Journal of Nano- and Electronic Physics
Vol. **13** No 3, 03019(4pp) (2021)

Журнал нано- та електронної фізики
Том **13** № 3, 03019(4cc) (2021)

Simulation study of Formamidinium Lead Halide ($FAPbX_3$; X = I and Br) Based Perovskite Solar Cells Using SCAPS-1D Device Simulator

R. Saha, K. Chakraborty, M.G. Choudhury, S. Paul

Advanced Materials Research and Energy Application Laboratory (AMREAL), Department of Energy Engineering, North-Eastern Hill University, Shillong-793022, India

(Received 10 January 2021; revised manuscript received 15 June 2021; published online 25 June 2021)

Perovskite nanomaterials have emerged as promising materials for its applications not only in solar energy field but also in optoelectronic devices. Now-a-days, simulation-based study of perovskite solar cells is gaining interest among the photovoltaic researchers to understand in depth the influences of material characteristics on the device performances of a solar cell. In this work, we have studied the simulation analysis of formamidinium lead halide ($FAPbX_3$; X = I and Br) based perovskite solar cells using SCAPS-1D device simulator namely $FAPbI_3$, and $FAPbBr_3$ based Perovskite solar cells. In this study, spiro-OMeTAD and TiO_2 was used as Electron Transport Layer (ETL) and Hole Transport Layer (HTL) in the solar cell configuration. We have evaluated the impact of various thicknesses of perovskite layers and working temperatures on the performance of the perovskite solar cells. The study provides the current-voltage (*I-V*) characteristic curves with respect to various thickness and temperature for each of the two Perovskite active materials, respectively.

Keywords: Perovskite, Solar cell, Formamidinium, SCAPS, Power conversion efficiency.

DOI: 10.21272/jnep.13(3).03019 PACS number: 88.40.jp

1. INTRODUCTION

In recent years, perovskite based solar cells (PSCs) have led to considerable development in the field of photovoltaic devices due to its outstanding characteristics such as high power conversion efficiency (PCE), ideal band gap, low-cost, easy fabrication, etc. Researchers and scientists are synthesizing new class of perovskite materials to find its suitable application in solar cells. Recently, formamidinium lead halide ($FAPbX_3$) based perovskite are considered to be a prominent material after methyl ammonium lead halide ($MAPbX_3$) based perovskite in the area of photovoltaic research. Formamidinium lead halide ($FAPbX_3$) has attracted greater attention in comparison to methylammonium lead halide $MAPbX_3$ due to its broad absorption and higher thermal stability [1]. A new method for the preparation of $MAPbX_3$ and $FAPbX_3$ (X = I and Br) perovskite based single crystals was introduced by applying microwave radiation in a in a microwave reactor. The synthesized perovskite crystals showed the same properties as compared to crystal prepared by the traditional ITC methods [2]. Kimball et al. reported the replacement of methylammonium cations by formamidinium cations using a solid-liquid-solid cation exchange reaction and found that the synthesized $FAPbX_3$ showed high photoluminescence quantum yields (PL QYs) of up to 69 % [3]. Yang et al. demonstrated the fabrication of formamidinium with multiple cations and mixed halide anions based high-performance PSCs by introducing additional iodide ions into organic cation solution to decrease the concentration of deep-level defects and they obtained PCE of over 22 % [4].

