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$MAGeI_3$ based Multi-dimensional Perovskite Solar Cells for Superior Stability and Efficiency

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Abstract: Perovskite solar cells (PSC)s have driven the photovoltaic technology to miles apart as a promising post-silicon photovoltaic technology. However, their decency in providing efficiency is quite intriguing but remains poor in stability. Advancement in lower dimension technology indicates shortcomings of 3D perovskite materials, which can be overcome by the introduction of 2D perovskites in an appropriate manner. 2D perovskites have piqued the interest in photovoltaic technology because of their remarkable structural and electrical properties which yield an increase in stability and enhance its light absorption properties. Therefore 2D/3D multi-dimensional perovskite solar cells are expected to provide substantial stability and higher efficiency. In this study, 2D perovskite materials such as $BA_2MA_2Pb_3I_{10}$ and $BA_2MA_2Pb_4I_{13}$ have been used as the capping layer on the 3D $MAGeI_3$ layer to fulfil the mixed-dimensionality. The band alignment of both 2D and 3D perovskite have been matched decently and other properties like defect tolerance and other IV characteristics on varying defect densities have been provided in this study. The mixed dimensional perovskite with n=4, has shown increased efficiency with respect to single 3D perovskite in decimals, yet more stable in harsh environments.

Keywords: 2D perovskite; Mixed-dimensional perovskite; Ruddlesden-Popper perovskite solar cell

1. Introduction

In perovskite solar cells (PSCs), a material with a perovskite crystal structure is used as the active layer. In recent years, perovskite materials, as they are promising candidates to provide high power conversion efficiency (PCE) in solar cells at low cost. Moreover, 2D PSCs use a thin layered perovskite structure instead of the traditional 3D structure. The layers are made up of organic cations and inorganic halides and are sandwiched between two electrode layers.

Tunability and higher stability make 2D perovskites attractive over traditional 3D perovskites. The thin layered architectures of 2D perovskites reduce the number of defects in the crystal structure, leading to increased stability. Additionally, the organic cations in 2D perovskites can be easily modified, allowing for adjustment of the optoelectronic properties of the material [1].

Despite these advantages, 2D PSCs still face challenges such as lower power conversion efficiencies compared to 3D perovskites and greater susceptibility to moisture and temperature changes. However, notable research efforts are focused on the improvement of the performance and stability of 2D perovskite solar cells [2].

One promising approach is the use of 2D perovskite material as a passivation layer to improve the stability of the 3D perovskite layer. *Zhang et al.* (2022) reported a higher efficiency of 2D/3D PSC using a 2D perovskite passivation layer. They have also obtained improved stability and a higher PCE with a value of 20.31% [3].

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Citation: Devi, H.; Lalat, G.; Midya, K.; $MAGeI_3$ based Multi-dimensional Perovskite Solar Cells for Superior Stability and Efficiency. *Eng. Proc.* **2023**, *1*, 0. https://doi.org/

Published:

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Furthermore, researchers have also investigated the use of composite layers of 2D and 3D perovskites to achieve high stability and efficiency. *Choi et al.* (2020) summarised the advantages of integration of a 2D perovskite layer as a cap on 3D perovskite and tested several organic compounds influencing the stability and overall performance [4]. In addition, several studies have focused on optimizing the composition and processing conditions of 2D perovskites to enrich their stability and device performance [5].

$BA_2MA_{n-1}PbnX_{3n+1}$ (such as $BA_2MA_2Pb_3I_{10}$ and $BA_2MA_3Pb_4I_{13}$) is a layered perovskite material which has been studied for its potential applications in optoelectronics, particularly in solar cells. It has a unique crystal structure that consists of alternating layers of organic and inorganic components, which provides interesting properties. The primary advantage of their properties is their high stability, both in ambient conditions and under high temperatures and humidity [5]. This is attributed to the presence of the bulky organic cations, which act as protective layers around the inorganic perovskite layers, preventing moisture and other contaminants from accessing the material. Better stability has been demonstrated in various studies. *Sidhik et at.*, (2023) reported the high stability of $BA_2MA_3Pb_4I_{13}$-based solar cells over 2000 hours of continuous operation and degraded for 25% of its initial PCE [6]. Another advantage of $BA_2MA_2Pb_3I_{10}$ is its high PCE in solar cells. Studies have shown that $BA_2MA_2Pb_4I_{13}$-based solar cells can achieve PCEs of up to 12.51 % and even up to 14.2% (*Ji et al.*, 2021) with optimization of the device structure and optimized fabrication process [5][7] [8]. The high efficiency is attributed to the unique crystal structure of $BA_2MA_2Pb_3I_{10}$, which allows for efficient charge transport and suppression of non-radiative recombination.

