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Hole Transport Layer Optimization for Mixed Halide Perovskite based Solar Cells to achieve Better Photovoltaic Performance

Shweta Dhakla[a*], Parvesh K Deendyal[a,b], Harpreet Singh[c], Monu Mishra[d], Sarvesh Kumar[e] & Manish K Kashyap[a]

[a]Renewable Energy Laboratory, School of Physical Sciences, Jawaharlal Nehru University, New Delhi 110 067, India

[b]Government Polytechnic for Women, Faridabad, Haryana 121 006, India

[c]Department of Physics, IIHS, Kurukshetra University, Kurukshetra, Haryana 136 119, India

[d]Department of Physics, Dyal Singh College, University of Delhi, New Delhi 110 003, India

[e]Inter-University Accelerator Centre (IUAC), Aruna Asaf Ali Marg, New Delhi 110 067, India

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Methylammonium lead iodide ($MAPbI_3$) has been emerged out as an efficient perovskite absorber material in solar cell applications and extensively studied on both experimental and theoretical fronts. However, these $MAPbI_3$-based perovskite solar cells (PSCs) undergo degradation due to thermal instability of $MAPbI_3$, which in-turn deteriorates the performance of PSC for a long-run. In this work, we deal with the computational analysis of mixed-halide $MAPb(I_{1-x}Cl_x)_3$ based PSCs by changing hole transport layers (HTLs) so that higher efficiency can be aimed. It has been observed that not only the appropriate band alignment of HTL with perovskite, but the mobility of HTL also play a pivotal role in achieving the better photovoltaic (PV) performance. Furthermore, it is noteworthy that Cu_2O exhibits a better PV performance in contrast to other HTLs considered in our study. Thus, the present simulation work paves a path for the experimentalists to design similar PSCs by cutting-down the cost of experimental trials.

Keywords: Methylammonium lead iodide; Perovskite absorber; Solar cells; Hole transport layers

1 Introduction

Hybrid halide perovskite (HHP) based solar cells have revolutionized the photovoltaic scenario with a miraculous advancement in power conversion efficiency (PCE) from just 3.8 %[1] in 2009 to 26.1 %[2] at present. Interestingly, the PCE shown by these perovskite solar cells (PSCs) has almost reached to the commercially available Silicon solar cells' efficiency mark. The outstanding photovoltaic (PV) performance of PSCs is ascribed to the inherent attributes of HHP materials such as high absorption coefficient, long carrier diffusion length, tunable band gap, high charge carrier mobility[3]. The exceptional performance of PSCs along with low-cost and facile solution-based fabrication process hold much potential for the futuristic solar cells.

Among all HHP materials, methylammonium lead iodide ($MAPbI_3$) has attracted unprecedented research scrutiny owing to its excellent PV performance in the PSCs. However, $MAPbI_3$ based PSCs have failed to cross the door of commercial market due to the compositional degradation and low crystallization energy which in turn result in the instability of $MAPbI_3$ against heat and light[3]. To overcome these issues, $MAPb(I_{3-x}Cl_x)$ has been employed as an absorber material in PSCs by partial replacement of I with Cl, resultingly, enhances the thermal stability and diffusion length and, also improves the film quality[4].

Moreover, the most commonly used hole transport layer (HTL), Spiro-OMeTAD, in the efficient PSCs has complicated synthesis process. It also requires ultra-purity which raises overall device's cost. Further, to work as an effective HTL, it requires additives and dopants such as t-BP, LiTFSI that impede the device stability[5]. In this regard, scientists have made continuous major efforts to search for dopant-free and inorganic HTL materials in order to attain low-cost and high stability of the resulting device.

After an extensive review of literature, we plan to optimize the HTL for the mixed HHP based solar cell along with ZnO as an electron transport layer (ETL). Here, we adopt the dry lab approach via numerical simulations; since it reduces the experimental trails to optimize HTL before utilizing them in final solar cell fabrication.