Formamidinium lead halide based perovskite possesses some unique properties that have gained interest as a potential material for in solar cells applications. Sun et al. studied the mechanical properties of formamidinium lead halide perovskites ($FAPbX_3$, X = Br or I) grown by inverse-temperature crystallization and observed that $FAPbX_3$ perovskites exhibited weaker hydrogen bond and lead to smaller Young's modulus as compared to $MAPbX_3$ perovskites [5]. Formamidinium lead iodide shows a narrower band gap (1.48 eV) than the methylammonium lead iodide (1.57 eV), making it suitable for planar heterojunction solar cells [6]. The optical and electrical properties of $FAPbX_3$ (where X = Br and I) single crystals was studied which showed that $FAPbI_3$ and $FAPbBr_3$ crystals possessed long carrier diffusion lengths of 6.6 μm and 19.0 μm, respectively much longer than $MAPbX_3$ crystal [7]. Simulation based study is a useful tool to evaluate the influences of material characteristics on the device performances of a solar cell. These studies could provide researchers a direction for choosing materials with suitable energy band properties, but in due time, the electron affinity and band gap of the studied materials are difficult to be used [8]. SCAPS, wxAMPS, GPVDM, PC1D, COMSOL MULTIPHYSICS, and SILVACO are the most used simulation software for understanding the device mechanism and performance of solar cells. It is a one-dimensional solar cell simulation program developed at the Department of Electronics and Information Systems of the University of Gent, Belgium. SCAPS is a Windows-oriented program, developed with Lab Windows/CVI of National Instruments [9]. Karthick et al. studied the experimental photovoltaic and numerical performance of fabricated FA-cation based solar cell using SCAPS-1D simulation, incorporating small amounts of cesium and bromide in it and demonstrated that the device exhibits a high efficiency of 15.1 % under 1 sun illumination [10].

In this study, we have used a simulation software called Solar Cell Capacitance Simulator (SCAPS) 1D for the simulation of formamidinium lead halide ($FAPbX_3$; X = Br and I) based PSCs with spiro-OMeTAD as HTL

The results were presented at the International Conference on Innovative Research in Renewable Energy Technologies (IRRET-2021)

and TiO_2 as ETL. We have employed SCAPS-1D to model the PSCs for modeling and to optimize the efficiency of the two perovskite-based solar cell configurations. The influence of device parameters such as thickness of the perovskite layer, device temperature, and quantum efficiency are highlighted in this paper.

2. MODELING OF PEROVSKITE SOLAR CELLS

A schematic flat band energy-level diagram of formamidinium lead halide ($FAPbX_3$) and schematic diagram of the proposed PSC model is shown in Fig. 1. In this figure, the Conduction Band (CB) and Valence Band (VB) edges of the layers and work function of the metal layer of the proposed PSCs are shown as reported from earlier reports [11-14]. From the figure, the two perovskite layers viz. $FAPbI_3$ (with HOMO level of – 5.4 eV and a LUMO level of – 4.0 eV), and $FAPbBr_3$ (with HOMO level of – 5.6 eV and a LUMO level of – 3.4 eV) acts as an absorbing layer, respectively. FTO on glass substrate serves as a bottom electrode, with TiO_2 (with HOMO level of – 7.2 eV and a LUMO level of – 4.0 eV) serves as an electron transport layer. Spiro-OMeTAD TiO_2 (with HOMO level of – 5.2 eV and a LUMO level of – 2.2 eV) works as a hole transport layer and Al (work function − 4.6 eV) works as a top metal electrode. The two formamidinium lead halide based PSCs layer configuration used for the simulation study are FTO/TiO_2/ $CH_3NH_3PbI_3$/ spiro-OMeTAD/Al and FTO/TiO_2/ $CH_3NH_3PbI_3$/ spiro-OMeTAD/Al. SCAPS-1D is the simulation software used in our study. The simulation of the proposed solar cells was performed under the illumination spectrum AM1.5G, 1000 W/m^2 at different sets of operating temperature and absorbing layer thickness. All the primary parameters of the formamidinium lead halide perovskite layers along with its ETL, HTL and glass substrate used in the simulation are summarized in Table 1 and Table 2. The device architecture of the proposed PSCs is shown in Fig. 2.

