

Numerical Assessment of Defect-Type, Resistance, and Spectral Sensitivity in a Lead-Free Cs_2PdBr_6 Double-Perovskite Solar Cell: A SCAPS-1D Simulation Study

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ABSTRACT

Cesium hexabromopalladate(IV) (Cs_2PdBr_6) is a lead-free, vacancy-ordered double perovskite with a near-optimal bandgap of 1.6–1.7 eV and excellent moisture and thermal stability, making it an attractive absorber for environmentally benign photovoltaics. This work presents a SCAPS-1D numerical investigation of a fullerene (C_{60}) electron-transport-layer device in the configuration $\text{FTO}/\text{C}_{60}/\text{Cs}_2\text{PdBr}_6/\text{MoO}_3/\text{Au}$, focusing on four robustness aspects: the influence of absorber defect density for three distinct trap charge states (neutral, single-donor and single-acceptor); the impact of parasitic series and shunt resistance; and the sensitivity of performance to the incident illumination spectrum. Simulation results show that the power conversion efficiency remains essentially unchanged below a bulk defect density of about 10^{14} cm^{-3} , but collapses beyond 10^{15} – 10^{16} cm^{-3} through Shockley–Read–Hall recombination, with donor-type traps proving markedly less damaging than acceptor-type or neutral traps. Series and shunt resistance losses follow classical one-diode-model trends, while spectral analysis confirms AM1.5G as the most reliable reference illumination and highlights a normalisation artefact in truncated spectra. The findings extend the physical understanding of Cs_2PdBr_6 device robustness and provide practical guidance for future fabrication and optimisation efforts.

Keywords: Cs_2PdBr_6 double perovskite; SCAPS-1D; lead-free solar cell; defect density and trap type; series and shunt resistance.

1. Introduction

Photovoltaic technology continues to diversify beyond conventional crystalline silicon toward perovskite absorbers, whose tunable bandgaps and high absorption coefficients have driven rapid efficiency gains over the past decade. However, the toxicity and moisture sensitivity of lead-halide perovskites have motivated growing interest in lead-free double perovskites such as $\text{Cs}_2\text{AgBiBr}_6$, Cs_2TiBr_6 , and Cs_2PdBr_6 . Among these, Cs_2PdBr_6 combines a favourable 1.6–1.7 eV bandgap with long-term structural, thermal and moisture stability [1], [2], positioning it as a particularly promising lead-free absorber for next-generation solar cells. Numerical simulation using SCAPS-1D has become the standard first step in evaluating new absorber and charge-transport-layer combinations before committing to costly experimental fabrication. Previous studies have applied this workflow to optimise hole- and electron-transport layers, absorber thickness and defect densities for lead-free perovskite solar cells, reporting simulated efficiencies as high as 22.78 % through systematic charge-transport-layer and buffer-layer engineering [3], [4], [5], [6]. Tariq, Kashif and Malik [1] recently reported the first SCAPS-1D study of Cs_2PdBr_6 -based solar cells, comparing an inorganic TiO_2 electron transport layer (ETL) against an organic C_{60} ETL in the configuration $\text{FTO}/\text{ETL}/\text{Cs}_2\text{PdBr}_6/\text{MoO}_3/\text{Au}$. After systematic optimisation of absorber thickness, ETL/HTL thickness, back-contact work function, operating temperature, and parasitic resistances, they reported optimised efficiencies of 26.00 % (TiO_2 ETL) and 23.36 % (C_{60} ETL).

Despite this progress, several robustness aspects of the C_{60} -based Cs_2PdBr_6 device remain unresolved. In particular, varying the absorber defect density does not resolve how the electronic character of the dominant trap — neutral, donor-like, or acceptor-like — independently governs the efficiency roll-off, a distinction shown to be significant in other perovskite absorber systems [7]. Likewise, the effect of the illumination spectrum itself, beyond a single AM1.5G reference case, has not been examined,

despite growing awareness that spectral normalisation errors are a common source of unrealistic SCAPS-1D efficiency claims [8].

