Perovskite Solar Cell design using Tin Halide and Cuprous Thiocyanate for Enhanced Efficiency

Shiva Sharma, Rajesh Mehra

Abstract: Utilization of Tin Halide as an absorber in Perovskite solar cells is immensely recognized as a substitute of lead halide absorber because of lead material’s toxicity. Also, Tin halide based Perovskites possess a potential for higher quantum efficiency because of their enhanced light absorption capability due to the wide-ranging absorption spectrum in the visible region with a comparatively lower bandgap of 1.3 eV than lead-based Perovskites. In the present work, glass/ transparent conductive oxide (TCO)/ titanium dioxide (buffer)/ tin halide Perovskite (Absorber)/ cuprous thiocyanate (HTM)/ Metal back solar cell structure has been designed and simulated by SCAPS software which yields Power Conversion Efficiency (PCE) of 28.32% and Fill Factor (FF) of 85.17%. The effect of total defect density, thickness, Valance Band Effective Density of States (VBEDS) and Conduction Band Effective Density of States (CBEDS) for an absorber layer has been analyzed. It has been observed that VBEDS variation has achieved PCE and FF to a significant extent i.e. up to 32.47% PCE and 85.86% FF.

Keywords: Recombination, Absorber, Solar Cell, Simulation.

I. INTRODUCTION

Energy demands have increased drastically. Relying on nonrenewable sources of energy is not an optimum solution to meet the growing energy demands. To fulfill the basic electric energy requirements solar cells with high Power Conversion Efficiency (PCE) and Fill Factor (FF) are desirable to get maximum output electric power from input received solar power. Perovskite solar cells, belonging to 3rd generation with a 3.8% efficiency record in 2009 which has hiked to 22.7% in 2017 [1] and 27.3% in 2018 [2] and is still on incremental stage with an unforeseeable upper limit. Out of two types of Perovskite materials viz. lead halide based Perovskites (CH3NH3PbI3) and tin halide based Perovskites (CH3NH3SnI3) (X=Br, I, Cl), Lead halide based Perovskites (CH3NH3PbI3) are malignant for the environment as it is composed of toxic element Pb. On the contrary, Tin Halide based Perovskites (CH3NH3SnI3) has greater quantum efficiency (QE) yielding ability due to its wide-ranging spectrum in the visible region with a comparatively lower bandgap of 1.3 eV [3]. This work proposes Tin Halide Perovskite Solar Cell design with glass/ transparent conductive oxide (TCO)/ TiO2 (Buffer)/ CH3NH3SnI3 (Absorber)/ CuSCN (HTM)/ Metal Back. The effect of input parameters viz. thickness, Valance Band and Conduction Band Effective Density of States of CH3NH3SnI3 Absorber has been inspected. It has been observed that the variation of VBEDS yields higher FF and PCE by simulation using SCAPS-1D.

II. PROPOSED TIN HALIDE PEROVSKITE SOLAR CELL STRUCTURE

Tin Halide Perovskite solar cell (THPSC) device design is presented in Figure 1 which depicts arrangement of different material layers viz. glass/ TCO (transparent conductive oxide)/ TiO2 (buffer) (ETM)/ CH3NH3SnI3 (Absorber)/ CuSCN (HTM)/ Metal back for the proposed design. Its 1-D numerical simulation and analysis have been made using SCAPS-1D software [4-13].

Photons are made to enter the Glass Substrate Material and passed through the transparent conductive oxide (TCO). Being a mesoporous layer, a pathway is provided by TiO2 for the movement of electrons from the Perovskite layer. Perovskite layer acts like an absorber layer which acts as an intermediating layer between the electron transport material layer (ETM) and hole transport material layer (HTM). The light is absorbed by Perovskite material to generate excited photoelectrons which are injected into the TiO2 (ETM). There is p-type CuI, HTM Layer, placed after Absorption Layer, HTM layer is made from CuI material and holes for conduction. The CuI (HTM) transports holes from a counter electrode (made from Silver or other metal which are deposited on HTM via thermal evaporation) and separates charges. Electrons and holes are then generated. Holes are infused into HTL whereas Electrons are interjected into an ETL. Movement of electrons towards the surface of photo-electrode through the TiO2 molecules creates an electrical current. In the external circuit, counter electrode excerts charge carriers via TCO.
Understanding of solar cell’s basics

