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Improved efficiency of $MAPbI_3$ perovskite solar cells through a numerical study utilizing TiO_2/ZnO as bilayer ETLs

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Abstract—**This paper utilized the SCAPS-1D Simulator to examine the photovoltaic characteristics of perovskite solar cells utilizing lead-free $MAPbI_3$.The theoretical device outputs obtained through the simulation were compared with experimental results, confirming the accuracy of the simulation. The study examined the influence of the thickness and carrier concentration of the absorber layer and different bilayer electron transport layer (ETL) options to optimize the device's performance and achieve higher efficiency. An improved efficiency of 29.26% was achieved at optimized thicknesses of 800nm for the absorber layer with the employment of bilayer ETL TiO_2/ZnO.**

Keywords—perovskite solar cell ; Device simulation; High efficiency; SCAPS-1D

List of Symbols and abbreviations

E_g (eV): Energy Band gap
χ (eV): Electron affinity
ε_r: Relative dielectric permittivity
Nc (cm^{-3}): CB effective density of states
Nv (cm^{-3}): VB effective density of states
PCE (%): Power Conversion Efficiency
FF (%): Fill Factor
FTO: Fluorine doped Tin Oxide
PSC: Perovskite Solar Cell
μ_n (cm^2/Vs): Electron mobility
μ_p (cm^2 /Vs): Hole mobility
N_D (cm^{-3}): Shallow uniform donor density
N_A (cm^{-3}): Shallow uniform acceptor density
Nt: Defect density
Voc (V): Open circuit voltage
Jsc (mA/cm^2): Short circuit current density
QE (%): Quantum efficiency
$MAPbI_3$: Methylammonium Lead Iodide
ETL: Electron Transport Layer
HTL: Hole Transport Layer
ZnO: Zinc oxide
TiO_2: Titanium dioxide
WO_3: Tungsten trioxide
SnO_2: Tin oxide

I. INTRODUCTION

The rapid progress in the power conversion efficiency (PCE) of perovskite solar cells (PSCs) has recently positioned them as a promising candidate for next-generation photovoltaic technology. This is attributed to their exceptionally high PCEs and favorable electrical properties. Additionally, PSCs offer advantages such as Low-cost fabrication techniques at moderate temperatures, a direct bandgap, reduced exciton binding energy, and enhanced light absorption characteristics [1]. The structure of a perovskite solar cell commonly consists of several key components, including a transparent anode layer such as indium tin oxides (ITO) or fluorine-doped tin oxides (FTO), an electron transport layer (ETL), a perovskite layer serving as the light-sensitive component, a hole transport layer (HTL), and a metal cathode such as gold (Au) or silver (Ag)

Methyl ammonium lead iodide ($MAPbI_3$ or $CH_3NH_3PbI_3$), an organic-inorganic halide perovskite compound lead, has attracted considerable interest as a promising material for solar cell fabrication. This is due to

its long carrier diffusion length, high carrier mobility, optimum band gap of 1.55 eV, high absorption coefficient, and exceptional energy harvesting efficiency, attributed to its low trap density[2].

The ETL plays a crucial role in achieving high power conversion efficiencies (PCEs) by facilitating the transport of light-induced electrons from the perovskite layer to the photoanode. This transport pathway helps to prevent recombination occurs when holes generated within the perovskite layer combine with other charge carriers. layer, thus improving overall device performance. This study focuses on simulating a solar cell with various bilayer electron transport configurations, such as TiO_2/SnO_2, TiO_2/ZnO, and TiO_2/WO_3, to enhance efficiency. The proposed structure significantly improved key solar cell metrics, including efficiency and fill factor (FF), leading to a peak efficiency of 29.26 % for the tested solar cell. Overall, the ZnO/TiO_2 bilayer ETL offers a promising approach to improve PSCs' efficiency, stability, and scalability, making it an important research area for perovskite solar cell technology.