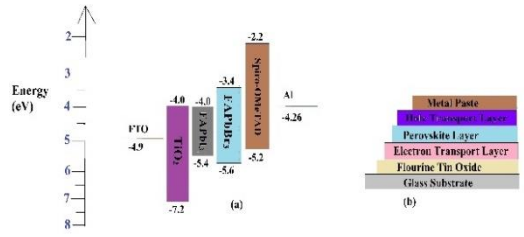


Fig. 1 – Band energy-level diagram and schematic diagram of the proposed perovskite solar cellmodel

Table 1 – The parameters set for $FAPbI_3$ and $FAPbBr_3$ based PSC at 300 K and at A.M1.5G

Sl. No.	Parameters	$FAPbI_3$	$FAPbBr_3$	Reference
1	Thickness (μm)	0.1, 0.5, 1, 2, 4	0.1, 0.5, 1, 2, 4	
2	Band gap (eV)	1.52	2.27	[15]
3	Electron affinity (eV)	4.74	4.51	[16]
4	Dielectric permittivity	11.4	8.6	[17], [18]
5	Donor density (1/cm^3)	3.9×10^9	1.5×10^9	[17], [18]
6	Acceptor density (1/cm^3)	3.9×10^9	1.5×10^9	[19]
7	Electron mobility (cm^2/Vs)	27	14	[19]
8	Hole mobility (cm^2/Vs)	27	14	[15]

Table 2 – The parameters set for different materials taken for the proposed PSC architecture

Sl. No.	Parameters	FTO [20]	TiO_2 [21, 22]	Spiro-OMeTAD [23]
1	Thickness (μm)	0.5	0.03	0.3
2	Band gap (eV)	3.5	3.2	3
3	Electron affinity (eV)	4	4.26	2.45
4	Dielectric permittivity	9	9	3
5	Donor density (1/cm^3)	2×10^{19}	6×10^{19}	0
6	Acceptor density (1/cm^3)	0	0	2×10^{18}
7	Electron mobility (cm^2/Vs)	20	4	2×10^{-4}
8	Hole mobility (cm^2/Vs)	10	2	2×10^{-4}

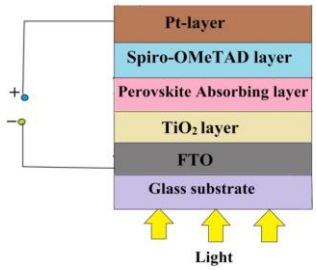


Fig. 2 – Diagram of the solar cell with all the layers and its architecture

3. RESULTS AND DISCUSSION

3.1 Influence of Thickness of Absorber Layer on Device Performance

The thickness of the perovskite absorbing layer is one of the important parameter that plays a role in improving the efficiency of the solar cells. The thickness was varied between 0.1 μm, 0.5 μm, 1.0 μm, 2.0 μm, and 4.0 μm for each of the perovskite materials in the simulation study to obtain the optimum thickness of PSCs. Tan et al. suggested that the efficiency of PSCs in simulation study is mainly dependent on two factors, photon absorption and carrier transport. Photon absorption results in thin absorber layer whereas the carrier transport is for thick absorber layer [20]. Fig. 3 shows the variation of PCE of the proposed devices with respect to the perovskite layer thickness. Here, Fig. 3 shows the *J-V* characteristics of $FAPbX_3$ perovskite layer. It is evident that the thickness of perovskite absorber has significant impact on the device performance. It is evident from Fig. 3 that the parameters viz open circuit voltage (V_{OC}), short circuit current density (J_{SC}), PCE and maximum power output (P_{MAX}) increases with increase in the thickness of the

perovskite active layers initially up to 1.0-2.0 μm. But it could be seen that the values of V_{OC} and J_{SC} do not change significantly, when the thickness of perovskite layer increases beyond 2.0 μm. This may be due to increase in the carrier diffusion lengths. The charge diffusion length is an important material and device property for PSCs that has a relation with the thickness of the absorbing layer. The simulation results reveals that the optimum thickness for $FAPbI_3$ and $FAPbBr_3$ layers are 2.0 μm. The PCE values at optimum thickness for the active perovskite materials, $FAPbI_3$ and $FAPbBr_3$ has been found to be 6.6 % and 20.59 %, respectively, for the proposed cell architecture.