- The rate of partial increase in space charge density is \( - \rho \frac{\partial n}{\partial t} \) (n)

- Hole current density \( J_p \) is mentioned below as:

\[
J_p = - \frac{\mu_p n \frac{\partial E_{Fp}}{\partial x}}{q}
\]

- For \( n \) and \( p \) where \( A \) and \( B \) are mentioned below:

\[
\frac{\partial^2 \psi}{\partial x^2} = -\frac{q}{\varepsilon_x} \left( p - n + N_D^+ - N_A^- + N_{def} \right)
\]

- The output parameters calculated are Fill Factor (FF), Power Conversion Efficiency (PCE), Short Circuit Current Density \( (J_{sc}) \) and Open Circuit Voltage \( (V_{oc}) \). The ratio of energy output from the solar cell to input energy from the sun is called PCE.

- FF determines the solar cell’s maximum power; it is the ratio of maximum power from the solar cell to the product of \( V_{oc} \) and \( I_{sc} \) (short circuit current). The maximum voltage extracted from the solar cell at zero current state is called \( V_{oc} \).

- Tin Halide Perovskite Solar Cell (THPSC) proposed design is presented in Table 1 [17-20].

This structure is made realistic by adding a band to band recombination to 10^{-7}cm/s for achieving equilibrium conditions. For checking the simulation method and working on SCAPS software, the CIGS solar cell has been simulated. The simulated value for fill factor (FF) is 61.4\%, it is in good agreement with the literature value of FF 61.4\% [5].

III. NUMERICAL SIMULATION

To simplify the understanding of solar cell’s basics numerical modeling is used which helps in determining major parameters; SCAPS-1D software [6] numerically solves the 1-dimensional equation that governs the semiconductor material under steady-state condition.

The equation relating space charge density \( (\rho) \) and the electric field of p-n junction \( (E) \) is given by poison’s equation:

\[
\frac{\partial^2 \psi}{\partial x^2} = -\frac{q}{\varepsilon_x} \left( p - n + N_D^+ - N_A^- + N_{def} \right)
\]

Where, electrostatic potential is denoted by \( \psi \), elementary charge is \( q \), static relative permittivity of the medium is \( \varepsilon_x \), density of ionized acceptors (donors) is \( N_A^+ \) (\( N_D^+ \)), hole(electron) density is \( p(n) \) and \( N_{def} \) represents the density of possible defects (Donor or Acceptor) [14,15].

Continuity equation of hole and electron in steady-state are mentioned below as:

\[
\frac{\partial p}{\partial t} - U_p + G = \frac{\partial n}{\partial t}
\]

\[
\frac{\partial n}{\partial t} - U_n + G = \frac{\partial p}{\partial t}
\]

Hole, electron current densities are \( J_p \) and \( J_n \); electron-hole generation rate is \( G = \frac{\partial n \partial p}{\partial t \partial t} \) is the rate of partial increase in electron(hole). \( U_p(U_n) \) is the net recombination rate for electron(hole).

IV. PROPOSED DEVICE SIMULATION

Simulated energy band diagram for the proposed Tin Halide Perovskite Solar Cell (THPSC) structure is illustrated in figure 3, which represents the transition energy of metastable defects and the defect level energy of the defects present. The solar cell device is a p-i-n-type structure. The absorption sub-model used for all layers of the THPSC structure is the sqrt(hv-Eg) law model. The optical absorption constant \( \alpha(\lambda) \) of a layer is given by the equation \( \alpha(\lambda) = \left( A + \frac{B}{h \nu} \right) \sqrt{h \nu - E_p} \) where \( A \) and \( B \) are the absorption constants in cm^{-1}eV^{1/2} & cm^{-1}eV^{1/2} respectively where \( h \) is the plank’s constant, \( \nu \) is the frequency, \( E_p \) is the actual bandgap of the material [6].

