

Unveiling architectural and optoelectronic synergies in lead-free perovskite/perovskite/kesterite triple-junction monolithic tandem solar cells

Md. Faiaad Rahman^a, Md. Ashaduzzaman Niloy^{a,b}, Ehsanur Rahman^{a,*},
Ahmed Zubair^{a,*}

^a*Department of Electrical and Electronic Engineering, Bangladesh University of Engineering and Technology, Dhaka, Dhaka-1205, Bangladesh*

^b*Department of Electrical and Electronic Engineering, Green University of Bangladesh, Dhaka, Narayanganj-1461, Bangladesh*

Abstract

The widespread use of lead-based materials in tandem solar cells raises critical environmental and health concerns due to their inherent toxicity and risk of contamination. To address this challenge, we focused on lead-free tandem architectures based on non-toxic, environmentally benign materials such as tin-based perovskites and kesterites, which are essential for advancing sustainable photovoltaic technologies. In this study, we present the proposition, design, and optimization of two distinct lead-free monolithic tandem solar cell architectures — an all-perovskite dual-junction device employing potassium tin iodide (KSnI₃) and formamidinium tin triiodide (FASnI₃) as absorbers for the top and bottom subcells, respectively, and a triple-junction monolithic tandem structure incorporating KSnI₃, FASnI₃, and Ag-doped copper zinc tin selenide (ACZTSe) as absorbers for the top, middle, and bottom subcells, respectively. We simulated the optical and electrical characteristics of these devices using the finite-difference time-domain and finite element methods, explicitly considering radiative, non-radiative, and surface recombination mechanisms. The optimized all-perovskite dual-junction solar cell achieved a power conversion efficiency (PCE) of 27.3%, with short-circuit current density (J_{sc}) of 14.74 mA/cm², open-circuit voltage (V_{oc}) of 2.227

*Corresponding authors:

Email addresses: ehsaneee@eee.buet.ac.bd (Ehsanur Rahman),
ahmedzubair@eee.buet.ac.bd (Ahmed Zubair)

V, and fill factor (FF) of 83.14%. Conversely, the optimized triple-junction hybrid perovskite–kesterite architecture secured an elevated PCE of 30.69%, along with J_{sc} of 13.184 mA/cm², V_{oc} of 2.766 V, and FF of 84.18%. These findings reveal the strong potential of lead-free perovskite and kesterite material based absorbers in promoting high-performance hybrid tandem solar cells, highlighting their importance in advancing sustainable and efficient photovoltaic technologies.

Keywords: Perovskite, Kesterite, Tandem, Monolithic, Hybrid Solar Cells

1. Introduction

The global energy industry is rapidly transitioning towards renewable energy as countries are making tremendous efforts to reduce their dependence on fossil fuels and curb climate change. Among renewable resources, solar energy stands out for its abundance, scalability, and accessibility. Solar photovoltaic (PV) technology has emerged as a key prospect in this shift, with ongoing innovations aimed at improving efficiency and lowering costs. Researchers are advancing solar performance by integrating existing PV technologies with novel materials, making high-efficiency solar power a key strategy for sustainable energy generation. In general, the evolution of solar PV technology can be grouped into three generations [1]. The first generation includes: crystalline silicon (c-Si), further classified into monocrystalline and polycrystalline types. Commercial monocrystalline modules typically exhibit 20–25% efficiency, while polycrystalline modules provide 18–21%, with recent research pushing monocrystalline silicon cell efficiency to 27.81% under laboratory conditions [2, 3]. Second-generation solar cells encompass a diverse class of thin-film technologies consisting of amorphous silicon (a-Si)[4], cadmium telluride (CdTe) [5], chalcogenide [6], and most importantly, group III-V materials [7]. A thoroughly studied III-V absorber, GaAs, with a direct bandgap of 1.42 eV, has demonstrated lab efficiencies up to 29.1% for single-junction devices [8], and even higher when used in multi-junction architectures. However, due to high production costs, III-V solar cells are typically reserved for niche applications such as aerospace and concentrator photovoltaics.

Third-generation solar cells represent a new wave of emerging photovoltaic technologies designed to overcome the efficiency and cost limitations

of first and second-generation devices. These technologies incorporate earth-abundant materials and structural innovations to enhance light absorption and charge separation. Notable examples of emerging photovoltaic technologies include plasmonic and metamaterials based solar cells, organic solar cells (OSCs), dye-sensitized solar cells (DSSCs), quantum dot solar cells (QDSCs), and perovskite solar cells (PSCs), as well as multi-junction tandem configurations [9–11]. These emerging photovoltaics possess unique advantages, notably mechanical flexibility, low-temperature solution-based fabrication process, semi-transparent operation, and compatibility with unconventional surfaces such as windows, fabrics, and portable electronic devices [12]. Yet, despite such positive aspects, third-generation devices face persistent hurdles like material instability, challenges with large-scale manufacturability, and interfacial degradation. PSCs are particularly vulnerable to environmental degradation and raise concerns about lead toxicity [13], whereas QDSCs struggle with surface traps, heavy-metal constituents, and long-term stability issues [14]. DSSCs are limited by electrolyte leakage and sealing problems [15], and OSCs suffer from lower efficiencies, rapid degradation, and morphological instability that impede large-area deployment [16]. Among third-generation cells, perovskite solar cells have emerged as frontrunners, reaching record efficiencies of 26.95% [8]. Perovskites adopt the ABX_3 crystal structure, where A is an organic or inorganic cation, B is a metal cation, and X is a halide. Perovskite materials offer a highly tunable bandgap ranging from 0.96 eV for cesium silver thallium bromide ($Cs_2AgTlBr_6$) to 2.82 eV for cesium lead chloride ($CsPbCl_3$), making them attractive for both single and multi-junction applications [17, 18]. Regardless of significant advances, single-junction solar cells remain constrained by the Shockley–Queisser theoretical efficiency limit, primarily due to excess carrier thermalization and sub-bandgap transmission losses. Multi-junction tandems mitigate these limitations by stacking absorbers with complementary bandgaps, and the compositional tunability of perovskites makes them particularly advantageous for achieving optimized spectral splitting and current matching.

For multi-junction tandem solar cells, the semiconducting materials employed must combine optimal bandgaps with high absorption coefficients, minimal recombination losses, and good lattice compatibility to suppress interface defects [19]. Dual-junction tandem structures such as III–V/Si [20], perovskite/Si [21], and all–III–V [22] or all–perovskite [23] material combinations were studied for their ability to surpass the Shockley–Queisser

limit of single-junction cells. More advanced triple-junction architectures, including perovskite/perovskite/Si [24] and III–V/III–V/Si [25] stacks, can further improve spectral utilization and reduce thermalization losses. Advancing beyond three junctions, devices with four or more junctions, typically composed of III–V materials, have demonstrated record-setting efficiencies. In 2020, Geisz *et al.* reported a six-junction III–V solar cell achieving 47.1% efficiency under 143 suns concentration through precise bandgap engineering and sophisticated epitaxial growth techniques [26]. Meanwhile, perovskite/Si tandem cells have attained over 30.6% efficiency in recent reports, offering a low-cost and scalable alternative to III–V systems [27]. All-perovskite tandem cells incorporating Sn-Pb-based narrow-bandgap absorbers have achieved 28.2% efficiency via improved grain passivation and interface quality [28]. In parallel, chalcogenide-based tandem solar cell architectures such as perovskite/CIGS [29] and perovskite/CZTS [30], have demonstrated considerable potential as sustainable, earth-abundant photovoltaic solutions. Nonetheless, multi-junction solar cells remain constrained by high fabrication complexity, current and lattice mismatch among sub-cells, and thermal-induced spectral instability under real-world illumination conditions. Moreover, the need for precise thickness and interface control during epitaxial or layer-by-layer growth imposes scalability challenges and cost barriers for industrial deployment [31]. Careful band alignment and interface engineering have pushed tandem perovskite devices to efficiencies near 30% [32]. Yet additional gains can be expected via targeted bandgap tuning, improved interface passivation, efficient optical management, and precise current matching among subcells [33]. Within the present framework, potassium tin iodide (KSnI_3) [34] and formamidinium tin triiodide (FASnI_3) [35] have emerged as two highly promising lead-free perovskite materials. Recent theoretical studies reported PCEs for single-junction KSnI_3 cells, reaching 23.85% [36]. In comparison, FASnI_3 -based devices have demonstrated even higher efficiency of 28.37% [37]. These findings point towards the possibility of coupling KSnI_3 and FASnI_3 absorbers in tandem configurations to extend device performance even further. On the chalcogenide front, silver-doped copper-zinc-tin-selenide (ACZTSe) stands out as an earth-abundant, non-toxic absorber with tunable bandgap values around 1.04–1.24 eV via the extent of Ag alloying [38]. All-chalcogenide tandem solar cells with ACZTSe as the bottom absorber attained PCEs of 24% in dual-junction and 36.04% in triple-junction designs [39]. Together, KSnI_3 with bandgap (~ 1.84 eV), FASnI_3 (1.3–1.4 eV), and ACZTSe (~ 1.0 eV) offer complementary optical

characteristics, making them ideal for multi-junction tandem cell integration. Their combined advantages position them as suitable contenders for advancing next-generation, high-efficiency, environmentally viable photovoltaic technologies.

In this study, we designed and assessed two novel lead-free tandem solar cells that adopted tin-based perovskite and kesterite materials, both renowned for their strong potential in prompting high-performance, non-toxic, and sustainable photovoltaic technology. Our initial proposed structure included an all-perovskite monolithic dual-junction tandem solar cell, incorporating KSnI_3 and FASnI_3 as absorber layers. Building on this architecture, a monolithic two-terminal triple-junction tandem architecture was originated, integrating KSnI_3 , FASnI_3 , and ACZTSe to enhance spectral utilization and overall device PCE. This research featured a comprehensive analysis of both the dual-junction ($\text{KSnI}_3/\text{FASnI}_3$) and triple-junction ($\text{KSnI}_3/\text{FASnI}_3/\text{ACZTSe}$) configurations. The investigation entailed detailed modeling and optimization of their respective single-junction subcells, followed by the sequential tuning of critical physical parameters, such as absorber layer thickness and doping densities, to maximize device performance. The insights derived from this work lay a foundation for the design and fabrication of high-efficiency next-generation multijunction solar cells with optimized interfacial and electronic properties.

2. Device architecture and simulation methodology

We designed and examined four distinct solar cell architectures based on KSnI_3 , FASnI_3 , and ACZTSe , encompassing single-junction (1-J), double-junction (2-J), and triple-junction (3-J) tandem configurations. The investigation expanded progressively from single-junction devices to fully integrated tandem configurations, with each contact and transport layer cautiously chosen in accordance with literature-guided performance and stability studies.

The first phase focused on designing and simulating a single-junction solar cell with an n/i/p configuration, utilizing KSnI_3 as the primary absorber, as shown in Fig. 1(a). With a direct bandgap of ~ 1.84 eV [34], KSnI_3 is well suited as a top absorber in tandem architectures. Fluorine-doped tin oxide (FTO) served as a transparent front contact owing to its high optical transmittance ($>85\%$), wide bandgap (~ 3.6 eV), and suitable work function

(4.4—4.7 eV) [40]. Titanium dioxide (TiO_2) in its anatase phase acted as an electron transport layer (ETL), offering a bandgap of 3.2 eV and favorable band alignment for electron transfer and hole blocking [36]. Copper(I) thiocyanate (CuSCN) functioned as the hole transport layer (HTL) with valence band (~ 5.3 eV), wide bandgap (~ 3.6 eV), high hole mobility, and strong optical transparency, enabling efficient hole extraction and suppressed recombination [38]. A thin magnesium fluoride (MgF_2) was introduced as an anti-reflection coating (ARC) to minimize reflection losses. For the back contact layer (BCL), both indium tin oxide (ITO) and gold (Au) were evaluated. ITO offers high transmittance and a suitable work function (4.7–5.0 eV), enabling near-ohmic contact and current continuity between stacked subcells [41].

Subsequently, another single-junction cell with an n/n+/p/p+ configuration, featured in Fig. 1(b), was modeled employing FASnI_3 as the absorber, with a direct bandgap of ~ 1.41 eV [35]. TiO_2 was retained as the ETL for consistency and compatibility, while a ZnO buffer layer was incorporated between TiO_2 and the absorber to improve electron transport. ZnO exhibits good mobility and favorable band alignment, although passivation may be required to reduce reactivity with iodide ions. For HTL, both CuSCN and Spiro-OMeTAD were examined. Spiro-OMeTAD, with a HOMO level of 5.1–5.3 eV, aligns closely with the FASnI_3 valence band (~ 5.2 eV), enabling efficient hole extraction and supporting open-circuit voltages above 0.9 V [38]. Au and ITO were also investigated as BCL to assess their electrical performance and optical transmittance.

After optimizing the single-junction devices, a double-junction (2-J) tandem solar cell was constructed by stacking the KSnI_3 cell as the top subcell over the FASnI_3 cell as the bottom subcell, as illustrated in Fig. 1(c). The same charge transport layers were retained, with TiO_2 serving as the ETL and CuSCN as the HTL. An ITO interlayer was introduced as the intermediate transparent conducting oxide (ITCO). Owing to its wide optical bandgap (3.5—4.3 eV), high visible transmittance, and strong electrical conductivity, ITO functions effectively as both a transparent electrode and a recombination layer [41]. To further improve band alignment and charge transfer, a ZnO buffer layer was incorporated between the TiO_2 ETL and the ITCO.

To extend the architecture further, a triple-junction (3-J) tandem so-

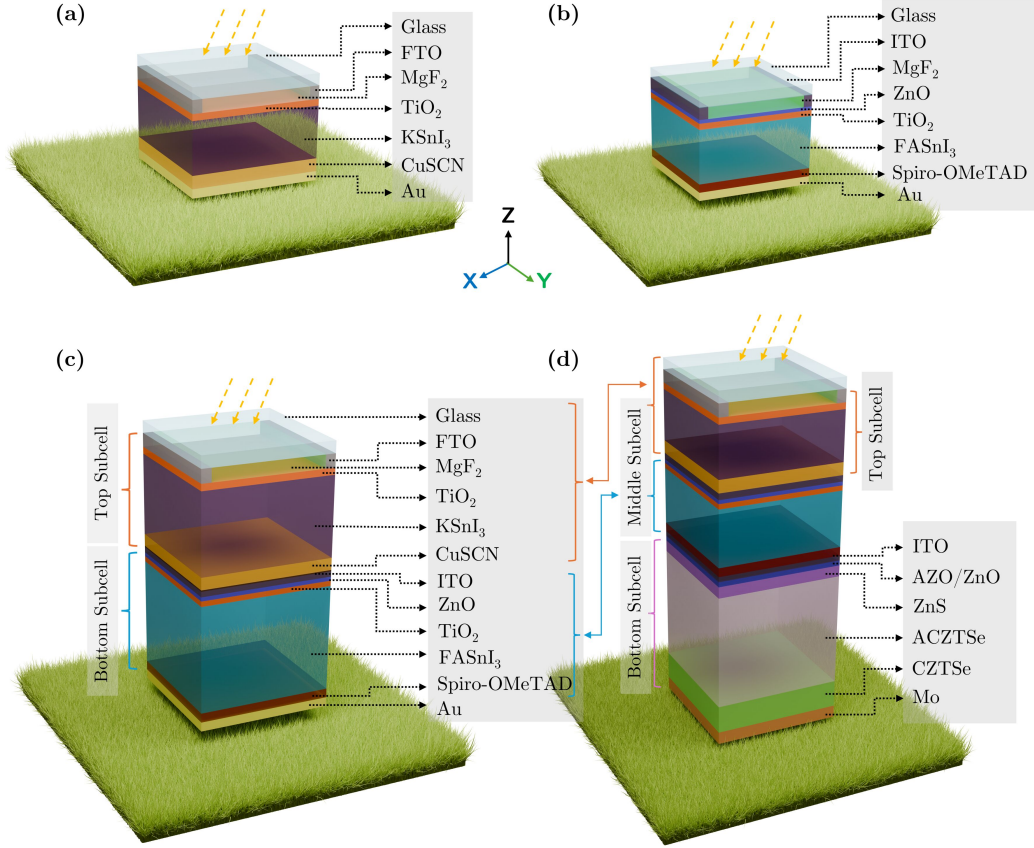


Figure 1: Device schematics of simulated modeled structures featuring: (a) 1-J KSnI_3 , (b) 1-J FASnI_3 , (c) 2-J monolithic $\text{KSnI}_3/\text{FASnI}_3$ tandem, and (d) 3-J monolithic $\text{KSnI}_3/\text{FASnI}_3/\text{ACZTSe}$ tandem solar cells.

lar cell was developed by introducing ACZTSe as the bottom absorber, as shown in Fig. 1(d). ACZTSe, a kesterite-type chalcogenide with a bandgap of 1.0 eV, exhibits high hole mobility and strong light absorption, with a valence band that aligns well with CZTSe. Zinc sulfide (ZnS) was employed as the ETL due to its wide bandgap (3.6—3.7 eV), suitable conduction band alignment, and high electron mobility (10—100 $\text{cm}^2/\text{V}\cdot\text{s}$), ensuring efficient electron extraction with minimal recombination [42]. A CZTSe back surface field (BSF) layer was incorporated below ACZTSe, leveraging its p-type conductivity, high hole mobility (7 $\text{cm}^2/\text{V}\cdot\text{s}$), and slightly deeper valence band to create a favorable gradient for hole transport while blocking electrons [43]. For the rear electrode, molybdenum (Mo) was selected owing to its

excellent conductivity, work function alignment, and infrared photon reflectivity, which enhances light harvesting [44]. Between the perovskite middle subcell (FASnI₃) and the kesterite bottom subcell (ACZTSe), a ZnO/AZO bilayer was introduced as a buffer and tunneling layer. Aluminum-doped ZnO (AZO) provides low resistivity, high transmittance, and chemical stability, while undoped ZnO improves energy alignment with ZnS and suppresses sulfur/selenium interdiffusion [45, 46]. Together, this bilayer supports efficient carrier extraction while preserving optical and structural integrity. This progressive simulation strategy, from single-junction cells to 3-J tandem structures, ensured optimized charge extraction, reduced interfacial recombination, and current matching across all subcells, with each layer chosen for compatibility, stability, and scalability.

We applied a hybrid optical-electrical simulation framework to design and optimize new proposed tandem solar cell architectures. The finite difference time domain (FDTD) was implemented to examine the optical behavior, whereas the finite element method (FEM) was employed to investigate the electrical performance behavior. Post-processing was conducted to interpret and analyze the results.

To analyze the optical responses of the proposed solar cell architectures, the FDTD method solves Maxwell’s wave equations to capture electromagnetic interactions within the device layers. FDTD was employed for its broadband capability, high accuracy, and extensive computational flexibility. For optical modeling, the complex refractive indices ($n+i\kappa$) of the constituent materials are utilized as optical input parameters, obtained from reported literature [34, 39, 47–54]. For modeling the proposed tandem cells in 2D, a periodic boundary condition was applied along the X-axis, while a perfectly matched layer (PML) was used along the Y-axis to prevent artificial reflection at the simulation boundary. The standard AM1.5G solar spectrum is incident along the Y-axis as an illumination source, covering a photon wavelength λ_{photon} ranging from 300 to 1200 nm. However, to exclude contributions from parasitic absorption and intra-band transitions, the photo-generation rate (G) was evaluated solely within the spectral range corresponding to inter-band optical transitions relevant to the solar cell architectures.

For electrical stimulation, key performance parameters, such as PCE (η), short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), and fill factor

(FF), were evaluated using the FEM method. The electrical simulations were performed by self-consistently solving the Poisson and drift-diffusion equations in two dimensions. Dirichlet boundary conditions were imposed at the metal-semiconductor interfaces, and Neumann conditions were used at the periodic boundaries. The electrical parameters used for theoretical calculations and performing the simulations for absorbers, transport layers, and contacts were extracted from prior studies [34, 35, 37, 39, 52, 55–64] and are summarized in Tables 1 and S1 of the Supplementary Material.

For parametric optimization of the monolithic tandem configurations, current matching was strictly enforced across all subcells to extract key photovoltaic performance parameters such as η , J_{sc} , V_{oc} , and FF, ensuring realistic performance evaluation. The definitions and governing equations for these parameters are detailed in the Supplementary Material. A complete workflow, from single-junction modeling to the final triple-junction configuration, is outlined in Fig. 2. The optimization workflow proceeded hierarchically. First, a single-junction cell was modeled, and parametric sweeps were performed until convergence to optimize its parameters. Each layer of the single-junction was then individually refined using the same approach, followed by an overall optimization of the full single-junction considering necessary trade-offs among performance metrics. Once all single-junctions were optimized, dual-junction cells were constructed and optimized with careful consideration of current matching between subcells. Finally, this strategy was extended to triple-junction architectures, applying the same layer and device-level opti-

Table 1: Electrical parameters of absorber materials (KSnI₃, FASnI₃, ACZTSe and CZTSe).

