

Next-Generation Tandem Photovoltaics: Role of Multinary Compound Semiconductors in Efficiency Enhancement

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Abstract

Tandem cells offer a direct path around the spectral utilisation limit of the single-junction PV devices, by using two absorbers with complementary bandgaps to capture high- and low-energy photons. This study critically explores the numerical dataset of a SCAPS-1D perovskite model in order to investigate the potential for the multinary, lead-free perovskite absorbers for enhancing the performance of two terminal monolithic tandem perovskite solar cells. The top absorber of two double perovskite designs was either a narrow-bandgap crystalline-silicon heterojunction bottom cell with a 1.3-eV bandgap (CsSnI_3) or a wide-bandgap quaternary-substituted double perovskite with a bandgap of ~ 1.8 eV. The model is solved for one dimensional Poisson, carrier-continuity and drift-diffusion equations, in AM1.5G illumination at 300K. SCAPS-1D is not capable of solving a full multijunction device, so the sub-cells were calibrated separately, the spectrum transmitted through the top stack was calculated and the absorber thicknesses were swept so that the two sub-cells were close to a common short-circuit current. The perovskite/silicon design has been optimized to achieve $\text{JSC} = 0.181068 \text{ mA cm}^{-2}$ @ 550 nm (top-absorber) and $\text{VOC} = 0.1812 \text{ V}$ @ 230 μm (bottom-absorber), resulting in a FF of 0.7918 and a PCE of 0.2595. The all-perovskite design reached its optimum at 700 nm and 65 nm, respectively, with $\text{JSC} = 16.91 \text{ mA cm}^{-2}$, $\text{VOC} = 1.73 \text{ V}$, $\text{FF} = 83.72\%$ and $\text{PCE} = 24.47\%$. The tandem configuration achieved a 19.81% and 50.12% improvement in PCE compared to the best stand-alone bottom-cell efficiencies analyzed, for the Si-based and all-perovskite systems, respectively. The results demonstrate the usefulness of multinary composition control for the elimination of lead and bandgap allocation, voltage addition and/or spectral complementarity. Principal unresolved issues include losses due to optical reflection, recombination-junction modelling, migration of ions, long term stability and experimental manufacturability.

Keywords: tandem photovoltaics; multinary semiconductors; lead-free perovskites; $\text{Cs}_2\text{AgBi0.75Sb0.25Br6}$; CsSnI_3 ; perovskite/silicon; current matching; SCAPS-1D; efficiency enhancement

1. Introduction

The PV power conversion relation between the incident solar spectrum and electronic band structure of the absorber is the fundamental control of PV power conversion. The photons with energy below the bandgap are transmitted while the energy of photons that are significantly higher than the bandgap of the cell is mostly lost through thermalisation of the carriers. The detailed-balance analysis of Shockley and Queisser therefore sets a practical limit on the efficiency of an ideal single absorber, and the tandem extension developed by Alexis De Vos demonstrates that this limit can be increased by stacking absorbers with different bandgaps [1,2].

Two terminal monolithic tandem cells are especially desirable since the sub-cells are optically stacked and electrically in series. The voltage of

them add up, the currents through them are the same. Consequently, tandem optimisation is not an easy task of finding the highest performing cell in the top or the bottom row separately. This is a coupled design challenge that involves interacting with main design components such as bandgap, thickness, absorption coefficient, transport layers, defect density, recombination contacts and parasitic losses all influencing the current-matching point. In recent years, perovskite/silicon research has shown that the optimization of interfaces, texture engineering, and optical coupling and scalable fabrication is as critical as the quality of the absorbers [4–12].

The use of chemical substitution allows for a much broader compositional design space for multinary compound semiconductors to be used in this coupled optimisation process than for elemental

and binary absorbers. The category contains chalcopyrite compounds (Cu(In,Ga)Se_2), kesterite compounds ($\text{Cu}_2\text{ZnSn(S,Se)}_4$), chalcogenide perovskites and complex halide perovskites. They affect the constituent ratios which can alter bandgap, defect chemistry, electron affinity, lattice strain, phase stability and carrier transport. In tandem devices, this tunability can be exploited to make the front sub-cell a material with a wide bandgap, and the rear sub-cell a material with a narrow bandgap, thus maximizing the spectral coverage and maintaining a high total voltage [20–25].

