Novel approach on Formamidinium tin iodide-based Perovskite solar cell for the best replacement materials of Hole Transport Layer and Electron Transport Layer by using Solar cell Capacitance simulation

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Research Article

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Abstract

Perovskite Solar Cells (PSC) research has increased noticeably in recent years as a result of significant improvements in their performance. Perovskite solar cells are a new development in Photovoltaic technology. Because PSCs are inexpensive, have the ability to tune the bandgap and are a promising future option for meeting cell efficiency limits and strong broad optical absorption. Toxicity is an important factor in the development of organic and inorganic Perovskite solar cells. PSCs based on Sn are a worthy competitor to PSCs based on Lead (Pb). The majority of them used Methylammonium tin iodide ($\text{CH}_3\text{NH}_3\text{SnI}_3$) material as an absorber layer because it has greater absorption but suffers from temperature instability. As a result, in this work, we use Formamidinium tin iodide (FASnI_3) absorber, which has higher temperature stability than $\text{CH}_3\text{NH}_3\text{SnI}_3$ Methylammonium tin iodide with a band gap of 1.4 eV. Solar cell Capacitance simulation used in this work to create FASnI_3 -based solar cells. This paper proposes a Power Conversion Efficiency of 24.22% for PSCs. For maximum Power Conversion Efficiency (PCE), the optimized thickness values are 10 nm for CuI (Hole transport layer), 850 nm for FASnI_3 , and 20 nm for ZnOS (Electron Transport Layer). The best temperature range for device performance is around 340K. SCAPS was used to calculate the current work. In addition, we demonstrated that ZnOS is the most promising ETL as a TiO_2 replacement in this work. The proposed work focuses on paving the way for novel eco-friendly non-toxic PSCs as well as investigating optimised Voltage output circuit (Voc), Current density (J_{sc}), Fill factor (FF), and Power conversion Efficiency (PCE) device characteristics.

1. Introduction

Nowadays Perovskite solar cell is the mainstream in PV technology with high PCE and PSCs is the best competitor for Si-wafer based solar cells [1]. Recently in perovskite solar cell, halides-based PSCs has the most promising absorber materials in PSCs [2]. The power conversion efficiency of PSCs which is based upon Pb-PSCs had a dramatic improvement from 3.08% to recently certify the value of 25.5% [3]. However, there are some shortcomings faced on Pb based PSCs. Like toxicity, thermal stability, air stability and non-ecofriendly to the humans. These are some of the disadvantages for Pb-based PSCs [4]. For that reasons pollution free and safety to humans, it is emergently needed to develop a nontoxic or low toxic metal to replace the Pb based ab-layer of PSCs. Therefore, research had to be made to replace Pb with some low and nontoxic materials in PSC_s [5]. Sn, Cu, Bi, and Sb have all been studied as potential replacements for Pb-PSCs [6]. Sn has emerged as the most promising immediate replacement material for Pb-based PSCs among these. Because Sn belongs to the same group as Pb in the periodic table, its electric and optical properties are too similar, making Sn the best replacement material in PSCs [7].

The general formula for organic and inorganic halide perovskite solar cells is ABX_3 , where A and B are cations and X is always a halogen. MA, FA, and Cs are the most commonly used cations (A) in PSC [8]. From 4th and 7th, group elements are widely used as a B cation and X halogen respectively [9]. In this work, we use Sn be the replacement of Pb. Sn is not only for the alternative ion to Pb, it is also to enhance ecofriendly of PSCs [10]. Due to temperature instability in CsSnI_3 and MASnI_3 we use the FASnI_3 as the

