Inorganic Lead-Free B-γ-CsSnI₃ Perovskite Solar Cells Using Diverse Electron-Transporting Materials: A Simulation Study

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ABSTRACT: B-γ-CsSnI₃ perovskite solar cells (PSCs) are simulated employing diverse electron-transporting layers (ETLs, including TiO₂, ZnO, SnO₂, GaN, C₆₀, and PCBM), and a comparative study has been made. Both regular and inverted planar structures are simulated. Effects of the thickness of absorbers and ETLs, doping of ETLs, and interface trap states on the photovoltaic performance are studied to optimize the device structures. The regular structures have larger short-circuit current density (Jₛ) than the inverted structures, but the inverted structures have larger fill factor (FF). All of the simulated optimal PSCs have similar open-circuit voltages (Vₒₒ) of ~0.96 V. The PSCs with TiO₂ ETLs have the best photovoltaic performance, and the optimum structure exhibits the highest efficiency of 20.2% with a Vₒₒ of 0.97 V, Jₛ of 29.67 mA/cm², and FF of 0.70. The optimal PSCs with ZnO, GaN, C₆₀, and PCBM ETLs exhibit efficiencies of 17.88, 18.09, 16.71, and 16.59%, respectively. The optimal PSC with SnO₂ ETL exhibits the lowest efficiency of 15.5% in all of the simulated PSCs due to its cliff-like band offset at the SnO₂/CsSnI₃ interface. Furthermore, the increase of interface trap density and capture cross section is found to reduce the photovoltaic performance of PSCs. This work contributes to designing and fabricating CsSnI₃ PSCs.

1. INTRODUCTION

Perovskite solar cells (PSCs) have emerged as a breakthrough photovoltaic (PV) technology, holding unprecedented promise for high-efficiency, low-cost solar cells due to their excellent photovoltaic properties. However, the potential toxicity associated with the lead-containing PSCs has become a major concern. The primary drawback of Pb perovskites for solar cells is that soluble Pb compounds are well known to be toxic to both the natural environment and humans. Therefore, there has been growing interest in the development of alternative perovskites that use Sn instead of Pb, such as CsSnI₃. Tin-based halide perovskite materials have been successfully employed in lead-free perovskite solar cells. Recently, several studies have revealed that the substitution of the methylammonium cation by cesium (Cs) in the perovskite structure could significantly enhance its thermal stability.

The B-γ-CsSnI₃ perovskite has been studied comprehensively and shown to possess favorable photovoltaic properties for PV application. B-γ-CsSnI₃ has a direct band gap of ~1.3 eV, which is suitable for solar radiation, high absorption coefficient (~10⁴ cm⁻¹ in the visible region), and high charge-carrier mobility (~400 cm²/(V·s) for holes). B-γ-CsSnI₃ has low exciton binding energy (10–20 meV), Wannier-type excitons, so the carrier separation in CsSnI₃ is easier than that in typical organic absorbers. In addition, several studies have shown that the CsSnI₃ has a melting point of 451 °C, and it can be synthesized via melting–solidification, implying superior intrinsic thermal stability. By contrast, the halide perovskite analogues of MASnI₃ and FASnI₃ start to decompose at ~300 °C. Most of the reported power conversion efficiencies (PCEs) for CsSnI₃-based PSCs are between 3 and 5%. Very recently, Li et al. reported a PCE of 8.2% (January 2021) for CsSnI₃ PSCs, and Ye et al. reported PCEs of 10.1 and 9.6% (March 2021). It is remarkable that the PCE of 10.1% is the highest one among all of the lead-free all-inorganic perovskite solar cells reported so far.