II. MATERIALS AND METHOD

A. Numerical simulation

The operational principle of SCAPS-1D involves solving the core equations governing one-dimensional semiconductors. These equations include Poisson's equation, as well as equations that govern the continuity of holes and electrons. The drift and diffusion equations are also included, as depicted below [3][4]:

$$\frac{\partial}{\partial x}\left(\varepsilon^0 \varepsilon_r \frac{\delta \Psi}{\delta x}\right) = -q(p - n + N_d^+ - N_A^- + \frac{\rho\, def}{q} \qquad (1)$$

$$-\frac{\partial J_n}{\partial x} - U_n + G = \frac{\partial n}{\partial t} \qquad (2)$$

$$-\frac{\partial J_p}{\partial x} - U_p + G = \frac{\partial p}{\partial t} \qquad (3)$$

$$j_n = -\frac{\mu_n n}{q}\frac{\partial E_{Fn}}{\partial x} \qquad (4)$$

$$j_p = +\frac{\mu_p p}{q}\frac{\partial E_{Fp}}{\partial x} \qquad (5)$$

In the given context
- ε represents the dielectric constant.
- q symbolizes the quantity of charge.
- N_d^+ and N_A^- denote donors and acceptors concentration, respectively
- Ψ represents the electrostatic potential.
- J_p indicates the photo-generated hole current density.
- J_n represents the photo-generated electron current density.
- G represents the rate of carrier generation.
- p signifies the density of free holes.
- n denotes the density of free electrons.
- E_{Fn}, and E_{FP}: represent the Fermi level energy of n-type carriers and p-type carrier Fermi level energies, respectively.

B. Device structure

The simulation employed a device structure consisting of $FTO/TiO_2/ZnO/MAPbI_3/Au$, as shown in Figure 1. The parameters for each layer were carefully chosen, considering both experimental and theoretical data [4]. The important parameters of the proposed structure used in the SCAPS simulation are summarized in Table 1 below.

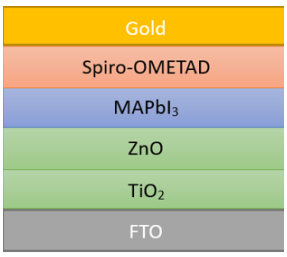

Figure 1. Structure solar cell

TABLE I. PHYSICAL PARAMETERS USED IN THIS SIMULATION STUDY [5][6][7][8]

Materials	**p-Spiro-OMETAD**	**p-Cu_2O**	**P-P3H**	**p-$MAPbI_3$**	**n-ZnO**	**n-TiO_2**	**n-SnO_2**	**n-WO_3**
Thickness (nm)	80	80	80	Varied	50	35	50	50
Band gap(eV)	2.9	2.17	1.85	1.55	3.3	3.2	3.5	3.15
Electron affinity(eV)	2.2	3.2	3.1	3.93	4	4	4	4.55
Dielectric permittivity (relative)	3	7.1	3.4	30	9	9	9	10
C_B effective density of states(cm^{-3})	$2.5x10^{20}$	$2.5x10^{18}$	10^{22}	$2.75x10^{18}$	$3.7x10^{18}$	10^{21}	$2.2x10^{18}$	$4.2x10^{18}$
V_B effective density of states(cm^{-3})	$2.5x10^{20}$	$1.8x10^{19}$	10^{22}	$3.9x10^{18}$	$1.8x10^{19}$	10^{20}	$1.8x10^{19}$	$9x10^{18}$
Electron mobility (cm^2/V.S)	$2.1x10^{-3}$	$2x10^2$	10^{-4}	10	$2x10^2$	20	20	20
Hole mobility (cm^2/V.S)	$2.6x10^{-3}$	$8.6x10^3$	10^{-3}	10	$2.5x10^{10}$	10	10	10
Shallow uniform donor dx10nsity ND (cm^{-3})	-	-	-	-	10^{18}	10^{18}	10^{19}	10^{18}
Shallow uniform donor density NA (cm^{-3})	10^{18}	10^{18}	10^{14}	10^{19}	-	-	-	-
Nt (cm^{-3})	10^{14}	10^{15}	10^{14}	10^{14}	10^{14}	10^{14}	10^{13}	10^{15}

III. VALIDATION OF THE DEVICE MODEL

Before commencing the simulation of the proposed device, we conducted validation work on existing perovskite solar cells, as detailed in the reference [5]. The architectures described in the reference[5] were recreated in SCAPS-1D using material physical parameters from previous literature. Fig. 2 compares the simulated current density-voltage (J-V) curve of the single-layer $CH_3NH_3PbI_3$ perovskite solar cell and the experimental results obtained in the published paper [5].