In the present numerical study, two multi-nary halide absorbers consisting of lead (Pb) with either selenium (Se) or tellurium (Te) are studied. A compositionally substituted double perovskite viz $\text{Cs}_2\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ is utilized as the ~ 1.8 eV top absorber. It has more than binary B-site chemistry which is relevant to lead-free band gap and defect engineering. In the all-perovskite construction, the rear absorber is approximately 1.3 eV CsSnI_3 , which is a strong absorber of near-infrared photons and sensitive to defect and Sn^{2+} oxidation that causes self-doping issues [15–20]. The tandem is made up of two types of cells: a double-perovskite top cell and a crystalline silicon heterojunction bottom cell. The second uses CsSnI_3 to replace silicon, which results in a configuration with two lead-free absorbers, all perovskite, 3-3-3. The second one uses CsSnI_3 to replace silicon to form an all perovskite configuration with two lead-free absorbers, 3-3-3.

The study aims at achieving four objectives as follows:

- (i) develop a repeatable numerical protocol based on the SCAPS-1D computer simulation;
- (iii) to measure the current matching requirements of the two tandem designs proposed; and
- (iii) to describe the simulated improvements for efficiency in terms of spectral allocation, voltage addition and absorber thickness;
- (iv) to recognize possible assumptions and constraints which must be dealt with prior to experimental translation.

The contribution is not an experimentally fabricated claim. Rather it offers the analysis and internal checks of a numerical set, with a review of recent papers as a benchmark and clear identification and correction of known inconsistent points in the transcription.

2. Multinary Semiconductors in Tandem Photovoltaics

2.1 Bandgap allocation and series-current constraint

In the case of the 2T tandem, it is desired that the front absorber transmit enough photons that cannot be efficiently converted in the WG sub-cell, but that it absorb a sufficient number of visible photons to contribute a similar current as does the rear cell. If the two cells are connected in series, the short circuit current of the combination is controlled by the lower value of the short circuit current of the two cells, while the open circuit voltage is close to the sum of the two. This generates a tradeoff between top-cell thickness, which increases the top-cell absorption and current, but decreases the amount of photons that can be sent to the bottom cell. When both thickness-dependent current curves intersect near the maximum current, the optimum current is located near the intersection of the two curves, not the same as the maximum current for either a single thickness thick current curve individually.

2.2 Why multinary absorbers are useful

Absorbers using the same material in more than one form (as for different applications) can be used in tandem design by tuning E_g and Gap of the composition. The bandgap in CIGS can be varied by changing the ratio of In to Ga at a given site, the absorption edge in CZTSSe can be changed by varying the S/Se ratio, and cation and anion substitution in chalcogenide perovskites changes both the bandgap and stability, whereas the ratio of mixed cations, mixed halides or mixed metal sites in halide perovskites allows wide- and narrow-bandgap formulations. These recent searches based on data have also extended the envisioned chalcogenide design space, but compounds must also be selected to be defect tolerant, abundant in

elements, and free from toxicity and phase impurities and be scalable for deposition.

Multinary double perovskites are realized by previously discussed partial substitution of Bi by Sb in $\text{Cs}_2\text{AgBi}_{1-x}\text{Sb}_x\text{Br}_6$ for the top-cell material discussed herein. This composition does not contain a lead and gives a high voltage contribution to the unit, but can have an indirect transition, deep defect and carrier mobility which is relatively low, so that the current is limited. CsSnI_3 is better for long wavelength absorption and has a lower bandgap, but requires the suppression of Sn vacancies, oxidation and the related non-radiative recombination for the bottom perovskite. The tandem design therefore capitalizes upon the synergistic abilities of both absorber families whilst also incorporating the weaknesses.

2.3 Research gap

Most of the high efficiency experimental tandem devices are based on mixed-halide perovskites containing lead on silicon or narrow-bandgap perovskites containing tin/lead in all perovskite structures. Most research on lead-free double perovskites has been used to look at them as single cells, or as a preliminary example. In contrast, there has been comparatively little work to evaluate the $\text{Cs}_2\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ top absorber coupled with a CsSnI_3 bottom absorber under the same filtered-spectrum and current-matching conditions. The paired analysis here gives a controlled comparison of the performance of a fixed wide-bandgap multinary absorber in combination with an already

proven silicon rear cell, and a compositionally tunable lead-free perovskite rear cell.

3. Numerical Methodology

3.1 Study design and data provenance

In this paper a numerical device-simulation method is used based on SCAPS-1D modelling of two PV architectures with two terminals. The simulation records that were available were all governing electrical equations, material parameters, defect models, contact settings, optical filtering procedure and the final outputs of the photovoltaics themselves, as well as sweeps of the thickness of the absorbers. All these inputs and outputs were compared and sorted into a single, analytically coherent flow and analyzed by applying the concise correlation between short-circuit current density, open-circuit voltage, fill factor and power-conversion efficiency. No experimental measurements were not reported that were added to the analysis.