The aforementioned considerations provide the rationale for the study of B-γ-CsSnI₃ as a promising inorganic, lead-free, thermally stable perovskite for next-generation PSCs. Dopant concentration, thickness, mobility and lifetime of carriers, surface properties, etc., of the electron-transporting layer (ETL) and hole transporting layer (HTL) and band alignment with perovskites are crucial factors for the transport of charge carriers to the front and back contacts.
many reports on HTL-free PSCs with high performance\textsuperscript{15–18} but very few reports on ETL-free PSCs with high performance have been found till now.\textsuperscript{19} Therefore, it seems that ETLs are essential and necessary for PSCs, at least so far.\textsuperscript{19} Theoretical and simulation studies could help to gain a deep insight into the transport mechanism of ETLs\textsuperscript{20} and select the appropriate electron-transporting materials for CsSnI\textsubscript{3} PSCs. In addition, simulations could help to find the optimal device structures and provide the theoretical possibilities of design for CsSnI\textsubscript{3} PSCs. However, to the best of our knowledge, there has been no theoretical and simulation literature on the comparative study of these conductor (TiO\textsubscript{2}, ZnO, SnO\textsubscript{2}, GaN, C\textsubscript{60}, and PCBM) are adopted as ETLs for CsSnI\textsubscript{3} PSCs. A comparative study on these electron-transporting materials for CsSnI\textsubscript{3} PSCs so far.

In this work, B-Y-CsSnI\textsubscript{3} is used as an all-inorganic lead-free absorber for PSCs. Several inorganic and organic semiconductors (TiO\textsubscript{2}, ZnO, SnO\textsubscript{2}, GaN, C\textsubscript{60}, and PCBM) are adopted as ETLs for CsSnI\textsubscript{3} PSCs. A comparative study on these electron-transporting materials has been made to select the appropriate electron-transporting materials. Both regular and inverted planar structures are simulated. Effects of the thickness of CsSnI\textsubscript{3} and ETLs and the dopant concentration of ETLs on the photovoltaic performance of PSCs are investigated to optimize the device structures for CsSnI\textsubscript{3} PSCs and provide the theoretical possibilities of design for CsSnI\textsubscript{3} PSCs. On the basis of comparing the photovoltaic performance of the optimal PSCs with those ETLs, the possible reasons (band structures, material, and device parameters) behind the discrepancies of performance are investigated and discussed in detail, to provide guidance for designing CsSnI\textsubscript{3} PSCs. In addition, the effects of interface trap density of states are also studied.

2. MODELING AND SIMULATIONS

The exciton in typical organic absorbers is of Frenkel-type; therefore, the exciton binding energy has to be taken into account for the carrier separation for accurate PV device modeling, which is rather complicated.\textsuperscript{21} In contrast, the exciton in the CsSnI\textsubscript{3} perovskite is of typical Wannier-type, so the photoexcited carriers can be dealt with in the same manner as with inorganic materials. The two facts, i.e., structural similarity without the mesoporous structure and the exciton type, enable us to apply an existing device simulator widely used in inorganic solar cells to the perovskite solar cells.\textsuperscript{22}

In this study, AMPS-1D software\textsuperscript{23} is used to simulate CsSnI\textsubscript{3} PSCs. AMPS-1D is a one-dimensional device physics code and based on the basic equations of semiconductors and solar cells.\textsuperscript{21} We have successfully applied this code in the simulation of In\textsubscript{1},Ga\textsubscript{1−}N/SnS and Al\textsubscript{1−},Ga\textsubscript{1−}N/SnS heterojunction solar cells.\textsuperscript{22} Here, we briefly describe the main equations used in the simulation as follows. Poisson’s equation:

\begin{equation}
\frac{d}{dx} \left( \varepsilon(x) \frac{d\varphi(x)}{dx} \right) = q \times \left[ p(x) - n(x) + N_0^+ (x) - N_0^- (x) + p_i (x) - n_i (x) \right]
\end{equation}

where the local vacuum level (in the unit of eV), absolute dielectric constant and the free electron \( n \), free hole \( p \), trapped electron \( n_t \), and trapped hole \( p_h \) as well as the ionized donor-like doping \( N_0^+ \) and ionized acceptor-like doping \( N_0^- \) concentrations are all functions of the position coordinate \( x \). \( q \) is the electron charge. The continuity equation for electrons is as follows:

\begin{equation}
\frac{1}{q} \left[ \frac{dJ_n(x)}{dx} \right] = - G_n(x) + R(x)
\end{equation}

