Numerical development of lead-free Cs$_2$TiI$_6$-based perovskite solar cell via SCAPS-1D

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Abstract. Because of the toxicity and stability concerns, commercialization of lead-based perovskite solar cells (PSCs) is limited. Solar cells made entirely of Ti-based all-inorganic perovskite could be a viable answer to these issues. This paper is a theoretical paper on a perovskite solar cell (PSC) based on Cs$_2$TiI$_6$ using all-inorganic charge transport materials. We proposed a high performance perovskite solar cell (PSC) according to variables such as charge transport materials and its optimal thicknesses, absorber thickness, absorber defect density and interface defect density and working temperature. The optimal absorber thickness, Hole transport layer (HTL) thickness, and Electron transport layer (ETL) thickness are 500 nm, 50 nm, and 10 nm, respectively. After analyzing the other factors, we ended up with a high-performance PSC with a power conversion efficiency of 22.5% at room temperature and 22.84% at 270 K. These results are useful for the conception and manufacture of PSCs.

1 Introduction

Organic-inorganic perovskite materials have been extensively explored in recent years as an alternative to silicon-based solar cells for improving device efficiency, and it are a potential alternative to silicon-based solar cells because of its lower manufacturing costs [1,2]. Since the 2012 breakthroughs [3,4], it is now generally agreed that the halide perovskite solar cells could have a significant practical influence in the future generation solar cells. The active material of a typical perovskite solar cell was an organic-inorganic halide material [5,6]. Jena et al. developed the perovskite solar cell (PSC) by employing an organic-inorganic lead halide as an absorber instead of an organic dye [7]. From 2009 to 2019, the PCE (power conversion efficiency) of a typical organic-inorganic hybrid PSC (perovskite solar cell) has risen from 3.8% to over 25%. Despite the excellent optoelectronic properties of organic-inorganic hybrid PSCs, they display three major problems. For starters, it reduce the shelf life of perovskitc compounds. Second, the presence of organic cations creates instability, and third, they use a lead (Pb)-based absorber, which is environmentally damaging [8]. As a result, all inorganic lead-free halide-based PSCs are now introduced [9]. As with organic-inorganic hybrid absorber, organic charge transport become unstable in oxygen, sunlight, and humidity [10,11]. Chen and his colleagues developed Cs$_2$TiBr$_6$ thin film Perovskites with a 3.28 % solar efficiency [12].

Because of its potential light harvesting properties, acceptable bandgap, good optical absorption, high stability, and benign defect property, Cs$_2$TiI$_6$ absorber material is shown to be a suitable alternate. The Cs$_2$TiI$_6$ absorber can be more resistant to environmental stress and have better tolerance [12,13]. Ahmed et al. reported an FTO/SnO$_2$/Cs$_2$TiBr$_6$/MoO$_3$/Au structure, with a stable PCE up to 11.49% [14]. Ahmad and his colleagues suggested an unique perovskite solar cell configuration of Au/CdTe/Cs$_2$TiI$_6$/TiO$_2$/ITO, which has a PCE of 15.06 % [15].

In this study, we propose an all-inorganic PSC with a new combination of FTO/ZnO/Cs$_2$TiI$_6$/MoO$_3$/Au using (SCAPS 1D-3.3.10). We analyzed the effects of various inorganic charge transport materials to find the optimum material for Cs$_2$TiI$_6$-based PSC, e.g., p- MoO$_3$, p-Cul, p-NiO, n-TiO$_2$, n-SnO$_2$, n-ZnO, n-CdS as HTL (Hole Transport Layer) and ETL (Electron Transport Layer) and optimized its thicknesses for achieving optimum performance. We also studied the effect of defect density of Cs$_2$TiI$_6$, defect density of the interface, and the effect of operating temperature on device performance. Upon comparing the performance parameters, e.g., short circuit J$_sc$ (current density), V$_oc$ (open-circuit voltage), PCE (power conversion efficiency), and FF (Fill Factor), we have obtained an optimized high-performance PSC with a novel combination of FTO/ZnO/Cs$_2$TiI$_6$/MoO$_3$/Au, that may aid in designing eco-friendly Ti-based perovskite solar cell for future technologies.
2 Theoretical simulation

FTO/ETL/perovskite/HTL/Au is a unidimensional n-i-p planar heterojunction composition in our modeled PSC structure (Fig. 1). In this structure, we used inorganic lead-free Cs₂TiI₆ as PAL (perovskite active layer), inorganic n-TiO₂, n-SnO₂, n-ZnO, and n-CdS as ETL (electron transport layer), and inorganic p-MoO₃, p-CuI, and p-NiO as HTL (hole transport layer). For validation of our simulation, we first simulated the FTO/Cs₂TiBr₆ (200 nm)/P3HT/Au structure using the SCAPS 1D simulation software and then compared it to the experimental results [12]. HTL and ETL thicknesses of 30 nm are determined by calibrating simulation data with experimental data.