Ab-layer in this simulated work [11]. FASnI_3 had a more solid perovskite structure, more stable and also the air stability is high compared with MASnI_3 . These are the reasons we use FASnI_3 having a perovskite layer for this simulated Device. Recently many approaches have been done to improve the FASnI_3 based device [12]. FASnI_3 has higher carrier mobility than MASnI_3 according to Milot et al [13]. Krishna moorthy groups published the first study on FASnI_3 -based PSCs in 2015, with an efficiency of 1.41 percent [14]. FASnI_3 with E_g (1.4eV) which is wider than MASnI_3 and CsSnI_3 (1.30eV) respectively and narrow to Pb based PSCs. Also, in FASnX_3 can be tuned the E_g with different halides [15]. Recently CuI had attained PCE 17.6% which is based upon spray deposition method [16]. CuI had a high electric conductivity compared with Spiro-MeOTD. For that reason, we use CuI as a HTM in this work. CuI [17]. Bansal had investigated MASnI_3 based solar cell with different ETL and HTL they achieved the PCE 21.1% in that ZnOS act as ETL [18]. Currently, before fabricating a cell or a device, the theoretical studies help more to understand and find the behavior of the cell Devices [19]. In recent days, many simulation devices are studied for PSCs with novel materials in SCAPS (1D) [20]. The initial structure is based on an 11.01% working PCE. After several optimizations on the thickness of the ab-layer, HTL and ETL, the simulated devices achieve an efficiency of 24.22%. At this present work, we optimized Novel FASnI_3 based Perovskite solar cell with CuI and ZnOS act as a very promising HTL and ETL respectively. Replacement of Spiro-MeOTD and TiO_2 and also FASnI_3 material is the best replacement for MASnI_3 based solar cell.

2. Methodology And Device Simulation

2.1. Basic Structure of the Device

The device's simulated configuration is FTO / ZnOS / FASnI_3 / CuI / Au. To date, three types of PSCs have been investigated: mesoporous planar, and inverted planar. The inverted planner configuration has been completed in this work and is shown in Fig. 1. (a) Device configurations of simulated PSCs, CuI as HTL and ZnOS as ETL. Energy band diagram for this device also shown in Fig (b). In Fig. 1(c, d) shown the initial J_{sc} , V_{oc} , and %PCE- wavelength studies. All the basic parameters are collected from various research papers and tabulated in Table.1 [3, 10, 15, 21, and 22]. Meng and his group report that at low temperature the inverted planar would exceed over 18% of efficiency flexibility with J-V hysteresis [23]. Table.2. Summaries the N_t , N_D , N_A , for the configuration of the device and the work function of front and back contact as FTO (4eV) and Au (5.1 eV) respectively [24, 25].

2.2. Methodology of Simulation

The simulated was performed in SCAPS 1D under AM 1.5G illumination. This simulation carries only inside the SCAPS-1D and mainstream to derive from the following fundamental equations Poisson, hole and electron continuity. All the simulated results were examined in separate sections through graphs for each stage. In initial structure results, values of is PCE 11.01%, after several optimization on the absorber layer, HTL, ETL, and temperature in this device we constructed a novel PSCs with high efficiency.

3. Results And Discussions

3.1. Optimization and Simulation of FASnI₃ PSC

3.1.1. Effect of Temperature

The temperature of the solar cell has an effect on its overall performance. In this simulation, we kept the temperature constant at 300K at first and then varied it from 300K to 400K to determine the effect of working temperature on PCE, Voc, Jsc, and FF for the best ab-layer thickness, as shown in fig. In this simulated device, it was discovered that as the temperature increased, the PCE, Voc, Jsc, and FF of the solar cell decreased, because of carrier concentrations, charge carrier mobility and the material's bandgap at high temperatures (mariSoucase, et.al.2016). The efficiency slightly decreased from 24.09–23.64% at (300-400) K. The diffusion length reduces and series resistance increases. The resultant is decreased in PCE and FF (Mandadapu and group at 2017), because of the increase in energy enhanced recombination, electrons become unstable at higher temperatures in a solar cell, resulting in a low PCE (Mandadapu 2017). For that 340k is the working temperature for this simulation work.

3.1.2. The Effect of the Ab-layer Thickness

The ab-layer in PSCs absorbs photons from sunlight whose energy is greater than the bandgap of the ab-layer, which causes excursions. These preciseions are essentially charged electron-hole pairs. To maximize PCE, it is necessary to understand the impact of defect density on PSC performance. Because the Ab-layer is a critical factor in determining the performance of PSCs, its effect on solar cell output parameters has been studied using simulation. The active layer thickness was increased from 100 nm to 1050 nm in 850 nm increments and the effect on performance parameters is shown in Fig. (3). The device's performance is excellent at 850 nm, and it achieves the maximum efficiency. At 850 nm, the device's performance is excellent, with the maximum efficiency of 24.22% achieved by using Voc 6.20 V, Jsc 30.77 mA/cm², and FF 12.68%.

3.1.3. Thickness optimized on Electron Transport Layer

Figure 4 illustrates the effect of ETL thickness and performance on ab-layer at 850 nm. When the ETL thickness is 20 nm, we get the best performance on the simulated device with the highest PCE of 24%, which is increased by 0.02% efficiency with FF of 12.68%, Voc of 6.20V, and Jsc of 30.77 mA/cm².