On simulating the proposed THPSC with input parameters as depicted in table 1 and 2, PCE of 28.30\%, FF of 85.08\%, \( V_{oc} \) of 0.9689 V and \( J_{sc} \) of 34.336mA/cm² are output parameters as per the J-V Curve in figure 4.

The optical reflectance of layers in this work is considered neither at the surface and nor at the interface.
Table- 1: The proposed design input parameters for its different layers [17-20]

<table>
<thead>
<tr>
<th>Parameters</th>
<th>TCO</th>
<th>TiO$_2$(buffer)</th>
<th>ETL</th>
<th>CH$_3$NH$_3$SnI$_3$ (Absorber)</th>
<th>CuSCN[HTL]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness (nm)</td>
<td>500</td>
<td>30</td>
<td>750</td>
<td>350</td>
<td></td>
</tr>
<tr>
<td>Band gap (eV)</td>
<td>3.5</td>
<td>3.20</td>
<td>1.3</td>
<td>3.4</td>
<td></td>
</tr>
<tr>
<td>Electron affinity $\chi$ (eV)</td>
<td>4</td>
<td>4.26</td>
<td>4.17</td>
<td>2.1</td>
<td></td>
</tr>
<tr>
<td>Relative permittivity $\varepsilon_r$</td>
<td>9</td>
<td>9</td>
<td>8.2</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td>Effective conduction Band Density $N_c$ (cm$^{-3}$)</td>
<td>2.20e+18</td>
<td>2.0e+18</td>
<td>1e+18</td>
<td>2.5e+18</td>
<td></td>
</tr>
<tr>
<td>Effective valence band density $N_v$ (cm$^{-3}$)</td>
<td>1.8e+19</td>
<td>1.8e+19</td>
<td>1e+18</td>
<td>1.8e+19</td>
<td></td>
</tr>
<tr>
<td>Electron mobility $\mu_e$(cm$^2$/V·s)</td>
<td>20</td>
<td>20</td>
<td>1.6</td>
<td>2e-4</td>
<td></td>
</tr>
<tr>
<td>Hole mobility $\mu_h$(cm$^2$/V·s)</td>
<td>10</td>
<td>10</td>
<td>1.6</td>
<td>1e+0</td>
<td></td>
</tr>
<tr>
<td>Shallow uniform acceptor density, $N_A$ (1/cm$^3$)</td>
<td>2e+19</td>
<td>1e+16</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>Shallow uniform donor density $N_D$ (1/cm$^3$)</td>
<td>2e+19</td>
<td>1e+16</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>Defect type</td>
<td>Neutral</td>
<td>Neutral</td>
<td>Neutral</td>
<td>Neutral</td>
<td></td>
</tr>
<tr>
<td>Capture cross section for Electrons (cm$^3$)</td>
<td>1e-15</td>
<td>1e-15</td>
<td>2e-14</td>
<td>1e-15</td>
<td></td>
</tr>
<tr>
<td>Capture cross section holes (cm$^3$)</td>
<td>1e-15</td>
<td>1e-15</td>
<td>2e-14</td>
<td>1e-15</td>
<td></td>
</tr>
<tr>
<td>Energetic distribution</td>
<td>Single</td>
<td>Single</td>
<td>Gaussian</td>
<td>Single</td>
<td></td>
</tr>
<tr>
<td>Energy level with respect to $E_V$ (above $E_V$, $E_V$)</td>
<td>0.6</td>
<td>0.6</td>
<td>0.65</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>Characteristic energy (Ev)</td>
<td>-</td>
<td>-</td>
<td>0.100</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$N_t$ total (1/cm$^3$) uniform</td>
<td>1e+15</td>
<td>1e+15</td>
<td>1e+10</td>
<td>1e+14</td>
<td></td>
</tr>
</tbody>
</table>

In this work, a study based on the consequences of the variation of the thickness, defect density $N_t$ (total), Valance Band Effective Density of States (VBEDS) and Conduction Band Effective Density of States (CBEDS) of the tin halide based Perovskite absorber layer by control variable method is conducted to optimize the proposed structure. The CBEDS ($N_c$) and $N_t$ (total) are the most influential parameters for the proposed solar cell efficiency enhancement.