Table 1. Input parameters for each layer of the PSC structure

|                | FTO       | TiO₂      | Cs₂TiBr₆  | P3HT     |
|----------------|-----------|-----------|-----------|----------|
| Thickness, d (nm) | 500       | 30        | 200       | 30       |
| Band gap, E₀ (eV)  | 3.5       | 3.2       | 1.8       | 2        |
| Electron affinity, χ (eV) | 4         | 4.1      | 4         | 3.2      |
| Permittivity, ε₀ | 9         | 9         | 10        | 3        |
| Effective density states at CB, Nₑ (cm⁻³) | 2.2E+18   | 2.2E+18   | 6E+19     | 1E+20    |
| Effective density states at VB, Nᵥ (cm⁻³) | 1.8E+19   | 1E+19     | 2.14E+19  | 1E+20    |
| Electron mobility, µₑ (cm²/V s) | 20        | 20        | 0.236     | 0.0001   |
| Hole mobility, µᵥ (cm²/V s) | 10        | 10        | 0.171     | 0.0001   |
| Density of n-type doping, N₀ (cm⁻³) | 1E+19     | 1E+18     | 3E+19     | 0        |
| Density of p-type doping, Nᵥ (cm⁻³) | 0         | 0         | 3E+18     | 1E+16    |
| Defect density, Nᵥ (cm⁻³) | -         | 1E+15     | 4.17E+15  | 1E+15    |
| Capture cross-section electrons (cm²) | -         | 2E-14     | 1E-15     | 1E-15    |
| Capture cross-section holes (cm²) | -         | 2E-14     | 1E-15     | 1E-15    |
| Reference       | [16]      | [12,17]   | [12]      | [12,16]  |

Table 2. Comparison between simulated and experimental Cs₂TiBr₆ based solar cells using P3HT as HTM

| Device Architecture | Jₑ (mA/cm²) | Vₑ (V) | FF (%) | PCE (%) |
|---------------------|-------------|--------|--------|---------|
| (Experiment) [12]   | 3.87        | 0.89   | 59.5   | 2.05    |
| (Simulation)        | 3.78        | 0.9    | 50.01  | 1.71    |

Fig. 1. Schematic of the simulated n-i-p perovskite solar cell

Fig. 2 depicts the energy band diagram and J-V plots of the simulated FTO/Cs₂TiBr₆/P3HT/Au structure. Table 2 shows the performance comparison between the simulated and experimental structure. The simulation results are clearly in agreement with the relevant experimental results. This also means that the simulation parameters are nearly identical to those of a real device.
3 Results and discussion

In this paper, analyzing several HTMs (Hole Transport Materials) and ETMs (Electron Transport Materials) by optimizing its thicknesses and, also by optimizing the density of PAL defects and interface defects, we propose a new structure FTO/ZnO/Cs₂TiI₆/MoO₃/Au.

3.1 HTL optimization

Instead of using organic P3HT as an HTM, we now select the most suited HTM and calculate its optimal thickness using inorganic CuI, MoO₃, and NiO. Table 3 shows the input values for several HTMs.