3.1.4. Thickness optimized in Hole Transport layer

From Fig. 5. Shows the impact on the HTL and the device performance at 850 nm Ab-layer. When the thickness of the Ab-layer is at 850nm, we reach the maximum PCE 24.22% at 10 nm thickness of HTL with FF of 12.68%, Voc of 6.20V, Jsc of 30.77 mA/cm².

3.1.5. Effect of Nt of the optimized device

In this simulation, the N_t of ab layered varied between 10^{12} cm^{-3} and 10^{18} cm^{-3} to find the best absorber thickness to find the variation in PCE, as shown in the Fig. 6,7,8 for the simulated device. PCE at other parameters of the simulated solar cell decreased as the absorber layer's N_t increased. As the defect density decreased, the efficiency stabilized at a certain point in this work, with the device performing well at 10^{16} cm^{-3} (N_t) of absorber layer. The device performs best at 10^{18} cm^{-3} and 10^{20} cm^{-3} for ETL and HTL, respectively.

3.1.6. Effect of Bandgap in the simulation device

Sn-based PSC has a tunable band gap ranging from 1.3eV to 2.15eV. (vedanayakam, mandadapu.,2017). In this simulation, the bandgap of the optimized solar cell ranged from 1.2eV to 2.0eV for the best ab-layer to determine the variation in efficiency and other parameters. As shown in the figure 9, as the bandgap increased, PCE, FF, and Jsc decreased slightly while Voc increased. Jsc decreased as the bandgap increased, owing to less electron generation. After several simulations, the device was tested at 1.4eV. Similarly, we optimised the bandgap of ETL and HTL until we found the best performance of Cul at 2.8 eV and ZnOS at 2.4eV after several attempts. Figure 10 depicts the external quantum efficiency curve for the device's best absorber layer.

4. Conclusion

1D SCAPS was used in this study to investigate the optimized and simulated behaviors of the Sn-based PSC with ETL as ZnOS and HTL as Cul configuration. The working temperature 340 k for this simulation study at standard illumination of Air mass at 1.5G, the perovskite layer from 150 nm to 1050 nm and the optimum layer range for HTL and ETL as 10-50 nm respectively both layers. The best layer thickness ranges from ZnOS are 20 nm and Cul is 10 nm respectively. The best defect density is performed at 10^{16} cm^{-3} for this simulated device. The band gap of ab-layer HTL, ETL were to change from after several simulation 1.4eV, 2.4 eV, 2.8 eV, respectively. The device performed well and good at when N_t of the ab-layer 10^{16} cm^{-3} with bandgap 1.4eV. When the optimized input parameter values are considered, the highest efficiency achieved is 24.22% (FF of 12.68%, Voc of 6.20V, Jsc of 30.77 mA/cm^2). When we use Cul as the hole transport layer and ZnOS as the electron transport layer in a FASnI_3 -based solar cell, we get the best results, and Cul and ZnOS are the best replacements for Spiro-OMeTAD and TiO_2 . The simulated result stands for only replacement materials for FAPbI_3 , Spiro-OMeTAD and TiO_2 because Pb based material had high toxicity for that reason we use Sn based ab-layer in this work. Spiro-OMeTAD and TiO_2 are too costly for that reason in this work we used lost cost Cul, ZnOS as HTL and ETL respectively.