The continuity equation for holes is as follows:

\begin{equation}
\frac{1}{q} \left[ \frac{dJ_p(x)}{dx} \right] = G_p(x) - R(x)
\end{equation}

\( \frac{dJ_n(x)}{dx} = n \mu_n \frac{d\varphi(x)}{dx} = J_n^0 + J_n^f \) and \( \frac{dJ_p(x)}{dx} = p \mu_p \frac{d\varphi(x)}{dx} = J_p^0 + J_p^f \) are the electron and hole current densities, respectively. \( \mu_n \) and \( \mu_p \) are the mobilities of the electron and hole, respectively. \( E_n^0 \) and \( E_p^0 \) are the electron and hole quasi-Fermi levels, respectively. \( R(x) \) is the net recombination rate resulting from direct (band-to-band) recombination and indirect (Shockley–Read–Hall) recombination through gap states. \( G_n(x) \) is the optical generation rate.

Determining transport characteristics then becomes a task of solving the three coupled nonlinear differential equations (eqs 1–3), each of which has two associated boundary conditions. In AMPS, these three coupled equations are solved simultaneously to obtain a set of three unknown state variables at each point in the device: the local vacuum level and the electron and hole quasi-Fermi levels. From these three state variables, the free carrier concentrations \( n(x) = \frac{2}{\sqrt{\pi}} N_i F_1/2 \left[ \frac{E_n(x) - E^0_n}{kT} \right] \) and \( p(x) = \frac{2}{\sqrt{\pi}} N_i F_1/2 \left[ \frac{E_p(x) - E^0_p}{kT} \right] \), where \( N_i \) and \( N_c \) are the effective density of states in the conduction band and valence band, respectively; \( k \) is the Boltzmann constant, \( T \) the temperature, and \( F_1/2(\varepsilon) \) the Fermionic integral, fields \((\varphi(x) = \frac{1}{q} \frac{dx}{dx})\), currents, etc. can then be computed.

The structures of regular and inverted planar CsSnI\textsubscript{3} PSCs are shown in Figure 1. The material parameters of CsSnI\textsubscript{3} ETLs, and HTLs used in the simulations are shown in Table S1. The

![Figure 1. Structures of CsSnI\textsubscript{3} perovskite solar cells.](image-url)
A lifetime model is adopted in this work except that the density of states (DOS) model is used in the simulation of interface trap density of states. The simulation is performed under AM1.5G illumination (100 mW/cm², 0.32–1.32 μm). Following are the simulation procedures carried out to understand the behavior and role of ETLs:

(a) The band structures.
(b) The effect of thickness of absorber.
(c) The effect of thickness and dopant concentration of ETLs.
(d) Comparison of the photovoltaic performance for the best structures.
(e) The effect of interface trap density of states.