This work addresses this gap by presenting a defect-type-resolved analysis of the $\text{Cs}_2\text{PdBBr}_6$ absorber alongside independent series-resistance, shunt-resistance and illumination-spectrum sweeps for the $\text{FTO}/\text{C}_{60}/\text{Cs}_2\text{PdBBr}_6/\text{MoO}_3/\text{Au}$ device, benchmarking each trend against the governing semiconductor transport equations and the wider perovskite-simulation literature. The remainder of the paper is organised as follows: Section 2 describes the device structure and the SCAPS-1D simulation methodology; Section 3 presents and discusses the simulated results; and Section 4 concludes the paper and outlines future prospects.

2. Simulation Methodology

The simulated device follows the planar n-i-p architecture $\text{FTO}/\text{C}_{60}/\text{Cs}_2\text{PdBBr}_6/\text{MoO}_3/\text{Au}$ reported by Tariq et al. [1], illustrated schematically in Fig. 1. Sunlight (AM1.5G, $100 \text{ mW}/\text{cm}^2$) is incident from the fluorine-doped tin oxide (FTO) front contact; C_{60} acts as the electron transport layer (ETL); $\text{Cs}_2\text{PdBBr}_6$ constitutes the light-absorbing double-perovskite layer; MoO_3 serves as the hole transport layer (HTL); and gold (Au) forms the back ohmic contact. Baseline layer thicknesses and electronic parameters (bandgap, electron affinity, doping and trap densities, carrier mobilities) are taken directly from Table 1 of the reference study [1], and only the parameter under investigation in each subsection of Section 3 is varied about this baseline.

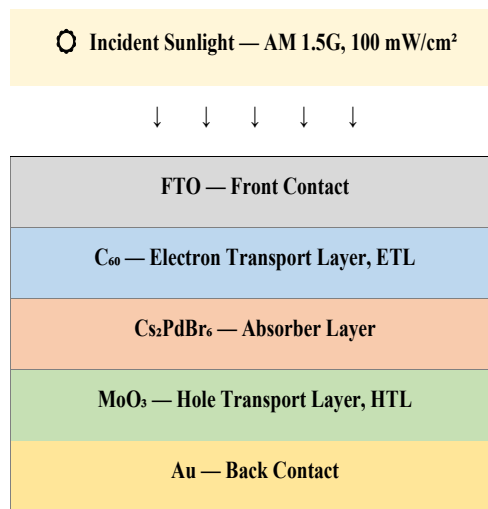


Fig.1: Schematic cross-section of the simulated $\text{FTO}/\text{C}_{60}/\text{Cs}_2\text{PdBBr}_6/\text{MoO}_3/\text{Au}$ double-perovskite solar cell.

The electrical behavior of the simulated device is obtained by solving the coupled Poisson equation, electron-continuity equation, hole-continuity equation, and carrier drift-diffusion equations[9], [10], [11].

The one-dimensional Poisson equation is expressed as

$$\frac{d}{dx} \left(\epsilon \frac{d\psi}{dx} \right) = -q(p - n + N_D^+ - N_A^- + \rho_{def}) \quad (1)$$

where ψ is the electrostatic potential, ϵ is the dielectric permittivity, q is the elementary charge, n and p are the electron and hole concentrations, N_D^+ and N_A^- are ionized donor and acceptor concentrations, respectively, and ρ_{def} represents the charge associated with defect states. The Poisson equation establishes the relationship between the electrostatic potential and the total space-charge density.

Under steady-state illumination, the carrier-continuity equations are

$$\frac{1}{q} \frac{dJ_n}{dx} = G - R \quad (2)$$

$$-\frac{1}{q} \frac{dJ_p}{dx} = G - R \quad (3)$$

where J_n and J_p are the electron and hole current densities, respectively, G is the carrier-generation rate and R is the total recombination rate.