Abbreviations

PSC- Perovskite solar cell

PV- Photovoltaic technology

SCAPS- Solar cell Capacitance simulation

FASnI₃ – Formamidinium tin iodide

MASnI₃- Methylammonium tin iodide

ZnOS- Zinc oxysulphide

FA- Formamidinium

Cs- Cesium

MA- Methylammonium

Sn- Tin

Pb-Lead

Cu- Copper

Copper iodide- CuI

ETL- Electron transport layer

HTL- Hole transport layer

TiO₂-Titanium Oxide

Spiro-OMeTAD - 2,2',7,7'-Tetrakis[N,N-di(4-methoxyphenyl)amino]-9

Voc- Voltage output circuit (Voc),

Jsc- Current density

FF- Fill factor

PCE- Power conversion Efficiency

N_t- Donor

N_A- Acceptor

References

1. P.Touni, S.O. Oseni, G.Sharma, Q.Yan, G.T.Mola, Perovskite photovoltaic solar cell: an overview of current status, *Renew.Sustain.Energy Rev.*91 (2018) 1025-1044, <http://doi.org/10.1016/j.rser.2018.04.069>.
2. L.Serrano-Lujan, N.Espinosa, T.T.Lasrsen-Olsen, J.Abad, A.Urbina, F.C.Krebs, Tin-and based perovskite solar cell under scrutiny: an environmental perspective, *Adv.Energy Matter*,5 (20) (2015), 1501119.
3. G.Xing, N. Mathews, S.Sun, S.S.Lim, Y.M.Lam et.al., Long-range balanced electron- and hole transport lengths in organic-inorganic $\text{CH}_3\text{NH}_3\text{PbI}_3$.*Science* 342 (6156), 344-347 (2013). <https://doi.org/10.1126/science.1243167>.
4. S.D.Stranks, G.E.Eperon, G.Eperon,G.Grancini, C.menelaou, M.J.P. Alcocer et al., Electron-Hole diffusion length exceeding 1 micrometer in an organometal trihalide perovskite absorber. *Science* 342 (6156), 341-344 (2013). <http://doi.org/10.1126/science.1243982>.
5. P.Docampo, J.M.Ball, M.Darwich, G.E. Eperon,H.J.Snaith, Efficient organometal trihalide perovskite planar heterojunction solar cells on flexiable polymer substrates. *Nat.Commun.* 4,2761-2766(2013).<https://doi.org/10.1038/ncomms3761>.
6. G.Schileo, G.Grancini, Lead or no lead Availability, toxicity, sustainability and environmental impact of lead free perovskite solar cell, *J.Mater.Chem.* C9(2021) 67-76, <https://doi.org/10.1039/D0TC04552G>.
7. M.Konstantakou, T.Stergiapolou, A critical review on tin halide perovskite solar cells, *J. Mater. Chem.*5 (2017) 11518-11549, <https://doi.org/10.1039/C7TA00929A>.
8. W.Zhou, Y.Zhou, X.Zhou. R. Fu, Q.Li, Y.Zhao, K. Liu, D.Yu.Q.Zhao, *J.phys.chem.Lett.*8(2017).4122-4128.
9. M.A.Green, E.D.Dunlop, J.Hohlebinger, M.Yoshita, N.Kopidakis, A.Hobaiillie, *Prog. Photovolt.*28 (2020) 3-15.
10. Minghao Wang, W.Wang, B.Ma, A.Shen, L.Liu, K.Cao, S.Chen, W.huang. Lead Free Perovskite Materials for Soalr Cells.*Nano-Micro Lett.* (2021).<https://doi.org/10.1007/s40820-020-00578-z>.
11. A.G.Kontos, A.kaltzoglou, E.Siranidi,D.Palles, G.K.Angeli, M.K.Arfanis,et.al,. Structural stability, vibrational propertyes and photoluminescence in CsSnI_3 perovskite upon the addition of SnF_2 , *Inorg. Chem.*56 (1) (2017) 84-91.
12. S.Abdelaziz, A.Zekry, A. Shaker, M.Abouelatta Investigating the performance of fomamidinium tin based perovskite solar cell by SCAPS device simulation.<https://doi.org/10.1016/j.optmat.2020.109738>.
13. R.L. Milot,G.E.Eperon, T.Green, H.J.Snaith radiative monomolecular recombination boosts amplified spontaneous emission in $\text{HC}(\text{NH}_2)_2\text{SnI}_3$ perovskite films, *J.Phys.chem.lett.*7 (20) (2016) 4178-4184,<https://doi.org/10.1021/acs.jpcllett.6b02030>.
14. T.M.Koh, T.Krishnamoorthy, N.Yantara, C.Shi. W.L.Leong. Formamidinium tin-based perovskite with low E_g for photovoltaic application, *J.mater.chem.*A3 (29) (2015) 14996-15000, <https://doi.org/10.1039/C5TA00190K>.

