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Design and efficiency enhancement of FTO/PC₆₀BM/CsSn₀.₅Ge₀.₅I₃/Spiro-OMeTAD/Au perovskite solar cell utilizing SCAPS-1D Simulator

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Abstract

The poisoning potential of lead, which is the main component of the absorber layer of lead halide (Pb) perovskites, as well as the stability problems of the manufactured devices, constitute a major obstacle to the industrialization of this technology. As a result, recent research is concentrating on lead-free metal halide perovskites. Unfortunately, current lead-free perovskites suffer from poor performance, hence the interest of our study. The research presented here shows that optimizing several variables related to the performance of each layer of a perovskite solar cell (PSC) constructed from lead-free inorganic materials provides an efficiency of 18.13%. We designed a structure with outstanding performance using the FTO/PC₆₀BM/CsSn₀.₅Ge₀.₅I₃/Spiro-OMeTAD/Au configuration. The impact of various relevant factors, such as the thickness and defect density of the absorber layer their doping densities, the back contact work, and the operating temperature, have been thoroughly investigated to boost the performance of the proposed device. The performance of cesium-tin-germanium triiodide (CsSn₀.₅Ge₀.₅I₃) solar cells with different electron transport materials, including ZnO, TiO₂, Cs₂CO₃, C₆₀, C₆₀, ZnO, IGZO, has also been examined. It has been demonstrated that using ZnO as an electron transport layer improves electron extraction and, therefore, performance. The best outcomes are obtained after optimizing all the factors mentioned above, namely: Jsc of 28.70 mA/cm², Voc of 1.115 V, FF of 87.86%, and PCE of 18.13%. Additionally, the explored structure may be an excellent candidate for the future development of lead-free perovskite solar cells.

1. Introduction

For solar cells, hybrid perovskite has become the ‘black gold.’ Indeed, hybrid perovskite applications have flourished in the latter’s boom as a low-cost material for photovoltaic cells. Their photovoltaic conversion efficiency has increased rapidly, rising from about 3.9 percent in 2009 to 25.2 percent in 2021 [1–3]. The perovskite solar cell’s marketing is limited by lead toxicity and stability [4–7]. Many low-toxicity replacements for Pb (II) in halogenated perovskites have been proposed, including Ag (I) [8], Ge (II) [9], and Sn (II) [10, 11]. One of these prospects is Sn (II) halide perovskites. On the other approach, Sn (II) is rapidly oxidized to Sn (IV), and in the CsSn₁₃ perovskite, phase destabilization exists, causing it to lose its properties [12]. The stability and efficiency of perovskites containing Ge and Sn are greater than pure Sn-based materials.

Indeed, perovskites FA₀.₂₅MA₀.₂₅Sn₁₃ were modified by Ito and co-workers [13] by adding 5% Ge to the composition, and they noticed that Ge atoms were located only on the surface of FA₀.₂₅MA₀.₂₅Sn₁₃, passivating perovskite traps, and surface defects. As a result, with 5% Ge doping, a PCE of 4.48 percent is obtained. The authors also doped FA₀.₇₅MA₀.₂₅Sn₀.₉₅Ge₀.₀₅I₃ with 5% Ge and reached 7.9% of PCE [14]. Chen and co-workers prepared a perovskite based on CsSn₀.₅Ge₀.₅I₃ and showed a promising efficiency of 7.11% [15]. Partial replacement of Sn (II) cations by Ge (II) cations resulted in a reduction in the density of Sn vacancies, allowing
this improvement [16, 17]. To enhance the performance of CsSn0.5Ge0.5I3-based devices, it is critical to understand the link between material parameters and device architecture.

The development of lead-free inorganic halide perovskites is still in its early stages and suffers from a weak PCE. Alloys of Ge and Sn represent promising materials, with an advantageous gap for optoelectronic applications and higher stability than pure Sn-based perovskites [18]. Numerous research based on numerical simulation has been carried out to better understand the impact of changes in the solar cell’s physical parameters on its performance and to put it into practice. Accordingly, we examined the optimization strategies of perovskite solar cells with the FTO/PC60BM/CsSn0.5Ge0.5I3/Spiro-OMeTAD/Au construction using SCAPS-1D software, as seen in figure 1. In this configuration, we used PC60BM as an electron transport material (ETM) and Spiro-OMeTAD (HTM) as a hole transport material.