The electron and hole current densities are described using the drift-diffusion equations:

$$J_n = q\mu_n nE + qD_n \frac{dn}{dx} \quad (4)$$

$$J_p = q\mu_p pE - qD_p \frac{dp}{dx} \quad (5)$$

where μ_n and μ_p are the electron and hole mobilities, D_n and D_p are the corresponding diffusion coefficients, and E is the electric field. The diffusion coefficients are related to the carrier mobilities through the Einstein relation,

$$D_{n,p} = \frac{k_B T}{q} \mu_{n,p} \quad (6)$$

where k_B is the Boltzmann constant and T is the absolute temperature.

The optical generation of carriers in the absorber is determined from the incident photon flux and the absorption coefficient.

In a simplified one-dimensional representation,

$$G(x) = \alpha(x)\Phi_0 \exp[-\alpha(x)x] \quad (7)$$

where α is the absorption coefficient and Φ_0 is the incident photon flux. The resulting carrier-generation profile is coupled to the electrical transport equations to calculate the illuminated current-voltage response.

Recombination through bulk and interface defect states is included in the SCAPS calculation. The Shockley–Read–Hall (SRH) recombination rate is represented as

$$R_{SRH} = \frac{np - n_i^2}{\tau_p(n + n_1) + \tau_n(p + p_1)} \quad (8)$$

where n_i is the intrinsic carrier concentration, τ_n and τ_p are electron and hole lifetimes, and n_1 and p_1 describe the equilibrium carrier concentrations associated with the defect energy level.

The principal photovoltaic parameters were obtained from the simulated J–V curve. The short-circuit current density is

$$J_{SC} = J(V = 0) \quad (9)$$

while the open-circuit voltage is determined from

$$J(V_{OC}) = 0 \quad (10)$$

The fill factor is calculated as

$$FF = \frac{V_{mp} J_{mp}}{V_{OC} J_{SC}} \quad (11)$$

where V_{mp} and J_{mp} are the voltage and current density at the maximum-power point.

Finally, the power conversion efficiency is given by

$$\eta = \frac{V_{OC} J_{SC} FF}{P_{in}} \quad (12)$$

where P_{in} is the incident solar power density.

3. Results and Discussion

3.1 Effect of Absorber Defect Density and Defect Type

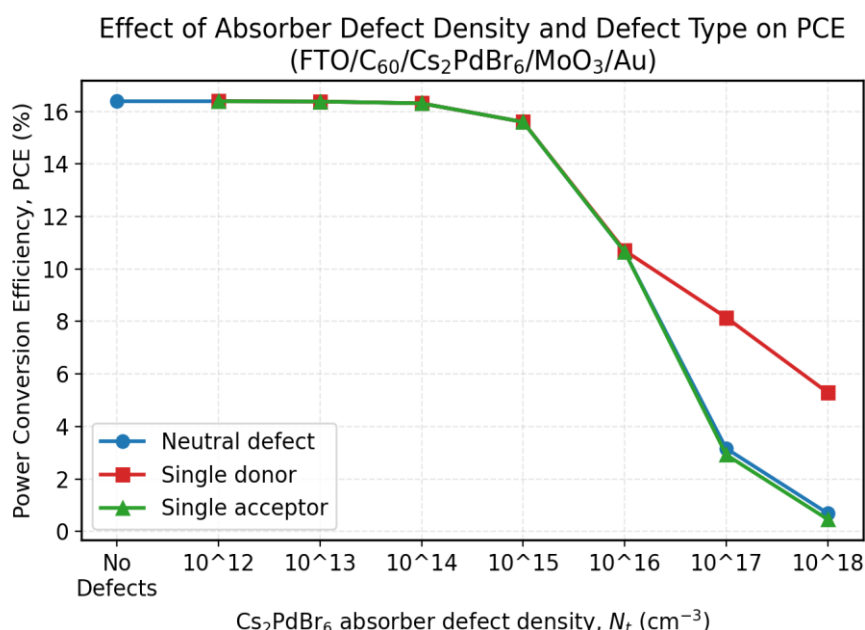


Fig. 2. PCE of the FTO/C₆₀/Cs₂PdBr₆/MoO₃/Au device as a function of the Cs₂PdBr₆ bulk defect density, for neutral, single-donor and single-acceptor trap states.