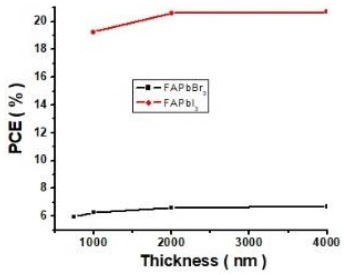


Fig. 3 – Variation of PCE of the proposed devices with respect to the perovskite layer thickness

3.2 Influence of Device Temperature on Perovskite Solar Cell Performance

The working temperature of the proposed model in our simulation study is changed from 15 °C to 75 °C, to determine the best working temperature for suitable electrical performance of the device. It is important to demonstrate a suitable device PCE of PSC at a lower temperature under 1 sun illumination. Fig. 4 shows the influence of temperature on PCE of PSCs for the two perovskite absorbing layers. The overall power conversion efficiency in all PSCs significantly remains unchanged as the operating temperature increases as seen in the fig. The relation between short circuit current density (J_{SC}) and open circuit voltage (V_{OC}) along with temperature (T) can be explained by using Eq. 1:

$$V_{OC} = \frac{kT}{q} \ln\left(\frac{J_{SC}}{J_0} + 1\right), \quad (1)$$

where J_{SC} is the current density, J_0 is the reverse saturation current, q is the electronic charge, n is the ideality factor, and K is the Boltzmann constant.

The values of J_{SC} and V_{OC} remain substantially constant with increase in the device temperature for each of the PSCs. The optimum device temperature for the $FAPbI_3$ and $FAPbBr_3$ perovskite active layers was found out to be 30 °C.

3.3 Influence of Quantum Efficiency

The External Quantum Efficiency (EQE) is an important measurement characteristic for PSCs. Fig. 5 is the QE curve of the $FAPbI_3$ and $FAPbBr_3$ based PSCs with various absorber thickness at different wavelengths of light. In the simulation work, we have studied the quantum efficiency effect of the two perovskite active layers for the wavelength range between 300 nm to 900 nm. $FAPbI_3$ active layer indicates a quantum efficiency of 90 % at 2.0 μm thickness and 98 % at 4.0 μm thickness, whereas $FAPbBr_3$ indicates a quantum efficiency of 98 % at 2.0 μm thickness and 87 % at 4.0 μm thickness. The best spectral response in the simulation study for $FAPbI_3$ was obtained from 300 nm to 820 nm, and 300 nm to 550 nm for $FAPbBr_3$ perovskite layer, respectively. The optimum performance for the $FAPbI_3$ and $FAPbBr_3$ perovskite materials was found to be approximately at 780 nm and 530 nm of solar spectrum, respectively. This suggests that all the perovskite materials are visible active and can be used to further develop commercial PSCs.

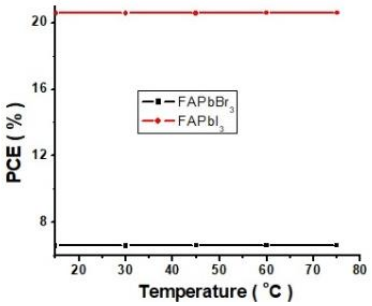
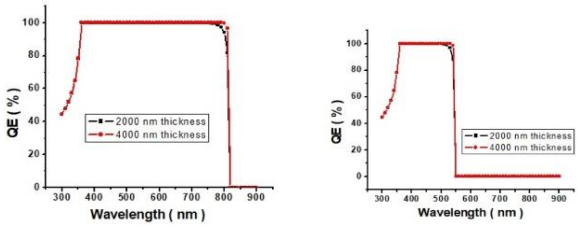


Fig. 4 – Influence of temperature on PCE of PSCs for the two perovskite absorbing layers



a b

Fig. 5 – QE curve of $FAPbI_3$ and $FAPbBr_3$ based PSCs with various absorber thickness