The numerical investigation was carried out in the following four steps. The calibration of the standalone Top and Bottom sub-cells was done independently. Secondly, the AM1.5G spectrum was sent through the top-cell stack with changing thicknesses of the top-absorbers. Third, the filtered spectrum was simulated for each bottom cell. Finally, the thicknesses of the absorbers have been changed to find the value where, the short circuit current densities of the top and bottom sub-cells are the same.

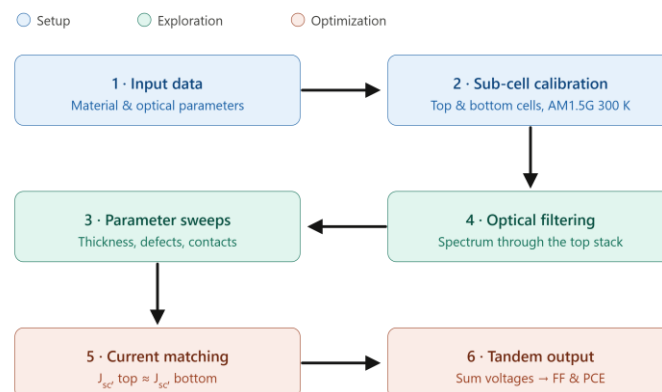


Figure 1. Numerical workflow used to calibrate, optically couple and current-match the tandem sub-cells.

3.2 SCAPS-1D device model

One-dimensional semiconductor simulator SCAPS-1D version 3.3.09 was used. It was initially created to analyze thin-film solar-cells and allows definition of multiple layers, bulk defects, interface defects as well as optical absorption files, front and rear contacts, series and shunt resistances, batch parameter sweeps and computation of J-V, C-V, C-f and quantum-efficiency characteristics [3]. Lateral uniformity of the properties was assumed in each sub-cell, representing it as an unidimensional stack. The electrical solution comprises of electrostatics and carrier conservation and drift-diffusion transport.

$$\frac{d}{dx} [\epsilon_0 \epsilon_r \frac{d\psi}{dx}] = -q (p - n + N_D^+ - N_A^- + p_t - n_t) \quad (1)$$

$$\frac{\partial n}{\partial t} = (1/q) \frac{\partial J_n}{\partial x} + G - U_n; \quad \frac{\partial p}{\partial t} = -(1/q) \frac{\partial J_p}{\partial x} + G - U_p \quad (2)$$

$$J_n = q \mu_n n E + q D_n \frac{dn}{dx}; \quad J_p = q \mu_p p E - q D_p \frac{dp}{dx} \quad (3)$$

ψ is electrostatic potential, n and p are free-electron and hole concentrations, N_D and N_A are the concentrations of ionised donors and acceptors, respectively, n_t and p_t are the concentrations of trapped carriers, respectively, J_n and J_p are current densities of free electrons and free holes, respectively, μ and D are the free-electron and free hole mobility, respectively, and G is the photogeneration and U is the net recombination rate. The defect distributions were considered in SCAPS as bulk and interface defects, and recombination could adjust to the density of

defects, energy level and capture cross section of defects.

3.3 Optical filtering and tandem reconstruction

The incident power density of AM1.5G spectrum was 1000 W m^{-2} and at the temperature of 300K in the top cell. The incident spectrum was attenuated by the top stack to obtain the spectrum delivered to the bottom cell. In this provided implementation, the intermediate reflection loss has been ignored, coherent interference and scattering loss is also ignored and only front contact transmission is taken into account in the input spectrum. The transmitted spectrum of a stack with i optical layers was given as:

$$S(\lambda) = S_0(\lambda) \exp [- \sum_i \alpha_i(\lambda) d_i] \quad (4)$$

where $S_0(\lambda)$ is the incident AM1.5G spectrum, $\alpha_i(\lambda)$ is the wavelength-dependent absorption coefficient and d_i is the thickness of layer i . The top and bottom cells were solved separately in SCAPS-1D since it does not calculate a complete two-terminal multijunction stack. The top-cell J-V curve presents the results from an AM1.5G spectrum, while the bottom-cell curve presents a calculation under each corresponding filtered AM1.5G spectrum. The tandem voltage $V_{\text{tandem}}(J)$ was recovered under a standard current J , where $V_{\text{tandem}}(J) = V_{\text{top}}(J) + V_{\text{bottom}}(J)$. To make the current matching condition, the absorber thickness is adjusted and the maximum common JSC is found.