Fig. 3. Energy band diagram for the proposed THPSC structure.

The Shockley–Read–Hall model affecting the performance of the defect density [21, 22] is represented as

![Fig. 3. Energy band diagram for the proposed THPSC structure.](image)

![Fig. 4. J-V Curve for the proposed THPSC design.](image)
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\[ R = \frac{np-n_i^2}{\tau_p \left( n + N_e (E_g - E_l)/kT \right) + \tau_n (p + N_h (E_g - E_l)/kT)} \]  

The solution of Poisson and continuity equations yields the concentration of holes/electrons represented as \( p(n) \). Appropriate forward bias due to \( qV > 3kT \) results in the abandonment of the \( n_i^2 \) term. And,

\[ \tau_{n,p} = \frac{1}{\sigma_{n,p} \tau \Phi \tau \tau} \]  

The lifetime of charge carriers is denoted by \( \tau_{n,p} \). The defect density’s energy level and concentration are depicted by \( E_t \) and \( N_t \). The thermal velocity and the capture cross-section is represented as \( V_{th} \) and \( \sigma_{n,p} \). By the following equation can be used to calculate the Diffusion length

\[ l = \sqrt{D \tau} \]  

The constant of diffusion is \( D \) and is represented as

\[ D = \mu kT/q \]

The above equations composition can be considered for the calculation of the diffusion length. The flat bands are essential to calculate work function. The absorber layer \( \text{CH}_3\text{NH}_3\text{SnI}_3 \) is used as a p-type of a semiconductor having \( 3.2 \times 10^{10} \text{ cm}^{-3} \) as the initial doping concentration.

V. RESULTS AND DISCUSSIONS

The simulated results obtained through SCAPS are presented in graphical and tabular form and discussed as given below

A. Effect of variation of total defect density of \( \text{CH}_3\text{NH}_3\text{SnI}_3 \) layer

When Total Defect Density \( N_t \) (total) of \( \text{CH}_3\text{NH}_3\text{SnI}_3 \) layer of the proposed Tin Halide Perovskite Solar Cell (THPSC) has been varied from \( 10^5 \) to \( 10^{11} \text{ cm}^{-3} \) keeping other input parameters same as mentioned in table 1. The simulation results are shown in figure 5. It has been observed that the value of PCE is 28.32% at \( 10^7 \text{ cm}^{-3} \) of total defect density \( N_t \) (total).

Fig. 5. Dependence of (a) PCE (b) FF (c) \( V_{oc} \) (d) \( J_{sc} \) on the variation of total defect density of \( \text{CH}_3\text{NH}_3\text{SnI}_3 \) layer of THPSC design.
With increasing the value of N_t (total), PCE decreases linearly and reaches to 28.311% at N_t (total) of 5X10^{10} cm^{-3} and 28.301% at 10^{11} cm^{-3} as shown in figure 5(a). Similarly, the values of FF, J_SC, and V_OC also decrease linearly with increasing the value of N_t (total) as shown in figure 5(b), (c) and (d). The values showing the amount of decrement at each point can be inferred from Table II.

B. Effect of thickness of CH_3NH_3SnI_3 absorber layer

On varying thickness of CH_3NH_3SnI_3 absorber layer of THPSC from 0.5 to 0.8 µm (keeping other input parameters same as mentioned in Table 1), it has been observed that at low thickness value very few charge carriers and less area will be available for conduction which leads to smaller value of PCE, whereas at optimum thickness of absorber layer 0.65 µm a steep increase in value of PCE is achieved due to more availability of charge carriers and their more movement due to more area availability and more conduction.