Parameters	KSnI ₃ [34, 58]	FASnI ₃ [35, 37, 59, 60]	ACZTSe [39, 61, 62]	CZTSe [39, 61]
Thickness (nm)	100–800	100–800	400–1400	50–300
Bandgap E_g (eV)	1.84	1.4	1.088	1.06
DC permittivity ϵ	10.4	8.2	8.5	7
Electron affinity χ (eV)	3.44	3.9	4.05	4.05
Mobility μ_n/μ_p (cm ² /Vs)	21.28 / 19.46	22 / 22	75 / 10	145 / 35
SRH lifetime τ_e/τ_h (ns)	7.78 / 8.135	7.93 / 7.93	1.79 / 1.79	0.52 / 0.52
Radiative recombination, $B_{n,p}$ (cm ³ s ⁻¹)	1.1×10^{-10}	4.37×10^{-10}	3.5×10^{-10}	1.08×10^{-10}
Auger recombination, $C_{n,p}$ (cm ⁶ s ⁻¹)	1×10^{-30}	9.3×10^{-30}	1×10^{-28}	1×10^{-28}
Effective conduction band density, N_C (cm ⁻³)	1×10^{18}	1.0×10^{18}	2.51×10^{19}	2.2×10^{18}
Effective valence band density, N_V (cm ⁻³)	1×10^{18}	1.0×10^{18}	1.79×10^{19}	1.8×10^{19}
Donor doping N_D (cm ⁻³)	1×10^{15}	–	–	–
Acceptor doping N_A (cm ⁻³)	1×10^{15}	1×10^{16}	1×10^{16}	5×10^{18}
Surface Recombination (cm/s)	–	–	–	(CZTSe/Mo) 1×10^7

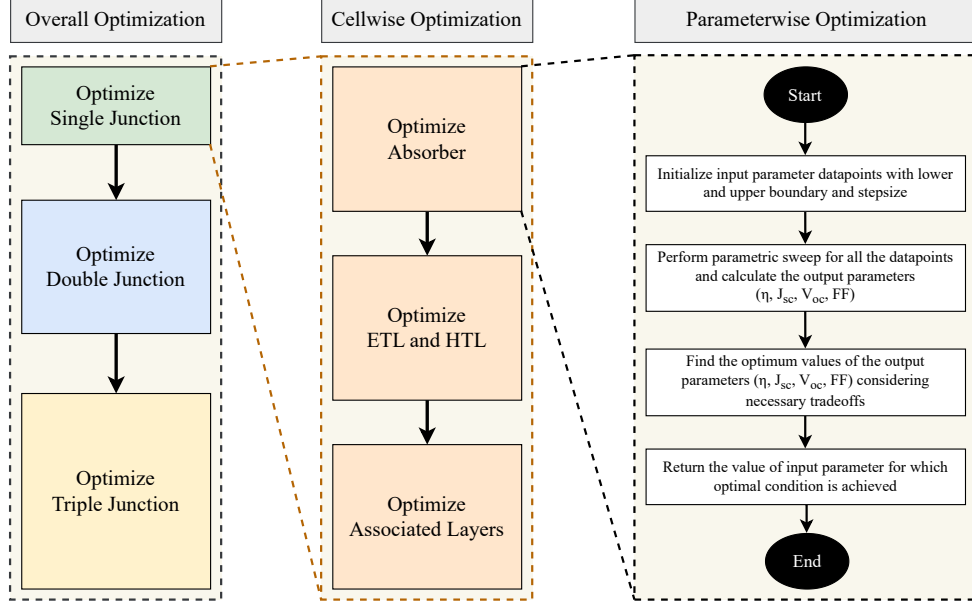


Figure 2: Sequential optimization of the modeled architectures from single junction cells to triple junction tandem cells.

mization while maintaining spectral complementarity and current matching across all subcells.

3. Results and discussion

3.1. Structural refinement and light-harvesting in 1-J KSnI_3 cells

To model the $\text{MgF}_2/\text{FTO}/\text{TiO}_2/\text{KSnI}_3/\text{CuSCN}/\text{ITO}$ configuration as an n/i/p single-junction solar cell architecture, as illustrated in Fig. 1(a), we considered MgF_2 as ARC, FTO as TCO, TiO_2 as ETL, KSnI_3 as absorber, CuSCN as HTL, and ITO as BCL with initial thickness of 100 nm, 100 nm, 90 nm, 200 nm, 50 nm, and 50 nm, respectively. Initially, the doping densities of ETL, absorber, and HTL were set to 5×10^{18} , 1×10^{15} , and $1 \times 10^{18} \text{ cm}^{-3}$, respectively. To establish the baseline device behavior, we performed preliminary thickness sweeps of the KSnI_3 absorber, TiO_2 ETL, and CuSCN HTL to identify performance-sensitive regions and define the parameter space for subsequent detailed optimization. The corresponding results are provided in Figs. S2 and S3 of the Supplementary Material.

Afterward, following the self-consistent optimization process as featured in Fig. 2, final iterative thickness optimization of the KSnI_3 absorber, TiO_2 ETL, and CuSCN HTL were performed to maximize device performance. First, the thickness of KSnI_3 was varied from 200 to 1200 nm while keeping the ETL and HTL thicknesses at 40 and 100 nm, respectively, as shown in Fig. 3(a). PCE increased steadily from 11.32% at 200 nm to a peak of 12.95% at 700 nm, after which further thickness increases led to marginal decreases in efficiency. This trend reflects the trade-off between enhanced photon absorption and increased carrier recombination in a thicker KSnI_3 absorber. J_{sc} increased with thickness due to improved light photon absorption in the 600–700 nm wavelength range, whereas V_{oc} and FF decreased slightly, indicating higher recombination and series resistance effects. This trend is observed in Figs. S4(b-d). Subsequently, a parametric sweep of the TiO_2 ETL (20–160 nm) and CuSCN HTL (60–160 nm) was conducted with the absorber thickness fixed at 700 nm as shown in Figs. 3(c-f). Analysis revealed that, a 40 nm thickness of TiO_2 and a 100 nm thickness of CuSCN yielded a peak PCE of 12.95%, with J_{sc} of 15.28 mA/cm^2 , V_{oc} of 1.247 V, and FF of 67.94%. Reducing the ETL thickness weakened the built-in electric field, thereby limiting electron extraction, while excessive thickness introduced additional series resistance and hindered charge transport. Similarly, optimizing the HTL thickness proved essential for maintaining efficient hole transport without compromising carrier collection at the interface. These findings highlight the importance of carefully balancing layer thicknesses to achieve optimal device performance.

We then explored doping density variations in the transport layers. The donor density (N_{D}) of TiO_2 and acceptor density (N_{A}) of CuSCN were varied from 5×10^{16} to $5 \times 10^{19} \text{ cm}^{-3}$ with the results visualized in Figs. S5(a-d). The highest PCE of 14.32% was achieved for N_{D} of $5 \times 10^{19} \text{ cm}^{-3}$ in TiO_2 and N_{A} of $2 \times 10^{19} \text{ cm}^{-3}$ in CuSCN , corresponding to J_{sc} of 15.29 mA/cm^2 , V_{oc} of 1.234 V, and FF of 75.86%. Higher donor doping in TiO_2 enhanced the built-in electric field, promoting electron extraction and reducing recombination at the absorber/ETL interface. Similarly, increasing acceptor doping in CuSCN improved hole transport; however, acceptor density exceeding 10^{19} cm^{-3} introduced significant defect-assisted recombination, limiting further efficiency gains [65]. Limiting N_{A} to $5 \times 10^{18} \text{ cm}^{-3}$ still yielded a comparable PCE of 14.31%, indicating the device’s robustness. Following that, we varied the thickness of MgF_2 ARC from 20-150 nm to reduce the reflection loss. A

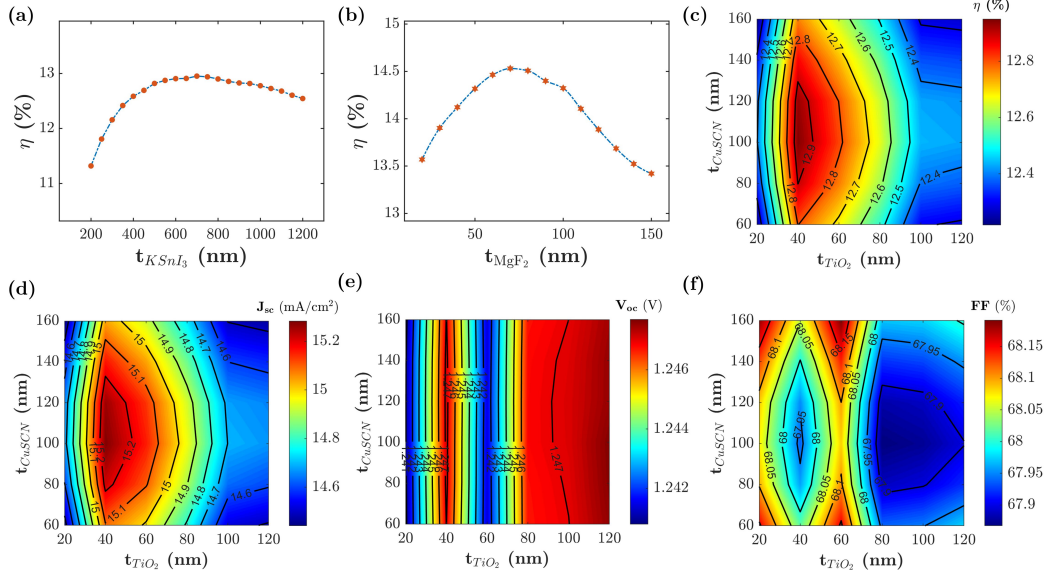


Figure 3: (a) Impact on PCE (η) of the 1-J KSnI₃ solar cell corresponding to the absorber thickness variation, presented for the final KSnI₃ thickness iterative sweep. (b) Influence of MgF₂ ARC thickness on PCE. Contour plots of key performance metrics (c) η , (d) J_{sc} , (e) V_{oc} , and (f) FF as functions of ETL (TiO₂) and HTL (CuSCN) thicknesses after final parametric sweeping in single junction KSnI₃ cell.

maximum PCE of 14.52% was achieved, as shown in Fig. 3(b). The optimal ARC thickness of 70 nm resulted in an average normalized reflection (R) of 4.45%, enhancing photon absorption in the 300–700 nm wavelength range, as highlighted in Fig. S6(a). Subsequently, a volumetric thicknesses sweep of absorber, ETL, and HTL confirmed an optimum PCE of 14.53% for absorber, ETL, and HTL thicknesses of 700 nm, 40 nm, and 100 nm, respectively, as shown in Fig. 4(a). This analysis demonstrates that, a self-consistent optimization of layer thicknesses and doping densities converges to the global performance maximum, highlighting the robustness and predictive accuracy of the design approach.

To reduce the interfacial band offset at the KSnI₃/TiO₂ junction, UV-ozone-treated TiO₂ was employed, shifting the TiO₂ work function (ϕ_{TiO_2}) from 5.3 eV (oxidized TiO₂ film) to 5.1 eV. Klasen *et al.* reported that UV-ozone treatment significantly increased the surface conductance of TiO₂ films by over two orders of magnitude, mitigated surface oxygen vacancies,

and enhanced charge extraction, resulting in a 2% increase in the average PCE of perovskite solar cells [66]. Implementing this treatment in the 1-J KSnI₃ device gained an improved PCE of 15.30%, as presented in Table S2. Next, instead of ITO as BCL, 50 nm Au BCL was utilized to further boost the single junction performance by achieving further absorption through back reflection in the wavelength range of 550–690 nm, as shown in Fig. S6(b). This additional absorption enhanced J_{sc} from 15.61 to 16.62 mA/cm². The final optimum PCE (η) was found 16.28% with J_{sc} of 16.62 mA/cm², V_{oc} of 1.23V, and FF of 79.50%.

Table 2 summarizes the gradually improved results from the initial to the final optimized structure of the 1-J KSnI₃ cell. Spatial generation rate profile, G across the X-Y cross-section, highlighting the maximum carrier generation regions in Fig. 4(b), where KSnI₃ spans from 150 to 850 nm (thickness 700

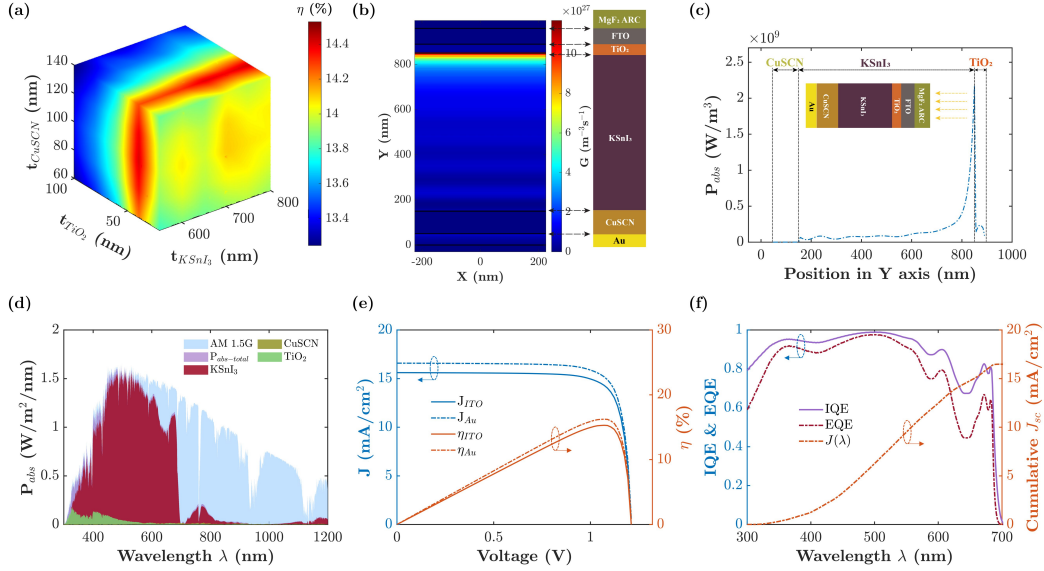


Figure 4: (a) Impact on PCE (η) through altering KSnI₃, ETL, and HTL thickness under different configurations. (b) Spatial carrier generation rate profile, G , highlighting the regions of maximum generation. (c) Absorbed power P_{abs} towards the illumination of sunlight in the Y direction. (d) Spectral power absorption of different layers 1-J KSnI₃ cell versus photon wavelength corresponding to AM 1.5G solar spectrum. (e) Comparison between J-V and P-V plot for incorporating Au and ITO as BCL. (f) Internal and external quantum efficiency (IQE, EQE) along with cumulative J_{sc} vs photon wavelength of the optimized 1-J KSnI₃ cell.

Table 2: Summary of the stepwise performance improvement of the single-junction KSnI₃ solar cell.

Optimization parameters	η (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)
Layers thickness - TiO ₂ /KSnI ₃ /CuSCN (40 nm/ 750 nm/ 100 nm)	12.95	15.28	1.247	67.94
ETL/HTL doping (ETL: 5×10^{19} cm ⁻³ , HTL: 5×10^{18} cm ⁻³)	14.31	15.29	1.234	75.86
MgF ₂ ARC thickness (70 nm)	14.53	15.62	1.235	75.37
UV-ozone treated TiO ₂ ETL (Work function: 5.1 eV)	15.30	15.62	1.228	79.79
Substitution of Au BCL with ITO (50 nm)	16.28	16.62	1.232	79.50

nm). Fig. 4(c) highlights absorbed power in different regions of the single junction cell corresponding to the direction of the sunlight in the Y direction. The spectral power absorption, P_{abs} and normalized IQE and EQE, along with cumulative J_{sc} as functions of photon wavelength λ , are presented in Figs. 4(d) and (f) respectively. For KSnI₃, photon energies above the optical bandgap of 1.84 eV effectively drive inter-band transitions, generating free electron-hole pairs that contribute to photocurrent. Beyond this threshold, photon energy becomes inadequate to induce inter-band transitions, resulting in weak optical absorption and the rapid non-radiative recombination of short-lived carriers, which therefore remain inactive in photovoltaic operation. Finally, replacing ITO with Au as the BCL significantly enhances device performance, yielding a relative PCE improvement of 6.4%, as evidenced by Table 2 and the J-V and η -V characteristics shown in Fig. 4(e).

3.2. Optoelectronic tuning and performance mapping of 1-J FASnI₃ cells

For modeling and optimization of the single junction (1-J) FASnI₃ solar cell, we employed two distinct hole transport layers, CuSCN and Spiro-

OMeTAD, to investigate their respective impacts on device performance along with other layers.

3.2.1. Incorporating CuSCN as hole transport layer

To model and perform opto-electronic simulation for 1-J FASnI₃ cell (MgF₂/ITO/ZnO/FASnI₃/CuSCN/ITO), the same optimization procedures described previously were followed. We modeled the initial architecture with MgF₂ (100 nm) as ARC, front ITO (100 nm) as TCO, ZnO (10 nm) as buffer layer, and TiO₂ (90 nm) as ETL with donor doping densities of 1.5×10^{17} and 5×10^{18} cm⁻³, respectively. FASnI₃ absorber (200 nm) was set to an acceptor doping density of 1×10^{16} cm⁻³, and CuSCN (50 nm) was used as HTL with an acceptor doping of 1×10^{18} cm⁻³. The bottom ITO (50 nm) acted as BCL. Preliminary thickness sweeps of the FASnI₃ absorber, TiO₂ ETL, and CuSCN HTL were performed to identify performance-sensitive regions and establish the baseline for subsequent optimization, as shown in Figs. S7 and S8 of the Supplementary Material.

After identifying the performance-sensitive region, we systematically varied the thicknesses of TiO₂, FASnI₃, and CuSCN to achieve self-consistent device performance, as depicted in Figs. 5(a) and Figs. S9(a–d). The absorber thickness was swept in the range of 200–1200 nm, during which the PCE gradually increased and reached a peak value of 21.88% at a thickness of 800 nm. The improvement with increasing thickness is primarily attributed to enhanced optical absorption and photo-carrier generation within the FASnI₃ layer. Beyond 800 nm, PCE exhibited a slight saturation, reflecting the competing effect of increased recombination losses once the absorber thickness exceeded the carrier diffusion length. Thickness sweeps of the TiO₂ ETL from 20 to 80 nm and CuSCN HTL from 140 to 200 nm yielded nearly invariant PCE, as Fig. S9 of Supplementary Material, indicating that both transport layers were sufficiently optimized to ensure effective charge extraction without introducing additional series resistance and optical interference. The best PCE of 21.88% was achieved for a 20 nm TiO₂ and a 160 nm CuSCN layer, confirming the robustness of the device against minor geometrical variations under constant doping conditions.

Subsequently, we tuned the doping densities of the ETL and HTL over a range of 1×10^{18} – 5×10^{19} cm⁻³, as shown in Figs. 5(c–f). Enhanced donor and acceptor doping densities improved the built-in potential and reduced

contact resistance, resulting in superior carrier selectivity and a higher fill factor. The highest efficiency of 22.05% was obtained for N_D of $5 \times 10^{19} \text{ cm}^{-3}$ for TiO_2 and N_A of $5 \times 10^{19} \text{ cm}^{-3}$ for CuSCN . However, considering interfacial stability, a moderate acceptor doping of $8 \times 10^{18} \text{ cm}^{-3}$ was selected for CuSCN , as it provided nearly identical efficiency with improved long-term reliability. Finally, N_A of absorber was varied between 10^{15} and 10^{17} cm^{-3} , revealing a clear optimum performance at $7 \times 10^{16} \text{ cm}^{-3}$. In this regime, the enhanced internal electric field strengthened charge separation and reduced interfacial

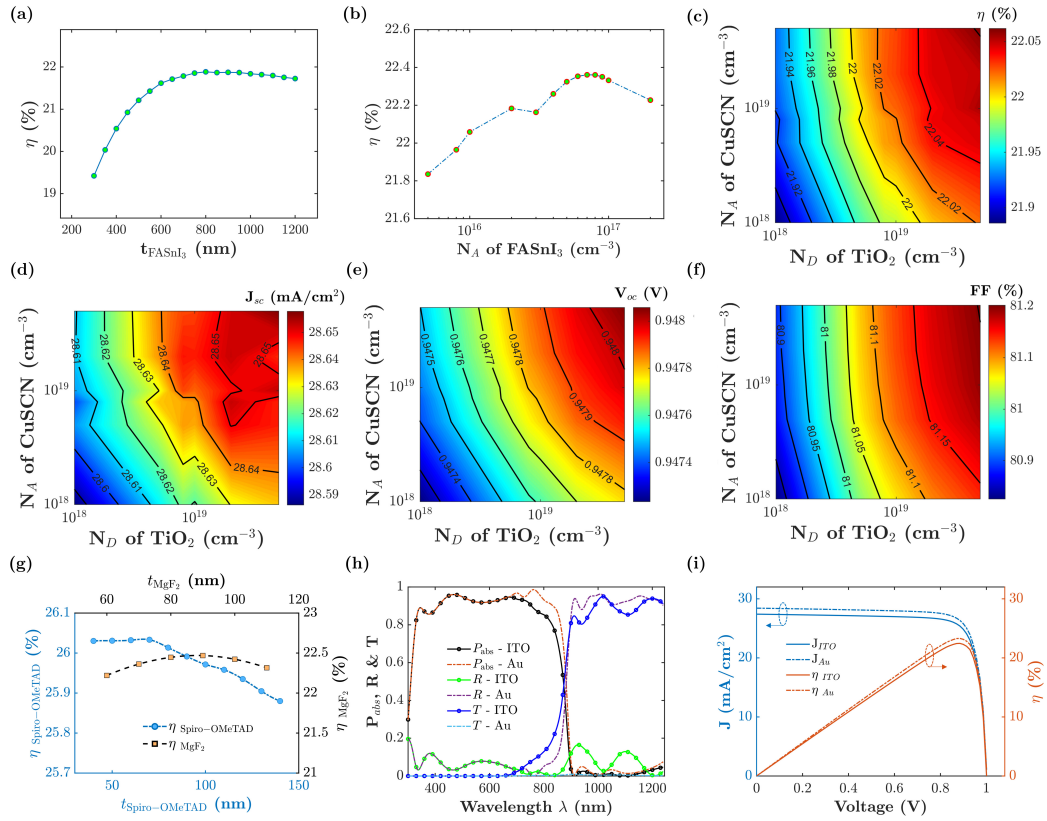


Figure 5: Impact on PCE (η) through altering (a) FASnI₃ absorber thickness and (b) FASnI₃ acceptor doping density N_A . Contour plot featuring performance matrices (c) η , (d) J_{sc} , (e) V_{oc} , and (f) FF corresponding to variation of donor doping density, N_D of TiO₂ ETL and acceptor doping density, N_A of CuSCN HTL for 1-J FASnI₃ cell. (g) Impact of MgF₂ ARC and Spiro-OMeTAD thickness on PCE of 1-J FASnI₃ cell. (h) Comparison between featuring normalized power absorption, reflection, and transmission corresponding to photon wavelength (i) J-V and η -V response for 1-J FASnI₃ with ITO and Au configurations.

recombination, yielding a peak PCE of 22.36%. To minimize optical reflection and maximize photon absorption, MgF_2 ARC thickness was systematically varied from 60 to 110 nm. The optimal PCE of 22.45% was found at 90 nm, as shown in Fig. 5(g), corresponding to the lowest overall reflectance within the device's active absorption range. Substituting ITO BCL with a 50 nm Au layer further enhanced the PCE to 23.31%, representing a relative 3.74% improvement over the ITO-based configuration, as evident from the J–V and η –V characteristics of Fig. 5(i). This enhancement stems from Au's higher conductivity and favorable energy alignment, which promote efficient carrier extraction and lower interfacial resistance. As shown in Fig. 5(h), Au BCL also suppressed reflection and improved near infrared (NIR) absorption in the 700–890 nm wavelength range, increasing J_{sc} by 1.01 mA/cm^2 , as summarized in Table 3. These combined optical and electrical enhancements demonstrate that the use of Au as BCL provides a more favorable balance between photon management and charge transport efficiency in the FASnI_3 device.