$$FF = (V_{mp} J_{mp}) / (V_{oc} J_{sc}); \quad PCE (\%) = \frac{V_{oc} J_{sc} FF}{P_{in}} \quad (5)$$

3.4 Device architectures

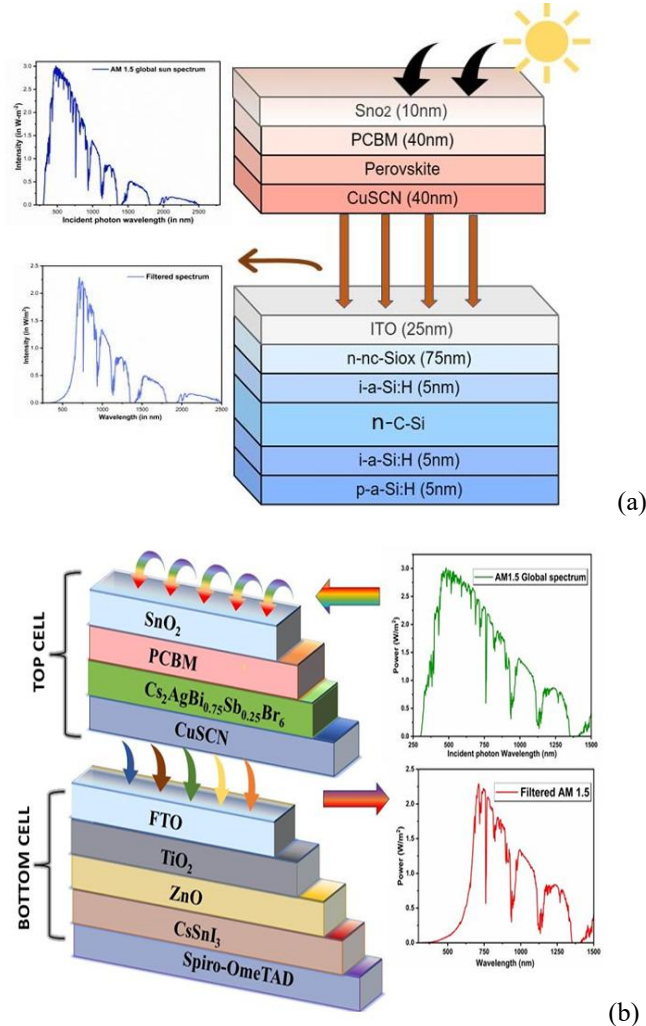


Figure 2. Reconstructed layer sequences of the two simulated two-terminal tandem architectures. Thicknesses correspond to the selected current-matched designs

Table 1. Layer sequences and functional roles of the investigated tandem cells.

Architecture	Top sub-cell	Bottom sub-cell	Electrical/optical role
Tandem 1	SnO ₂ / PCBM / Cs ₂ AgBi _{0.75} Sb _{0.25} Br ₆ / CuSCN	ITO / n-nc-SiO _x / i-a-Si:H / n-c-Si / i-a-Si:H / p-a-Si:H	Wide-bandgap visible absorption coupled to a high-quality near-infrared silicon heterojunction
Tandem 2	SnO ₂ / PCBM / Cs ₂ AgBi _{0.75} Sb _{0.25} Br ₆ / CuSCN	FTO / TiO ₂ -ZnO / CsSnI ₃ / Spiro-OMeTAD	Fully perovskite spectral division using approximately 1.8 eV and 1.3 eV lead-free absorbers

3.5 Silicon bottom-cell material parameters

The bottom cell consisted of an n-type crystalline-silicon wafer sandwiched in between intrinsic and doped amorphous-silicon passivation and contact layers, respectively, for the silicon heterojunction bottom cell. Table 2 gives an overview of the key

materials used in the source data set. During the tandem matching, the silicon thickness was changed, but the single silicon thickness was reported as 150 μm and the optimal tandem thickness as 230 μm . Changes in the preliminary result would be recorded in an earlier descriptive statement as a preliminary design value.

Table 2. Electrical parameters used for the silicon heterojunction bottom cell.