The optimization is based on the analysis of the variations of the parameters of the cell according to several factors such as the influence of the donor density of the PC60BM (electron transport layer) and the acceptor density of Spiro-OMeTAD (hole transport layer (HTL)) as well as the impact of thickness and defect density of CsSn0.5Ge0.5I3 perovskite, on the device’s overall efficiency, were studied using SCAPS-1D. Numerical analysis was also performed on the effect of the back contact work function on the cell efficiency. PSC efficiency was examined as a function of different electron transport layers (ETLs) and temperatures. This paper also recommends an electron transport material (ETM) to improve the effectiveness of perovskite solar cells (PSCs), based on Sn and Ge alloys. The working temperature was also explored to complete this study.

Our model was validated by comparing the results to the experimental values supplied by Chen and co-workers [15].

Based on our findings, the lead-free structure investigated in this research looks to be a feasible and attractive competitor to lead-based photovoltaic cells.

2. Methodology

2.1. SCAPS-1D modeling
Professor Marc Burgelman of the Department of ELIS (Electronics and Information Systems) the University of Ghent in Belgium created the SCAPS computer platform application [19]. Simulation programs necessitate the solution of basic semiconductor equations such as Poisson’s equation and electron continuity equations.

Figure 1. Design of FTO/PC60BM/CsSn0.5Ge0.5I3/Spiro-OMeTAD/Au device configuration.
The following are the electron and hole continuity equations:

\[
\frac{\partial J_n}{\partial x} - U_n + G_n = 0
\]

(1)

\[
\frac{\partial J_p}{\partial x} - U_p + G_p = 0
\]

(2)

For electrons and holes, the drift-diffusion equations are given as follows:

\[
J_n = qn(x)\mu_nE(x) + qD_n\frac{dn}{dx}
\]

(3)

\[
J_p = qp(x)\mu_pE(x) + qD_p\frac{dp}{dx}
\]

(4)

The Poisson equation is written as follows:

\[
\frac{d}{dx}\left(\frac{d\Psi}{dx}\right) = -\frac{q}{\varepsilon}\left[p - n + N^+_D(x) - N^-_A(x) + p_i(x) - n_i(x)\right]
\]

(5)

where \(J_n\) and \(J_p\) are the current densities of electrons and holes, respectively. Recombination rates are denoted by \(U_n\) and \(U_p\), respectively, and the rate of photo generated electron-hole pairs is denoted by \(G\). An electronic charge is denoted by \(q\). The electrostatic potential is denoted by \(\Psi\). Free-hole and free-electron are represented by \(p\) and \(n\), respectively. The electron and hole mobility are represented by \(\mu_n\) and \(\mu_p\), respectively, whereas the electron and hole diffusion coefficients are represented by \(D_n\) and \(D_p\), respectively. The type doping ionized donor and acceptor concentrations are \(N^+_D\) and \(N^-_A\), respectively, whilst the trapped hole and electron concentrations are \(p_i\) and \(n_i\).

2.2. Simulation of devices and numerical modeling

The physical parameters used to simulate the non-inverted planar architecture of solar cells: FTO/PC_{60}BM/CsSn_{0.5}Ge_{0.5}I_{3}/Spiro-OMeTAD/Au are listed in table 1. At a 300 K working temperature, total simulations were run with a 5.6 \(\Omega\) series resistor and a 4202 \(\Omega\) shunt resistor, and the entering lighting is the standard AM1.5G spectrum. The values of the data of the devices and materials utilized for the simulation by the SCAPS-1D software are extracted from experimental data and the literature [15, 22–25]. Figure 1 shows the device structure as FTO/PC_{60}BM/CsSn_{0.5}Ge_{0.5}I_{3}/Spiro-OMeTAD/Au.

3. The experimental perovskite solar cell’s numerical validation

The experimental data presented by Chen [15] is used to confirm the numerically calculated J-V characteristic and external quantum efficiency (EQE) [15].

The PSC of Chen [15], constructed on fluorine-doped tin oxide (FTO), performs poorly (PCE = 7.11\%, FF = 60.6\%, Voc = 0.63 V and Jsc = 18.61 mA cm\(^{-2}\)). Since defect density has such a significant impact on cell efficiency, determining the defect density that reproduces the experimental results is crucial, to use it in our calculations [26, 27]. For this reason, the impact of absorber defect density (N) on solar cell properties is first studied theoretically, utilizing the data in table 1, as shown in figure 2.
In the considered structure, a CsSn_{0.5}Ge_{0.5}I_{3} absorber film (200 nm) is positioned between PC_{60}BM (20 nm) as the electron transport layer (ETL) and Spiro-OMeTAD (200 nm) as the hole transport layer (HTL). As shown in figure 2, there is a good agreement between the J-V characteristics of the simulated PSC and the experimental results once the defect density is fixed at 10^{17} \text{cm}^{-3}.