Table 3: Sequential optimization of layer thickness, doping, and interfaces in the 1-J FASnI_3 solar cell.

Optimization parameters	η (%)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)
Optimized $\text{TiO}_2/\text{FASnI}_3/\text{CuSCN}$ thickness (20 nm/800 nm/160 nm)	21.88	28.59	0.947	80.83
Optimized doping in $\text{TiO}_2/\text{FASnI}_3/\text{CuSCN}$ ($N_{\text{D},\text{TiO}_2}$: $5 \times 10^{19} \text{ cm}^{-3}$, $N_{\text{A},\text{FASnI}_3}$: $7 \times 10^{16} \text{ cm}^{-3}$, $N_{\text{A},\text{CuSCN}}$: $8 \times 10^{18} \text{ cm}^{-3}$)	22.36	27.28	1.001	81.89
Optimized MgF_2 ARC thickness (90 nm)	22.47	27.40	1.001	81.92
Substituting ITO BCL with Au layer (50 nm)	23.31	28.42	1.002	81.90
Integrated Spiro-OMeTAD as HTL (70 nm)	26.03	29.80	1.029	84.84

3.2.2. Impact of Spiro-OMeTAD for hole extraction

To enhance the effective hole transportation in the FASnI₃/HTL interface, we incorporated Spiro-OMeTAD instead of CuSCN as HTL in 1-J FASnI₃ cell. The thickness of Spiro-OMeTAD was varied from 40 to 140 nm under a constant acceptor doping density of $8 \times 10^{18} \text{ cm}^{-3}$. The PCE showed a steady increase with Spiro-OMeTAD thickness, reaching a maximum of 26.03% at 70 nm. Beyond this point, additional thickness led to a decline in performance due to an increase in carrier recombination, as shown in Fig. 5(g). Fig. S10 of the Supplementary Material presents the band diagrams of the 1-J FASnI₃ cell at short-circuit ($V = 0 \text{ V}$) and maximum power point ($V = V_{\text{mpp}}$) conditions for both CuSCN and Spiro-OMeTAD configurations. Analyzing Figs. S10(a) and (c), it is evident that the FASnI₃/Spiro-OMeTAD interface exhibits more favorable energy level alignment compared to the FASnI₃/CuSCN interface. Specifically, Spiro-OMeTAD introduces a slight

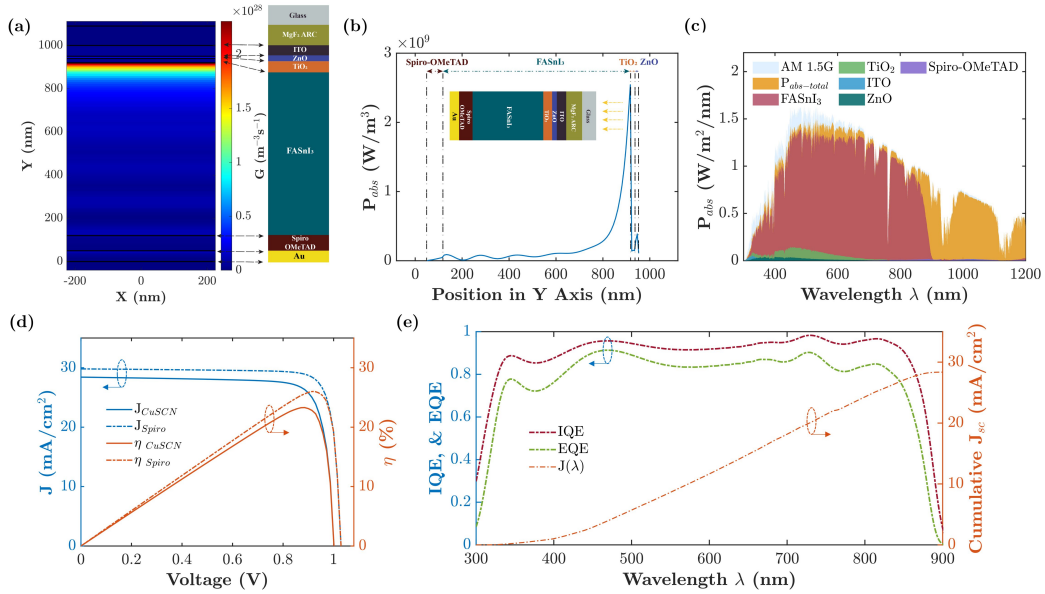


Figure 6: (a) Spatial carrier generation rate profile, G in X-Y cross-section, highlighting the region of maximum generation. (b) Absorbed power, P_{abs} , towards the illumination of sunlight in the Y-direction. (c) Spectral power absorption, P_{abs} of individual layers in the 1-J FASnI₃ solar cell as a function of photon wavelength under the AM1.5G illumination spectrum. (d) Comparison between J - V and η - V curves for incorporating CuSCN and Spiro-OMeTAD as HTL. (e) Response of IQE, EQE, and cumulative J_{sc} vs photon wavelength for the optimized 1-J FASnI₃ cell.

negative valence band offset (VBO) ≈ -0.06 eV, forming a downhill potential that facilitates hole drift from the absorber to the HTL, while maintaining a sufficient conduction band offset (CBO) ≈ 1.56 eV, to block electron back-injection. In contrast, CuSCN provides a nearly flat VBO ≈ 0 eV, offering minimal driving force for hole transport despite its higher CBO ≈ 2.0 eV, which restricts charge extraction efficiency.

Furthermore, the difference in layer thickness amplifies the effect of carrier drift and diffusion. The thinner 70 nm Spiro-OMeTAD layer supports a stronger built-in electric field across the FASnI₃/HTL junction, promoting efficient hole drift while minimizing diffusion-induced recombination. Conversely, the thicker 160 nm CuSCN layer imposes higher series resistance and longer diffusion paths, causing significant carrier loss and reduced hole mobility. The combined influence of favorable band alignment, stronger built-in potential, and reduced transport resistance in Spiro-OMeTAD leads to more pronounced band bending and quasi-Fermi level splitting, as evident in Figs. S10 (c) and (d).

Table 3 shows the gradual improvement of the performance of the 1-J FASnI₃ cell. Incorporation of Spiro-OMeTAD with FASnI₃ enhanced the PCE by 2.72%. Therefore, we considered FASnI₃ with Spiro-OMeTAD configuration as the final optimized 1-J FASnI₃ cell. The spatial generation rate profile G, highlighting the region of maximum carrier generation in Fig. 6(a). In Fig. 6(b) features the absorbed power density, P_{abs} in different layers of 1-J FASnI₃. Spectral power absorption in different layers corresponding to AM 1.5G solar spectrum, comparison between η -V and J-V for CuSCN and Spiro-OMeTAD as HTL, and normalized IQE, EQE with cumulative J_{sc} with respect to photon wavelength of optimized 1-J FASnI₃ with Spiro-OMeTAD configuration are illustrated in Figs. 6(c-e). Together, these outcomes offer a detailed understanding of the optical and electrical behavior of the optimized 1-J FASnI₃ cell.

3.3. Tandem integration and current matching in 2-J KSnI₃/FASnI₃ cells

To model and optimize the dual-junction monolithic tandem solar cell, we constructed the top subcell as MgF₂/FTO/TiO₂/KSnI₃/CuSCN/ITO, where ITO served as the ITCO and recombination layer, and the bottom subcell as ZnO/TiO₂/FASnI₃/Spiro-OMeTAD/Au. Initially, we fixed the top subcell absorber KSnI₃ thickness at 700 nm, along with a 40 nm ETL and 100 nm

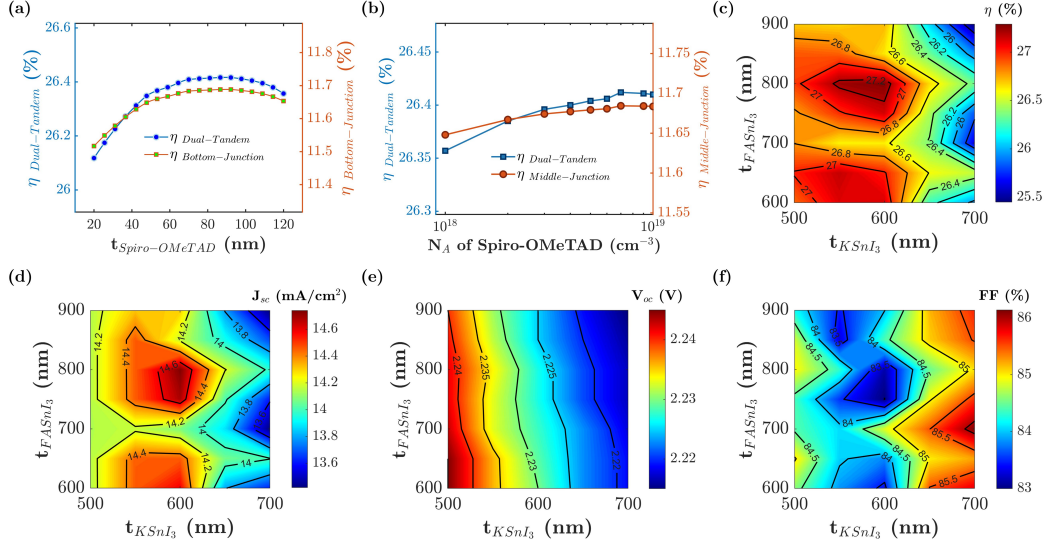


Figure 7: Impact on PCE (η) of 2-J KSnI₃/FASnI₃ tandem cell by (a) altering Spiro-OMeTAD HTL (bottom cell) layer's thickness and (b) acceptor doping density. Contour plot of parametric sweep featuring performance metrics – (c) η , (d) J_{sc} , (e) V_{oc} , and (f) FF corresponding to variation of TiO₂ ETL and CuSCN HTL thickness.

HTL, while the ITO recombination layer was 50 nm thick. The doping profile of the top subcell followed the optimized parameters from the single-junction KSnI₃ cell. For the bottom subcell in the 2-J tandem configuration, we varied the Spiro-OMeTAD thickness and acceptor doping density, as shown in Figs. 7(a) and (b). The highest PCE of 26.415% was achieved for a Spiro-OMeTAD thickness of 80 nm and an acceptor density N_A of $7 \times 10^{18} \text{ cm}^{-3}$, demonstrating the crucial role of hole transport layer optimization in facilitating efficient carrier extraction via drift and diffusion mechanisms.

To further maximize the performance, we systematically varied the absorber thickness of the top and bottom subcells within 500–700 nm and 600–900 nm ranges, respectively, as featured in Figs. 7(c–f). Observing Figs. 7(c) and (d), both η and J_{sc} attain higher values for KSnI₃ thicknesses of 550–600 nm and FASnI₃ thicknesses of 700–850 nm, indicating minimal current mismatch and efficient electron-hole transport across the ITCO/recombination interface. This behavior is consistent with reduced bulk and interfacial recombination due to optimal drift-driven carrier extraction in the built-in electric fields of each subcell. Although V_{oc} is higher for KSnI₃ thicknesses of 500–550 nm and FASnI₃ thicknesses of 600–800 nm, in the two-terminal (2-T) monolithic

tandem configuration, the overall V_{oc} is predominantly governed by the bottom FASnI₃ subcell. Increasing the KSnI₃ thickness beyond this range leads to a gradual decrease in V_{oc} , attributable to enhanced bulk recombination and series resistance effects. Meanwhile, FF increases with thicker KSnI₃ and FASnI₃ layers, as seen in Fig. 7(f). The optimized 2-J KSnI₃/FASnI₃ tandem cell achieves a PCE of 27.29%, with J_{sc} of 14.74 mA/cm², V_{oc} of 2.227 V, and FF of 83.14%.

Table 4 presents the progressive enhancement in the performance of the two-terminal (2-T) monolithic tandem solar cell, achieved through systematic optimization of the absorber layer thicknesses in each subcell. Fig. 8(a-f) provides a detailed optoelectronic analysis of the optimized tandem cell. Figs. 8(a-b) feature the carrier generation rate, highlighting the maximum generation regions, and absorbed power density along the device thickness under the illumination of sunlight in the Y direction. Fig. 8(c) compares the spectral absorbed power of the individual subcells and the complete 2-J tandem cell with the AM 1.5G solar spectrum. The complementary absorption of KSnI₃ and FASnI₃ enables efficient spectrum splitting from 300 to 900 nm, maximizing photon utilization and minimizing optical losses in the tandem stack. Fig. 8(d) presents the normalized IQE and EQE vs. photon wavelength of both subcells. The broad and overlapping IQE-EQE responses confirm strong carrier generation and collection across the respective absorption regions, with smooth spectral transitions indicating balanced cur-

Table 4: Summary of performance metrics of the 2-J KSnI₃/FASnI₃ tandem cell as a function of thickness and doping density variations in the bottom subcell.

Solar cell	Parameters	η (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	P_{mpp} (W/m ²)	V_{mpp} (V)
1-J FASnI ₃ cell	Spiro-OMeTAD (80 nm and 7×10^{18} cm ⁻³)	26.03	29.81	1.029	84.84	260.33	0.96
2-J tandem cell	Spiro-OMeTAD (Thickness: 80 nm)	26.41	13.98	2.217	85.26	264.14	1.994
2-J tandem cell	Spiro-OMeTAD (Doping: 7×10^{18} cm ⁻³)	26.41	13.98	2.217	85.24	264.14	1.994
2-J tandem cell	KSnI ₃ and FASnI ₃ (Thickness: 600 nm and 800 nm)	27.29	14.74	2.227	83.14	272.89	1.981

rent matching between the top and bottom subcells as depicted in Fig. 8(e). These results collectively validate efficient optical coupling and carrier extraction within the optimized 2-J tandem architecture. Additionally, Fig. 8(f) includes η -V characteristics of the top and bottom subcell following the optimized 2-J tandem cell. These analyses collectively elucidate the interplay of optical absorption, drift-diffusion-driven carrier transport, and interfacial recombination, providing a detailed understanding of the superior performance of the optimized monolithic 2-J tandem architecture.

3.4. Triple-junction architecture: layer-wise optimization and device efficiency

Before building the triple-junction $\text{KSnI}_3/\text{FASnI}_3/\text{ACZTSe}$ tandem cell, we introduced ITO as the ITCO and BCL in the middle FASnI_3 subcell to facilitate the efficient passage of excess low-energy photon injection to the bottom cell. This modification improved optical transmission across the

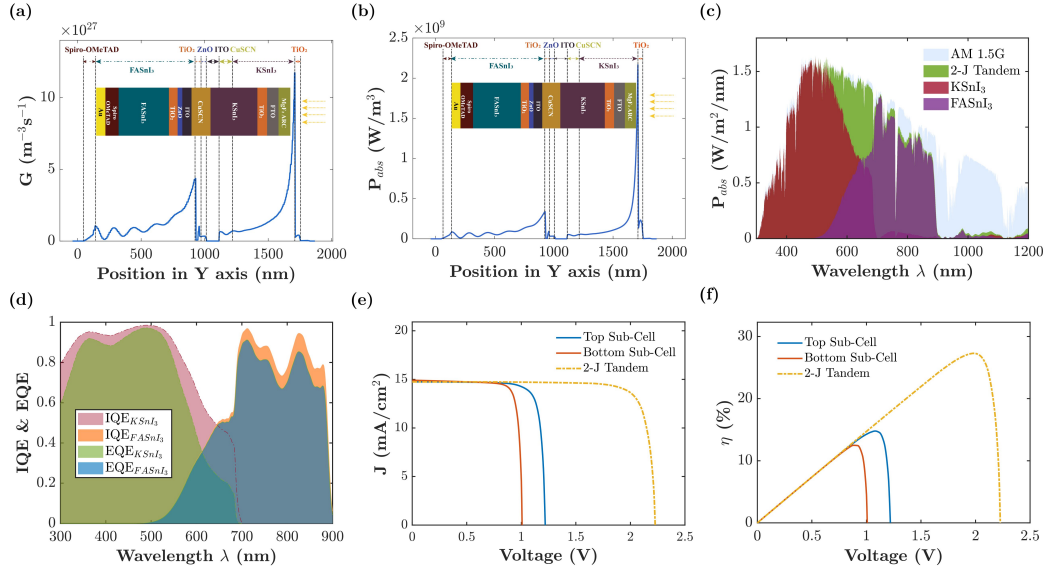


Figure 8: (a) Carrier generation rate, G , towards the illumination of sunlight in the Y direction, highlighting the maximum carrier generation regions. (b) Absorbed power density, P_{abs} , of different layers in a 2-J tandem cell towards the illumination of sunlight in the Y direction. (c) Spectral power absorption of the absorber layers of a 2-J tandem cell as a function of photon wavelength corresponding to the AM 1.5G spectrum. (d) IQE and EQE vs photon wavelength, (e) J-V, and (f) η -V plots of top, and bottom subcells, along with the optimized 2-T $\text{KSnI}_3/\text{FASnI}_3$ based tandem cell.

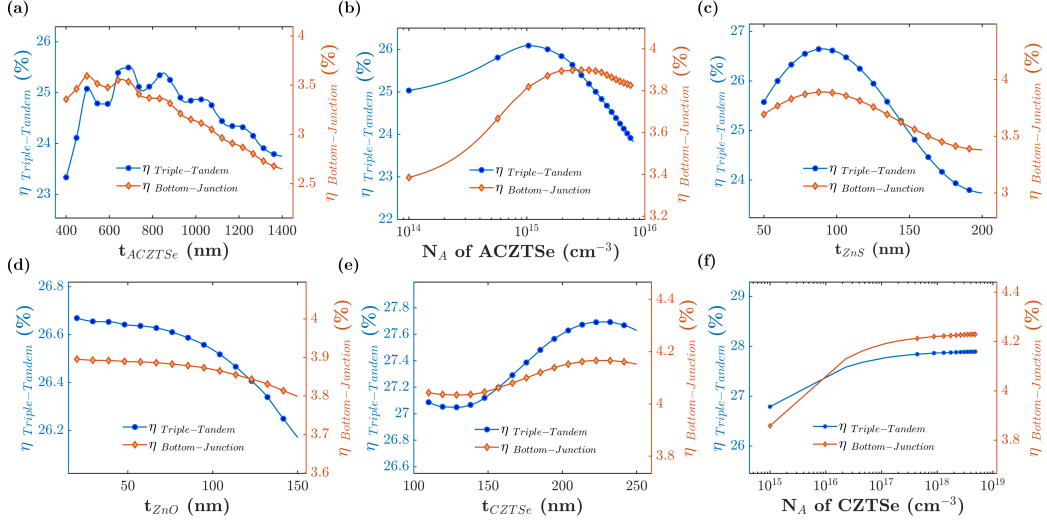


Figure 9: Effect of thickness and doping density variations in the key layers of the ACZTSe bottom subcell on PCE (η) of a monolithic triple-junction tandem solar cell. The analysis highlights both the overall tandem cell performance and the specific contributions of the bottom subcell. The parameters investigated include: (a) ACZTSe absorber layer thickness, (b) ACZTSe acceptor doping density, (c) ZnS buffer layer thickness, (d) ZnO window layer thickness, (e) CZTSe back surface field (BSF) layer thickness, and (f) CZTSe acceptor doping density.

850—1240 nm wavelength range, as illustrated in Fig. S11 of the Supplementary Material. However, an initial drop in performance parameters occurred due to reduced optical back reflection and photon reabsorption, as summarized in Table S3.

Next, we modeled the third subcell as follows: AZO/ZnO/ZnS/ACZTSe/CZTSe/Mo. For optimization purposes, the initial thickness and doping density were taken as shown in Table S4. At first, the ACZTSe absorber thickness was varied from 400 to 1400 nm, under constant acceptor doping. The results showed a peak PCE of 25.77% for a 750 nm thickness of ACZTSe as Fig. 9(a). Subsequently, N_A of ACZTSe absorber layer was varied from 1×10^{14} to $1 \times 10^{15} \text{ cm}^{-3}$. The maximum PCE of 26.08% was achieved at N_A of $1 \times 10^{15} \text{ cm}^{-3}$, as shown in Fig. 9(b). However, beyond a N_A value of $9 \times 10^{14} \text{ cm}^{-3}$, a declining trend in J_{sc} was noticed from Table S5 of Supplementary Material, indicating increased electron-hole recombination at the ACZTSe/CZTSe junction. Therefore, N_A of $9 \times 10^{14} \text{ cm}^{-3}$ was selected for

further optimization. Afterwards, n-doped ZnS and n-doped ZnO layer thicknesses were varied as shown in Figs. 9(c) and 9(d). For a ZnS buffer layer thickness of 90 nm and a ZnO layer thickness of 20 nm, the highest PCE of 26.67% was achieved. Subsequently, to evaluate the influence of the CZTSe back surface field (BSF) layer, its thickness was varied from 110 to 250 nm, as shown in Fig. 9(e). The results indicate a rising trend in PCE, increasing from 27.1% and peaking at 27.69% for a BSF thickness of 220 nm. This improvement suggests enhanced lateral absorption in the ACZTSe layer, contributing to better device performance. N_A of the ACZTSe layer was varied from 1×10^{16} to $1 \times 10^{17} \text{ cm}^{-3}$. As illustrated in Fig. 9(f), the PCE exhibits a steady increase from 1×10^{16} to $1 \times 10^{17} \text{ cm}^{-3}$ and continues to rise with higher doping densities. However, Li *et al.* reported that increasing the N_A of CZTSe from 10^{15} to 10^{18} cm^{-3} significantly reduced the trap-assisted carrier lifetime from 5 ns to 13 ps, indicating higher recombination losses [67]. Therefore, N_A of $6 \times 10^{16} \text{ cm}^{-3}$ was selected as a trade-off, yielding a PCE of 27.71%, with J_{sc} of 12.11 mA/cm², V_{oc} of 2.726 V, and FF of 83.98% for the 3-J tandem cell.