4. CONCLUSIONS

These simulation-based studies demonstrate the optimized thickness of active perovskite layer, device temperature and internal quantum efficiency using the SCAPS-1D software. The proposed device architecture FTO/TiO_2/$FAPbX_3$/ spiro-OMeTAD/Al (where X represents I and Br) based PSCs were investigated in this simulation-based study. We can conclude from the results that the device performance depends on the thickness and device temperature. The optimized thickness for $FAPbI_3$ and $FAPbBr_3$ layers was found out to be 2.0 μm, whereas the optimum device temperature for the $FAPbI_3$ and $FAPbBr_3$ perovskite active layers was found out to be 30 °C. The spectral response for the $FAPbI_3$ and $FAPbBr_3$ perovskite materials was found to be approximately at 780 nm and 530 nm of solar spectrum, respectively. It could be concluded that all the perovskite materials are active in the visible region of the solar spectrum. The results exhibited that by optimizing the necessary electrical parameters, the PCE of PSCs could be increased to a higher efficiency.

ACKNOWLEDGEMENTS

The authors would like to thank Professor Marc Burgelman, Department of Electronics and Information Systems at the University of Gent, Belgium, who permitted the authors to use their simulation software (SCAPS-1D). The authors also acknowledge SERB, DST, New Delhi (File No. SERB/F/9654/2016-17 and PDF/2016/002390) for the financial support to carry out this work.

REFERENCES

1. A. Perumal, S. Shendre, M. Li, Y.K.E. Tay, V.K. Sharma, S. Chen, Z. Wei, Q. Liu, Y. Gao, P.J.S. Buenconsejo, S.T. Tan, C.L. Gan, Q. Xiong, T.C. Sum, H.V. Demir, *Sci. Rep.* **6**, 1 (2016).
2. J. Jancik, A.J. Prochazkova, M.C. Scharber, A. Kovalenko, J. Másilko, N.S. Sariciftci, M. Weiter, J. Krajcovic, *Cryst. Growth Des.* **20** No 3, 1388 (2020).
3. K. Hills-Kimball, Y. Nagaoka, C. Cao, E. Chaykovsky, O. Chen, *J. Mater. Chem. C* **5** No 23, 5680 (2017).
4. W.S. Yang, B-W. Park, E.H. Jung, N.J. Jeon, Y.C. Kim, D.U. Lee, S.S. Shin, J. Seo, E.K. Kim, J.H. Noh, S. Seok, *Science* **356** No 6345, 1376 (2017).
5. S. Sun, F.H. Isikgor, Z. Deng, F. Wei, G. Kieslich, D.B. Paul, J. Ouyang, A.K. Cheetham, *ChemSusChem* **10** No 19, 3740 (2017).
6. G.E. Eperon, S.D. Stranks, C. Menelaou, M.B. Johnston, L.M. Herz, H.J. Snaith, *Energy Environ. Sci.* **7** No 3, 982 (2014).
7. A.A. Zhumekenov, M.I. Saidaminov, M.A. Haque, E. Alarousu, S.P. Sarmah, B. Murali, I. Dursun, X-H. Miao, A.L. Abdelhady, T. Wu, Mohammed F. Omar, O.M. Bakr, *ACS Energy Lett.* **1** No 1, 32 (2016).
8. N. Cheng, W. Li, S. Sun, Z. Zhao, Z. Xiao, Z. Sun, W. Zi, L. Fang, *Mater. Sci. Semicond Proc.* **90**, 59 (2019).
9. M. Burgelman, K. Decock, S. Khelifi, A. Abass, *Thin Solid Films* **535**, 296 (2013).
10. S. Karthick, S. Velumani, J. Boucle, *Sol. Energ.* **205**, 349 (2020).
11. Y. Wu, W. Chen, G. Chen, L. Liu, Z. He, R. Liu, *Nanomaterials* **8** No 6, 356 (2018).
12. C-C. Chueh, C-Z. Li, A.K.Y. Jen, *Energy Environ. Sci.* **8** No 4, 1160 (2015).
13. M.A. Haque, A.D. Sheikh, X. Guan, T. Wu, *Adv. Energy Mater.* **7** No 20, 1602803 (2017).
14. M. Pegu, L. Calio, M. Ahmadpour, H-G. Rubahn, S. Kazim, M. Madsen, *Emergent Mater.* **3**, 109 (2020).
15. T.J. Jacobsson, J.P. Correa-Baena, M. Pazoki, M. Saliba, K. Schenk, M. Grätzel, A. Hagfeldt, *Energy Environ. Sci.* **9** No 5, 1706 (2016).
16. S. Tao, I. Schmidt, G. Brocks, J. Jiang, I. Tranca, K. Meerholz, S. Olthof, *Nat. Commun.* **10**, 2560 (2019).
17. N-G. Park, *Nano Converg.* **3** No 15, 1 (2016).
18. O. Gunawan, S.R. Pae, D. Bishop, Y. Lee, Y. Virgus, N. Jeon, J. Noh, X. Shao, T. Todorov, D. Mitzi, B. Shin, *arXiv: Applied Physics* (2018).
19. W. Rehman, R.L. Milot, G.E. Eperon, C. Wehrenfennig, J.L. Boland, H.J. Snaith, M.B. Johnston, L.M. Herz, *Adv. Mater.* **27** No 48, 7938 (2015).
20. K. Tan, P. Lin, G. Wang, Y. Liu, Z. Xu, Y. Lin, *Solid State Electron.* **126**, 75 (2016).
21. S.Z. Haider, H. Anwar, M.A. Wang, *Semicond. Sci. Technol.* **33** No 3, 035001 (2018).
22. A. Husainat, W. Ali, P. Cofie, J. Attia, J. Fuller, *Am. J. Opt. Photon.* **7** No 2, 33 (2019).
23. N. Devi, K.A. Parrey, A. Aziz, S. Datta, *J. Vac. Sci. Technol. B* **36** No 04G105 (2018).
24. A.B. Coulibaly, S.O. Oyedele, N.R. Kre, B. Aka, *Model. Num. Simul. Mater. Sci.* **9** No 4, 97 (2019).