Figure 3(a) provides the J-V plots of the experimental and theoretical calculations, showing good concordance between the results of reference [15] and our calculations. Similarly, in both solar cells (simulated and experimental), the EQE spans the whole visible spectrum and charge carrier photogeneration starts at 840 nm, as shown in figure 3(b). This shows good agreement between the experimental and theoretical results. Based on illustrations figures 3(a) and (b), we can conclude that our work has been correctly confirmed, as summarized in table 2.

4. Optimization of performance parameters

4.1. Effect of PC_{60}BM layer donor density (N_D)

It is important to raise the conductivity of the PC_{60}BM to improve the short circuit current density (J_{sc}), which is limited by the PC_{60}BM’s low electron mobility and inherent electrical conductivity. This is possible by using an optimal doping concentration to improve the electrical characteristics of the PC_{60}BM layer. An adequate amount of PC_{60}BM doping results in a profound energy state at heterojunction interfaces, which minimizes non-radiative recombination and enhances efficiency [28]. A strong electric field is generated at high and reasonable doping concentrations, effectively collecting electrons while minority carriers are being pushed aside at the ETL/perovskite interface, reducing recombination at the interface [28].

Figure 4 indicates the impact of PC_{60}BM doping concentrations on PSC efficiency (PCE, FF, Voc, and J_{sc}) all improved when the N_D of the ETL is increased. This layer has a variable donor density of 10^{16} to 10^{20} \text{cm}^{-3}. It was observed that J_{sc} rises initially with increased doping concentration but reaches a value of 18.94 \text{mA cm}^{-2} with a doping concentration of about 10^{19} \text{cm}^{-3}. However, when doping concentrations rise, net carrier generation rates rise as well, eventually reaching a steady state with a doping concentration of about 10^{20} \text{cm}^{-3}. The conductivity of the PC_{60}BM layer is improved by doping. Nevertheless, if the doping concentration is high enough, the undesirable effect of impurity diffusion could outweigh the favorable effect of doping, leading to a decrease in carrier mobility. Indeed, higher doping can result in deep coulomb traps, reducing charge carrier mobility and affect the short-current density [29, 30]. Other parameters like FF and PCE follow the same trend as J_{sc}, as seen in figure 4. As a result of the dopant’s increased conductivity, fewer carriers aggregate in the CsSn_{0.5}Ge_{0.5}I_{3}/PC_{60}BM contact zone, resulting in a reduction in leakage current and consequently, a monotonic increase in the PSC’s FF and PCE [31]. As the doping concentration in the PCBM rises from 10^{16} \text{cm}^{-3} to 10^{17} \text{cm}^{-3}, Voc increases from 0.636 to 0.66 V and then remains constant at 0.660 V for doping amounts above 10^{17} \text{cm}^{-3}. The optimal doping value is selected depending on the difficulty to reach high doping practically and the fact that cell parameters do not vary much from the 10^{19} \text{cm}^{-3} concentrations. When the doping concentration grows from 10^{19} \text{cm}^{-3} to 10^{20} \text{cm}^{-3}, PCE enhances only by 0.53%, and to prevent deep
Coulomb traps, we must avoid doping above $10^{19}$ cm$^{-3}$. Finally, the optimal doping concentration seems to be $10^{19}$ cm$^{-3}$.

4.2. Effect of Spiro-OMeTAD layer acceptor density ($N_A$)

The hole transport material has two purposes: the first is to ensure that holes are transported without recombination, and the second is to keep the device free from moisture and oxygen in the air. Spiro-OMeTAD is usually utilized as an HTM in the n-i-p PSCs. Unfortunately, the latter suffers from low conductivity. For this reason, doping of the HTL is required [31]. Indeed, we will be able to minimize the series by varying the hole transport layer’s doping, which will increase both charge extraction and transport [32].

As shown in figure 3, the parameters (PCE, FF, Voc, and Jsc) rise as the concentration of HTL rises, since the conductivity of the HTLs has been enhanced. After a certain amount of doping, this improvement reaches a saturation point. As a result, $10^{19}$ is determined to be a viable value for HTL doping concentration.