$MAGeI_3$ (M = Methylammonium, A = Cesium, Ge = germanium, I = iodine) is a promising perovskite material that has been recently investigated for its potential use in solar cells. $MAGeI_3$ has a unique crystal structure and similar properties as $MAPbI_3$ perovskite material, which allows for efficient charge transport and improved stability compared to other perovskite materials, theoretically proven by Density Functional Theory (DFT) methods [9][10]. One study reported that $MAGeI_3$ solar cells had a PCE of 13.1%, which is comparable to other high-performance perovskite solar cells [10] [11]. Another study found that doping with p-type and n-type to $MAGeI_3$ absorber layer reached a maximum efficiency of 19.16% with equal bulk defect densities [12]. $MAGeI_3$ also exhibits good stability under varying environmental conditions, including high temperature and humidity.

In summary, it can be predicted that the combination of $MAGeI_3$ with 2D perovskites as encapsulation layer (i.e., $BA_2MA_2Pb_3I_{10}$ and $BA_2MA_2Pb_4I_{13}$) might be proved as a perovskite material with high-performance and stable solar cells. Therefore, detailed research is needed to optimize the device architecture and improve the efficiency and stability of both $MAGeI_3$, $BA_2MA_2Pb_3I_{10}$ and $BA_2MA_2Pb_4I_{13}$-rooted solar cells.

2. Materials and Methods

2.1. Simulations

SCAPS-1D is a widely used solar cell simulator that simulates the performance of solar cells under various operating conditions. It is a one-dimensional simulation tool that can predict the behaviour of solar cells based on their material properties and device architecture. SCAPS-1D uses a detailed physics-based model to simulate the generation, transport, and recombination of charge carriers within the solar cell structure. SCAPS-1D is a widely used tool for solar cell researchers and engineers to design, optimize, and evaluate the performance analysis of a solar cell. Its accuracy and flexibility make it a valuable resource in the development of new solar cell technologies [13].

The continuity equation of electrons and holes:

$$q\frac{\partial n}{\partial t} = \frac{\partial J_n}{\partial x} + qG - aR \quad (1)$$

$$q\frac{\partial p}{\partial t} = -\frac{\partial J_p}{\partial x} + qG - aR \tag{2}$$

The Poisson equation:

$$\frac{\partial^2}{\partial x^2}\varphi(x_i) = \frac{q}{\epsilon}[n(x_i) - p(x_i)] \tag{3}$$

For PSCs, The electrons and holes move by diffusion, which is the tendency of particles to spread out evenly, and by drift, which is the tendency of particles to move in the direction of an electric field:

$$J_{(electron)} = J_{df} + J_{dr} = q[n\mu_n\frac{\partial \varphi}{\partial x} + D_n\frac{\partial n}{\partial x}] \tag{4}$$

$$J_{(hole)} = J_{dfn} + J_{dr} = -q[p\mu_p\frac{\partial \varphi}{\partial x} + D_p\frac{\partial p}{\partial x}] \tag{5}$$

J_{df}, J_{dr} are the diffusion and drift current.

Einstein's relation for a semiconductor:

$$D_{p,n} = \mu_{p,n}\frac{k_B T}{q} = V_t \tag{6}$$

It also accounts for optical generation, recombination mechanisms, contact properties and external parameters such as temperature and illumination. SCAPS-1D can be used to optimize the design and performance of the device (solar cell) and modules based on different materials and structures.

2.2. Structure

The device architecture of HTL/3D/2D/ETL/ITO Fig.1 has been simulated using the SCAPS-1D solar simulator. The back contact layer is a thin film of metal or transparent conductive oxide (TCO) that serves as the bottom electrode and provides good electrical contact with the substrate. The HTL layer is a hole transport layer that facilitates the extraction of positive charges from the perovskite layer and blocks the recombination of electrons. The 3D layer is a three-dimensional perovskite layer that absorbs sunlight and generates electron-hole pairs. The 2D layer is a two-dimensional perovskite layer that acts as a passivation layer and prevents the degradation of the 3D layer. The ETL layer is an electron transport layer that facilitates the extraction of negative charges from the perovskite layer and blocks the recombination of holes. The ITO or glass layer is a transparent top electrode that allows light to enter the device and collects the current.

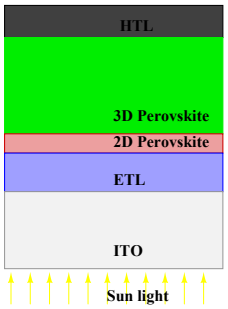

Figure 1. Device architecture of 2D/3D perovskite solar cell

In short, SCAPS-1D can be used to simulate various device architectures for different types of solar cells, by adjusting the parameters such as thickness, carrier concentration, defect density, work function and temperature of each layer. By doing so, performance and stability optimization can be performed by exploring new materials and device structures.