### Table 3. Parameter settings for several HTMs, ETMs and for the PSC structure’s absorber

| Parameter                              | p-CuI  | p-NiO  | p-MoO₃ | Cs₂TiI₆ | n-SnO₂ | n-ZnO  | n-CdS  |
|----------------------------------------|--------|--------|--------|---------|--------|--------|--------|
| Thickness, d (nm)                      | 30     | 30     | 30     | 200     | 30     | 30     | 30     |
| Band gap, E_g (eV)                     | 3.1    | 3.8    | 3.5    | 1.80    | 4.1    | 4.18   |
| Electron affinity, χ (eV)              | 2.1    | 1.46   | 2.5    | 4.20    | 4.1    | 4.18   |
| Permittivity, ε_r                      | 6.5    | 10.7   | 12.5   | 18.0    | 9      | 10     |
| Effective density states at CB, N_c (cm⁻³) | 2.8E+19 | 2.8E+19 | 1.0E+19 | 2.2E+17 | 2.2E+18 |
| Effective density states at VB, N_v (cm⁻³) | 1E+19  | 1E+19  | 1.0E+19 | 2.2E+16 | 1.9E+19 |
| Electron mobility, μ_n (cm²/V s)       | 100    | 12     | 100    | 25      | 100    | 100    |
| Density of p-type doping, N_A(p) (cm⁻³) | 43.9   | 2.8    | 100    | 25      | 100    | 100    |
| Density of n-type doping, N_A(n) (cm⁻³) | 0      | 0      | 0      | 1E+19   | 1E+18  | 1E+18  |
| Capture cross-section electrons (cm²)   | 1E+18  | 1E+18  | 1E+18  | 1E+18   | 1E+18  | 1E+18  |
| Capture cross-section holes (cm²)       | 1E+15  | 1E+15  | 1E+15  | 1E+15   | 1E+15  | 1E+15  |

The measurements for several HTMs are shown in table 4, and fig 3 shows the corresponding J-V characteristics for each HTM employed in the device. When compared to the performances of other HTMs, MoO₃ has better J-V characteristics, a higher V_oc, and a higher PCE.

### Table 4. Performance parameters of several HTLs

| Hole Transport Material | J_sc (mA/cm²) | V_oc (V) | FF (%) | PCE (%) |
|------------------------|---------------|----------|--------|---------|
| MoO₃                   | 11.53         | 0.97     | 83.88  | 9.4     |
| CuI                    | 11.53         | 0.69     | 80.76  | 6.42    |
| NiO                    | 11.52         | 0.75     | 81.92  | 7.08    |

Fig. 3. J-V characteristics curve for several HTLs

Fig. 4. Energy level of Cs₂TiI₆ and HTMs

The Valance Band Offset (VBO) for all HTMs is negative, as seen in Fig. 4. When VBO is negative, there is no barrier to photo-generated holes flowing toward the back electrode, hence J_sc is nearly constant. J_sc is found to be 11.52 mA/cm² for NiO, 11.53 mA/cm² for CuI and...
MoO$_3$. However, the increase in negative VBO causes an increase in interface recombination, which reduces $V_{oc}$ [23]. We find $V_{oc}$ is 0.97 V, 0.69 V, and 0.75 V for MoO$_3$, CuI, and NiO respectively. For MoO$_3$, CuI, and NiO, the $V_{oc}$ is 0.97 V, 0.69 V, and 0.75 V, respectively. When it is increased, negative VBO decreases FF from 83.88 % to 81.92 %. MoO$_3$ has the highest PCE = 9.4 % because it has the lowest negative VBO, i.e. best band alignment with Cs$_2$TiI$_6$. PCE declines to 7.08 % when negative VBO increases in NiO. CuI, on the other hand, has the largest negative VBO, indicating poor band alignment, and lowest PCE of 6.42 % [23].

![Fig. 5. PCE for several HTLs](image)

The structure of MoO$_3$ has the best PCE, as shown in Fig. 5, so we picked MoO$_3$ to optimize the thickness. We changed the HTL thickness from 20 nm to 50 nm to optimize the HTL thickness taking in mind the regular feasibility of the perovskite solar cell construction process [24], and its effect is shown in Fig. 6. It can be found that with the increase in the HTL thickness, the values of $J_{sc}$ decreases gradually. $V_{oc}$ increases when the thickness is smaller than 35 nm and then it remained almost unchanged. The FF and PCE of the device first increase when the HTL thickness is less than 50 nm and then decrease slightly with further increasing the HTL thickness. This is because when the HTL is too thin, it may lead to low shunt resistance and current leakage, thus resulting in a low FF [25]. However, when the HTL is too thick, the series resistance will increase, leading to a reduction in FF. The structure using MoO$_3$ as HTM provides the best performance at 50 nm having $V_{oc}$ $0.97$ V, $J_{sc} = 11.54$ mA/cm$^2$, FF = 84.06%, and PCE = 9.46 %. Therefore, we select MoO$_3$ as the optimum HTM for Cs$_2$TiI$_6$ based PSC with an optimized thickness of 50 nm.