![Figure 3](image)

Figure 3. (a) JV plots of the experimental (reference [15]) and theoretical verification. (b) Quantum efficiency of theoretical and experimental verification.

### Table 2.

| Parameters | Values | Unit | Values | Unit | Values | Unit |
|------------|--------|------|--------|------|--------|------|
| PCE (%)    | 7.11   |      | 7.22   |      | 7.11   |      |
| FF (%)     | 60.60  |      | 59.97  |      | 60.60  |      |
| Voc (V)    | 0.63   |      | 0.64   |      | 0.63   |      |
| Jsc (mA cm$^{-2}$) | 18.61 |      | 18.06  |      | 18.61  |      |

Coulomb traps, we must avoid doping above $10^{19}$ cm$^{-3}$. Finally, the optimal doping concentration seems to be $10^{19}$ cm$^{-3}$.
4.3. Influence of the parameters change of the absorbing layer

The absorber’s parameters must be carefully chosen if trying to design efficient solar cells. The active layer thickness is a critical component in maximizing solar cell performance, so it must be properly chosen. Since the light absorption will be minimal if the active layer is extremely thin; nevertheless, if the active layer is thick, charge carrier recombination will occur, which is accentuated before to charge carriers exiting the absorber layer (due to the large distance the charge carriers have to travel).

Perovskite thickness impacts solar cell performance and is also investigated using numerical models with thicknesses ranging from 100 to 700 nm. As illustrated in figure 6, the device’s PCE increases exponentially with thickness, rising from 4.47% for a thickness of 100 nm to 7.97% for a thickness of 480 nm. The Jsc follows the...
same trend as the PCE, as it is explicitly responsible for the PCE’s improvement, as the Voc and FF decrease when the thickness is increased. The PCE decreases when the thickness exceeds 480 nm, as the charge carrier recombination process leads to a decrease in PCE since the photo-generated charges must travel longer distances. At 200 nm thickness, both Voc and FF lose their starting values, although the device Jsc increasing exponentially.

The Voc value decreases as the active layer thickness languishes due to an increase in saturation currents caused by charge carrier recombination losses. Judging from our observations, a thickness of 480 nm will provide the optimal performance. Indeed, the large gain in device efficiency is related to the quick increase in Jsc (an increase of about 19.5% over the initial value).

The defect density \( N_t \) seems to be an essential element that controls the electrical properties and can thus assist in improvements in solar cell efficiency. It is observed that the device’s performance improves as the defect density decreases (see figure 2). Despite the decrease in defect density from \( 10^{15} \) to \( 10^{13} \) cm\(^{-3} \), Jsc and Voc remained stable. However, it is difficult to synthesize a material with a density of defects as low as \( 10^{13} \) cm\(^{-3} \) in an experimental setting, while a value of \( N_t = 10^{15} \) cm\(^{-3} \) allows the solar cell to attain optimum efficiency.

4.4. Back contact work effect
The work function of the contacts has a considerable influence on the ability of the cathode or the anode to collect charge carriers. By altering the work function of the rear contact (anode) from 4.6 to 5.6 eV while keeping the work function of the front contact (FTO) constant at 4.4 eV, the correct collection of the dominant charge carriers may be achieved. The work function of the metal has an impact on the evolution of the parameters related to the performance of CsSn\(_{0.5}\)Ge\(_{0.5}\)I\(_3\) based photovoltaic cells, as seen in figure 7. PCE, FF, Voc, and Jsc rise as the work function approaches 5.0 eV and stay the same at high work function values (> 5.0 eV). Due to this increasing trend, a larger electric field is required on the anode/HTM to improve carrier collection and, consequently, solar yield. Given the need for ohmic contact for higher efficiencies, our research highlights the importance of obtaining a material with a higher work function (> 5.0 eV).

4.5. Effect of different electron transport materials
The performance of the CsSn\(_{0.5}\)Ge\(_{0.5}\)I\(_3\) based PSC was evaluated using several ETMs, including ZnO, TiO\(_2\), Cds, C\(_{60}\), Cds\(_0.5\)Zn\(_{0.5}\)S, as well as IGZO, and then the results were compared to PC\(_{60}\)BM-based ETM.
Figure 8 shows the J-V characteristics of each electron transport material, and figure 9 illustrates the PCE associated with each transport material, the simulation parameters are listed in table 3. A summary of important performance parameters corresponding to each ETM is provided in table 4. In comparison to the other ETMs studied, the results show that ZnO has a better efficiency (figure 9), due to the enhancement of Jsc (figure 8). The conduction band offset between the ETL/perovskite absorber interface and higher carrier mobility can be used to explain these results.