Afterward, the thickness of the ITO tunnel layer between the middle and bottom subcell was varied as shown in Figs. 10(a) and (b). As the ITO thickness increases, enhanced reflection of the overall tandem cell and carrier recombination are observed. This phenomenon led to a decrease in the PCE of both the middle and bottom subcells, along with the overall 3-J tandem cell. Based on this analysis, a 20 nm ITO thickness was selected as an optimal trade-off, balancing device stability with efficient charge transport.

To determine the overall optimal performance of the 3-J tandem cell, the thicknesses of the primary absorber layers in each subcell were systematically varied, while maintaining constant thicknesses for the transport and recombination layers. Specifically, KSnI₃ thickness was varied from 350 to 550 nm, FASnI₃ from 400 to 700 nm, and ACZTSe from 800 to 1150 nm. Figs. 10(c-f) present the performance metrics corresponding to the volumetric parametric thickness sweep for the KSnI₃, FASnI₃, and ACZTSe. As shown in Fig. 10(c), the PCE of the 3-J tandem cell was highly sensitive to the thickness of the top subcell absorber KSnI₃ (350–450 nm) and the middle subcell absorber FASnI₃ (400–550 nm), particularly when the bottom subcell absorber ACZTSe had a thickness in the range of 850–1100 nm. This behavior can be attributed to the monolithic 2-T configuration of the tandem structure, which requires a uniform J_{sc} across all subcells. Any mismatch in the photocurrent among the

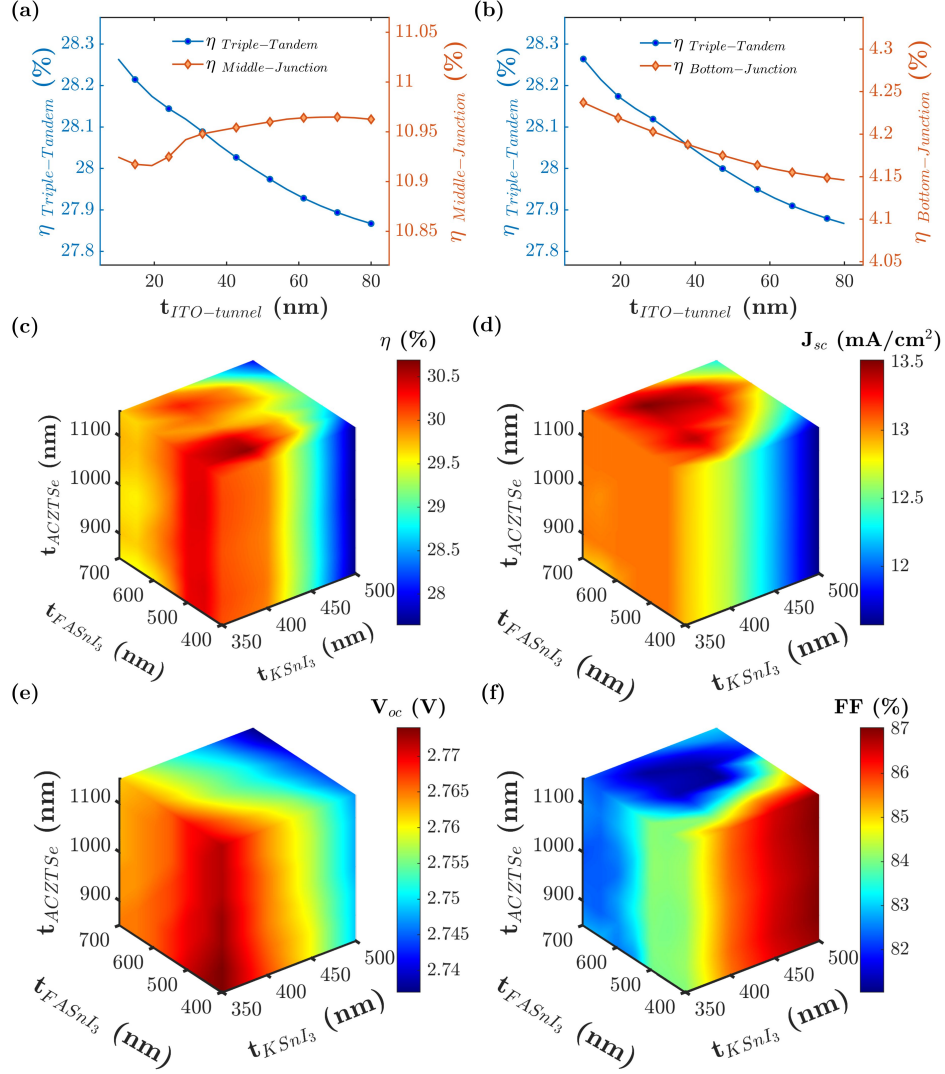


Figure 10: Influence of ITO tunnel junction thickness on PCE (η) of the device featuring (a) overall tandem PCE and middle junction, and (b) overall tandem PCE and bottom junction. Volumetric contour plot of (c) η , (d) J_{sc} , (e) V_{oc} , and (f) FF featuring the performance of triple junction tandem cell corresponding to various thickness ranges of $KSnI_3$, $FASnI_3$, and $ACZTSe$.

subcells leads to increased internal carrier recombination, thereby reducing the overall device efficiency. For $KSnI_3$ thickness ranging from 400 to 450 nm and $FASnI_3$ thickness ranging from 550 to 650 nm, elevated J_{sc} values

are observed, peaking at 13.51 mA/cm² for thicknesses of 400 nm of KSnI₃, 650 nm of FASnI₃, and 1100 nm of ACZTSe, as depicted in Fig. 10(d). However, V_{oc} was observed to be higher for thinner regions of KSnI₃ and FASnI₃ as shown in Fig. 10(e). This trend arises from increased carrier recombination associated with a larger absorber thickness, which in turn elevates the dark saturation current density (J₀) and, consequently, reduces V_{oc}. The FF shows a consistent increase with ACZTSe thickness, likely due to enhanced carrier collection and reduced recombination. For KSnI₃, FF improved as thickness increased, providing better charge transport and current matching. However, FASnI₃ demonstrated an optimal FF at an intermediate thickness, beyond which resistive losses became dominant. This consistent behaviour underscores the crucial role of KSnI₃ in maintaining a high FF as illustrated

Table 5: Summary of performance metrics of the 3-J tandem cell as a function of thickness and doping density variations in the bottom subcell.

Optimization Parameters	η (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	P_{mpp} (W/m ²)	V_{mpp} (V)
ACZTSe thickness (750 nm)	25.77	11.74	2.695	81.46	257.68	2.343
ACZTSe doping (9.00×10^{14} cm ⁻³)	26.06	11.46	2.714	83.77	260.60	2.414
ZnS thickness (90 nm)	26.64	11.78	2.716	83.30	266.45	2.402
ZnO thickness (20 nm)	26.67	11.79	2.716	83.29	266.69	2.402
CZTSe Thickness (220 nm)	27.69	12.10	2.725	83.95	276.89	2.410
CZTSe doping (6.00×10^{16} cm ⁻³)	27.71	12.11	2.726	83.98	277.11	2.411
Tandem absorbers thickness KSnI ₃ /FASnI ₃ /ACZTSe (400 nm/450 nm/900 nm)	30.69	13.18	2.766	84.18	30.692	2.432

in Fig. 10(f). An optimal PCE of 30.69% was achieved with J_{sc} of 13.184 mA/cm^2 , V_{oc} of 2.766 V, and FF of 84.18%, corresponding to absorber thicknesses of 400 nm for KSnI_3 , 450 nm for FASnI_3 , and 900 nm for ACZTSe. A 2D contour plot, as shown in Figs. S12(a-d), along with the data in Table S6, supports these findings. A summary of the progressive enhancement in performance metrics of the ACZTSe subcell within the 3-J tandem configuration, as a result of volumetric absorber thickness optimization across all subcells, is presented in Table 5.

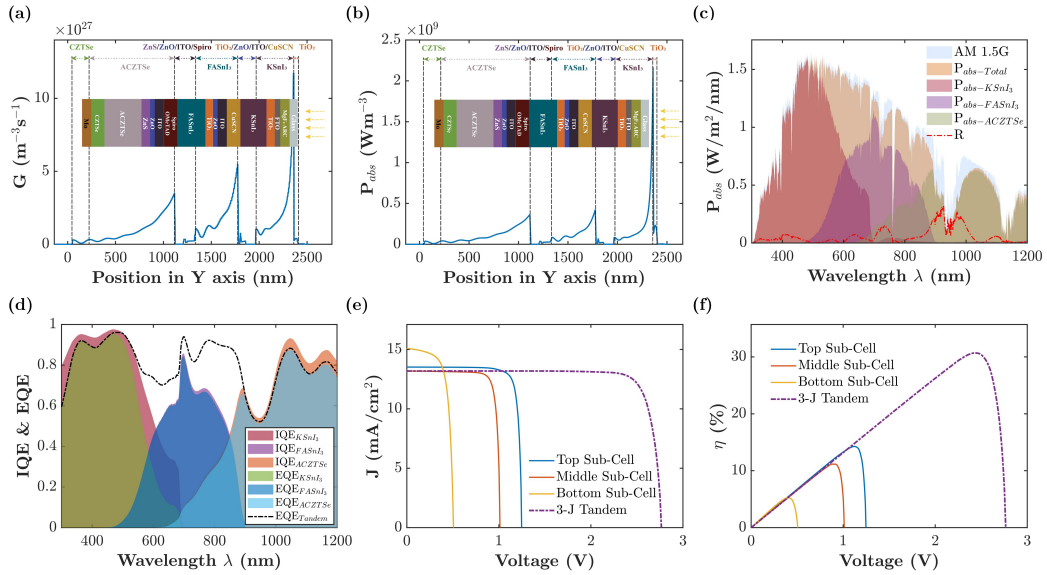


Figure 11: (a) Carrier generation rate, G as a function of depth (Y-axis), under solar illumination, highlighting the maximum generation regions. (b) Absorbed power density, P_{abs} , along the Y-direction, for different layers in 3-J tandem cell. (c) Normalized optical power absorption, (d) spectral optical power absorption across the primary absorber layers of the 3-J tandem cell as a function of photon wavelength corresponding to AM 1.5G solar spectrum, (e) IQE, and EQE versus photon wavelength λ , (e) J -V, and (f) η -V curve of the optimized 2-T $\text{KSnI}_3/\text{FASnI}_3/\text{ACZTSe}$ -based tandem cell and its different subcells.

Fig. 11 presents a comprehensive opto-electronic analysis of the optimized 2-T $\text{KSnI}_3/\text{FASnI}_3/\text{ACZTSe}$ -based tandem solar cell. Fig. 11(a) showcases the generation rate profile, G , highlighting the carrier generation regions as a function of depth (Y-axis) under solar illumination. Fig. 11(b) highlights the absorbed power density, P_{abs} , in different layers of the tandem cell, along the Y-direction. Fig. 11(c) compares the wavelength-resolved spectral absorbed

power, P_{abs} , of the optimized triple-junction tandem with the AM 1.5G spectrum. The distinct absorption regions of each subcell demonstrate effective spectral splitting from 300–1200 nm, while the low reflectance verifies strong optical confinement and minimal photon loss. The high-energy photons are predominantly absorbed in the KSnI_3 top cell, intermediate photons in FASnI_3 , and near-infrared photons in the ACZTSe bottom cell. Fig. 11(d) presents the corresponding EQE and IQE spectra, showing high carrier collection efficiency within each subcell’s absorption range. The smooth spectral transitions and broad EQE response confirm efficient current matching and complementary photon harvesting across the solar spectrum. Finally, Figs. 11(e–f) represent the J–V and corresponding η –V characteristics of the optimized 3-J tandem solar cell. The individual subcells exhibit distinct V_{oc} determined by their bandgaps, while the combined 3-J tandem curve demonstrates series-connected behavior with the total current limited by the lowest photocurrent. The close current matching among subcells confirms efficient carrier extraction, minimal interfacial recombination, and balanced photo-response across the device stack.

Table 6 and a bar plot as shown in Fig. 12 highlight the progressive enhancement in device performance as the architecture evolved from single to tandem structures, demonstrating the effectiveness of absorber stacking and junction engineering. After optimization, 1-J KSnI_3 and 1-J FASnI_3 solar

Table 6: Summary of the performance matrices from single junction cells to triple junction tandem cells.

Solar cell types	η (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	P_{mpp} (mW/cm ²)	V_{mpp} (V)
1-J KSnI_3 Cell	16.28	16.63	1.232	79.50	16.28	1.08
1-J FASnI_3 Cell	26.03	29.81	1.030	84.84	26.03	0.92
2-J tandem cell ($\text{KSnI}_3/\text{FASnI}_3$)	27.29	14.74	2.227	83.14	27.29	1.98
3-J tandem cell ($\text{KSnI}_3/\text{FASnI}_3/\text{ACZTSe}$)	30.69	13.18	2.766	84.18	30.69	2.43

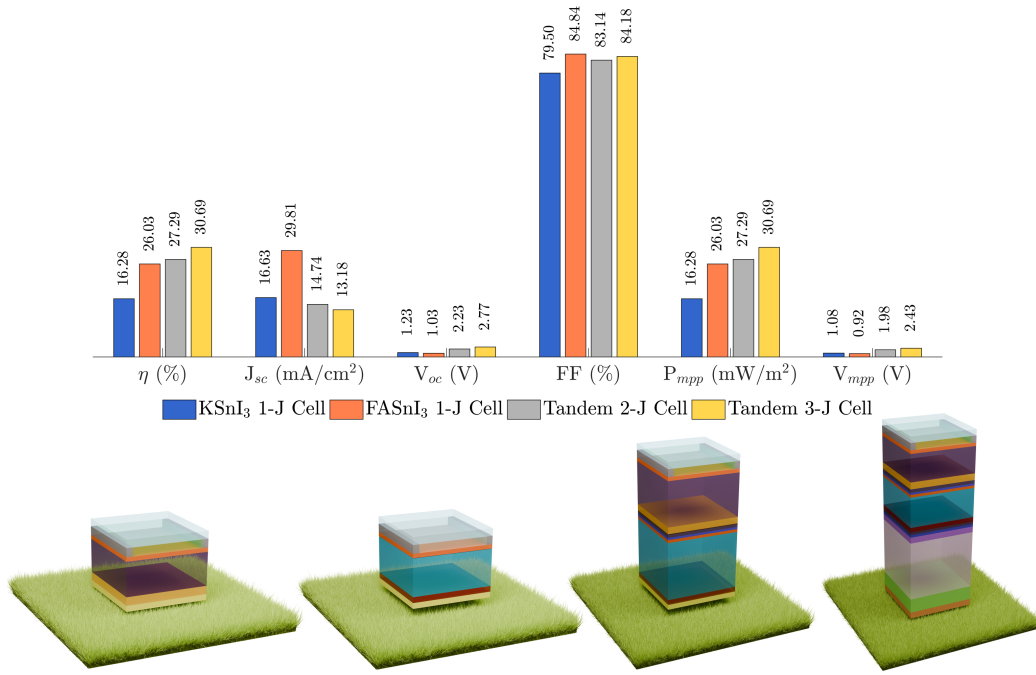


Figure 12: Bar plot illustrating the optimized performance metrics of both single-junction and different multi-junction tandem solar cells shown in the bottom row.

cells secured 16.28% and 26.03% respectively, whereas the 2-J and 3-J tandem cell secured 27.29% and 30.69% accordingly. Table 7 showcases the photon flux, ϕ , and absorbed optical power per unit cross-sectional area for the subcells of 2-J and 3-J tandem cells. In the case of the 2-J tandem cell, as the top subcell is responsible for absorbing higher-energy photons, it necessi-

Table 7: Photon flux (ϕ) and power consumption (P_{consump}) in 2-J and 3-J tandem solar cells.

2T KSnI ₃ /FASnI ₃ Tandem Cell		
Subcells	ϕ (cm ⁻² s ⁻¹)	P_{consump} (Wm ⁻²)
Top KSnI ₃ subcell	1.136×10^{21}	387.36
Bottom FASnI ₃ Subcell	1.1665×10^{21}	270.63
2T KSnI ₃ /FASnI ₃ /ACZTSe Tandem Cell		
Top KSnI ₃ subcell	1.19×10^{21}	349.90
Middle FASnI ₃ subcell	1.03×10^{21}	238.31
Bottom ACZTSe subcell	1.28×10^{21}	204.17

tates a larger portion of the incident optical power to ensure proper current matching with the bottom subcell. Moreover, the photon fluxes across the two subcells are closely aligned, with the top subcell ultimately determining the current limitation of the overall tandem device as seen in Fig. 8(e). For the 3-J tandem cell, the top subcell primarily absorbs near-UV and visible photons, the middle subcell absorbs visible and near-infrared (NIR) photons, and the bottom subcell predominantly absorbs NIR photons from the incident spectrum to facilitate current matching with the subsequent subcells as featured in Fig. 11(c). From the absorbed photon fluxes among the three subcells, the middle subcell emerged as the current-limiting junction in the 3-J tandem cell, as evidenced in Table 7.

3.5. Performance benchmarking with literature

Table 8 presents a comparative analysis of key performance metrics from this work alongside previously reported perovskite-based tandem solar cells. Liu *et al.* demonstrated an all-perovskite tandem solar cell achieving 28.1% efficiency by combining a wide-bandgap absorber, formamidinium–cesium lead iodide–bromide, $(\text{FA}_{0.8}\text{Cs}_{0.2}\text{Pb}(\text{I}_{0.62}\text{Br}_{0.38})_3)$, with a narrow-bandgap absorber, formamidinium–methylammonium lead–tin iodide, $(\text{FA}_{0.7}\text{MA}_{0.3}\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_3)$, and employing metal-oxide nanocrystal hole transport layers to minimize optical losses [68]. Despite the impressive efficiency and stability, the device’s reliance on lead and tin poses concerns due to toxicity and the instability of tin-based perovskites. Sun *et al.* reported scalable solution processing of hybrid fullerene electron transport layers in an all-perovskite tandem module, reaching 23.3% efficiency. Their approach improves energy-level alignment and reduces interfacial recombination but faces challenges related to the complexity of fullerene blends and slightly lower performance [69]. Lin *et al.* introduced a bilayer heterojunction design with mixed-dimensional Pb–Sn perovskites to suppress surface recombination and facilitate charge extraction, achieving a certified efficiency of 28.0% [23]. However, the mixed-dimensional phases can introduce charge transport losses, limiting fill factor and complicating fabrication. Pan *et al.* developed a surface reconstruction method using diammonium halide salts to passivate Sn–Pb mixed perovskites, minimizing nonradiative recombination and achieving a certified 28.49% efficiency for two-junction tandems [70]. Their work advances film quality and stability but still contends with tin oxidation challenges inherent to Sn–Pb perovskites. Wang *et al.* tackled halide heterogeneity in Br-rich perovskites to reduce recombination and realized a 25.1% efficient

Table 8: Comparative analysis of previously reported performance matrices of different perovskite (PVSK) material-based tandem solar cells with this work.