3. RESULTS AND DISCUSSION

3.1. Band Structures. The band alignment of all contact materials and CsSnI₃ adopted in this study is shown in Figure S1, and the initial thickness and donor concentrations of ETLs are shown in Table S1. The band structures of all of the simulated PSCs are shown in Figure 2. As the band gap ($E_g$) and electron affinity ($\chi$) of TiO₂, ZnO, and GaN are very close, the band structures of PSCs with TiO₂, ZnO, and GaN are similar. For all of the simulated PSCs, there is no spike-like band offset in the conduction band at the ETL/CsSnI₃ interface, which can form an electron barrier to block the drift of photogenerated electrons from CsSnI₃ absorbers and lead to the degraded collection efficiency. Besides, the valence band offset at the ETL/CsSnI₃ interface can generate a hole barrier to prevent the hole in CsSnI₃ from moving to ETLs. However, as the electron affinity of SnO₂ is too large for the PSC with SnO₂, there are cliff-like band offsets in the conduction band at the SnO₂/CsSnI₃ interface ($\Delta E_c = \chi(\text{CsSnI}_3) - \chi(\text{SnO}_2) = 3.62 - 4.5 = -0.88$ eV, $\Delta E_v = \Delta E_c + E_g(\text{CsSnI}_3) - E_g(\text{SnO}_2) = -0.88 + 1.3 - 3.5 = -3.08$ eV), which can give rise to high interface-related recombination.²³
3.2. Effect of the Thickness of Absorbers. Variation of PSC performance with the thickness of CsSnI₃ absorbers is shown in Figure 3. The shape of efficiency curves is similar for all of the ETLs (TiO₂, ZnO, SnO₂, GaN, C₆₀, and PCBM), and the same is true for open-circuit voltages ($V_{oc}$) curves and short-circuit current density ($J_{sc}$) curves. The efficiencies increase more or less steeply up to 100−200 nm and then decrease up to 800 nm (the efficiencies for SnO₂, C₆₀, and PCBM decrease sharply up to 800 nm). The PSCs with SnO₂ exhibit the lowest efficiencies in all of the simulated PSCs. It is attributed to their cliff-like band offsets at the SnO₂/CsSnI₃ interface (see Figure 2), which give rise to high interface-related recombination and low fill factor (FF). When the thickness of absorbers (CsSnI₃) increases, recombination in the absorbers and reverse saturated current increases. Therefore, $V_{oc}$ decreases with increasing CsSnI₃ thickness. The regular structures have larger $J_{sc}$. The $J_{sc}$ increases more or less steeply up to 100−200 nm, which is attributed to the increased optical absorption and photogenerated carriers. When the thickness of absorbers increases further (>200 nm), $J_{sc}$ tends to be saturated (for the regular structures) or decreases (for the inverted structures). It is because the recombination and series resistance increases when the thickness of absorbers increases. For the regular structures, FF decreases with the increasing thickness of absorbers, which is also due to the increased recombination and series resistance. The simulated results show that the inverted structures have larger FF than the regular structures. The optimal thickness of PSCs with TiO₂, ZnO, SnO₂, GaN, C₆₀, and PCBM is 150, 200, 100, 200, 150, and 200 nm, respectively. The optimal thickness of absorbers is in the range of 100−200 nm, which is consistent with some experimental reports. In this work, the hole mobility of 400 cm²/V/s was used for CsSnI₃, which was obtained from transport property measurements. If the calculated hole mobility of 585 cm²/V/s is used, the optimal thickness of CsSnI₃ absorbers should be longer. In the following sections, the thickness of CsSnI₃ absorbers is set as the optimal thickness for all of the simulated PSCs.

3.3. Effect of the Thickness and Dopant Concentrations of ETLs. Figure 4 is the plot of the efficiency and $J_{sc}$ versus the thickness of ETLs. The figure shows that the efficiency and $J_{sc}$ are not very sensitive to the thickness of the ETLs. For example, the efficiency of PSCs with TiO₂ gradually decreases from 19.61 to 19.534% and $J_{sc}$ gradually decreases from 29.689 to 29.532 mA/cm² due to the fractional absorption of incident light by the ETLs and increase in series resistance. $V_{oc}$ and FF remain almost the same when the thickness of ETLs increases. For simplicity, the thickness of the ETLs is set as 30 nm for all of the simulated PSCs.
Figure 4. Efficiency (a) and $J_{sc}$ (b) of CsSnI$_3$ PSCs as functions of the thickness of ETLs.

Figure 5 shows the PV cell parameters as functions of dopant concentrations ($N_D$). In the figure, $N_D$ is set below the density of states in the conduction band of ETLs, and the barrier height of the ETL/electrode contact ($q\phi_m = E_c - E_f$) is set to make a flat band. For the regular structures, the PV parameters (efficiency, $J_{sc}$ and FF) are enhanced (slightly) with increasing dopant concentrations due to the increase in the conductivity of ETLs. For the inverted structures, the PV parameters (efficiency, $V_{oc}$ and FF) increase slightly first and then decrease. The decrease of PV parameters may be attributed to the change of band structures caused by the increased dopant concentrations and has been observed in the experiment for the inverted MAPbI$_3$ PSCs. In the following sections, the dopant concentrations are set as the optimal concentrations (see Table S1) for all of the simulated PSCs. For the inverted CsSnI$_3$ PSCs, the optimal donor concentrations of ETLs are about $5 \times 10^{18}$ cm$^{-3}$.