3. Results

3.1. Energy band gap in 2D/3D perovskite solar cells

In a 2D/3D perovskite solar cell, the 2D perovskite layer (such as $BA_2MA_2Pb_3I_{10}$ and $BA_2MA_3Pb_4I_{13}$) operates as a passivation layer, while the 3D perovskite layer (such as $MAGeI_3$) acts as the light-absorbing layer. Each layer's bandgap controls the energy levels at which photon absorption takes place. The bandgap for both the 2D and 3D perovskite

Parameters	$MAGeI_3$[14]	$BA_2MA_2Pb_3I_{10}$[15]	$BA_2MA_3Pb_4I_{13}$[15]	C_{60} [14]	Cu_2O[15]	ITO[14]
L (nm)	850	50	50	50	250	100
E_G (eV)	1.9	1.85	1.60	1.7	3.3	3.65
χ (eV)	3.98	3.53	3.87	3.9	4.8	
ϵ_r	10	5.7	5.8	4.2	9	8.9
N_C (cm^{-3})	1×10^{16}	7.6×10^{17}	7.24×10^{17}	8×10^{19}	2×10^{13}	5.8×10^{18}
N_V (cm^{-3})	1×10^{15}	1.33×10^{18}	1.5×10^{18}	8×10^{19}	2×10^{13}	1×10^{18}
μ_e (cm^2/Vs)	16.2	0.8	1.4	8×10^{-2}	100	10
μ_h (cm^2/Vs)	10.1	0.8	1.4	3.5×10^{-3}	25	10
N_D (cm^{-3})	10×10^{19}	1×10^{10}	1×10^{10}	2.6×10^{18}	1×10^{18}	1×10^{20}
N_A (cm^{-3})	10×10^{9}	-	-	-	-	
N_t (cm^{-3})	Varied	Varied	Varied	-	-	-

Table 1. Input parameters for various perovskite materials.

layers of a 2D/3D perovskite solar cell is determined by several factors, including device performance, stability, and materials selected. Optimising the bandgap of each layer results in better light absorption, less charge recombination, and overall device performance [17]. The bandgap of the proposed device has been depicted in Fig.2(a)

Rudlesden-Popper (RP) 2D perovskites have piqued the interest of photovoltaic researchers because of their remarkable structural and electrical properties. Two notable RP 2D perovskites are $BA_2MA_2Pb_3I_{10}$ and $BA_2MA_3Pb_4I_{13}$. This study is focused on the performance analysis of solar cells with the bandgap variation to find the most suitable value.

For optimal light absorption, the bandgap of the 3D perovskite layer should be optimised to match the solar spectrum. It should be wide enough to absorb a considerable percentage of the solar spectrum but not so wide that it causes significant losses from photons with energies below the bandgap. Tuning the bandgap of the 3D perovskite layer, such as utilising mixed cations or halide composition engineering, can assist in achieving optimum absorption and maximising device efficiency.

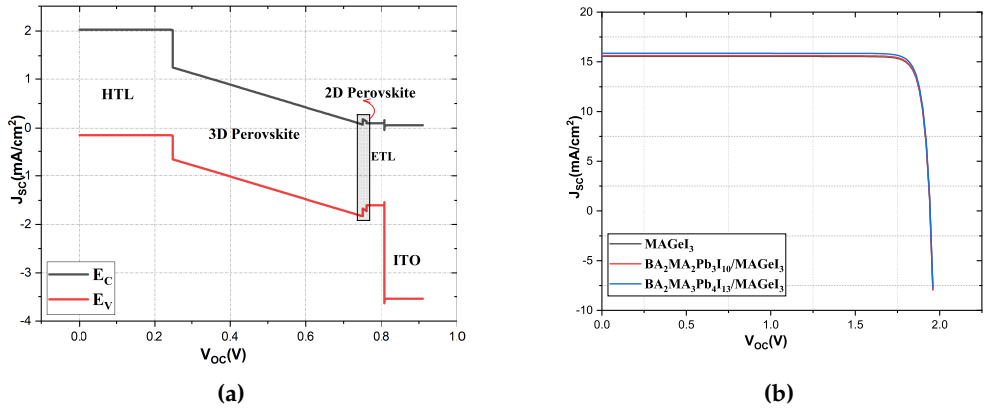

Figure 2. Schematic representation of device architecture (a) band diagram and (b) J-V curve