Up to a defect density of $\sim 10^{14} \text{ cm}^{-3}$, the efficiency of the device is essentially unaffected ($16.39\% \rightarrow 16.31\%$) regardless of the assumed trap type (Fig. 2). Beyond 10^{15} cm^{-3} the PCE collapses steeply, falling by roughly an order of magnitude between 10^{15} and 10^{17} cm^{-3} . This behaviour is the classic signature of Shockley–Read–Hall (SRH) trap-assisted recombination, in which the minority-carrier lifetime scales as $\tau = 1/(\sigma v_{\text{th}} N_t)$, so that the diffusion length $L = \sqrt{D\tau}$ falls as $N_t^{-1/2}$. Once L becomes comparable to or shorter than the absorber thickness, photogenerated carriers recombine before reaching the ETL/HTL and both J_{SC} and V_{OC} degrade sharply — precisely the trend captured here and widely reported for perovskite absorbers by Chowdhury et al., who identified $N_t < 10^{14} \text{ cm}^{-3}$ as the threshold required to sustain $\text{PCE} > 25\%$ in a comparable p-i-n perovskite stack [12], and by Alghamdi et al. for the double-perovskite Cs_2PtI_6 absorber [13]].

The three trap types diverge only at high N_t ($\geq 10^{16} \text{ cm}^{-3}$): the single-donor trap degrades the device far more gently (8.15% and 5.29% at 10^{17} and 10^{18} cm^{-3}) than the neutral or single-acceptor traps ($2.9\text{--}3.2\%$ and $0.5\text{--}0.7\%$, respectively). This is consistent with the charge-state dependence of SRH recombination built into SCAPS-1D: a donor-type trap is positively charged when empty and neutral once it has captured an electron, whereas an acceptor-type trap is neutral when empty and negatively charged once occupied. Because the $\text{Cs}_2\text{PdBBr}_6$ absorber in this stack is a p-type layer ($N_A = 1 \times 10^{17} \text{ cm}^{-3}$ [1]), donor-like traps partially compensate the fixed acceptor charge and act preferentially as hole traps for the majority carriers rather than as efficient recombination centres for the photogenerated minority electrons, whereas acceptor-like traps add to the net negative space charge and recombine minority electrons more efficiently. Saeed et al. observed an analogous asymmetry when comparing single-donor, single-acceptor and double-donor/acceptor defect statistics in a perovskite absorber, reporting markedly different efficiency roll-offs for each trap type at equal trap density [7], which corroborates the trend obtained here. Practically, this indicates that not only the concentration but also the electronic character of the dominant native defect in melt-/solution-grown $\text{Cs}_2\text{PdBBr}_6$ crystals must be controlled (e.g., through Pd-to-Br stoichiometry and halide-rich growth conditions) to preserve device performance.

3.2 Effect of Series Resistance

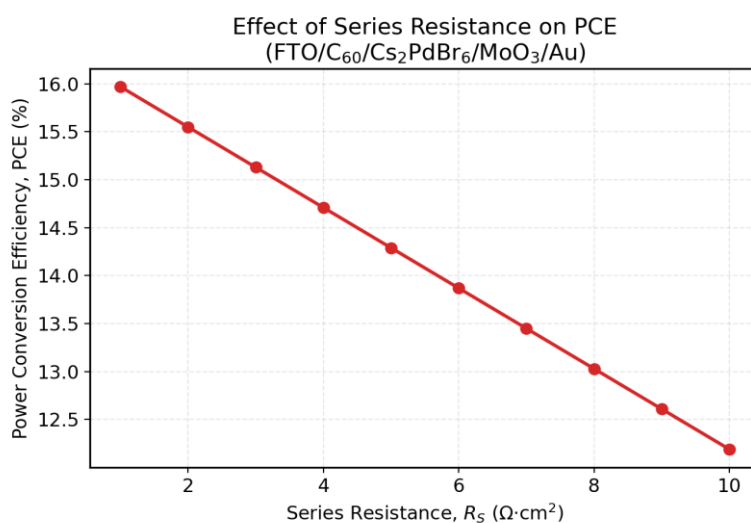


Fig. 3. PCE of the FTO/C₆₀/Cs₂PdBBr₆/MoO₃/Au device as a function of series resistance, R_s (defect-free absorber, 100 mW/cm^2 AM1.5G, 300 K).