3.4. Comparison of Photovoltaic Performance for the Best Structures. The best photovoltaic performance and optimal device parameters of all of the simulated PSCs are shown in Figure 6 and Table 1, respectively. From the figure and table, the regular structures have higher $J_{sc}$ than the inverted structures, but the inverted structures have a larger FF. All of the simulated optimal PSCs have similar $V_{oc}$. The optimal PSC with TiO$_2$ exhibits the highest efficiency of 20.131%, and the optimal PSC with SnO$_2$ exhibits the lowest efficiency of 15.504%. It could be concluded that there should not be spike-like or cliff-like band offsets in the conduction band at the ETL/CsSnI$_3$ interface to obtain high performance. The band gap and electron affinity of TiO$_2$, ZnO, and GaN are very close, but it is noted that TiO$_2$ has a large dielectric constant of 55, which is much higher than that of ZnO (8.656$^{28}$) and GaN (9.5$^{29}$). Some groups$^{31-33}$ have investigated experimentally how ETLs with different dielectric constants affect the performance of organic−inorganic hybrid PSCs. They found that the dielectric constant of ETLs plays an important role in preventing electron−hole recombination in PSCs and the device performance could be improved with the increased dielectric constant of ETLs.$^{31-33}$ The photovoltaic performance of PSC with C$_{60}$ is slightly better than that of PSC with PCBM. It is partly attributed to the fact that the carrier mobility of C$_{60}$ (1.6 cm$^2$/V/s$^{34}$) is higher than that of PCBM (0.01 cm$^2$/V/s$^{35}$).

All of the simulated optimal PSCs with diverse electron-transporting materials (both regular and inverted planar structures) exhibit a similar $V_{oc}$ of ~0.96 V. However, in practice, low $V_{oc}$ (<0.6 V) was reported in most of the experimental CsSnI$_3$ PSCs$^{36-39}$ which is far behind the band gap (1.3 eV) of CsSnI$_3$ absorber. For comparison, the $V_{oc}$ exceeding 1.26 V could be achieved with its band gap of 1.55 eV for MAPbI$_3$ PSCs.$^{39}$ In the research field of CsSnI$_3$ PSCs, the most crucial goal is to develop efficient strategy to improve $V_{oc}$ of the devices, and more fundamental studies are required to understand the origin of low $V_{oc}$.$^{36}$ It can be seen from Figure 9b that the interface trap at the ETLs/CsSnI$_3$ interface could cause a significant drop in $V_{oc}$ when the interface trap density and capture cross section are large. From the simulation results and other experimental reports,$^{12,13,38}$ it could be concluded that the low crystal quality of CsSnI$_3$ thin films and the interface quality may mainly be responsible for the low $V_{oc}$ in CsSnI$_3$ PSCs. Defects in the CsSnI$_3$ films and interface trap could increase recombination and reverse saturation current, and thus reduce $V_{oc}$. Li et al.$^{12}$ reported (January 2021) that an obvious $V_{oc}$ enhancement from 0.47 to 0.63 V is achieved by reducing deep level trap-state density with the surface passivation of thiosemicarbazide. Ye et al.$^{13}$ (March 2021) found that the undesirable oxidation in CsSnI$_3$ is restricted by engineering the localized electron density with a pthalimide (PTM) additive and demonstrated that the lone electron pairs of NH and two CO functional groups in the PTM form coordination interactions with Sn$^{2+}$ in the CsSnI$_3$ and protect it from oxidation to Sn$^{4+}$. Their CsSnI$_3$-PTM device exhibited an overall PCE of 10.1% with a $V_{oc}$ of 0.64 V.