Solar Cell Type	Structure	η (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	Ref
PVSK/PVSK	Glass/ITO/SAM-NiO _x /FA _{0.8} CS _{0.2} Pb(I _{0.62} Br _{0.38}) ₃ / C ₆₀ /ALD-SnO ₂ /IC-CH/FA _{0.7} MA _{0.3} Pb _{0.5} Sn _{0.5} I ₃ / C ₆₀ /ALD-SnO ₂ /Cu	28.10	16.70	2.11	79.50	[68]
PVSK/PVSK	Glass/ITO/SAMs-NiO _x /CS _{0.35} FA _{0.65} PbI _{1.8} Br _{1.2} / HF(C ₆₀ :PCBM:ICBA)/ALD-SnO ₂ /Au/PEDOT:PSS/ FA _{0.7} MA _{0.3} Pb _{0.5} Sn _{0.5} I ₃ /HF/ALD-SnO ₂ /Cu	27.40	15.90	2.09	82.40	[69]
PVSK/PVSK	ITO/PEDOT:PSS/FA _{0.7} CS _{0.3} Pb(I _{0.85} Br _{0.14}) ₃ / FL-FA _{0.8} CS _{0.2} Pb(I _{0.62} Br _{0.38}) ₃ /C ₆₀ /BCP/Cu	28.5	16.5	0.873	82.6	[23]
PVSK/PVSK	Glass/ITO/Me-4PACz/Al ₂ O ₃ /FA _{0.8} CS _{0.2} Pb(I _{0.6} Br _{0.4}) ₃ / PDAI ₂ /C ₆₀ /ALD-SnO ₂ /Au/PEDOT:PSS/ Al ₂ O ₃ /FA _{0.6} MA _{0.3} CS _{0.1} Pb _{0.5} Sn _{0.5} I ₃ / BDA-EDA ₂ /C ₆₀ /BCP/Ag	28.49	16.02	2.12	83.88	[70]
PVSK/PVSK/PVSK	Glass/IOH/NiO _x /Me-4PACz/CS _{0.15} FA _{0.85} Pb(I _{0.4} Br _{0.6}) ₃ / PCBM/PEI/SnO _x /ITO/NiO _x /Me-PACz/ CS _{0.05} FA _{0.9} MA _{0.05} Pb(I _{0.85} Br _{0.15}) ₃ /C ₆₀ /SnO _x /Au/ PEDOT:PSS/CS _{0.05} FA _{0.7} MA _{0.25} Pb _{0.5} Sn _{0.5} I ₃ / C ₆₀ /SnO _x /Ag	25.10	9.70	3.33	78.00	[71]
PVSK/PVSK/Si	SHJ/IZO/MeO ₂ PACz/Rb _{0.05} CS _{0.1} FA _{0.85} PbI ₃ /C ₆₀ / SnO ₂ /IZO/NiO _x /2PACz/CS _{0.1} FA _{0.9} PbI _{0.9} Br _{2.1} / C ₆₀ /SnO ₂ /IZO/MgF ₂ /Ag	26.40	11.90	3.04	72.90	[72]
PVSK/PVSK/Si	Si/ITO/Me-4PACz/FA _{0.9} CS _{0.1} PbI ₃ /C ₆₀ /SnO ₂ /ITO /NiO _x /Me-4PACz/FA _{0.6} MA _{0.15} CS _{0.25} Pb(I _{0.45} Br _{0.5} OCN _{0.05}) ₃ /C ₆₀ /SnO ₂ /ITO/LiF	27.60	11.58	3.13	76.30	[24]
PVSK/PVSK	Glass/FTO/TiO ₂ /KSnI ₃ /CuSCN/ITO/ZnO/ TiO ₂ /FASnI ₃ /Spiro-OMeTAD/Au	27.29	14.74	2.227	83.14	This work
PVSK/PVSK/ Kesterite	Glass/FTO/TiO ₂ /KSnI ₃ /CuSCN/ITO /ZnO/TiO ₂ /FASnI ₃ /Spiro-OMeTAD/ITO/ AZO/ZnO/ZnS/ACZTSe/CZTSe/Mo	30.69	13.184	2.766	84.18	This work

triple-junction all-perovskite tandem, though challenges remain in balancing halide distribution and improving device stability [71]. Xu *et al.* combined co-additives to stabilize wide-bandgap perovskites integrated in perovskite-silicon triple-junction tandems, achieving 26.4% efficiency, but the inclusion of silicon introduces additional processing complexity and cost [72]. In contrast, our work proposes novel double and triple-junction tandem architectures integrating lead-free, tin-based perovskites (KSnI₃ and FASnI₃) and an earth-abundant kesterite absorber (ACZTSe). This design achieved competitive PCEs of 27.29% for 2-J tandem and 30.69% for 3-J tandem cells, respectively, while simultaneously addressing critical issues such as toxicity,

material scarcity, and infrared spectral coverage. The absorption edge of the ACZTSe layer boosted infrared photon absorption, enhancing current-matching and device efficiency. Moreover, band alignment and intermediate transportation layers were optimized to provide efficient charge extraction as depicted from high fill factors exceeding several reported perovskite tandems. Even though the oxidation of tin is still an issue, it can be alleviated by using encapsulation and interface engineering methods. By avoiding toxic lead and scarce dopants like rubidium or bromide, our tandem cells offer an environmentally friendly, scalable, and high-performance platform that balances efficiency, stability, and sustainability more effectively than many existing perovskite tandem designs.

4. Feasibility of the proposed 3-J tandem cell architecture

The fabrication of the proposed triple junction tandem cell architecture requires precise and sequential deposition techniques optimized according to the thermal and chemical sensitivities of each constituent layer. The process commences with the deposition of a thin MgF_2 ARC via thermal or electron beam evaporation, designed to reduce optical losses at the air/glass interface [73]. Subsequently, highly conductive FTO films are deposited using radio frequency (RF) magnetron sputtering, which offers strong adhesion, uniformity, and scalability [74]. Anatase-phase TiO_2 thin films are then prepared on the FTO surface through spin-coating followed by annealing. Post-annealing, UV-ozone treatment is employed to enhance surface properties for subsequent layer integration [66]. The deposition of KSnI_3 absorber layer, though still limited experimentally, can be achieved via spin-coating a precursor solution of potassium iodide (KI) and tin(II) iodide (SnI_2) in a solvent mixture of N,N-dimethylformamide (DMF) and dimethyl sulfoxide (DMSO). This is followed by thermal annealing to promote crystallization, following methodologies adapted from tin-based perovskite literature [75]. Afterwards, CuSCN HTL is applied through spin-coating from a dipropyl sulfide solution and mildly annealed to form a compact and hydrophobic interface [76]. ITO interlayer is then deposited by low-temperature sputtering to preserve the integrity of the underlying layers [77]. ZnO is introduced as ETL layer either by atomic layer deposition (ALD) or low-temperature sol-gel techniques, followed by a second compact TiO_2 layer. The FASnI_3 absorber is deposited using an antisolvent-assisted spin-coating method, incorporating SnF_2 as an additive to improve phase stability and reduce hysteresis [78]. Subsequently,

Spiro-OMeTAD is spin-coated from a chlorobenzene solution containing Li-TFSI and 4-tert-butylpyridine (tBP) as dopants to enhance hole mobility [79]. A second transparent ITO layer (20 nm) is sputtered, followed by an Al-doped ZnO (AZO) layer to reduce sheet resistance and enhance optical transmission. ZnO and ZnS buffer layers are deposited via ALD or chemical bath deposition (CBD), ensuring favorable band alignment and reduced interfacial recombination [80]. ACZTSe/CZTSe absorber stack is subsequently formed through co-sputtering of Cu, Zn, Sn, Se, and Ag, followed by high-temperature selenization to achieve the desired kesterite phases and interface quality [81]. Molybdenum (Mo) back contact layer is deposited via DC magnetron sputtering to serve as a chemically stable, low-resistance electrode. To ensure both environmental and operational stability, the device is encapsulated with a UV-curable epoxy and sealed with a glass barrier.

5. Conclusion

The design and optimization of high-efficiency tandem solar cells calls for a holistic approach, that encompasses several interconnected aspects, such as the selection and incorporation of complementary absorber materials, precise tunnel junction engineering, and accurate matching of photocurrent among the subcells. In this paper, we have addressed these challenges by introducing two new lead-free tandem solar cell architectures, an all-perovskite dual-junction solar cell ($\text{KSnI}_3/\text{FASnI}_3$) and a hybrid perovskite-kesterite triple-junction solar cell ($\text{KSnI}_3/\text{FASnI}_3/\text{ACZTSe}$). The chosen absorbers effectively cover from near-UV to NIR spectral region, reducing bandgap disparities and enhancing overall photon utilization. By systematically optimizing the absorber thickness, doping density, and tunnel-junction band alignment, the proposed architectures achieved effective current matching and enhanced charge transport throughout the device. The dual-junction cell delivered a PCE of 27.29% with J_{sc} of 14.74 mA/cm^2 , V_{oc} of 2.227 V, and FF of 83.14%. Subsequently, after integration and careful optimization of the triple junction subcell, the optimized triple-junction configuration reached a champion PCE of 30.69% with J_{sc} of 13.184 mA/cm^2 , V_{oc} of 2.766 V, and FF of 84.18%, corresponding to a relative PCE gain of 12.5% over the dual-junction architecture. These results and insights demonstrated the ability of hybrid tandem architectures to exceed the efficiency limits of conventional designs while employing sustainable, earth-abundant, and non-toxic materials.

6. CRediT authorship contribution statement

Md. Faiaad Rahman: conceptualization, methodology, visualization, simulation, investigation, writing – original draft, and writing – review & editing. **Md. Ashaduzzaman Niloy:** conceptualization, visualization, investigation, writing – original draft, and writing – review & editing. **Ehsanur Rahman:** methodology, visualization, supervision, writing – original draft, and writing – review & editing, **Ahmed Zubair:** conceptualization, methodology, visualization, resources, supervision, writing – original draft, and writing – review & editing.

7. Data availability statement

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

8. Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

9. Acknowledgments

The authors express their sincere gratitude to the Department of Electrical and Electronic Engineering at Bangladesh University of Engineering and Technology (BUET) for providing access to the Ansys Lumerical software and the necessary technical support.

References

- [1] S. Almosni, A. Delamarre, Z. Jehl, D. Suchet, L. Cojocaru, M. Giteau, B. Behaghel, A. Julian, C. Ibrahim, L. Tatry, et al., Material challenges for solar cells in the twenty-first century: directions in emerging technologies, *Science and Technology of Advanced Materials* 19 (1) (2018) 336–369.

- [2] M. A. Green, E. D. Dunlop, M. Yoshita, N. Kopidakis, K. Bothe, G. Siefer, X. Hao, J. Y. Jiang, Solar cell efficiency tables (version 65), Progress in photovoltaics: research and applications (2024).
- [3] G. Masson, M. de l’Epine, I. Kaizuka, Trends in photovoltaic applications 2024, Tech. Rep. IEA-PVPS T1-43:2024, International Energy Agency Photovoltaic Power Systems Programme (IEA PVPS), Paris, France (2024).
- [4] S. Saravanan, R. S. Dubey, Study of ultrathin-film amorphous silicon solar cell performance using photonic and plasmonic nanostructure, International Journal of Energy Research 46 (3) (2022) 2558–2566.
- [5] S. R. Kavanagh, A. Walsh, D. O. Scanlon, Rapid recombination by cadmium vacancies in cdte, ACS Energy Letters 6 (4) (2021) 1392–1398.
- [6] D. R. Sajitha, B. Stephen, A. Nakamura, M. Selvaraj, S. T. Salammal, S. Hussain, The emergence of chalcogenides: A new era for thin film solar absorbers, Progress in Solid State Chemistry 76 (2024) 100490.
- [7] S. Tomasulo, M. L. Lee, Advances in wide-bandgap iii-v solar cells, Applied Physics Reviews 12 (3) (2025) 031309.
- [8] National Renewable Energy Laboratory, Best research-cell efficiency chart, <https://www.nrel.gov/pv/cell-efficiency.html>, accessed: 2025-07-17 (2025).
- [9] M. F. Rahman, A. Sur, A. Zubair, Hierarchically structured, high-efficiency and thermally robust perovskite solar cells with band-engineered double-hole layers, arXiv (2025) 2509.03146.
- [10] P. P. Nakti, D. Sarker, M. I. Tahmid, A. Zubair, Ultra-broadband near-perfect metamaterial absorber for photovoltaic applications, Nanoscale Advances 5 (2023) 6858–6869.
- [11] J. Pastuszak, P. Wegierek, Photovoltaic cell generations and current research directions for their development, Materials 15 (16) (2022) 5542.
- [12] A. A. Ugochukwu, F. Ahmad, M. Khalid, M. A. Ali, I. Mamoon, S. C. Udensi, U. Ogbonna, A. Iqbal, R. Ali, M. Usman, J. Khan, I. Khurshid, U. Zahoor, A. H. shah, A. U. Rahman, F. Rehman, Recent enhancement

in photovoltaic cell efficiency performance, stability, and cost reduction: a review, *Solar Energy* 300 (2025) 113853.

- [13] J. Han, K. Park, S. Tan, Y. Vaynzof, J. Xue, E. W.-G. Diau, M. G. Bawendi, J.-W. Lee, I. Jeon, Perovskite solar cells, *Nature Reviews Methods Primers* 5 (1) (2025) 3.
- [14] A. S. Najm, V. Selvanathan, T. M. Aljuwaya, L. S. Sabri, M. S. Jamal, A. Abdullah Al-Zahrani, A. M. Holi, I. Jaber, A. Al Ghamdi, M. T. Amin, K. Sopian, R. A. Ismail, H. Moria, B. Bais, H. S. Majdi, S. Chowdhury, A. J. Sultan, T. M. Alhuzaymi, Recent progress in performance improvement strategies for quantum dot sensitization methods: Challenges, achievements, and future prospects, *APL Materials* 11 (9) (2023) 090601.
- [15] A. Ebenezer Anitha, M. Dotter, A review on liquid electrolyte stability issues for commercialization of dye-sensitized solar cells (dssc), *Energies* 16 (13) (2023).
- [16] J. Yi, G. Zhang, H. Yu, H. Yan, Advantages, challenges and molecular design of different material types used in organic solar cells, *Nature Reviews Materials* 9 (1) (2024) 46–62.
- [17] A. H. Slavney, L. Leppert, A. Saldivar Valdes, D. Bartesaghi, T. J. Savenije, J. B. Neaton, H. I. Karunadasa, Small-band-gap halide double perovskites, *Angewandte Chemie* 130 (39) (2018) 12947–12952.
- [18] M. Noman, Z. Khan, S. T. Jan, A comprehensive review on the advancements and challenges in perovskite solar cell technology, *RSC Advances* 14 (8) (2024) 5085–5131.
- [19] N. Rono, C. C. Ahia, E. L. Meyer, Recent advances in transition metal dichalcogenides-based materials for fourth-generation perovskite solar cell devices, *AIP Advances* 14 (7) (2024).
- [20] S. Essig, C. Allebé, T. Remo, J. F. Geisz, M. A. Steiner, K. Horowitz, L. Barraud, J. S. Ward, M. Schnabel, A. Descoeur, D. L. Young, M. Woodhouse, M. Despeisse, C. Ballif, A. Tamboli, Raising the one-sun conversion efficiency of III–V/Si solar cells to 32.8% for two junctions and 35.9% for three junctions, *Nature Energy* 2 (2017) 17144.

- [21] J. Liu, Y. He, L. Ding, H. Zhang, Q. Li, L. Jia, J. Yu, T. W. Lau, M. Li, Y. Qin, X. Gu, F. Zhang, Q. Li, Y. Yang, S. Zhao, X. Wu, J. Liu, T. Liu, Y. Gao, Y. Wang, X. Dong, H. Chen, P. Li, T. Zhou, M. Yang, X. Ru, F. Peng, S. Yin, M. Qu, D. Zhao, Z. Zhao, M. Li, P. Guo, H. Yan, C. Xiao, P. Xiao, J. Yin, X. Zhang, Z. Li, B. He, X. Xu, Perovskite/silicon tandem solar cells with bilayer interface passivation, *Nature* 635 (2024) 596–603.
- [22] M. A. Steiner, R. M. France, J. Buencuerpo, J. F. Geisz, M. P. Nielsen, A. Pusch, W. J. Olavarria, M. Young, N. J. Ekins-Daukes, High efficiency inverted GaAs and GaInP/GaAs solar cells with strain-balanced GaInAs/GaAsP quantum wells, *Advanced Energy Materials* 11 (1 2021).
- [23] R. Lin, Y. Wang, Q. Lu, B. Tang, J. Li, H. Gao, Y. Gao, H. Li, C. Ding, J. Wen, P. Wu, C. Liu, S. Zhao, K. Xiao, Z. Liu, C. Ma, Y. Deng, L. Li, F. Fan, H. Tan, All-perovskite tandem solar cells with 3d/3d bilayer perovskite heterojunction, *Nature* 620 (2023) 994–1000.
- [24] S. Liu, Y. Lu, C. Yu, J. Li, R. Luo, R. Guo, H. Liang, X. Jia, X. Guo, Y.-D. Wang, et al., Triple-junction solar cells with cyanate in ultrawide-bandgap perovskites, *Nature* 628 (8007) (2024) 306–312.
- [25] P. Schygulla, R. Müller, O. Höhn, M. Schachtner, D. Chojniak, A. Cordaro, S. Tabernig, B. Bläsi, A. Polman, G. Siefer, D. Lackner, F. Dimroth, Wafer-bonded two-terminal III-V//Si triple-junction solar cell with power conversion efficiency of 36.1% at am1.5g, *Progress in Photovoltaics: Research and Applications* 33 (2025) 100–108.
- [26] J. F. Geisz, R. M. France, K. L. Schulte, M. A. Steiner, A. G. Norman, H. L. Guthrey, M. R. Young, T. Song, T. Moriarty, Six-junction iii–v solar cells with 47.1% conversion efficiency under 143 suns concentration, *Nature Energy* 5 (2020) 326–335.
- [27] M. A. Green, E. D. Dunlop, M. Yoshita, N. Kopidakis, K. Bothe, G. Siefer, X. Hao, J. Y. Jiang, Solar cell efficiency tables (version 66), *Progress in Photovoltaics: Research and Applications* 33 (2025) 795–810.
- [28] R. Lin, J. Xu, M. Wei, Y. Wang, Z. Qin, Z. Liu, J. Wu, K. Xiao, B. Chen, S. M. Park, G. Chen, H. R. Atapattu, K. R. Graham, J. Xu, J. Zhu,

- L. Li, C. Zhang, E. H. Sargent, H. Tan, All-perovskite tandem solar cells with improved grain surface passivation, *Nature* 603 (2022) 73–78.
- [29] M. Jošt, E. Köhnen, A. Al-Ashouri, T. Bertram, Špela Tomšič, A. Magomedov, E. Kasparavicius, T. Kodalle, B. Lipovšek, V. Getautis, R. Schlatmann, C. A. Kaufmann, S. Albrecht, M. Topič, Perovskite/CIGS tandem solar cells: From certified 24.2% toward 30% and beyond, *ACS Energy Letters* 7 (2022) 1298–1307.
- [30] H. Ferhati, F. Djeflal, B. Drissi, Performance improvement of Perovskite/CZTS tandem solar cell using low-cost ZnS/Ag/ITO multilayer spectrum splitter, *Superlattices and Microstructures* 148 (2020) 106727.
- [31] V. C. Karade, M. He, Z. Song, A. Abudulimu, Y. Park, D. Song, Y. Yan, J. H. Kim, R. J. Ellingson, J. H. Yun, X. Hao, S. W. Shin, M. P. Suryawanshi, Opportunities and challenges for emerging inorganic chalcogenide–silicon tandem solar cells, *Energy Environ. Sci.* 18 (2025) 6899–6933.
- [32] F. Pei, S. Lin, J. Tang, X. Huang, Y. Han, Q. Sun, S. Lin, H. Wang, Z. Zhang, X. Zhuang, K. Li, C. Zhu, Y. Chen, T. Song, T. Cheng, Y. Cui, Y. Zhang, H. Hou, W. Zhou, Y. Lin, B. Li, S. Hong, H. Zhou, X. Wang, Y. Zhou, D. Zhong, Q. Chen, Y. Jiang, Perovskite/CIGS tandem solar cells with over 1000 h operational stability through interconnection stress relief, *Journal of the American Chemical Society* 147 (40) (2025) 36815–36824.
- [33] G. Farias-Basulto, T. Mehlhop, N. J. Otto, T. Bertram, K. Jäger, S. Gall, N. Weinberger, R. Schlatmann, I. Lauermann, R. Klenk, E. List-Kratochvil, C. A. Kaufmann, Improving perovskite/cigs tandem solar cells for higher power conversion efficiency through light management and bandgap engineering, *ACS Applied Materials & Interfaces* 17 (40) (2025) 56250–56255.
- [34] G. Pindolia, S. M. Shinde, P. K. Jha, Non-leaded, KSnI_3 based perovskite solar cell: A dft study along with scaps simulation, *Materials Chemistry and Physics* 297 (2023) 127426.
- [35] J. D. Hutchinson, E. Ruggeri, J. M. Woolley, G. Delport, S. D. Stranks, R. L. Milot, Resolving the ultrafast charge carrier dynamics of 2d and 3d

- domains within a mixed 2d/3d lead-tin perovskite, *Advanced Functional Materials* 33 (50) (2023) 2305736.
- [36] M. Belarbi, Numerical analysis of performance enhancement in KSnI_3 -based perovskite solar cells with Sb_2Se_3 as a hole transport layer, *Materials Science and Engineering: B* 313 (2025) 117874.
 - [37] S. Pachori, R. Agrawal, B. L. Choudhary, A. S. Verma, et al., An efficient and stable lead-free organic-inorganic tin iodide perovskite for photovoltaic device: Progress and challenges, *Energy Reports* 8 (2022) 5753–5763.
 - [38] C. J. Hages, M. J. Koeper, R. Agrawal, Optoelectronic and material properties of nanocrystal-based CZTSe absorbers with ag-alloying, *Solar Energy Materials and Solar Cells* 145 (2016) 342–348.
 - [39] U. Saha, M. K. Alam, Proposition of an environment friendly triple junction solar cell based on earth abundant CBTSSe/CZTS/ACZTSe materials, *physica status solidi (RRL)–Rapid Research Letters* 12 (1) (2018) 1700335.
 - [40] Q. C. Bui, V. Consonni, S. Boubenia, G. Gay, C. Perret, M. Zeghouane, S. Labau, H. Roussel, X. Mescot, G. Ardila, et al., High figure-of-merit in Al-doped ZnO thin films grown by ald through the Al content adjustment, *Materialia* 31 (2023) 101863.
 - [41] H. Hempel, C. J. Hages, R. Eichberger, I. Repins, T. Unold, Minority and majority charge carrier mobility in $\text{Cu}_2\text{ZnSnSe}_4$ revealed by terahertz spectroscopy, *Scientific reports* 8 (1) (2018) 14476.
 - [42] P. Saha, S. Singh, S. Bhattacharya, FASnI_3 -based eco-friendly heterojunction perovskite solar cell with high efficiency, *Micro and Nanostructures* 186 (2024) 207739.
 - [43] I. B. Abdul Ghani, M. Khalid, H. Yan, M. Kashif, B. Nawaz, J. Wang, Boosting KSnI_3 -based perovskite solar cell efficiency by 22.78% through optimized charge transport and eco-friendly buffer layer, *Journal of Materials Science* 59 (31) (2024) 14547–14572.