2 Simulation Methodology

The numerical simulation was accomplished via Solar Cell Capacitance Simulator simulation (SCAPS-

*Corresponding author: (E-mail: shweta.psc1@gmail.com)

Table 1 — Input parameters of FTO, absorber layer, ETL and various HTLs.

Simulation Parameter	FTO[6,7]	ETL[8]	Absorber layer[4]	HTL[9-10]				
		ZnO	$MAPb(I_{1-x}Cl_x)_3$	Spiro-OMeTAD	CuI	Cu_2O	CuSCN	MEH-PPV
Thickness (nm) (t)	300	120	440	200	200	200	200	200
Band gap (eV)(E_g)	3.5	3.3	1.55	3	3.1	2.17	3.6	2.1
Electron affinity (eV) (χ)	4	4	3.9	2.2	2.1	3.2	1.7	2.8
Dielectric Permittivity (ϵ)	9	9	6.5	3	6.5	7.1	10	3
Conduction Band Density of State(cm^{-3}) (N_c)	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}	2.8×10^{19}	2.02×10^{17}	2.2×10^{19}	2.5×10^{19}
Valence Band Density of States (cm^{-3}) (N_V)	1.8×10^{19}	1.8×10^{19}	1.8×10^{19}	1.8×10^{19}	1×10^{19}	1.1×10^{19}	1.8×10^{18}	2.5×10^{19}
Electron thermal velocity (cm/s) (V_e)	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}
Hole thermal velocity (cm/s)(V_h)	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}
Electron mobility (cm^2/Vs) (μ_e)	20	100	2	2.1×10^{-3}	100	200	100	0.5×10^{-4}
Hole mobility(cm^2/Vs) (μ_h)	10	25	2	2.16×10^{-3}	43.9	80	25	2.5×10^{-4}
Donor defect density (cm^{-3}) (N_D)	1×10^{19}	2×10^{18}	-	0	0	0	0	0
Acceptor defect density (cm^{-3}) (N_A)	0	0	-	1×10^{18}	1×10^{18}	9×10^{21}	1×10^{18}	1×10^{18}
Defect density in bulk (cm^{-3}) (N_t)	1×10^{15}	1×10^{15}	1×10^{14}	1×10^{15}	1×10^{15}	1×10^{15}	1×10^{15}	1×10^{15}

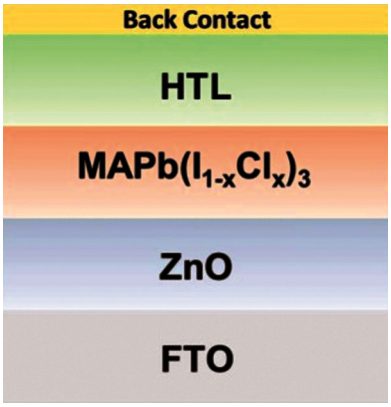

Fig. 1 — Basic device architecture of simulated PSC.

1D) software. It works on three basic equations *i.e.*, Poisson's equation, continuity equation for electrons and continuity equation for holes. The simulated device architecture is shown in Fig. 1. All the input parameters for various layers of simulated structure were extracted directly from the previous studies[4,6-11] and are listed in Table 1.

The simulated device was exposed under one sun AM 1.5G (100mW cm^{-2}) solar spectrum at 300 K. The work function for back contact electrodes (Au) was 5.1 eV. All the simulation work was carried out without taken into consideration the optical reflectance of each layer at surfaces and interfaces.

3 Results and Discussions

With an aim to replace the costly organic HTL, Spiro-OMeTAD, the various materials were investigated. The proposed materials for this replacement were CuI, Cu_2O, CuSCN, MEH-PPV.

Table 2 — Output performance parameters of mixed-halide perovskite with different proposed HTLs.