4.6. Impact of temperature on quality of PSC
A solar cell’s performance is critically affected by its working temperature. Even though the normal working temperature of a solar cell is 300 K, the temperature of the cell will be significantly greater during operation. As shown in figure 10, temperature change significantly impacts the JV curves of PV cells. The Voc of PV cells decreases considerably as temperature increases, while Jsc increases only slightly. With rising temperature, a rise in reverse saturation current can induce a reduction in open-circuit voltage. Also, with rising temperature, the

Figure 7. (a) PCE, (b) FF, (c) Voc, and (d) Jsc of PSC according to the back contact work function.

Figure 8. J-V curve for various electron transport materials.
bandgap of the semiconductor reduces, resulting in a slight rise in Jsc. According to the JV graph, PV cells’ energy production decreases when the temperature rises, mainly due to the decrease in Voc. Furthermore, the transport carriers, charge mobility, resistance, and bandgap of the materials are all modified by increasing temperatures, which changes the PV characteristics.

Overall, we obtained promising results such as Jsc of 28.70 mA cm\(^{-2}\), PCE of 18.13%, FF of 87.86%, and Voc of 1.115 V after integrating all optimized parameters. The final PSC optimization parameters are shown in the table inserted in the figure 11. Through this work, we were able to improve the performance-related parameters of the starting device, such as PCE, FF, Jsc, and Voc, by more than 60%, 31%, 37%, and 42%, respectively.
The SCAPS-1D tool is included in this research to numerically model the FTO/PC60BM/CsSn0.5Ge0.5I3/Spiro-OMeTAD/Au perovskite solar cell structure, attempting to equal or outperform Methylammonium lead triiodide perovskite competitors in terms of performance and stability. We have examined the different factors that influence the optimization of the photovoltaic parameters, including the absorber layer thickness, the density of defects, donor density of the ETL, acceptor density of the HTL, the work function of the anode materials, and operating temperature. ETL and HTL doping concentrations have a major effect on PSC performance. Increasing the doping of these charge transport layers improves the Voc and PCE of PSCs due to improved conductivity and quasi-Fermi separation levels. Concentrations that are optimal for exhibiting increased efficiency were found to be $10^{19}$ cm$^{-3}$ for PC60BM-based ETM and Spiro-OMeTADs-based HTM. By optimizing the perovskites thickness of 480 nm, the PCE gains a great deal of performance (18.13 percent). According to the results obtained, the elaboration of an absorber layer with a minimal and realizable defect density, as well as the selection of appropriate work functions for the anode, leads to increased performance. The

**5. Conclusion**

The SCAPS-1D tool is included in this research to numerically model the FTO/PC60BM/CsSn0.5Ge0.5I3/Spiro-OMeTAD/Au perovskite solar cell structure, attempting to equal or outperform Methylammonium lead triiodide perovskite competitors in terms of performance and stability. We have examined the different factors that influence the optimization of the photovoltaic parameters, including the absorber layer thickness, the density of defects, donor density of the ETL, acceptor density of the HTL, the work function of the anode materials, and operating temperature. ETL and HTL doping concentrations have a major effect on PSC performance. Increasing the doping of these charge transport layers improves the Voc and PCE of PSCs due to improved conductivity and quasi-Fermi separation levels. Concentrations that are optimal for exhibiting increased efficiency were found to be $10^{19}$ cm$^{-3}$ for PC60BM-based ETM and Spiro-OMeTADs-based HTM. By optimizing the perovskites thickness of 480 nm, the PCE gains a great deal of performance (18.13 percent). According to the results obtained, the elaboration of an absorber layer with a minimal and realizable defect density, as well as the selection of appropriate work functions for the anode, leads to increased performance. The
greatest efficiency was obtained with an anode work function of more than 5.5 eV. Temperature was also a factor in this study in the sense that as it grows from 277 K to 410 K, the efficacy drops.

The best outcomes are obtained after optimizing all the factors mentioned above, namely: PCE of 18.13%, FF of 87.86%, 28.70 mA cm$^{-2}$, and Voc of 1.115 V.

Ecologically sustainable, low-cost, and high-performance PSCs can be made using the structure of FTO/PC$_{60}$BM/CsSn$_{0.5}$Ge$_{0.5}$I$_3$/Spiro-OMeTAD/Au.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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