On the enhancement of the absorber layer’s thickness from 0.65 µm, there is a small increase in PCE. Despite there is an increment in the number of charge carriers because a larger distance is required to be traveled by these charge carriers for conduction through larger absorber layer area which results in a small increase of PCE. A similar effect can be observed for V_OC and J_SC. But FF decreases linearly as illustrated in figure 6 and the values are depicted in Table II.

![Graphs showing PCE, FF, V_OC, and J_SC vs. thickness of CH_3NH_3SnI_3 absorber layer](image)

Fig. 6. Dependence of (a) PCE (b) FF (c) V_OC (d) J_SC on the variation of thickness of CH_3NH_3SnI_3 absorber layer of THPSC design.

C. Effect of variation of Valance Band Effective Density of States (VBEDS) of CH_3NH_3SnI_3 layer

The number of holes for a unit volume per unit energy for a given energy level is VBEDS denoted by N_v and is defined as the number of states which are available to be occupied per energy interval around the top of the valance band.

\[ N_v = 4.82 \times 10^{15} \times (m_p/m)^{3/2} T^{1/2} \]  

\[ m_p = h \pi / (d^2 E_V / dK^2) \]  

\[ h = h/2\pi \]  

Where m_p is the hole effective mass.
mass, \( m \) is the rest mass of hole i.e. \( 9.11 \times 10^{-31} \) Kg, 
\( h \) is the planks constant i.e. \( 6.626 \times 10^{-34} \) J.s or \( 4.135667 \times 10^{-15} \) eV.s,
\( E_V \) is the Valance band energy level,
\( K \) is the Wave vector.

\[
N_\beta = 2 \left( \frac{n_\beta kT}{2\pi \hbar^2} \right)^{3/2}
\]  

Equation (13) is not valid for heavily doped materials [23].

The effective density of states \( (N_\beta) \) is given by

\[
N_\beta = 2 \left( \frac{n_\beta kT}{2\pi \hbar^2} \right)^{3/2}
\]

\( n_\beta \) represents the valence band or conduction band effective mass. In heavily doped materials, (13) is not valid in general [23].

The variation of Fill Factor (FF), Power Conversion Efficiency (PCE), Short Circuit Current Density (J_{SC}) and Open Circuit Voltage (V_{OC}) with the variation of VBEDS i.e. \( N_v \) value is shown in figure 7. When \( N_v \) is varied from \( 10^8 \) to \( 10^{18} \) cm\(^{-3}\) PCE, V_{OC} and FF decreases sharply up to \( N_v \) value of \( 5 \times 10^{17} \) cm\(^{-3}\) and beyond this value there is a small decrease in their value but J_{SC} increases linearly up to \( N_v \) value of \( 5 \times 10^{17} \) and remains constant on even on further increase as depicted in figure 8 and values in Table II.

Fig. 7. Dependence of (a) PCE (b) FF (c) V_{OC} (d) J_{SC} on the variation of VBEDS of CH\(_3\)NH\(_3\)SnI\(_3\) layer of THPSC design.

D. Effect of variation of Conduction Band Effective Density of States (CBEDS) of CH\(_3\)NH\(_3\)SnI\(_3\) layer

CBEDS \( (N_c) \) is varied from \( 10^8 \) to \( 10^{18} \) cm\(^{-3}\) keeping other parameters as mentioned in Table I. The graphs of simulated results are represented in Figure 8. CBEDS \( (N_c) \) is given by

\[
N_c = 2 \left( \frac{n_c kT}{2\pi \hbar^2} \right)^{3/2}
\]

Equation (14) is not valid for heavily doped materials [23]. Where \( N_c \) is the CBEDS, 
\( k \) is the wave vector, \( T \) is temperature, \( m_c \) is an effective mass of electrons.
occupying the conduction band effective density of states.

Fig. 8. Dependence of (a) PCE (b) FF (c) V_{OC} (d) J_{SC} on the variation of CBEDS of CH$_3$NH$_3$SnI$_3$ layer of THPSC design.