- [44] J. Patel, R. K. Sharpe, M. A. Quijada, M. M. Rana, A review of transparent conducting films (tcfs): Prospective ITO and AZO deposition methods and applications, *Nanomaterials* 14 (24) (2024) 2013.
- [45] P. Zhang, C. Li, M. He, Z. Liu, X. Hao, The intermediate connection of subcells in si-based tandem solar cells, *Small Methods* 8 (2) (2024) 2300432.
- [46] A. Razzaq, T. G. Allen, W. Liu, Z. Liu, S. De Wolf, Silicon heterojunction solar cells: Techno-economic assessment and opportunities, *Joule* 6 (3) (2022) 514–542.
- [47] L. V. Rodríguez-de Marcos, J. I. Larruquert, J. A. Méndez, J. A. Aznárez, Self-consistent optical constants of MgF_2 , LaF_3 , and CeF_3 films, *Optical Materials Express* 7 (3) (2017) 989–1006.
- [48] J. M. Ball, S. D. Stranks, M. T. Hörlantner, S. Hüttner, W. Zhang, E. J. Crossland, I. Ramirez, M. Riede, M. B. Johnston, R. H. Friend, et al., Optical properties and limiting photocurrent of thin-film perovskite solar cells, *Energy & Environmental Science* 8 (2) (2015) 602–609.
- [49] T. A. König, P. A. Ledin, J. Kerszulis, M. A. Mahmoud, M. A. El-Sayed, J. R. Reynolds, V. V. Tsukruk, Electrically tunable plasmonic behavior of nanocube–polymer nanomaterials induced by a redox-active electrochromic polymer, *ACS Nano* 8 (6) (2014) 6182–6192.
- [50] R. Treharne, A. Seymour-Pierce, K. Durose, K. Hutchings, S. Roncallo, D. Lane, Optical design and fabrication of fully sputtered CdTe/CdS solar cells, in: *Journal of Physics: Conference Series*, Vol. 286, IOP Publishing, 2011, p. 012038.
- [51] S. Sarkar, V. Gupta, M. Kumar, J. Schubert, P. T. Probst, J. Joseph, T. A. König, Hybridized guided-mode resonances via colloidal plasmonic self-assembled grating, *ACS Applied Materials & Interfaces* 11 (14) (2019) 13752–13760.
- [52] A. Tooghi, D. Fathi, M. Eskandari, High-performance perovskite solar cell using photonic–plasmonic nanostructure, *Scientific Reports* 10 (1) (2020) 11248.

- [53] E. Raoult, R. Bodeux, S. Jutteau, S. Rives, A. Yaiche, D. Coutancier, J. Rousset, S. Collin, Optical characterizations and modelling of semi-transparent perovskite solar cells for tandem applications, in: 36th European photovoltaic solar energy conference and exhibition, 2019, pp. 757–763.
- [54] K. Ghimire, D. Zhao, Y. Yan, N. J. Podraza, Optical response of mixed methylammonium lead iodide and formamidinium tin iodide perovskite thin films, *AIP Advances* 7 (7) (2017).
- [55] S.-Y. Kuo, M.-Y. Hsieh, Efficiency enhancement in $\text{Cu}_2\text{ZnSnS}_4$ solar cells with subwavelength grating nanostructures, *Nanoscale* 6 (13) (2014) 7553–7559.
- [56] U. Saha, M. K. Alam, Proposition and computational analysis of a kesterite/kesterite tandem solar cell with enhanced efficiency, *RSC Advances* 7 (8) (2017) 4806–4814.
- [57] S. W. Tabernig, L. Yuan, A. Cordaro, Z. L. Teh, Y. Gao, R. J. Patterson, A. Pusch, S. Huang, A. Polman, Optically resonant bulk heterojunction PbS quantum dot solar cell, *ACS Nano* 16 (9) (2022) 13750–13760.
- [58] F. B. Sumona, M. Kashif, E. Danladi, A. M. Tighezza, N. Al-Mahmud, G. F. Toki, R. Pandey, M. K. Hossain, Optimization of perovskite- KSnI_3 solar cell by using different hole and electron transport layers: a numerical scaps-1d simulation, *Energy & Fuels* 37 (23) (2023) 19207–19219.
- [59] R. L. Milot, G. E. Eperon, T. Green, H. J. Snaith, M. B. Johnston, L. M. Herz, Radiative monomolecular recombination boosts amplified spontaneous emission in $\text{HC}(\text{NH}_2)_2\text{SnI}_3$ perovskite films, *The journal of physical chemistry letters* 7 (20) (2016) 4178–4184.
- [60] M. Kumar, A. Raj, A. Kumar, A. Anshul, An optimized lead-free formamidinium sn-based perovskite solar cell design for high power conversion efficiency by scaps simulation, *Optical Materials* 108 (2020) 110213.
- [61] X. Tan, J. Hu, W. Zhu, F. Wu, X. Han, Improved performance of kesterite $\text{Cu}_2\text{ZnSn}(\text{S},\text{Se})_4$ thin film solar cells by Ag/Ge co-doping, *Journal of Alloys and Compounds* 981 (2024) 173645.

- [62] T. Enkhbat, E. Enkhbayar, N. Otgontamir, M. H. Sharif, M. S. Mina, S. Y. Kim, J. Kim, High efficiency CZTSSe solar cells enabled by dual Ag-passivation approach via aqueous solution process, *Journal of Energy Chemistry* 77 (2023) 239–246.
- [63] S. B. Ivriq, M. H. Mohammadi, R. S. Davidsen, Enhancing photovoltaic efficiency in half-tandem MAPbI₃/MASnI₃ perovskite solar cells with triple core-shell plasmonic nanoparticles, *Scientific Reports* 15 (1) (2025) 1478.
- [64] J. Xu, J. Wu, Q. Chen, Y. Wang, R. Li, X. Chen, Z. Lan, W. Sun, Spiro-OMeTAD doped with iodine pentoxide to enhance planar perovskite solar cell performance, *Journal of Alloys and Compounds* 970 (2024) 172749.
- [65] I. S. Jin, J. H. Lee, Y. W. Noh, S. H. Park, J. W. Jung, Molecular doping of cuscN for hole transporting layers in inverted-type planar perovskite solar cells, *Inorg. Chem. Front.* 6 (2019) 2158–2166.
- [66] A. Klasen, P. Baumli, Q. Sheng, E. Johannes, S. A. Bretschneider, I. M. Hermes, V. W. Bergmann, C. Gort, A. Axt, S. A. Weber, et al., Removal of surface oxygen vacancies increases conductance through TiO₂ thin films for perovskite solar cells, *The Journal of Physical Chemistry C* 123 (22) (2019) 13458–13466.
- [67] S. Li, M. A. Lloyd, B. E. McCandless, J. B. Baxter, Effects of cation composition on carrier dynamics and photovoltaic performance in Cu₂ZnSnS₄ monocrystal solar cells, *Solar Energy Materials and Solar Cells* 205 (2020) 110255.
- [68] C. Liu, R. Lin, Y. Wang, H. Gao, P. Wu, H. Luo, X. Zheng, B. Tang, Z. Huang, H. Sun, et al., Efficient all-perovskite tandem solar cells with low-optical-loss carbazolyl interconnecting layers, *Angewandte Chemie International Edition* 62 (51) (2023) e202313374.
- [69] H. Sun, K. Xiao, H. Gao, C. Duan, S. Zhao, J. Wen, Y. Wang, R. Lin, X. Zheng, H. Luo, et al., Scalable solution-processed hybrid electron transport layers for efficient all-perovskite tandem solar modules, *Advanced Materials* 36 (2) (2024) 2308706.

- [70] Y. Pan, J. Wang, Z. Sun, J. Zhang, Z. Zhou, C. Shi, S. Liu, F. Ren, R. Chen, Y. Cai, H. Sun, B. Liu, Z. Zhang, Z. Zhao, Z. Cai, X. Qin, Z. Zhao, Y. Ji, N. Li, W. Huang, Z. Liu, W. Chen, Surface chemical polishing and passivation minimize non-radiative recombination for all-perovskite tandem solar cells, *Nature Communications* 15 (2024) 7335.
- [71] J. Wang, L. Zeng, D. Zhang, A. Maxwell, H. Chen, K. Datta, A. Caiazzo, W. H. Remmerswaal, N. R. Schipper, Z. Chen, et al., Halide homogenization for low energy loss in 2-ev-bandgap perovskites and increased efficiency in all-perovskite triple-junction solar cells, *Nature Energy* 9 (1) (2024) 70–80.
- [72] F. Xu, E. Aydin, J. Liu, E. Ugur, G. T. Harrison, L. Xu, B. Vishal, B. K. Yildirim, M. Wang, R. Ali, et al., Monolithic perovskite/perovskite/silicon triple-junction solar cells with cation double displacement enabled 2.0 ev perovskites, *Joule* 8 (1) (2024) 224–240.
- [73] W. Li, W. Cao, H. Zhou, X. Zhang, K. Wang, An amorphous MgF_2 anti-reflective thin film for enhanced performance of inverted organic–inorganic perovskite solar cells, *RSC Advances* 14 (2024) 2757–2762.
- [74] M. Ahmadipour, W. K. Cheah, M. F. Ain, K. V. Rao, Z. A. Ahmad, Effects of deposition temperatures and substrates on microstructure and optical properties of sputtered CCTO thin film, *Materials Letters* 210 (2018) 4–7.
- [75] X. Wang, J. Yang, J. Zhong, J. Yu, X. Pan, Innovative materials for high-performance tin-based perovskite solar cells: A review, *Polymers* 16 (2024) 3053.
- [76] N. Arora, M. I. Dar, A. Hinderhofer, N. Pellet, F. Schreiber, S. M. Za-keeruddin, M. Grätzel, Perovskite solar cells with cusen hole extraction layers yield stabilized efficiencies greater than 20%, *Science* 358 (2017) 768–771.
- [77] K. A. Bush, A. F. Palmstrom, Z. J. Yu, M. Boccard, R. Cheacharoen, J. P. Mailoa, D. P. McMeekin, R. L. Z. Hoyer, C. D. Bailie, T. Lei-jtens, I. M. Peters, M. C. Minichetti, N. Rolston, R. Prasanna, S. Sofia, D. Harwood, W. Ma, F. Moghadam, H. J. Snaith, T. Buonassisi, Z. C.

- Holman, S. F. Bent, M. D. McGehee, 23.6%-efficient monolithic perovskite/silicon tandem solar cells with improved stability, *Nature Energy* 2 (2017) 17009.
- [78] T. Wu, X. Liu, X. Luo, H. Segawa, G. Tong, Y. Zhang, L. K. Ono, Y. Qi, L. Han, Heterogeneous FASnI_3 absorber with enhanced electric field for high-performance lead-free perovskite solar cells, *Nano-Micro Letters* 14 (2022) 99.
- [79] E. Kasparavicius, M. Franckevičius, V. Malinauskiene, K. Genevičius, V. Getautis, T. Malinauskas, Oxidized Spiro-OMeTAD: Investigation of stability in contact with various perovskite compositions, *ACS Applied Energy Materials* 4 (2021) 13696–13705.
- [80] V. Hernández-Calderón, O. Vigil-Galán, M. Guc, A. Carrillo-Osuna, S. Ramírez-Velasco, F. J. Sánchez-Rodríguez, P. Vidal-Fuentes, S. Giraldo, E. Saucedo, Y. Sánchez, CdS/ZnS bilayer thin films used as buffer layer in 10%-efficient $\text{Cu}_2\text{ZnSnSe}_4$ solar cells, *ACS Applied Energy Materials* 3 (2020) 6815–6823.
- [81] J. Shi, J. Wang, F. Meng, J. Zhou, X. Xu, K. Yin, L. Lou, M. Jiao, B. Zhang, H. Wu, Y. Luo, D. Li, Q. Meng, Multinary alloying for facilitated cation exchange and suppressed defect formation in kesterite solar cells with above 14% certified efficiency, *Nature Energy* 9 (9) (2024) 1095–1104.

Supplementary Material: Unveiling architectural and optoelectronic synergies in lead-free perovskite/perovskite/kesterite triple-junction monolithic tandem solar cells

Md. Faiaad Rahman^a, Md. Ashaduzzaman Niloy^{a,b}, Ehsanur Rahman^{a,*},
Ahmed Zubair^{a,*}

^a*Department of Electrical and Electronic Engineering, Bangladesh University of Engineering and Technology, Dhaka, Dhaka-1205, Bangladesh*

^b*Department of Electrical and Electronic Engineering, Green University of Bangladesh, Dhaka, Narayanganj-1461, Bangladesh*

Contents

S1 Simulation methodology	S2
S2 Optimization procedure and electrical parameters	S5
S3 Optimization results: from single junction to triple junction	S6
S3.1 Single Junction KSnI ₃ Cell	S6
S3.1.1 Thickness optimization of layers	S6
S3.1.2 Doping optimization of ETL and HTL	S9
S3.1.3 Impact of MgF ₂ ARC thickness	S10
S3.1.4 Incorporation of UV-ozone treated ETL and Au BCL .	S11
S3.2 Single Junction FASnI ₃ Cell	S12
S3.2.1 Thickness optimization of layers	S12
S3.2.2 Incorporation of Spiro-OMeTAD HTL	S15
S3.3 Dual Junction KSnI ₃ /FASnI ₃ Tandem Cell	S16
S3.4 Triple Junction KSnI ₃ /FASnI ₃ /ACZTSe Tandem Cell	S17
S3.4.1 Impact of ACZTSe doping on 3-J tandem cell.	S17
S3.4.2 Volumetric thickness optimization of absorbers	S18

*Corresponding authors:

Email addresses: ehsaneee@eee.buet.ac.bd (Ehsanur Rahman),
ahmedzubair@eee.buet.ac.bd (Ahmed Zubair)

S1. Simulation methodology

First, absorption and optical generation needs to be calculated by solving Maxwell's curl electromagnetic wave equation using Finite Difference Time Domain (FDTD) analysis for optical electric field distribution in different layers:

$$\frac{\partial \vec{D}}{\partial t} = \nabla \times \vec{H} \quad (1)$$

$$\vec{D}(\omega) = \epsilon_o \epsilon_r(\vec{r}, \omega) \vec{E}_{op} \quad (2)$$

$$\frac{\partial \vec{H}}{\partial t} = -\frac{1}{\mu_o} \nabla \times \vec{E}_{op} \quad (3)$$

Where, $\vec{H}$, $\vec{E}_{op}$ and $\vec{D}(\omega)$ are the magnetic, electric and displacement fields respectively. $\epsilon_r(\vec{r}, \omega)$ is the complex relative dielectric constant, where, ω is the angular frequency. Each material has been modeled according to its respective refractive index (n) and extinction coefficient (k). The absorbed power (P_{abs}) is then calculated by the following equation:

$$P_{abs} = -\frac{1}{2} \omega |\vec{E}_{op}(\vec{r}, \omega)|^2 \Im\{\epsilon(\vec{r}, \omega)\} \quad (4)$$

Afterwards, the generation rate is calculated as:

$$G(\vec{r}) = \int g(\vec{r}, \omega) d\omega \quad (5)$$

$$g(\vec{r}, \omega) = \frac{P_{abs}}{\hbar \omega} = -\frac{\pi}{\hbar} |\vec{E}_{op}(\vec{r}, \omega)|^2 \Im\{\epsilon(\vec{r}, \omega)\} \quad (6)$$

Where, h is the Planck's constant. In X-axis, periodic boundary condition and in Y-axis, perfectly matched layer (PML) boundary condition is maintained. To perform optical simulation, the AM 1.5G solar spectrum has been set as the input irradiation source.

Performance metrics like efficiency (η), open circuit voltage (V_{oc}), short circuit current (J_{sc}), and fill factor FF can be found from the J-V characteristics of each cell independently by solving Poisson's equation, drift-diffusion equations, and continuity equations as mentioned below:

$$-\nabla \cdot (\epsilon_{dc} \nabla V) = q\rho \quad (7)$$

$$\vec{J}_n = q\mu_n n \vec{E} + qD_n \nabla n \quad (8)$$

$$\vec{J}_p = q\mu_p p \vec{E} - qD_p \nabla p \quad (9)$$

$$\frac{\partial n}{\partial t} = \frac{1}{q} \nabla \cdot \vec{J}_n - R_n \quad (10)$$

$$\frac{\partial p}{\partial t} = -\frac{1}{q} \nabla \cdot \vec{J}_p - R_p \quad (11)$$

Where, ϵ_{dc} is the dielectric dc permittivity, V is the electrostatic potential (electric field, $\vec{E} = -\nabla V$), ρ is the net charge density ($\rho = p - n + C$, C =ionized impurity density), $\vec{J}_{n(p)}$ is the electron (hole) current density, q is the positive electron charge, $\mu_{n(p)}$ is the mobility of electron (hole), $D_{n(p)}$ is the diffusivity of electron (hole) ($D_{n(p)} = \mu_{n(p)} \frac{k_B T}{q}$), n and p are electron and hole densities respectively, $R_{n(p)}$ is the net recombination rate, k_B is the Boltzmann constant and T is the temperature. The computed generation rate retrieved from the optical simulation is then employed as an input in the continuity equations, and eqs. (7) to (11) are solved by implementing Dirichlet boundary conditions at contacts, and Neumann boundary conditions at the insulating boundaries and interfaces, respectively.

To quantify the charge collection efficiency, the external quantum efficiency (EQE) is evaluated as the ratio between the photocurrent density and the incident photon flux at each wavelength. Under illumination, EQE at a given wavelength λ_0 is calculated using:

$$EQE(\lambda_0) = \frac{J(\lambda_0)}{q \cdot \Phi_{inc}(\lambda_0)} \quad (12)$$

whereas, $J(\lambda_0)$ is the photocurrent density (mA/cm²), $\phi(\lambda_0)$ is the incident photon flux density (photons/cm²·s), q is the elementary charge. Again, the photon flux ϕ_λ for a given wavelength, λ_0 (nm) can be expressed as:

$$\Phi_{inc}(\lambda) = \frac{P_{inc}(\lambda)}{hc/\lambda} = \frac{P_{inc}(\lambda) \cdot \lambda}{hc} \quad (13)$$

Combining the above, the EQE becomes:

$$EQE(\lambda) = \frac{hc}{q\lambda} \cdot \frac{J(\lambda)}{P_{inc}(\lambda)} \quad (14)$$

The obtained EQE is a unitless quantity. Unlike EQE, IQE considers only those photons that are absorbed in the photoactive layer, $P_{abs}^{absorber}$, excluding

optical losses due to reflection, transmission, and parasitic absorption in non-active layers (e.g., electrodes, transport layers). IQE is defined at a specific wavelength λ_0 as:

$$IQE(\lambda_0) = \frac{EQE(\lambda_0)}{A(\lambda_0)} \quad (15)$$

$$A(\lambda_0) = \frac{P_{abs}^{absorber}(\lambda_0)}{P_{inc}(\lambda_0)} \quad (16)$$

Here, $A(\lambda_0)$ is the absorbance of the photoactive layer at given wavelength λ_0 . These overall frameworks allow assessment of device performance under spectrally resolved illumination and are essential for optimizing the design of high-efficiency single junction solar cells.

In the case of calculating the performance parameters of a tandem solar cell, the short-circuit current density is determined by the subcell that delivers the lowest current density, due to the monolithic architecture:

$$J_{sc}^{Tandem} = \min(J_{sc}^1, J_{sc}^2, \dots, J_{sc}^m) \quad (17)$$

The open-circuit voltage, V_{oc} of the tandem cell is the sum of the open-circuit voltages of all subcells if the current density J is the same in all the subcells.

$$V_{oc}^{Tandem} = V_{oc}^1 + V_{oc}^2 + \dots + V_{oc}^m \quad (18)$$

Alternatively, at a matched current J , the total operating voltage of the tandem cell is given by:

$$V_{Tandem}(J) = V_1(J) + V_2(J) + \dots + V_m(J) \quad (19)$$

Here, m denotes the number of subcells in the tandem configuration, and $V_i(J)$ is the voltage of subcell i when operating at the common current J .

S2. Optimization procedure and electrical parameters

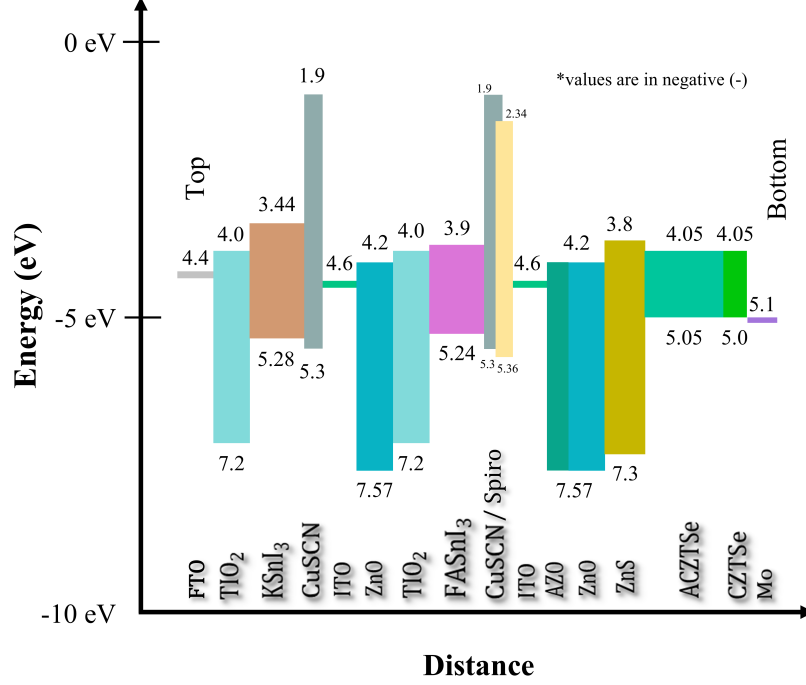


Figure S1: Energy (eV) vs space diagram of proposed 2-T monolithic triple junction tandem solar cell architecture.

For the modeling and simulation of the solar cell architectures, we considered the work functions of FTO and ITO as 4.4 eV and 4.6 eV, respectively.

Table S1: Electrical parameters of TiO₂, ZnS, ZnO, AZO, CuSCN and Spiro-OMeTAD.