As shown in Fig. 3 the PCE falls almost linearly from 15.97% at $R_s = 1 \Omega \cdot \text{cm}^2$ to 12.19% at $R_s = 10 \Omega \cdot \text{cm}^2$ — a relative loss of $\approx 24\%$ in efficiency for a ten-fold increase in R_s . In the single-diode model, series resistance enters the J–V characteristic as an extra ohmic voltage drop, $J = J_{\text{PH}} - J_0[\exp(q(V+JR_s)/nkT) - 1] - (V+JR_s)/R_{\text{sh}}$, so that increasing R_s reduces the voltage delivered to the external load at any given current, which principally lowers the fill factor while leaving J_{SC} and V_{OC} almost unchanged (both are limiting-condition parameters measured at zero net current or zero voltage, where the JR_s drop is small) [14]. This is exactly the trend reported for the reference $\text{TiO}_2/\text{C}_{60}$ $\text{Cs}_2\text{PdBBr}_6$ device, where FF fell from $\sim 90\%$ to $\sim 80\%$ as R_s was raised from 0 to $6.5 \Omega \cdot \text{cm}^2$ while J_{SC} stayed essentially flat [1], and it agrees with the general resistive-loss analysis reported for other SCAPS-1D perovskite structures [13]. Physically, R_s in a real device originates from the bulk resistivity of the transport layers, contact resistance at the FTO/C₆₀ and MoO₃/Au interfaces, and the sheet resistance of the transparent front

electrode; minimising it (e.g., via a thinner/more heavily doped ETL, low-resistance ohmic back contact, and finger/bus-bar metallisation) is therefore essential to realise the ~89–90 % fill factors predicted at the ideal $R_s \rightarrow 0$ limit.

3.3 Effect of Shunt Resistance

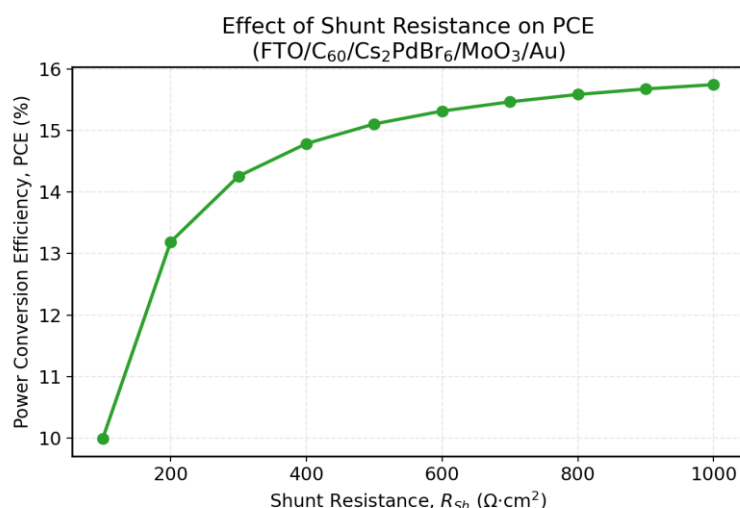


Fig. 4. PCE of the FTO/C₆₀/Cs₂PdBBr₆/MoO₃/Au device as a function of shunt resistance, R_{sh} (defect-free absorber, 100 mW/cm² AM1.5G, 300 K).

