perovskite without compromising the performance. Therefore, the proposed 2D-$MAPbI_3$ capping layer is expected to enhance the stability of 3D $MAPbI_3$-based hybrid cell by providing additional resistance to environmental degradation. However, a comprehensive investigation of stability enhancements through experimental validation would be beneficial. Based on the findings of this numerical study, future experiment studies can be built to confirm the stability of enhancement afforded by 2D-$MAPbI_3$.

IV. Conclusion

In this research, the 2D/3D mixed dimensional solar with device architecture of $Au/Cu_2O/2D\text{-}MAPbI_3/3D\text{-}MAPbI_3/TiO_2/FTO$ was numerically simulated using SCAPS-1D software. The 2D-$MAPbI_3$ layer is capped on top of the 3D-$MAPbI_3$ layer to boost the long-term operational stability of 3D perovskite. The performance of the PSC was optimized by varying the thickness and the defect density of the 3D-$MAPbI_3$ layer. At the optimum thickness of 1µm and defect density range of 10^{10}-10^{12} cm^3, the proposed device achieved a PCE of 24.52%. This study offers valuable insight into the designing and optimization of 2D/3D mixed dimensional PSCs with balanced efficiency and long-term stability. Ultimately, it enables laboratory-scale fabrication of PSCs with minimal effort and cost-effective manner.

Acknowledgment

The authors sincerely recognize Prof. Marc Burgelman and his team at the University of Gent, Belgium, for creating, maintaining and freely distributing the SCAPS-1D software, which was essential in performing the solar cell simulations in this research.

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