The quantum efficiencies (QEs) or spectral responses of all of the simulated PSCs are shown in Figure 7. In the near-infrared region, the quantum efficiencies of the PSCs decrease sharply at about 950 nm, which corresponds to the band gap of CsSnI$_3$ thin film. PSC with TiO$_2$ exhibits the highest quantum efficiency (~0.88) in the visible region. The inverted structures with C$_{60}$ and PCBM exhibit the lowest quantum efficiency (~0.59) in the visible region and thus the lowest $J_{sc}$.

Figure 8 shows the quantum efficiencies of TiO$_2$/CsSnI$_3$/ Spiro-OMeTAD PSCs versus the thickness of CsSnI$_3$ absorbers. The quantum efficiencies increase steeply when the thickness of the absorber increases from 50 to 100 nm due to the increased optical absorption and photogenerated carriers. It tends to be saturated when the thickness of the absorber increases above 200 nm, which is because the recombination increases (so the collection efficiency decreases) and series resistance increases. When the thickness of CsSnI$_3$ is 150 nm, the optimal TiO$_2$/ CsSnI$_3$/Spiro-OMeTAD PSC exhibits the highest efficiency of 20.131% ($J_{sc} = 29.673$ mA/cm$^2$).

3.5. Effect of the Interface Trap Density of States. The nanoscale interface states play an important role in the device performance.$^{39}$ In the device simulation, the effects of the interface trap density of states are also studied by introducing a $\delta$
nm interface defect layer (IDL) at the TiO2/CsSnI3 interface for TiO2/CsSnI3/Spiro-OMeTAD PSCs. Figure 9 shows the plot of PV cell parameters as functions of the interface trap density and capture cross section. From the figure, the interface trap has an important influence on efficiency, $V_{oc}$, $J_{sc}$, and FF. It is clear that efficiency, $V_{oc}$, $J_{sc}$, and FF decrease with increasing interface trap density and capture cross section. When the capture cross sections are $10^{-18}$ and $10^{-17}$ cm$^2$, the efficiencies decrease slowly.
from 19.825 to 19.424% and 16.798%, respectively. However, when the capture cross section increases to $10^{-15}$ and $10^{-14}$ cm$^2$, the efficiency decreases sharply to 3.232 and 0.154%, respectively. Interface trap and large capture cross section increase the recombination centers and hence change in shunt resistance. Optimizing the doping of the ETL and formation of a flat, smooth, and homogeneous surface will significantly reduce the interface trap density and hence enhance the performance. The recombination mechanisms of photocarriers at the ETL/perovskite interfaces could be found in the literature, such as refs 41, 42.

4. CONCLUSIONS

B-γ-CsSnI$_3$ has attracted considerable attention as a promising inorganic, lead-free, thermally stable perovskite for next-generation PSCs due to its favorable photoelectric properties. In this work, CsSnI$_3$ PSCs with diverse ETLs (TiO$_2$, ZnO, SnO$_2$, GaN, C$_{60}$, and PCBM) were simulated. Effects of the thickness of CsSnI$_3$ absorbers and ETLs, dopant concentration of ETLs, and interface trap density of states on the photovoltaic performance of PSCs were investigated to find the optimal device structures. The optimal thickness of CsSnI$_3$ absorbers is in the range of 100–200 nm (the hole mobility of 400 cm$^2$/V/s for CsSnI$_3$ was used for CsSnI$_3$-E, which was obtained from transport property measurements). The regular structures have larger $J_{sc}$ than the inverted structures, but the inverted structures have larger FF. All of the simulated optimal PSCs exhibit a similar $V_{oc}$ of $\sim 0.96$ V.