HTL	V_{oc} (V)	J_{sc} ($mAcm^{-2}$)	FF (%)	PCE (%)
Spiro-OMeTAD	1.20	22.95	83.44	22.94
CuI	1.20	22.95	83.42	22.94
Cu_2O	1.20	23.03	84.51	23.35
CuSCN	1.20	22.95	84.35	23.16
MEH-PPV	1.20	22.95	69.17	19.04

Here, ZnO is chosen as common ETL for all HTLs in PSCs, owing to its suitable band alignment with perovskite, high electron mobility, comparable band gap to most efficient ETL (TiO_2) and low deposition temperature. The input parameters of the proposed HTL materials were taken from reported literature studies[9-11] as summarized in Table 1. Concurrently, a thickness of 200 nm was chosen for each HTL material. The output photovoltaic parameters of PSC with different HTLs are exhibited in Table 2. It is worth to mention that Cu_2O displays better performance among all the proposed HTL, yielding an impressive PCE of 23.35 % with a short circuit current density (J_{sc}) of 23.03 mA. cm^{-2}, open circuit voltage (V_{oc}) of 1.20 V and fill factor (FF) of 84.51 % (Table 2). The outstanding performance can be attributed to a high value of hole mobility (Table 1) in contrast to other HTL materials. Interestingly, CuSCN performed almost comparable to Cu_2O. Spiro-OMeTAD and CuI, on the other hand, also showcased comparable and better performance in terms of PV parameters. Furthermore, there is an appropriate band alignment between highest occupied molecular orbital (HOMO) of perovskite and the valence band of

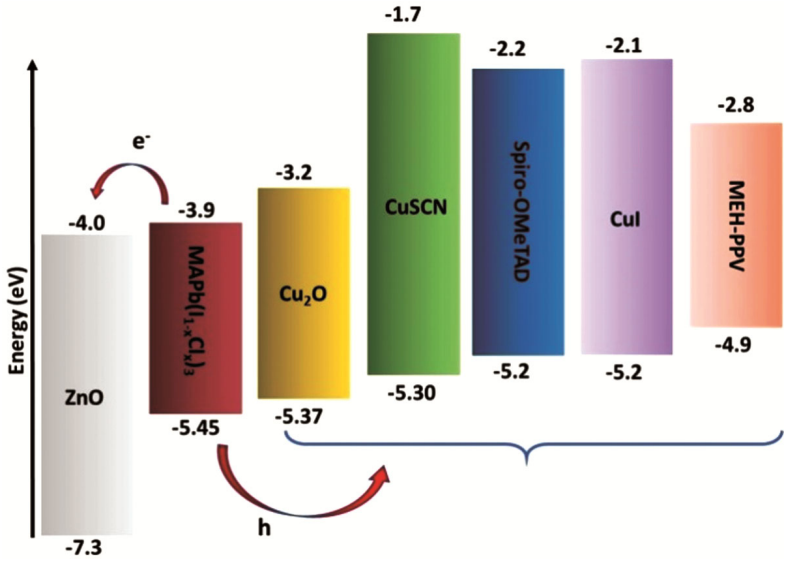

Fig. 2 — Band alignments of ZnO, perovskite absorber and various proposed HTL materials.

proposed HTLs, as shown in Fig. 2. Despite of the good band alignment in case of MEH-PPV with perovskite, it exhibited poor PV performance. One of the reasons for this may be the poor carrier mobility for this HTL material, which in turn affects the charge collection at respective electrodes and hence PCE. In the nutshell, the better performance of Cu_2O establishes it as an appropriate HTL for the chosen perovskite in present study. It should also be favored since it is an inorganic material with merits such as low cost, simple preparation, high hole mobility and good chemical stability.

4 Conclusion

The present study deals with the optimization of HTL for mixed-halide perovskite based solar cells by fixing ZnO as ETL using numerical simulations through SCAPS-1D software. It is concluded that Cu_2O, exhibits better photovoltaic performance among all the proposed HTL materials showing a PCE of 23.35 %. It has been found that not only the proper band alignment, but the mobility of HTL plays a crucial role in a better photovoltaic performance. These simulated results will be helpful for researchers to design similar PSCs by reducing the experimental trials for HTL optimization.

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