Parameters	TiO ₂ [52]	ZnS [55, 56]	ZnO [55–57]	AZO [55, 56]	CuSCN [52]	Spiro-OMeTAD [63, 64]
Thickness (nm)	20–150	20–100	20–100	10	20–150	20–150
Bandgap E_g (eV)	3.2	3.58	3.37	3.37	3.4	3.02
DC permittivity ϵ	9	9	9	9	10	3.2
Electron affinity χ (eV)	4.0	3.8	4.0	4.0	1.9	2.34
Mobility μ_n/μ_p (cm ² /Vs)	20 / 10	230 / 40	150 / 50	50 / 5	1×10^{-4} / 1×10^{-2}	2×10^{-4} / 2×10^{-4}
SRH lifetime τ_e/τ_h (ns)	5 / 2	0.5 / 0.5	1 / 1	–	5 / 5	5 / 5
Radiative recomb. (cm ³ s ⁻¹)	–	1.5×10^{-10}	–	–	–	–
Effective conduction band density, N_C (cm ⁻³)	1×10^{19}	2.7×10^{18}	2.2×10^{18}	2.2×10^{18}	2.51×10^{19}	1×10^{19}
Effective valence band density, N_V (cm ⁻³)	1×10^{19}	1.7×10^{19}	1.8×10^{19}	1.9×10^{19}	1.79×10^{19}	1×10^{19}
Donor doping N_D (cm ⁻³)	5×10^{18}	5×10^{16}	1.5×10^{17}	8×10^{18}	–	–
Acceptor doping N_A (cm ⁻³)	–	1.79×10^{19}	–	1×10^{15}	5×10^{18}	2×10^{18}
Surface Recombination (cm/s)	(FTO/TiO ₂) & (MgF ₂ /TiO ₂) 1×10^7	–	–	(AZO/ITO) 1×10^7	–	–

S3. Optimization results: from single junction to triple junction

S3.1. Single Junction KSnI_3 Cell

S3.1.1. Thickness optimization of layers

To establish the baseline device behavior, we first performed a preliminary thickness sweep of the KSnI_3 absorber from 200 to 1200 nm, keeping TiO_2 ETL, and CuSCN HTL layer thickness fixed. As shown in Fig. S2(b), increasing the KSnI_3 absorber thickness from 150 to 1200 nm led to a steady rise in J_{sc} due to enhanced optical absorption and improved photon harvesting within the bulk region. However, as the absorber thickness increases, carriers generated deeper inside the absorber experience longer transport paths and higher recombination probability, especially beyond 750 nm for our case. This resulted in a gradual decrease in V_{oc} and FF as shown in Figs. S2(c) and (d), attributed to increased bulk recombination and reduced internal electric field strength that hindered efficient carrier extraction. Consequently, the

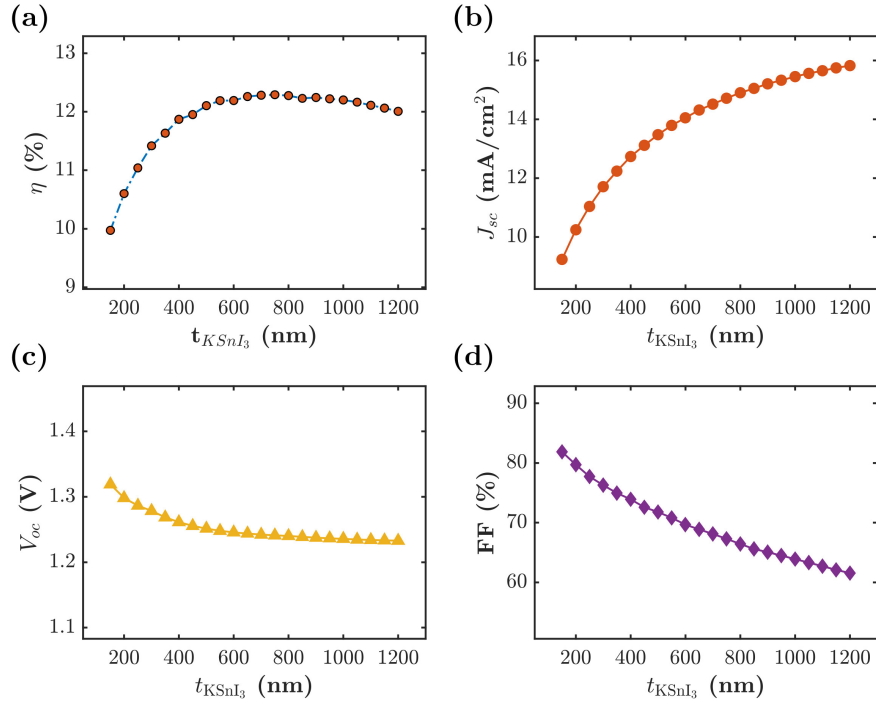


Figure S2: Initial iterative analysis showing the impact on performance metrics—(a) η , (b) J_{sc} , (c) V_{oc} , and (d) FF—of the single-junction KSnI_3 cell corresponding to the thickness variation of the KSnI_3 absorber. The peak PCE (η) was obtained at a thickness of 750 nm.

PCE (η) exhibited a saturation trend, reaching its peak value of 12.29% at 750 nm, where the trade-off between optical absorption and carrier transport was best balanced.

Keeping the KSnI_3 absorber fixed at 750 nm, we analyzed the influence of ETL (TiO_2) and HTL (CuSCN) thicknesses on device performance as shown in Fig. S3(a-d). From Figs. S3(a) and (b), increasing the TiO_2 thickness from 40 to 60 nm improved electron extraction by providing more uniform coverage and better contact at the ETL/absorber interface, reducing interface recombination and enhancing J_{sc} and PCE. Beyond 60 nm, further thickening slightly increased the carrier transport path and series resistance, leading to marginal reductions in performance. For the HTL, thinner CuSCN layers led

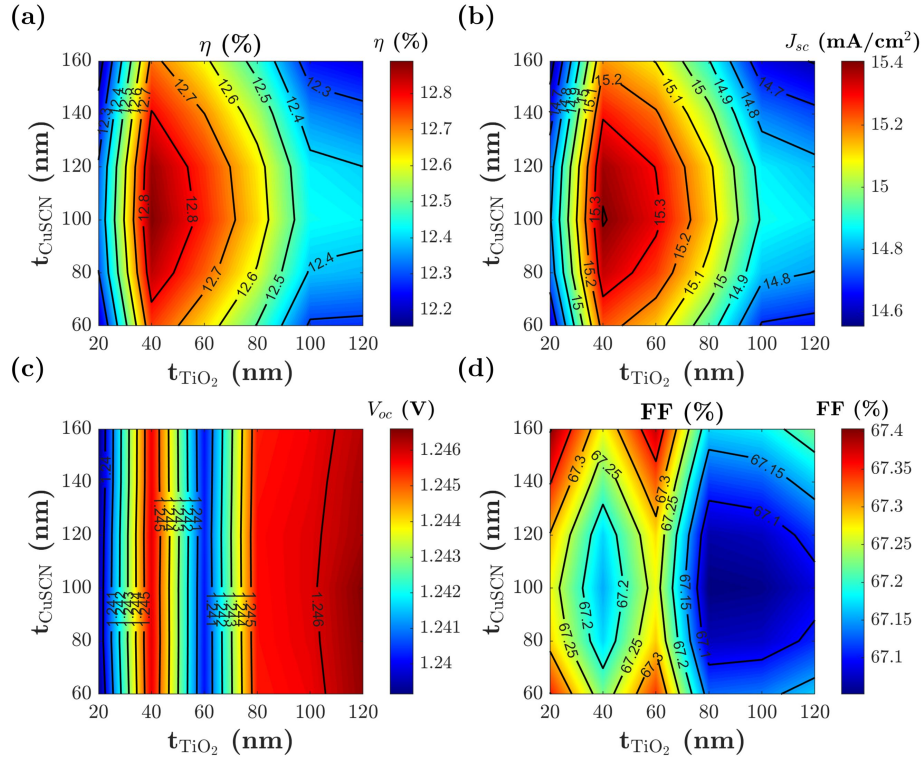


Figure S3: Initial parametric sweep showing the impact on performance metrics—(a) η , (b) J_{sc} , (c) V_{oc} , and (d) FF of 1-J KSnI_3 cell corresponding to the thickness variation of TiO_2 ETL and CuSCN HTL layers. The peak η was secured 12.89% for 40 nm thickness of TiO_2 ETL and 100 nm CuSCN HTL.

to insufficient hole extraction and higher recombination at the absorber/HTL interface, whereas thicker layers increased the transport distance for holes, slightly reducing V_{oc} and FF due to slower charge collection, as seen from Fig. S3(c) and (d). The highest PCE (η) of 12.89% was obtained for TiO_2 of 40 nm and CuSCN of 100 nm, reflecting a balance between effective carrier extraction, minimized interface recombination, and favorable electric field distribution across the junction.

The final iterative analysis of KSnI_3 thickness, as shown in Fig. S4, confirms the trends from our preliminary study. Increasing the absorber up to 700 nm steadily improves J_{sc} and PCE, while further thickening offers only marginal J_{sc} gains that are offset by additional recombination losses and resistive effects, resulting in a modest decrease in PCE. The maximum PCE of 12.95% was obtained for KSnI_3 thickness of 700 nm.

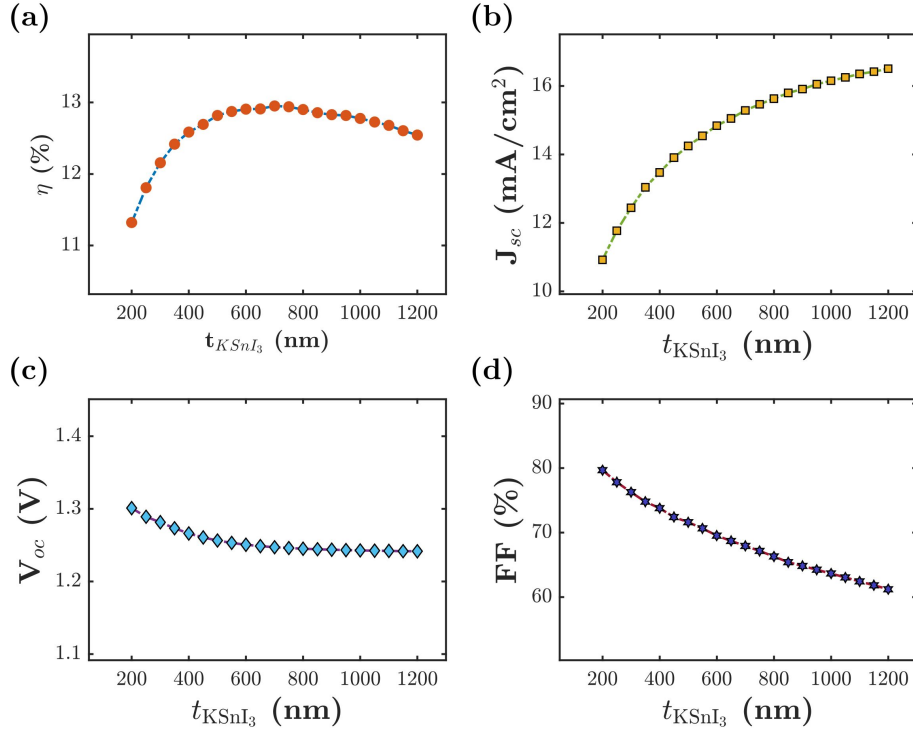


Figure S4: Final iterative analysis showing the impact on performance metrics—(a) PCE (η), (b) J_{sc} , (c) V_{oc} , and (d) FF—of the single-junction KSnI_3 cell corresponding to the thickness variation of the KSnI_3 absorber. The peak PCE is obtained at a thickness of 700 nm.

is achieved at 700 nm, aligning with our preliminary estimate.

S3.1.2. Doping optimization of ETL and HTL

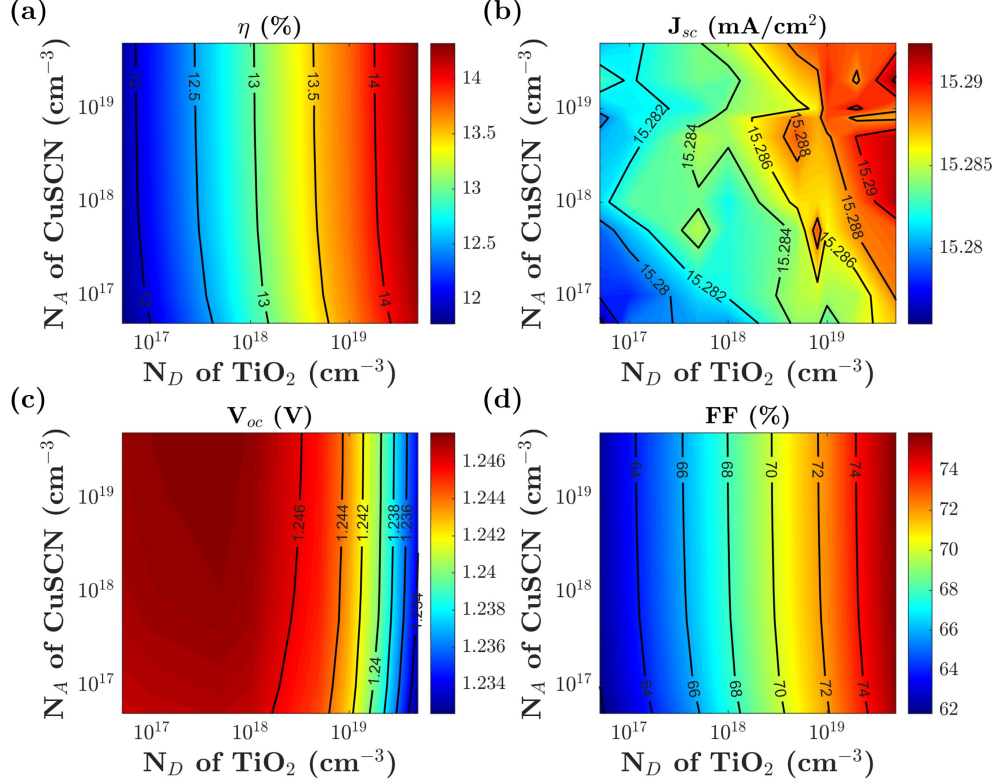


Figure S5: Contour plot of parametric sweep of the performance metrics – (a) η , (b) J_{sc} , (c) V_{oc} , and (d) FF of 1-J KSnI₃ cell corresponding to variation of donor density, N_D of TiO_2 ETL and acceptor density, N_A of CuSCN HTL.

We varied the donor doping density, N_D in the TiO_2 ETL from 5×10^{16} to $5 \times 10^{19} \text{ cm}^{-3}$, while we adjusted the acceptor doping density, N_A in the CuSCN HTL over the same range, from 5×10^{16} to $5 \times 10^{19} \text{ cm}^{-3}$. The results indicate that increasing the acceptor doping in CuSCN and donor doping in TiO_2 improves the overall device performance. Higher doping enhances the built-in electric field, promoting more efficient carrier drift and reducing recombination losses, which is reflected in the gradual increase of PCE, J_{sc} , V_{oc} , and FF. The maximum PCE of 14.32% is achieved for TiO_2 ETL and

CuSCN HTL doping densities of $5 \times 10^{19} \text{ cm}^{-3}$, demonstrating the critical role of optimized doping in facilitating charge extraction and improving optical-to-electrical conversion in the KSnI₃ solar cell.

S3.1.3. Impact of MgF₂ ARC thickness

To reduce the optical reflection, we varied the thickness of MgF₂ ARC from 20 to 150 nm. A minimum average reflection of 4.48% has been attained for photon wavelength of 300-700 nm range as depicted from Fig. S6(a). This reduced reflection boost the carrier-generation rate, resulting increase in PCE from 14.31% to 14.51%.

As shown in Figs. S6(c-e), the spatial distribution of the electric field and the band diagram collectively illustrate the charge transport mechanism across the FTO/TiO₂/KSnI₃/CuSCN/Au stack. The electric field profile exhibits a pronounced peak at the KSnI₃/CuSCN interface, originating from the built-in potential, V_{bi} between the p-type CuSCN and the intrinsic KSnI₃ ab-

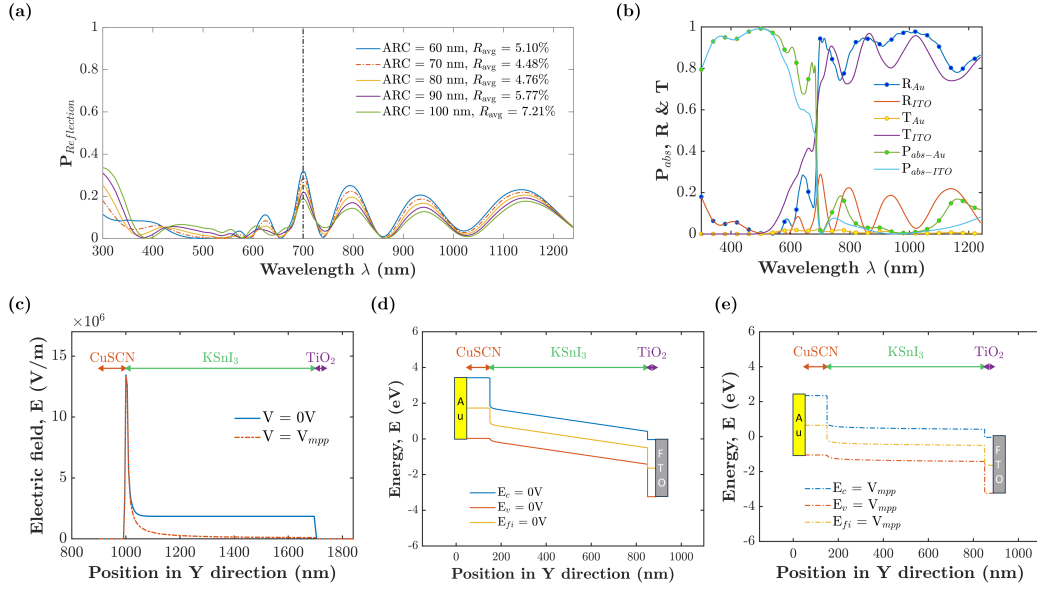


Figure S6: (a) Impact on overall reflection from 1-J KSnI₃ cell via altering MgF₂ ARC thickness. (b) Normalized power absorption (P_{abs}), reflection (R), and transmission (T) corresponding to photon wavelength (λ). (c) Spatial E-field profile, and (d) band-diagram of ITO/TiO₂/KSnI₃/CuSCN/Au corresponding to device thickness at thermal equilibrium at short circuit condition ($V = 0V$) and maximum power point ($V = V_{mpp}$).

sorber. This high-field region corresponds to the strong band bending seen in the thermal-equilibrium band diagram, which drives drift-dominated carrier transport, promoting efficient hole extraction toward CuSCN. Across the KSnI₃ layer, the field gradually diminishes, consistent with the band flattening that leads to a diffusion-dominated regime governed by carrier concentration gradients. Near the TiO₂/KSnI₃ interface, the bands align smoothly with a weak and flat field, indicating minimal energy barriers for electron extraction. Under operating bias at $V = V_{\text{mpp}}$, partial band flattening and a reduced field magnitude confirm the transition toward balanced drift–diffusion transport, ensuring efficient charge collection and limited interfacial recombination.

S3.1.4. Incorporation of UV-ozone treated ETL and Au BCL

At the FTO/TiO₂/KSnI₃ interface, the conduction band of TiO₂ lies above that of KSnI₃, having a conduction band offset (CBO) of ≈ 0.56 eV, enabling efficient electron extraction while blocking holes. To further optimize this interface, UV-ozone treatment was applied to TiO₂, lowering its work function from 5.3 eV to 5.1 eV. This reduced interfacial recombination by mitigating surface defects and improving electron transport, resulting in enhanced charge collection, achieving a PCE of 15.30% as shown in Table S2.

Incorporating Au as the back contact layer (BCL) enhances photon absorption in the 550–690 nm range via back-reflection, increasing carrier generation and collection. This leads to higher J_{sc} and an overall 8.6% relative improvement in PCE, reaching 16.28% for the optimized 1-J KSnI₃ cell.

Table S2: Impact of UV-ozone treated TiO₂ ETL on single-junction KSnI₃ solar cell performance.

Back Contact Layer (BCL)	TiO ₂ ETL	ϕ_{TiO_2} (eV)	η (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)
ITO	Oxidized	5.3	14.53	15.62	1.235	75.37
ITO	UV-ozone treated	5.1	15.30	15.62	1.228	79.79
Au	UV-ozone treated	5.1	16.28	16.62	1.232	79.50

S3.2. Single Junction FASnI_3 Cell

S3.2.1. Thickness optimization of layers

To identify performance-sensitive regions and optimize the 1-J FASnI_3 cell ($\text{MgF}_2/\text{ITO}/\text{ZnO}/\text{FASnI}_3/\text{CuSCN}/\text{ITO}$), we initialized an iterative thickness sweep of the FASnI_3 absorber from 300 to 1200 nm as shown in Fig. S7. Increasing the absorber thickness enhances photon absorption across the visible–NIR spectrum, raising J_{sc} from 24.52 to 28.53 mA/cm^2 , which saturates around 900 nm, indicating efficient collection of photogenerated carriers at the selective contacts. As the absorber thickens, longer carrier diffusion paths increase bulk recombination, leading to a gradual reduction in V_{oc} and FF as noticed in Fig. S7(c–d). Consequently, the PCE rises with thickness, reaching a maximum of 21.69% at 900 nm, beyond which the trade-off between enhanced absorption and recombination causes a slight decline.