It was found that the photovoltaic performance of CsSnI$_3$ PSCs is not very sensitive to the thickness of the ETLs. The optimal thickness of ETLs is about 20–30 nm. For the regular structures, the PV parameters (efficiency, $J_{sc}$, and FF) are enhanced (slightly) with increasing dopant concentrations of ETLs due to the increase in the conductivity of ETLs. For the inverted structures, the PV parameters (efficiency, $V_{oc}$, and FF) increase slightly first and then decrease with increasing dopant concentrations of ETLs. The optimal donor concentrations of ETLs are about $5 \times 10^{18}$ cm$^{-3}$ for the inverted structures. It was found that TiO$_2$ is the best electron-transporting material in all of the simulated ETLs. When the thickness of CsSnI$_3$ is 150 nm, the optimal TiO$_2$/CsSnI$_3$/Spiro-OMeTAD PSC exhibits the highest efficiency of 20.2% with a $V_{oc}$ of 0.97 V, $J_{sc}$ of 29.67 mA/cm$^2$, and FF of 0.70. The optimal PSCs with ZnO, GaN, C$_{60}$, and PCBM ETLs exhibit efficiencies of 17.88, 18.09, 16.71, and...

Table 1. Best Photovoltaic Performance and Optimal Device Parameters of CsSnI$_3$ PSCs

| PSC with ETL | TiO$_2$ | ZnO | SnO$_2$ | GaN | C$_{60}$ | PCBM |
|--------------|---------|-----|---------|-----|---------|------|
| thickness of CsSnI$_3$ absorber (nm) | 150 | 200 | 100 | 200 | 150 | 200 |
| thickness of ETL (nm) | 30 | 30 | 30 | 30 | 30 | 30 |
| donor concentration $N_{D}$ (cm$^{-3}$) | $1.00 \times 10^{21}$ | $1.00 \times 10^{18}$ | $1.00 \times 10^{18}$ | $1.00 \times 10^{18}$ | $5.00 \times 10^{18}$ | $5.00 \times 10^{18}$ |
| eff (%) | 20.131 | 17.882 | 15.504 | 18.091 | 16.711 | 16.589 |
| $V_{oc}$ (V) | 0.967 | 0.952 | 0.976 | 0.954 | 0.959 | 0.949 |
| $J_{sc}$ (mA/cm$^2$) | 29.673 | 25.874 | 24.158 | 26.273 | 20.037 | 20.177 |
| FF | 0.702 | 0.726 | 0.658 | 0.722 | 0.87 | 0.866 |

Figure 6. Photovoltaic performance of optimal CsSnI$_3$ PSCs.

Figure 7. Quantum efficiencies of optimal CsSnI$_3$ PSCs.

Figure 8. Quantum efficiencies of TiO$_2$/CsSnI$_3$/Spiro-OMeTAD PSCs versus the thickness of CsSnI$_3$ absorbers.
16.59%, respectively. The optimal PSC with SnO2 ETL exhibits the lowest efficiency of 15.5% in all of the simulated PSCs due to its cliff-like band offset at the SnO2/CsSnI3 interface. It could be concluded that there should not be spike-like or cliff-like band offsets in the conduction band at the ETL/CsSnI3 interface to obtain high performance. Furthermore, the photovoltaic performance of PSCs was found to degrade with increasing interface trap density and capture cross section.

**Figure 9.** Photovoltaic performance of TiO2/CsSnI3/Spiro-OMeTAD PSCs as functions of the interface trap density and capture cross section $\varepsilon$ (a), $V_{oc}$ (b), $J_{sc}$ (c), and FF (d).

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c04096.

Material parameters of CsSnI3, ETLs, and HTLs used in the simulations (Table S1) and band alignment of all contact materials and CsSnI3 adopted in this study (Figure S1) (PDF)

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The manuscript was written through the contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS
This work is supported by the Xiamen University Malaysia Research Fund (No. XUMURF/2019-C3/10IR/0001), the Education Department of Fujian Province Jk Project (No. JK2017033), Minnan Normal University Project (Nos. HX2019077 and XJZ18046), National Natural Science Foundation of China (No. 61975072), Industry-University-Research Collaboration Foundation of the Fujian Province (No. 2020H6017), and Program for Innovative Research Team in Science and Technology in Fujian Province University (Optoelectronic Materials and Device Application). The authors would like to thank the Fonash Research Group in the Pennsylvania State University for providing the AMPS-1D simulation package.

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