In contrast to R_s , the PCE increases monotonically with R_{sh} , from 9.99 % at 100 $\Omega\cdot\text{cm}^2$ to 15.75 % at 1000 $\Omega\cdot\text{cm}^2$ as shown in Fig. 4, with the largest gains at low R_{sh} (+3.20 pp between 100 and 200 $\Omega\cdot\text{cm}^2$) and a clearly diminishing slope thereafter (+0.07 pp between 900 and 1000 $\Omega\cdot\text{cm}^2$). The $(V+J R_s)/R_{sh}$ leakage term in the diode equation diverts part of the photogenerated current away from the external circuit through parasitic shunting paths — pinholes, grain-boundary leakage or local absorber shorts — and this loss is largest, in absolute terms, at low R_{sh} , which explains both the steep initial rise and the $1/R_{sh}$ -type saturation observed at high R_{sh} [15]. A low R_{sh} primarily depresses V_{OC} and FF because the shunting current is comparatively most significant near open circuit, where the photocurrent is small; this is the same qualitative behaviour reported for the TiO₂/C₆₀ Cs₂PdBBr₆ device swept from 1000 to 6000 $\Omega\cdot\text{cm}^2$ [1] and for other SCAPS-simulated perovskite cells in which the FF and PCE were found to saturate only once R_{sh} exceeded a few k $\Omega\cdot\text{cm}^2$ [13]. The practical implication is that shunt paths — rather than series resistance — are the more forgiving loss mechanism once R_{sh} exceeds roughly 10³ $\Omega\cdot\text{cm}^2$, but at low values (<500 $\Omega\cdot\text{cm}^2$, e.g., from pinholes in a thin, solution-processed Cs₂PdBBr₆ film) they can be the dominant efficiency-limiting factor, underscoring the importance of a pinhole-free, fully coalesced absorber film.

3.4 Effect of Illumination Spectrum

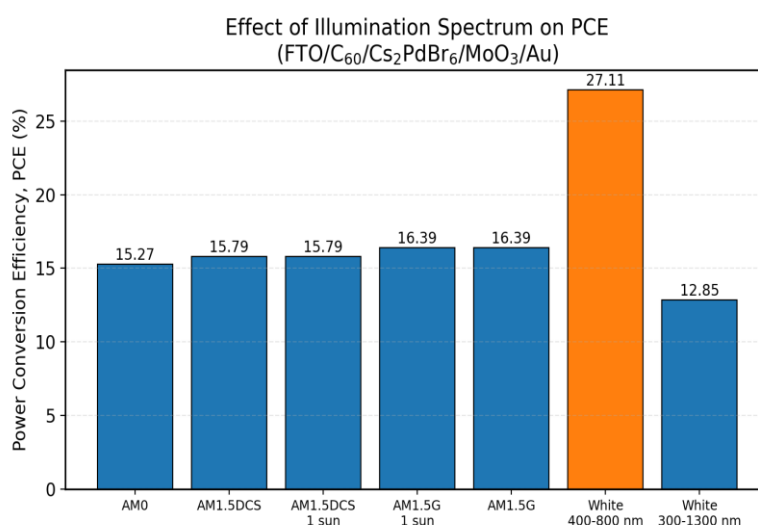


Fig. 5. PCE of the FTO/C₆₀/Cs₂PdBBr₆/MoO₃/Au device under different illumination spectra (defect-free absorber, ideal R_s/R_{sh} , 300 K).

Under the two standard terrestrial references, AM1.5G (16.39%) outperforms AM1.5 direct/circumsolar (AM1.5DCS, 15.79%), and both outperform the extraterrestrial AM0 spectrum (15.27%) (Fig. 5). This ordering matches the physical composition of each spectrum: AM1.5G includes the diffuse-sky component in addition to the direct beam, giving a modestly higher usable photon flux in the visible/near-infrared band that overlaps the 1.6–1.7 eV $\text{Cs}_2\text{PdBBr}_6$ absorption edge, whereas AM1.5D contains only the direct component and therefore slightly less integrated flux in that band. AM0 lacks atmospheric filtering altogether and is comparatively enriched in high-energy UV/blue photons; the excess photon energy above the bandgap is lost as heat via rapid thermalisation of hot carriers, and the associated absorption very close to the front $\text{C}_{60}/\text{Cs}_2\text{PdBBr}_6$ interface increases the carriers' exposure to interface recombination — both effects lower the fraction of incident power converted into extractable current relative to the better-matched AM1.5G spectrum. An essentially identical spectral ranking (AM1.5G 1-sun > AM1.0 > AM0, etc.) was recently reported for a SCAPS-1D-simulated lead-free tin-based perovskite cell, where the authors attributed the AM1.5G advantage to its "optimal balance of visible and near-infrared photons" relative to the absorber's absorption window [16].