3.2. Efficiency of the device

Liu et al. (2019) investigated the photovoltaic performance of 2D/3D PSCs using 2D passivation layers of $BA_2MA_2Pb_4I_{13}$ and $BA_2MA_2Pb_3I_{10}$. They discovered that the device containing $BA_2MA_2Pb_4I_{13}$ had a greater power conversion efficiency and better stability during light soaking than the device containing $BA_2MA_2Pb_3I_{10}$. The JV curve of various perovskite solar cells has been depicted in Fig.2(b), including a 3D perovskite solar cell. The researchers ascribed this improvement to $BA_2MA_2Pb_4I_{13}$'s properties such as high carrier mobility and more organised crystal structure is responsible for the higher efficiency [6]. They demonstrated that the surface roughness of $BA_2MA_3Pb_4I_{13}$ was less

responsive to annealing temperature than $BA_2MA_3Pb_3I_{10}$, implying higher film quality and the possibility of device stability.

Parameters				
Absorber layer 2D/3D	**Voc**	**Jsc**	**FF**	**PCE**
$MAGeI_3$ (Only 3D)	1.9430	15.548	89.40	27.01 %
$BA_2MA_2Pb_3I_{10}$/$MAGeI_3$	1.9439	15.919	89.34	27.65 %
$BA_2MA_2Pb_4I_{13}$/$MAGeI_3$	1.9466	17.006	89.52	29.64 %
Glass/ITO/PEDOT:PSS/$BA_2MA_3Pb_4I_{13}$/$PC_{61}BM$/Ag (Experimental [[17]])	0.9540	16.13	69.51	10.70 %

Table 2. Simulated parameters for various devices with different absorber materials with an architecture of CuO_2/Perovskite 3D/2D/C_{60}/TCO as well as the experimental work.

3.3. Deffects in 2D perovskites

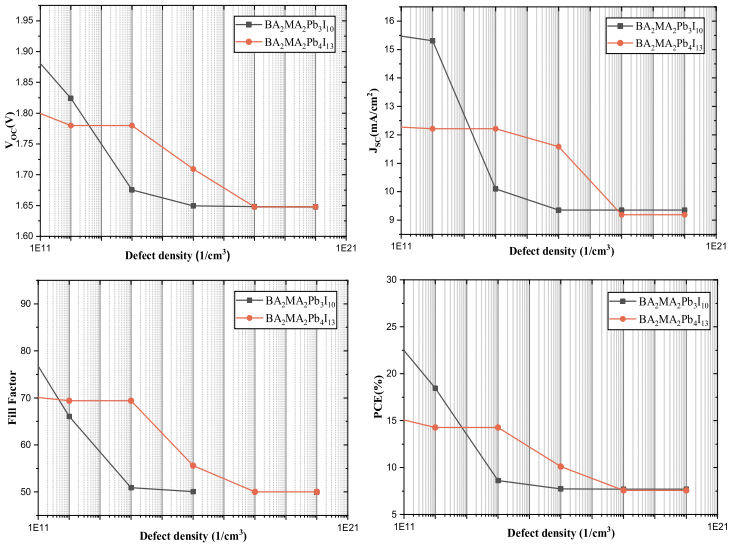

Figure 3. The effect of defects in 2D in IV characteristics

The defect densities in 2D perovskites $BA_2MA_2Pb_3I_{10}$ and $BA_2MA_2Pb_4I_{13}$ have a considerable influence on the functioning of 2D/3D PSCs. Defects in perovskite materials can occur due to a variety of factors such as crystal lattice defects, impurities and structural disorders. These defects have the potential to inject energy levels into the bandgap, influencing charge transport, recombination processes, and the overall performance of the device. In this paper, we will look at the significance of the defect densities in 2D perovskites $BA_2MA_2Pb_3I_{10}$ and $BA_2MA_2Pb_4I_{13}$ in the context of 2D/3D multi-dimensional PSCs.

4. Conclusions

In this work, mixed-dimensional (2D/3D) PSCs employing Ruddlesden-Popper (RP) 2D perovskites as the capping layer onto the 3D $MAGeI_3$ perovskite material have been examined. The band alignment of 2D Perovskite along with 3D Perovskite was well matched, and the defect tolerance and other IV features on changing defect concentrations were also presented. The mixed dimensional perovskite with n=4 showed greater efficiency in numbers over a single 3D perovskite while remaining more robust in demanding conditions. These findings indicate that mixed-dimensional PSCs have the capability to be significantly more stable and efficient than conventional 3D PSCs. However, further study is needed

to increase the efficiency of these cells and understand the underlying principles of their greater stability.

Acknowledgments: The authors would like to appreciate the usage of the SCAPS 1D software tool in the modelling and analysis of the solar cells reported in this research study. The software was crucial in anticipating device performance and optimising device architecture. The authors would like to thank the SCAPS 1D developers for creating such a robust and user-friendly simulation tool for the scientific community.

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