15. Y.Zhao, K.Zhu. Organic-inorganic hybrid lead perovskite for optoelectronic and electronic applications. Chem, Soc. Rev.45(3), 655-689 (2016). <https://doi.org/10.1039/c4cs00458b>.
16. Xin Li, Junyou Yang, Qinghui Jiang, Synergistic effect to high performance perovskite solar cell with reduced hysteresis and improved stability by the introduction of Na treated TiO₂ and spraying deposited CuI as transport layers, ACS Appl. Mater. Interfaces 9 (47) (2017) 41354-41362.
17. Mostafa M. salah. Kamel M.Hassan, Mohamed Abouelatta, Ahmed shaker, A comparative study of different ETM's in perovskite solar cell with inorganic copper iodide as HTM, <https://doi.org/10.1016/j.ijleo.2018.10.052>.
18. S.Bansal, P.Aryal, evaluation of new materials for electron and hole transport layer in perovskite based solar cell through SCAPS-1D simulations, in; IEEE 43rd photovoltaic Specialists conference (PVSC), 2016, <https://doi.org/10.1109/PVSC.2016.7749702>, 0747-0750.
19. Mandadapu U, Victor Vedanayakam S, Thyagarajan K (2017) Numerical simulation of CH₃NH₃PbI₃-xCl_x perovskite solar cell using SCAPS-1D. Int J Eng Sci Invention 2:40–45
20. Mare Burgelman, Koen Decock, Samira Khelifi., Aimi Abass, Advanced electrical stimulation of thin film solar cell, Thin solid film, 535 (2013) 296-301.
21. Hechavarría-Difura, F.J. Sánchez-Rodríguezb, Maykel Courelc, Hailin Hu Optimization of CH₃NH₃PbI₃ perovskite solar cells: A theoretical and experimental study. Solar Energy 199 (2020) 198–205.
22. M. I. Hossain, F. H. Alharbi, and N. Tabet, "Copper oxide as inorganic hole transport material for lead halide perovskite-based solar cells," Sol. Energy 120, 370–380 (2015).
23. K.Deepthi Jayan, Varkey Sebastian. Comprehensive device modeling and performance analysis of MASnI₃ based perovskite solar cell with diverse ETL, HTL, and back metal contacts, Solar cell, 2021.
24. Y. Ogomi, A. Morita, S. Tsukamoto, T. Saitho, N. Fujikawa, Q. Shen, et al., CH₃NH₃SnxPb(1-x)I₃ perovskite solar cells covering up to 1060 nm, J. Phys. Chem. Lett. 5 (6) (2014) 1004–1011.
25. Zhao P, Liu Z, Lin Z, Chen D, Jie S, Zhang C, Zhang J, Chang J, Hao Y (2018) Device simulation of inverted CH₃NH₃PbI₃-xCl_x perovskite solar cells based on PCBM electron transport layer and NiO hole transport layer. Sol Energy 169:11–18

Tables

Table.1 [3,10,15,20,21,22]

Material property	ZnOS	FASnI ₃	CuI
Thickness 't'(nm)	Varied	Varied	Varied
Bandgap 'Eg' (eV)	2.8	1.4	2.6
Electron affinity 'χ' (eV)	3.9	3.52	3.9
Relative Dielectric permittivity 'ε _r '	3	8.2	4
CB effective density of state 'N _c ' (cm ⁻³)	1E+20	1.0E+18	1E+21
VB effective density of state 'N _v ' (cm ⁻³)	1E+20	1.0E+18	2E+20
Electron mobility 'μ _n ' (cm ² /V.s)	1E-4	22	0.01
Hole mobility 'μ _p ' (cm ² /V.s)	1E-4	22	0.01
Donor concentration 'N _D '(cm ⁻³)	1E+7	0	1E+20
Acceptor concentration 'N _A '(cm ⁻³)	1E+7	7.0E+16	–
Defect density 'N _t ' (cm ⁻³)	-	2.0E+15	–

Table.2: Parameters for back and front contact. [23,24,25]

Parameters	Back contact	Front contact
Surface recombination velocity of electrons (cm/s)	1.00E+7	1.000E+5
Surface recombination velocity of holes (cm/s)	1.000E+5	1.000E+7
Metal work function (eV)	5.144	4.43
Majority carrier barrier height relative to E _f (eV)	-0.0444	-0.0600
Majority carrier barrier height relative to E _v (eV)	-0.0000	-0.0028

Figures

Figure 1

(a). Cell configuration of FASnI₃ designed solar cell

Figure 2

Temperature VS Voc, Jsc, FF, PCE

Figure 3

Thickness of active layer Vs V_{oc} , J_{sc} , FF, P

Figure 4

Thickness of CuI Vs Voc, Jsc, FF, PCE

Figure 5

Thickness of ZnOS Vs Voc, Jsc, FF, PCE

Figure 6

HTL Vs Doping density

Figure 7

ETL Vs Doping density

Figure 8

Defect density Vs PCE

Figure 9

Bandgap for best perovskite layer, HTL and ETL

Figure 10

QE vs Wavelength