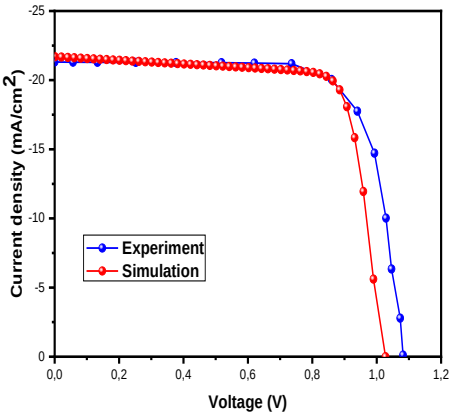

Figure 2. J-V curve of simulated and experimental results

Table 2 shows that the SCAPS-1D findings closely align with the experimental results, with any small discrepancies ascribed to materials, interface defects, and impurities introduced by the deposition processes utilized in the experiments.

TABLE II. THEORETICAL CHARACTERISTICS OF THE SOLAR CELL TESTED VERSUS EXPERIMENTAL RESULTS

Study type	Voc(V)	Jsc (mA/cm^2)	FF (%)	PCE (%)
Experiment	1.084	21.03	75.00	17.17
Simulation	1.012	21.69	78.43	17.23

IV. RESULTS AND DISCUSSION

A. *The influence of the absorber layer thickness and carrier concentration*

To assess how the performance of the device is affected, the thickness and carrier concentration (Na) of the $MAPbI_3$ absorber layer were examined, these parameters were systematically varied within the ranges of 0.2 to 1.2 µm and 10^{14} to 10^{20} cm^{-3}, respectively.

As the thickness of the absorber film increases, it absorbs more light, leading to higher V_{oc} and J_{sc} values. This is primarily due to the increased number of absorbed photons, which results in greater electron-hole pair generation and enhanced quantum efficiency.

In this simulation study, a thickness of 800 nm was determined to be the optimal choice for the perovskite absorber layer. This choice was reached using a variety of parameters, including the overall photovoltaic performance of the studied solar cells, material costs, and consumption. The optimized thickness of 800 nm for the $MAPbI_3$ absorber layer will be utilized for subsequent calculations.

In this part of the work, we examined the influence of the carrier concentration in the absorber layer on the photovoltaic outputs, and the findings are illustrated in Figure 3b. The carrier concentration was systematically varied in the range of 10^{11} to 10^{20} cm^{-3} while maintaining a constant optimized thickness of 800 nm for the absorber layer.

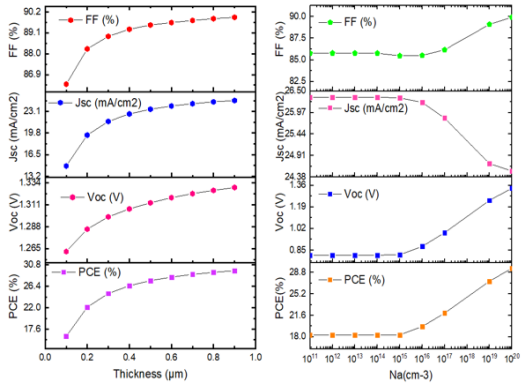

Figure 3. Effect of thickness and Na of $MAPbI_3$ absorber layer

The photovoltaic output parameters, such as V_{oc}, J_{sc}, FF, and PCE, remained relatively stable up to 10^{19} cm^{-3}. Beyond this concentration, V_{oc}, FF, and PCE increased, while J_{sc} slightly decreased with increasing doping concentration. This decrease in J_{SC} can be attributed to the less photo-generated carrier recombination at higher doping concentrations. By meticulously increasing the concentration in the absorber layer, it is possible to introduce enough charge carriers to enhance the photovoltaic outputs of the perovskite solar cells (PSCs). These findings underline the importance of choosing the right absorber doping density for optimal device performance. This trend is consistent with previous studies [6][7]. An absorber doping concentration of 10^{19} cm^{-3} is optimal for achieving the best results in the studied solar cell performance. A PCE of 29.26% was achieved at this doping density, with V_{oc} of 1.32 V, J_{sc} of 24.55 mA cm^{-2}, and FF of 89.85%.

B. *Effect of bilayer ETL*

To obtain better performance of the perovskite solar cell a bilayer ETL materials are suggested (TiO_2/SnO_2, TiO_2/ZnO, and TiO_2/WO_3).

The obtained J-V characteristics are shown in Fig. 4 and the extracted output parameters are summarized in Table 3. According to the obtained results, the bilayer ETL consisting of TiO_2/ZnO is an efficient configuration that creates a compact interfacial layer, effectively preventing direct contact between the FTO substrate and the perovskite layer. This setup helps to minimize carrier accumulation at the interface between the ETL and active layer, resulting in reduced charge recombination and improved device performance. [8].