The two "white-light" cases, which restrict the incident spectrum to a fixed wavelength window, must be interpreted with more caution. The narrow 400–800 nm window — closely bracketing the $\text{Cs}_2\text{PdBBr}_6$ absorption range — yields an anomalously high PCE of 27.11 %, exceeding even the fully resistance- and defect-optimised C_{60} -based device reported in the reference study (23.36 %) [1]. This is very unlikely to represent a genuine physical gain and is instead consistent with a well-documented artefact of ad hoc spectral truncation in SCAPS-1D: unless the total incident irradiance is manually re-normalised after narrowing the wavelength window, the solver can retain the nominal 100 mW/cm² (one-sun) power reference while concentrating essentially all of that power into the band the absorber converts most efficiently, artificially inflating the apparent PCE. Saidarsan et al., in a systematic audit of more than 250 published SCAPS-1D perovskite studies, explicitly flag non-standard or improperly normalised illumination spectra as one of the leading causes of efficiencies that are not physically attainable, in several cases exceeding the Shockley–Queisser limit [8]. Consistent with this interpretation, broadening the window instead to 300–1300 nm — which now also admits deep-UV and, more importantly, a large sub-bandgap (>750 nm) near-infrared range that $\text{Cs}_2\text{PdBBr}_6$ cannot absorb — drives the apparent efficiency down to the lowest value in the whole data set, 12.85 %, because that unabsorbed power still contributes to the denominator, P_{in} , of the $\text{PCE} = (J_{\text{sc}} \cdot V_{\text{oc}} \cdot \text{FF}) / P_{\text{in}}$ expression. We therefore report the white-light results for completeness but recommend that only the standard AM1.5G (one-sun) spectrum be used for headline efficiency claims, in line with the recommendations of [8] and standard photovoltaic reporting convention.

4. Conclusion

This SCAPS-1D study of the $\text{FTO}/\text{C}_{60}/\text{Cs}_2\text{PdBBr}_6/\text{MoO}_3/\text{Au}$ double-perovskite solar cell shows that the absorber tolerates bulk defect densities up to about 10^{14} cm^{-3} without efficiency penalty, but suffers severe Shockley–Read–Hall recombination losses above $10^{15}–10^{16} \text{ cm}^{-3}$, with donor-type traps consistently less damaging than acceptor-type or neutral traps at equal density — a distinction that should guide future defect-engineering and growth-condition studies of $\text{Cs}_2\text{PdBBr}_6$ crystals and thin films [7], [12]. Series resistance imposes an almost linear fill-factor penalty and must be minimised through low-resistance contacts and transport layers, while shunt-resistance losses, dominant below about $500 \Omega \cdot \text{cm}^2$, underline the importance of pinhole-free absorber coalescence [1], [13]. Spectral analysis confirms that AM1.5G remains the appropriate reference illumination, whereas ad hoc truncated white-light spectra can generate numerically striking but physically unreliable efficiencies unless the incident power is properly re-normalised [8], [16]. Looking ahead, several avenues merit further investigation. Experimental growth of single-crystal or thin-film $\text{Cs}_2\text{PdBBr}_6$ with controlled, donor-dominated defect chemistry could help realise the high efficiencies predicted here, while temperature-dependent defect spectroscopy (e.g., deep-level transient spectroscopy) would help identify the dominant native trap species and validate the SRH parameters assumed in this model. Coupling the present electrical simulation with optical transfer-matrix modelling would refine the photogeneration profile and quantum-efficiency predictions, and extending the parametric study to alternative, non-toxic hole- and electron-transport-layer combinations — following the charge-transport-layer engineering strategy previously applied to lead-free KSnI_3 solar cells [3], [4], [5] — could further improve the fill factor and long-term stability of $\text{Cs}_2\text{PdBBr}_6$ double-perovskite devices *en route* to experimental fabrication and eventual module-level deployment.

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