![Fig. 6. HTL thickness optimization curve for MoO$_3$](image)

### 3.2 Absorber thickness optimization

We have optimized the absorber thickness by using optimized MoO$_3$ as HTL. We have varied the absorber thickness from 300-520 nm using inorganic TiO$_2$, SnO$_2$, ZnO, and CdS as ETMs underneath the perovskite. Tables 1 and 3 provide the input parameters of the ETMs. Fig. 7 displays the optimization of the absorber thickness for different ETMs. It shows that the use of TiO$_2$ and ZnO provides both the best results than other ETMs, with a PCE of 12.01%, but using ZnO gives a better $J_{sc}$ than using TiO$_2$. The other parameters; $V_{oc}$ and FF are almost the same for all ETMs. So we chose ZnO as ETL, with optimal absorber thickness of 500 nm. The reason for this better PCE of ZnO can be elicited from Fig. 8.

![Fig. 7. PCE curve against thickness variation of the Cs$_2$TiI$_6$ absorber for different ETMs.](image)
Table 5. Output parameters for ETMs of the PSC structure

| Electron Transport Material | J<sub>sc</sub> (mA/cm<sup>2</sup>) | V<sub>oc</sub> (V) | FF (%) | PCE (%) |
|-----------------------------|---------------------------------|-----------------|--------|--------|
| ZnO                         | 16.402363                       | 0.9805          | 74.68  | 12.01  |
| TiO<sub>2</sub>              | 16.402336                       | 0.9805          | 74.68  | 12.01  |
| CdS                         | 16.415707                       | 0.9804          | 74.55  | 12.00  |
| SnO<sub>2</sub>              | 16.301709                       | 0.9804          | 74.75  | 11.95  |

Fig. 8. Energy level diagram of the absorber and different ETMs

In Fig. 8, the conduction band offset (CBO) is negative for all ETMs. When CBO is negative, V<sub>oc</sub> decreases monotonically. The negative CBO being very small there is no effect on the value of V<sub>oc</sub> and FF [14]. According to literature, all the ETMs have a good band alignment with the absorber. So we should have obtained a constant J<sub>sc</sub> [23].

However, we obtain J<sub>sc</sub> 16.415707 mA/cm<sup>2</sup> for CdS, 16.402336 mA/cm<sup>2</sup> for TiO<sub>2</sub>, 16.402363 mA/cm<sup>2</sup> for ZnO and, 16.301709 mA/cm<sup>2</sup> for SnO<sub>2</sub> due to the variation in its quantum efficiency (QE) which is shown in Fig. 9. As J<sub>sc</sub> varies, the PCE of structure using different ETMs also varies.

Fig. 9. Quantum efficiency curves for various ETMs at optimal absorber thickness.

From Fig. 9, we find the CdS, TiO<sub>2</sub>, SnO<sub>2</sub>, and ZnO exhibit as much as 98.75 %, 98.72 %, 98.69%, and 98.72 % QE respectively in the visible range. Therefore, ZnO possesses J<sub>sc</sub> of 16.402363 mA/cm<sup>2</sup> and thus the best PCE. Therefore, we select the optimized Perovskite Active Layer (PAL) thickness to be 500 nm for ZnO as ETL.

3.3 ETL thickness optimization

We optimized the ZnO thickness by varying it from 10 nm to 55 nm as ETL using the optimal HTL and PAL thicknesses. As shown in Fig. 10, there was no significant improvement in the output parameters when the ETL thickness was varied. As a result, we choose the optimized thickness of 10 nm with the highest PCE 12.02 %, V<sub>oc</sub> 0.9805 V, J<sub>sc</sub> 16.402430 mA/cm<sup>2</sup>, and FF 74.71%.
3.4 Absorber defect density optimization

We have studied the impacts of defect density by varying it from $10^{11}$ cm$^{-3}$ to $10^{16}$ cm$^{-3}$ using selected ETM and HTM, as well as optimal HTL, PAL, and ETL thicknesses. The J-V curves at various defect densities are seen in Fig. 11. The $J_{sc}$ of PSC degrades from 16.50 mA/cm$^2$ for $10^{12}$ cm$^{-3}$ to 16.26 mA/cm$^2$ for $10^{16}$ cm$^{-3}$. The increase in the recombination rate as defect density increases is the source of this degradation [26]. When the defect density is less than $10^{12}$ cm$^{-3}$, however, it remains almost constant. As a result, we chose a defect density of $10^{12}$ cm$^{-3}$, with a $V_{oc}$ of 0.9886 V, and $J_{sc}$ of 16.50 mA/cm$^2$.