Імітаційне дослідження перовскітних сонячних елементів на основі галогеніду свинцю і формамідію ($FAPbX_3$; X = I та Br) з використанням симулятора SCAPS-1D

R. Saha, K. Chakraborty, M.G. Choudhury, S. Paul

Advanced Materials Research and Energy Application Laboratory (AMREAL), Department of Energy Engineering, North-Eastern Hill University, Shillong-793022, India

Перовскітні наноматеріали стали перспективними матеріалами для застосування не тільки в галузі сонячної енергетики, а й в оптоелектронних пристроях. Дослідження перовскітних сонячних елементів за допомогою моделювання викликають все більший інтерес серед дослідників фотоелектричних систем для глибшого розуміння впливу параметрів матеріалу на характеристики пристроїв на основі сонячних елементів. У роботі проведено імітаційний аналіз перовскітних сонячних елементів на основі галогеніду свинцю і формамідію ($FAPbX_3$; X = I та Br), а саме $FAPbI_3$ і $FAPbBr_3$, за допомогою симулятора SCAPS-1D. У дослідженні spiro-OMeTAD і TiO_2 були використані відповідно як електронний транспортний шар (ETL) і дірковий транспортний шар (HTL) в конфігурації сонячних елементів. Ми оцінили вплив різних товщин перовскітних шарів та робочих температур на продуктивність перовскітних сонячних елементів. У роботі представлені вольт-амперні характеристики в залежності від товщини і температури для кожного з двох перовскітних активних матеріалів.

Ключові слова: Перовскіт, Сонячний елемент, Формамідій, SCAPS, Ефективність перетворення енергії.