TABLE III. PHOTOVOLTAIC PARAMETERS OF TESTED PSCS WITH VARIOUS BILAYER ETL

Used Bilayer ETL	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
TiO_2/ZnO	1.32	24.55	89.85	29.26
TiO_2/SnO_2	1.26	24.46	87.67	27.06
$TiO_2/WO3$	0.97	24.66	52.24	12.46

However, studying alternative ETL materials yields the best QE of above 90 %. As a result, TiO_2/ZnO is the best

bilayer ETL material to achieve the highest device efficiency of 29.26%.

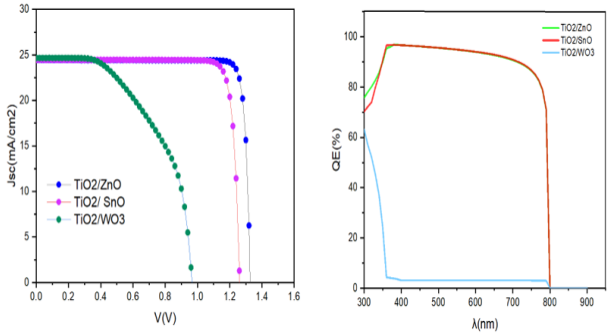

Figure 4. Effect of the different bilayer ETL materials on J-V characteristics of studied PSCs

C. *Effect of HTL on the PV parameters of tested PSCs*

The previous section, TiO_2/ZnO was identified as the optimal bilayer material for the electron transport layer (ETL). Similarly, in this section, several materials are being investigated for use as hole transport layers (HTLs), such as P3HT, Cu_2O, and spiro-OMETAD.

The best performance is obtained with spiro-OMETAD because the hole mobility in P3HT and Cu_2O is considerably lower than that of spiro-OMETAD. The J-V curves show significant improvements in the performance of the perovskite solar cell, as shown in Figure 5.

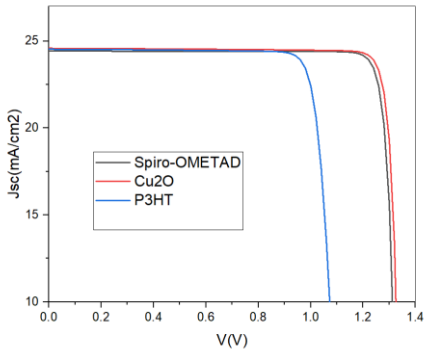

Figure 5. Impact of various HTL materials on J-V characteristics of studied PSCs

D. *Comparative study*

In this section, we provide a comprehensive comparison between this current study and several previous research efforts in this area, intending to highlight the unique contributions, advances, and novel findings of the present work within the broader context of the existing literature.[6][7] [9].

TABLE IV. COMPARISON OF OUR WORK WITH PREVIOUS STUDIES

STRUCTURE	STUDY TYPE	J_{SC} (MA/CM^2)	V_{OC} (V)	FF (%)	PCE (%)	REF
AU/SPIRO/MAPBI$_3$/ZNO/TIO$_2$/FTO	EXPERIMENTAL	21.03	1.084	75.00	17.17	[5]
AU/SPIRO/MAPBI$_3$/ZNO/TIO$_2$/FTO	SIMULATION	24.55	1.32	89.85	29.26	THIS WORK
AU/SPIRO/MAPBI$_3$/TIO$_2$/FTO	SIMULATION	24.42	1.16	87.73	24.77	[9]
AG/SPIRO/MAPBI$_3$/WO$_3$/TIO$_2$/FTO	EXPERIMENTAL	23.42	1.06	72.41	17.96	[6]
AG/SPIRO/MAPB$_{I3}$/CDS /TIO$_2$/FTO	EXPERIMENTAL	23.65	0.99	61.00	14.64	[7]

V. CONCLUSION

This study focuses on improving a perovskite solar cell of the configuration FTO/TiO_2/ZnO/MAPbI$_3$/Spiro-OMETAD/Au using the SCAPS 1D. The insertion of a bilayer electron transport layer of TiO_2 and ZnO in studied solar cells led to a remarkable power conversion efficiency of 29.26%, indicating its significant potential for enhancing the device's photovoltaic parameters. This research is a crucial step in developing more cost-effective and efficient solar cell technology, which might allow considerable advancements in the field.

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