3.5 Optimization of the defect density at the interface

J-V characteristics for different defect densities at the interfaces for values between $10^4$ cm$^{-3}$ and $10^9$ cm$^{-3}$ are shown in Fig. 12. $V_{oc}$ saturates at defect densities below $10^5$ cm$^{-3}$, as shown in this diagram. $V_{oc}$, on the other hand, decreases from 1.63V to 1.4V above $10^5$cm$^{-3}$ to $10^9$ cm$^{-3}$. As a result, at $10^5$ cm$^{-3}$, we choose the optimum interface defect with $V_{oc}$ of 1.63 V, $J_{sc}$ of 16.50 mA/cm$^2$, FF of 83.42 % and PCE of 22.50 %.

3.6 Optimized device

We achieved a high performance PSC with a new combination of FTO/ZnO/CsTiI$_6$/MoO$_3$/Au after optimizing HTL, PAL, and ETL. The new structure's schematic is shown in Figure 13.

![Fig. 10. Variations in $J_{sc}$ and FF as a function of ETL thickness](image)

![Fig. 11. J-V curve comparison for various absorber defect densities](image)

![Fig. 12. Comparison of J-V curve for different interface defects](image)

![Fig. 13. The optimized novel structure FTO/ZnO/Cs$_2$TiI$_6$/MoO$_3$/Au](image)
The J-V characteristics curve and the QE curve for the optimized device are shown in Fig. 14. The device offers a PCE of 22.5%, $J_{sc}$ of 16.5 mA/cm$^2$, $V_{oc}$ of 1.63 V, and FF of 83.42% with a 99.28% QE at 360 nm, in the visible range. Table 6 compares the performance of different lead-free PSC studies. It can be seen that our device produces comparatively promising results, it’s eco-friendly because it is made entirely of inorganic Ti-based perovskite materials, and that it is expected to be highly stable in the environment due to the use of all inorganic charge transport layers.

**Table 6. Performance of several lead-free PSC**

| Absorber     | PCE  | Ref   |
|--------------|------|-------|
| CsSnI$_3$    | 4.81 | [27]  |
| CsSnBr$_3$   | 3.04 | [26]  |
| CsSnCl$_3$   | 9.66 | [28]  |
| CsSnBr$_3$   | 10.46| [28]  |
| CsGeI$_3$    | 4.94 | [29]  |
| CsGeBr$_3$   | 4.92 | [29]  |
| CsGeCl$_3$   | 2.57 | [28]  |
| CsSm$_{0.5}$Ge$_{0.5}$I$_3$ | 7.11 | [30]  |
| CsSnBr$_2$   | 3.2  | [31]  |
| CsSnI$_3$    | 2.3  | [32]  |
| Cs$_2$TiBr$_6$ | 11.49| [14]  |
| Cs$_2$TiI$_6$ | 15.06| [15]  |
| Cs$_2$TiI$_6$ | **22.5** | **This work** |

**3.8 Working Temperature effect**

To understand the effect of temperature on our new device, and the suitable temperature to get the best PCE, we changed the working temperature from 270 K to 360 K. Fig. 16 shows that as the working temperature rises from 270K to 360 K, the $V_{oc}$ decreases from 1.6585 V to 1.5790 V. This is due to the fact that when the temperature rises, the saturation current and recombination rate increase. Additionally, due to the thermal generation of carriers and the reduction of the band gap, $J_{sc}$ increases from 16.503854 mA/cm$^2$ to 16.504397 mA/cm$^2$ [33]. The FF decreases from 83.43 % to 81.63 % as the rate of increase in $J_{sc}$ is less than the rate of decrease in $V_{oc}$, and the PCE reduces from 22.84 % to 21.27 %. The overall performance of the PSC degrades when PCE decreases [34]. So the device gives a better performance for 270K, it can be used especially in cold regions.

![Fig. 14. (a) J-V characteristics curve, (b) QE curve, for optimized FTO/ZnO/Cs$_2$TiI$_6$/MoO$_3$/Au structure](image)

![Fig. 15. Output parameters variations for various working temperatures.](image)
4 Conclusion

FTO/ZnO/Cs₂TiI₆/MoO₃/Au has been presented as a new PSC configuration. We presented a high-performance PSC with a PCE of 22.5 % at ambient temperature and 22.84 % at 270 K based on factors such as charge transport materials and thickness, absorber thickness, absorber defect density, and interface defect density. For a temperature of 270K, the device performs better, making it ideal for usage in cold areas. The results indicate that Cs₂TiI₆ could be useful as an absorber perovskite in highly efficient lead-free inorganic perovskite solar cell technology.

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