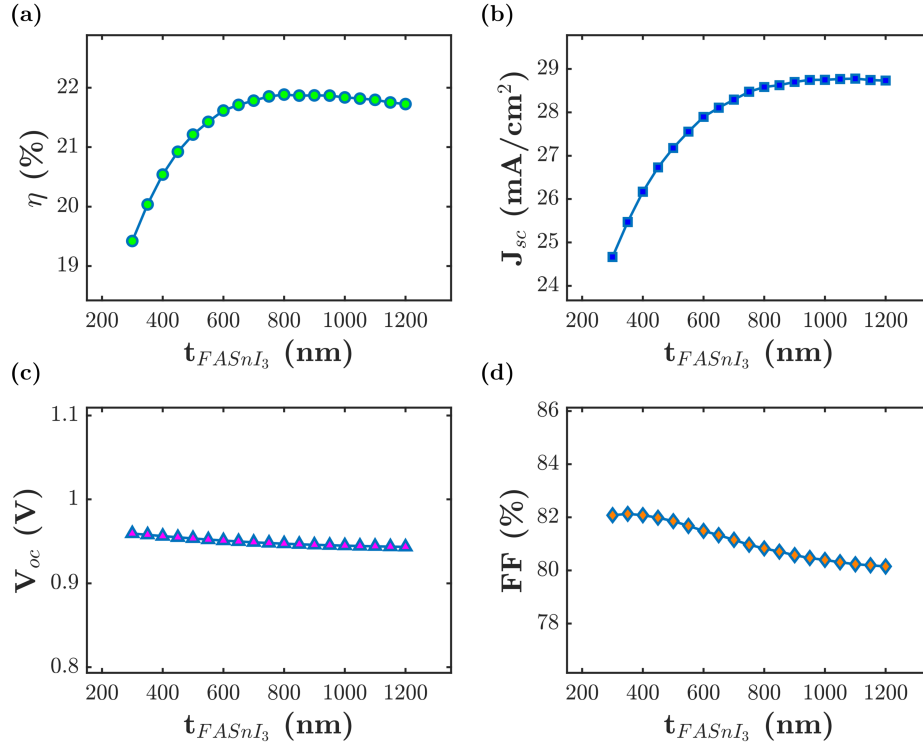


Figure S7: Initial iterative analysis showing the impact on performance metrics—(a) η , (b) J_{sc} , (c) V_{oc} , and (d) FF of 1-J FASnI_3 cell corresponding to the initial thickness variation of FASnI_3 absorber.

After optimizing the FASnI_3 absorber at 900 nm, we performed iterative thickness sweeps of the TiO_2 ETL from 20 to 160 nm and CuSCN HTL from 60 to 200 nm as featured in Fig. S8. The analysis shows that a thin ETL of 20 nm is sufficient for efficient electron extraction, minimizing series resistance and preserving the built-in field, whereas a thicker HTL in 160–180 nm range improves hole collection and reduces interfacial recombination at the back contact. J_{sc} remains nearly saturated, and both V_{oc} and FF exhibit minor variations, indicating that the absorber governs photo-generation while the transport layers optimize charge extraction. The peak PCE of 21.87% is achieved for this configuration, confirming that thinner ETL and thicker HTL layers provide optimal carrier collection without compromising overall device efficiency.

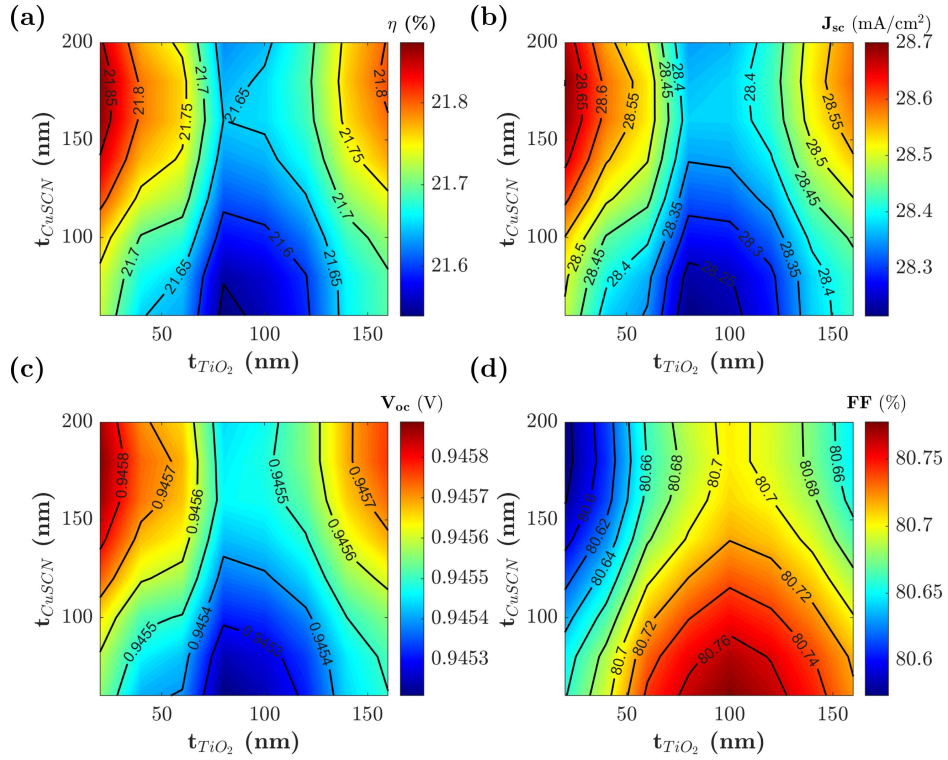
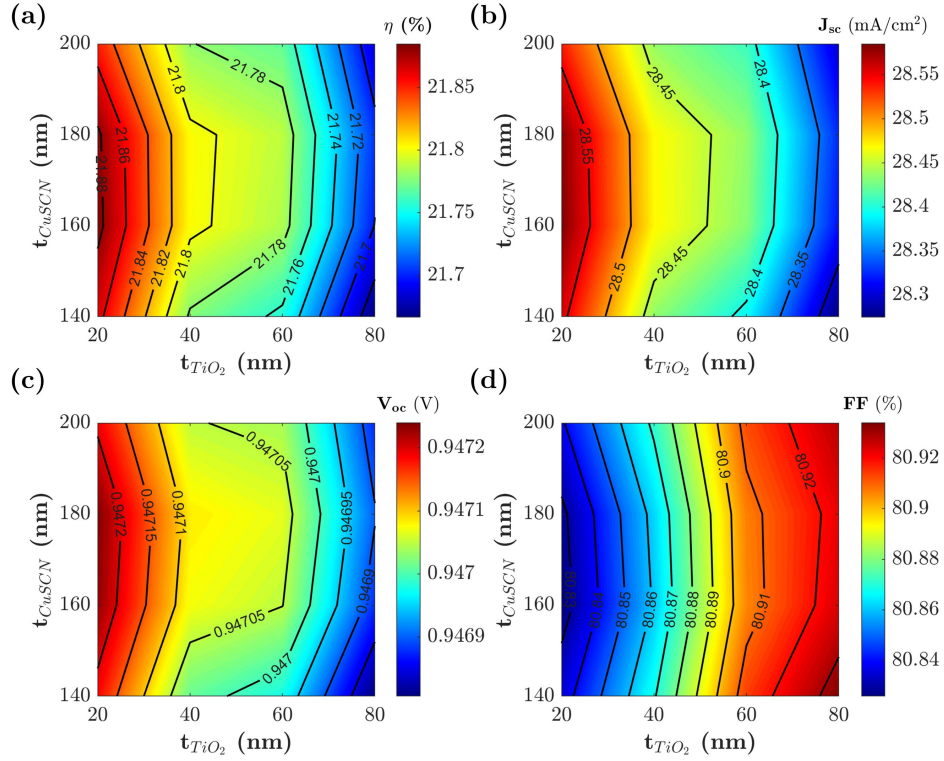


Figure S8: Initial parametric sweep showing the impact on performance metrics—(a) η , (b) J_{sc} , (c) V_{oc} , and (d) FF of 1-J FASnI_3 cell—by varying thickness of TiO_2 ETL and CuSCN HTL layers for ITO/ ZnO / TiO_2 / FASnI_3 / CuSCN /ITO architecture.

After obtaining optimum FASnI₃ absorber thickness of 800 nm, the final parametric sweep of transport layers demonstrates that the device performance exhibits a clear plateau across variations in TiO₂ ETL and CuSCN HTL thickness. The maximum performance was achieved for 20 nm and 160 nm thickness of TiO₂ ETL and CuSCN HTL, confirming that the simulation has converged to a self-consistent optimum. The performance matrices shows minimal changes beyond these ETL and HTL values, indicating that the device has reached an optimized state where optical absorption, carrier generation, and drift–diffusion transport are balanced.



S3.2.2. Incorporation of Spiro-OMeTAD HTL

Fig. S10 presents the band diagrams of the 1-J FASnI₃ cell (ITO/ZnO/FASnI₃/HTL/Au) using CuSCN and Spiro-OMeTAD as HTLs at short-circuit condition ($V = 0$ V) and maximum power point condition ($V = V_{mpp}$). The FASnI₃/Spiro-OMeTAD interface exhibits a slightly negative VBO ≈ -0.06 eV, providing a downhill potential that promotes hole drift toward the Au contact while maintaining a sufficient CBO ≈ 1.56 eV, to suppress electron back-injection. In contrast, the FASnI₃/CuSCN interface shows a nearly flat VBO of ≈ 0 eV, resulting in a weaker hole driving potential despite its larger CBO of ≈ 2.0 eV. The thinner Spiro-OMeTAD layer of 70 nm, induces a

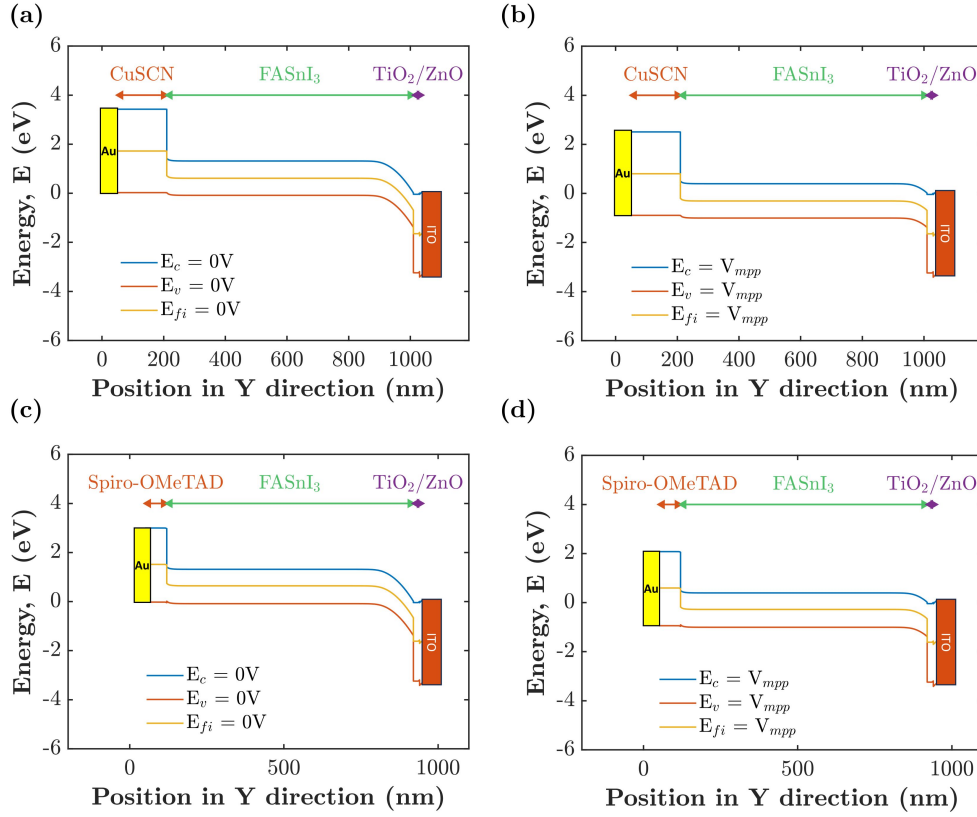


Figure S10: Band-diagram of ITO/ZnO/TiO₂/FASnI₃/CuSCN/Au at thermal equilibrium at (a) short circuit condition ($V = 0$ V) and (b) maximum power point condition ($V = V_{mpp}$). Band-diagram of ITO/ZnO/TiO₂/FASnI₃/Spiro-OMeTAD/Au at thermal equilibrium at (c) short-circuit condition ($V = 0$ V) and (d) maximum power point ($V = V_{mpp}$).

stronger built-in electric field, enhancing hole drift and reducing recombination, whereas the thicker CuSCN layer of 160 nm, increases series resistance and diffusion length, leading to higher carrier loss. Consequently, the Spiro-OMeTAD-based cell exhibits stronger band bending and quasi-Fermi level splitting, confirming more efficient field-assisted charge extraction and superior photovoltaic performance.

S3.3. Dual Junction $\text{KSnI}_3/\text{FASnI}_3$ Tandem Cell

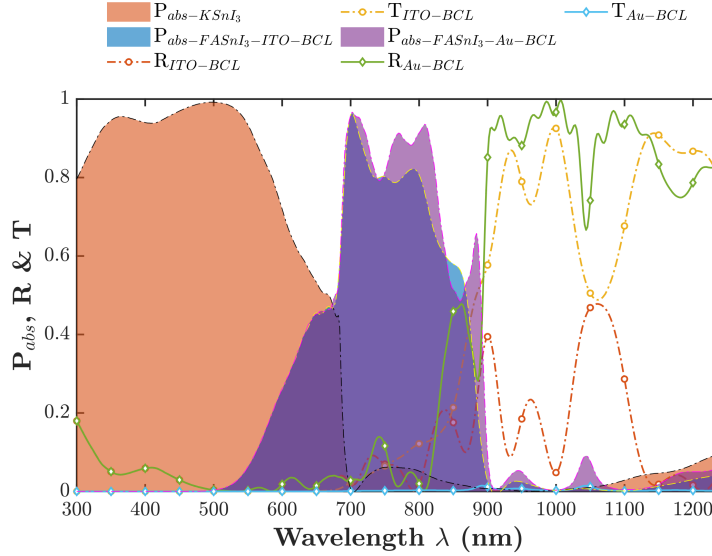


Figure S11: Normalized power absorption (P_{abs}), reflection (R), and transmission (T) as a function of photon wavelength (λ), illustrating the effect of Au and ITO back contacts on bottom absorber (FASnI_3) absorption, total device reflection, and transmission through the bottom cell in the 2-J $\text{KSnI}_3/\text{FASnI}_3$ tandem configuration.

Table S3: Impact of incorporating ITO instead of Au as BCL in 2-J bottom subcell's BCL.

Bottom subcell's BCL	η (%)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)	P_{mpp} (W/m^2)	V_{mpp} (V)
Au BCL	27.29	14.74	2.227	83.14	272.89	1.981
ITO BCL	24.89	12.97	2.224	86.31	248.89	2.012

S3.4. Triple Junction $\text{KSnI}_3/\text{FASnI}_3/\text{ACZTSe}$ Tandem Cell

To construct the initial triple-junction (3-J) tandem architecture, the bottom subcell was designed as AZO/ZnO/ZnS/ACZTSe/CZTSe/Mo, while the optimized 1-J KSnI_3 and 1-J FASnI_3 layers were employed as the top and middle subcell, respectively. The initial layer thicknesses and doping concentrations used for each subcell are summarized in Table S4.

Table S4: Initial thickness and doping density of KSnI_3 , FASnI_3 , and ACZTSe-based triple junction tandem solar cell.

Material	Thickness (nm)	Doping density (cm^{-3})	Doping type
FTO	100	—	—
MgF_2	90	—	—
TiO_2	40	5×10^{19}	n
KSnI_3	600	1×10^{15}	i
CuSCN	100	5×10^{18}	p
ITO	50	—	—
ZnO	10	8×10^{18}	n
TiO_2	20	5×10^{19}	n
FASnI_3	800	7×10^{16}	p
Spiro-OMeTAD	80	7×10^{18}	p
ITO	20	—	—
AZO	10	8×10^{18}	n
ZnO	60	1.5×10^{17}	n
ZnS	100	5×10^{16}	n
ACZTSe	400	5×10^{14}	p
CZTSe	200	1×10^{16}	p
Mo	100	—	—

S3.4.1. Impact of ACZTSe doping on 3-J tandem cell.

Table S5 summarizes the effect of ACZTSe doping concentration on the photovoltaic performance of the 3-J tandem cell. As the acceptor density increases from 5×10^{14} to $1 \times 10^{15} \text{ cm}^{-3}$, PCE improves slightly from 25.72% to 26.08%, primarily due to enhanced built-in potential and improved carrier

extraction, leading to higher V_{oc} and FF. Beyond this optimum range, further increase in doping reduces J_{sc} and overall efficiency owing to increased carrier scattering and recombination losses in the highly doped absorber. The optimal doping concentration for ACZTSe is identified at $9 \times 10^{14} \text{ cm}^{-3}$, where the balance between electric field strength and carrier transport yields maximum PCE.

Table S5: Impact of acceptor doping, N_A of ACZTSe on 3-J tandem cell.

ACZTSe acceptor density $N_A \text{ (cm}^{-3}\text{)}$	η (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	P_{mpp} (W/m ²)	V_{mpp} (V)
5.00×10^{14}	25.72	11.44	2.705	83.17	257.25	2.392
6.00×10^{14}	25.84	11.45	2.707	83.39	258.45	2.394
7.00×10^{14}	25.93	11.46	2.710	83.56	259.39	2.396
8.00×10^{14}	26.01	11.46	2.712	83.68	260.11	2.412
9.00×10^{14}	26.06	11.46	2.714	83.77	260.60	2.414
1.00×10^{15}	26.08	11.45	2.717	83.82	260.89	2.416
2.00×10^{15}	25.82	11.27	2.733	83.89	258.28	2.431
3.00×10^{15}	25.34	10.99	2.745	83.99	253.47	2.455
4.00×10^{15}	24.93	10.76	2.752	84.24	249.39	2.461
5.00×10^{15}	24.60	10.56	2.757	84.46	246.00	2.466

S3.4.2. Volumetric thickness optimization of absorbers

To identify the optimum configuration of the 3-J tandem device, we carried out an extensive volumetric optimization analysis, where the absorber thicknesses of KSnI₃, FASnI₃, and ACZTSe were systematically tuned to achieve optimal current matching and maximize device efficiency. Specifically, the KSnI₃ thickness was varied from 350 to 500 nm, and for each selected KSnI₃ thickness, a detailed two-dimensional sweep of FASnI₃ from 400 to 700 nm and ACZTSe from 850 to 1150 nm, was carried out. This comprehensive approach allowed us to capture the performance-sensitive regions and identify the most balanced combinations of subcell thicknesses. The summarized results in Table S6 present the best-performing configurations corresponding to each KSnI₃ region, highlighting the depth of our optimization analysis and the considerable effort devoted to achieving a finely tuned, high-efficiency 3-J tandem design. Fig. S12 features the contour plots of the parametric sweep, showing the variation of performance metrics with FASnI₃ and ACZTSe thicknesses while keeping the top cell's KSnI₃ thickness fixed at 400 nm. The plot highlights the most efficient region of the parameter space, where a champion PCE(η) of 30.69% was achieved.

Table S6: Tandem 3D thickness sweep (KSnI₃, FASnI₃ & ACZTSe)

KSnI ₃ (nm)	FASnI ₃ (nm)	ACZTSe (nm)	η (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	P_{mpp} (mW/cm ²)	V_{mpp} (V)
350	450	900	30.40	13.05	2.772	84.07	304.04	2.438
400	450	900	30.69	13.18	2.766	84.18	306.92	2.432
450	600	1100	29.99	13.45	2.751	81.04	299.94	2.405
500	550	1100	29.17	12.64	2.743	84.16	291.70	2.412
550	500	850	28.44	12.14	2.738	85.55	284.40	2.422

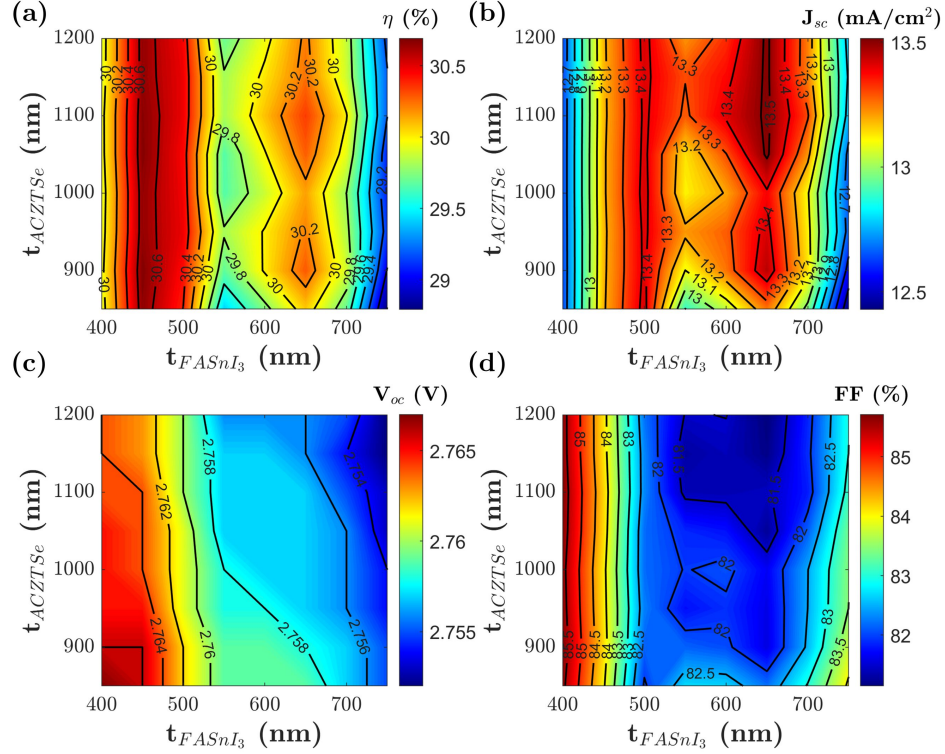


Figure S12: Contour plot of parametric sweep showing performance metrics—(a) η (b) J_{sc} (c) V_{oc} (d) FF corresponding to thickness variation of FASnI₃ and ACZTSe thickness keeping top cell's KSnI₃ absorber thickness 400 nm highlighting the optimized performance of the 3-J tandem cell with a champion power conversion efficiency (η) of 30.69%.