On varying N$_c$ from $10^8$ to $10^{18}$ cm$^-3$, PCE and FF increase sharply up to $5 \times 10^{17}$ cm$^-3$ and beyond this there the increase in their value is small whereas the conflicting effect is shown by $V_{OC}$ and $J_{SC}$ increases linearly on another hand.

Figure 9 represents the ratio of the number of charge carriers collected by the proposed cell to the number of incident photons i.e. Quantum Efficiency (QE). The range of QE 300 to 900 nm in which constant value of around 100% QE lies from 360-660 nm.

The comparison between Refered designs [3, 20, 24-27] with the simulated Proposed Design 1 is presented in Table III and the bar graph in figure 10. P1 represents the proposed simulated designs.

Fig. 9. Quantum Efficiency graph for the proposed Design.
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Fig. 10. Bar Graph showing comparison of proposed designs with literature designs.

Table- II: Output parameters for the proposed design obtained by varying input parameters of the absorber layer

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Variation range</th>
<th>Values</th>
<th>PCE(n)(%)</th>
<th>FF(%)</th>
<th>V oc (V)</th>
<th>J sc (mA/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total defect density (Nₖ) (total) (cm⁻³)</td>
<td>10⁵-10¹¹</td>
<td>10⁷</td>
<td>28.32</td>
<td>85.17</td>
<td>0.9684</td>
<td>34.335</td>
</tr>
<tr>
<td>Thickness (µm)</td>
<td>0.5-0.8</td>
<td>0.5</td>
<td>27.53</td>
<td>86.206</td>
<td>0.967</td>
<td>33.025</td>
</tr>
<tr>
<td>VBEDS (Nₙ) (cm⁻³)</td>
<td>10³-10¹⁸</td>
<td>10⁸</td>
<td>32.47</td>
<td>85.86</td>
<td>1.1077</td>
<td>34.141</td>
</tr>
<tr>
<td>CBEDS (Nₙ) (cm⁻³)</td>
<td>10³-10¹⁸</td>
<td>10⁸</td>
<td>27.978</td>
<td>83.678</td>
<td>0.9738</td>
<td>34.335</td>
</tr>
</tbody>
</table>

Table- III: Comparison of Output parameters of proposed designs of Solar Cell with literature designs

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>PCE(n)(%)</td>
<td>24.1</td>
<td>25.68</td>
<td>25.7</td>
<td>26.11</td>
<td>27.6</td>
<td>29</td>
<td>32.47</td>
</tr>
<tr>
<td>FF(%)</td>
<td>81.1</td>
<td>84.11</td>
<td>88.54</td>
<td>87.96</td>
<td>-</td>
<td>-</td>
<td>85.86</td>
</tr>
<tr>
<td>V oc (V)</td>
<td>0.93</td>
<td>1.21</td>
<td>1.056</td>
<td>1.2439</td>
<td>-</td>
<td>-</td>
<td>1.1077</td>
</tr>
<tr>
<td>J sc (mA/cm²)</td>
<td>31.7</td>
<td>25.33</td>
<td>25.483</td>
<td>23.778</td>
<td>-</td>
<td>-</td>
<td>34.141137</td>
</tr>
</tbody>
</table>

VI. CONCLUSION

In this paper Tin Halide Perovskite solar cell (THPSC) structure has been designed and simulated using SCAPS-1D software to determine PCE, FF, V oc and J sc. It has been observed that PCE of 28.32%, FF of 85.17%, V oc 0.9684 V, J sc 34.33532 mA/cm² are obtained. On optimizing the structure with the variation of total defect density Nₖ (total), thickness, valance band effective density of states i.e. Nₙ and conduction band effective density of states i.e. Nₖ of Perovskite (CH₃NH₃SnI₃) absorber layer the maximum PCE of 32.47%, FF 85.86%, V oc 1.1077 V and J sc 34.141137 mA/cm² have been achieved. The graphs are explained properly. Results are presented in the tabular form and are compared with the literature values obtained from different designs.
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