Parameter	ITO	n-nc-SiOx	i-a-Si:H	n-c-Si	p-a-Si:H
Thickness	25 nm	75 nm	5 nm	Variable	5 nm
Bandgap E_g (eV)	3.72	1.40	1.72	1.12	1.72
Electron affinity χ (eV)	3.60	3.94	3.80	4.05	3.80
Relative permittivity ϵ_r	9.4	11.9	11.9	11.9	11.9
N_c (cm ⁻³)	2.2×10^{19}	2.8×10^{19}	2.5×10^{20}	2.8×10^{19}	2.5×10^{20}
N_v (cm ⁻³)	1.8×10^{19}	1.4×10^{19}	2.5×10^{20}	1.04×10^{19}	2.5×10^{20}
Electron mobility (cm ² V ⁻¹ s ⁻¹)	10	1.4×10^3	20	1.4×10^3	20
Hole mobility (cm ² V ⁻¹ s ⁻¹)	10	4.5×10^2	5	4.5×10^2	5
Donor density N_D (cm ⁻³)	1×10^{19}	1×10^{20}	0	5×10^{16}	0
Acceptor density N_A (cm ⁻³)	—	—	0	—	1×10^{20}

Tandem current matching was a procedure used to change Si thickness. Stand alone calibration was done with the silicon thickness of 150 μm while the optimum tandem calibration was found to be 230 μm . A preliminary value of 250 μm was found in the design record, and is assumed to be an initial design configuration, and not the current matched thickness.

3.6 Defects, contacts and sweep conditions

Cs₂AgBi_{0.75}Sb_{0.25}Br₆ absorber was given a bulk defect density of $2.5 \times 10^{13} \text{ cm}^{-3}$, a Gaussian distribution of defects and a characteristic energy

of 0.1 eV. The silicon heterojunction contained intrinsic a-Si:H with amphoteric Gaussian defects and band-tail states, while the doped a-Si:H layers has lower densities of amphoteric defects and band-tail states were present. Single-level centres (neutral defects) were used to represent interface defects. The Tandem 2 was assumed to have flat-band contacts. The 107 and 105 cm s⁻¹ surface recombination velocities for electrons and holes were at the front contact and the carrier selectivity was inverted at the back contact.

Table 3. Simulation conditions and thickness sweeps used for tandem optimisation.

Model element	Tandem 1 setting	Tandem 2 setting	Purpose
Illumination	AM1.5G, 1000 W m ⁻² , 300 K	AM1.5G, 1000 W m ⁻² , 300 K	Standardised input and direct comparison
Top absorber sweep	Approximately 80–700 nm; selected 550 nm	100–900/1000 nm; selected 700 nm	Control top-cell absorption and transmitted flux
Bottom absorber sweep	c-Si approximately 50–300 μm ; selected 230 μm	CsSnI ₃ approximately 50–350 nm; selected 65 nm	Locate the common-current intersection
Top absorber defects	$2.5 \times 10^{13} \text{ cm}^{-3}$, Gaussian, 0.1 eV	Same wide-bandgap absorber model	Account for non-radiative recombination
Optical coupling	23 filtered-spectrum files reported	Thickness-specific filtered spectra	Provide bottom-cell illumination
Tandem connection	Series reconstruction at matched JSC	Series reconstruction at matched JSC	Add voltages while enforcing one current

3.7 Validation and internal consistency checks

The validation and internal consistency checks are included. A standalone sub-cell was calibrated prior to optical and electrical tandem reconstruction to experimentally reported or previously reported benchmark cell behaviour. For each of the efficiencies reported, the equation 5 was used to

confirm it. This test of arithmetic found three definite discrepancies on the numbers available from the records of the simulated experiments. The first is that Tandem 1's filtered-spectrum silicon voltage should not be 7044 V, but instead 0.7044 V. The second one is not possible for a single silicon sub-cell and is not consistent with the tandem voltage.

Second, the standalone CsSnI₃ voltage is about 0.704 V (not 0.073 V) when taking into account the $JSC = 33.31 \text{ mA cm}^{-2}$, $VOC \approx 0.704 \text{ V}$ and $FF = 69.5\%$, which is consistent with the reported standalone efficiency. Third, the current-matching calculation and performance table show 550 nm as the thickness selected for the top-absorber in Tandem 1 while one summary record states 500 nm. Thickness-dependent current intersection and

final output parameters are taken as being 550 nm, as it is the value that corresponds to the above mentioned parameters. The above corrections have no impact on the relative findings of the study. They enhance the numerical consistency and provide control of the reported current, voltage, fill factor and efficiency that fulfill the governing photovoltaic relations.

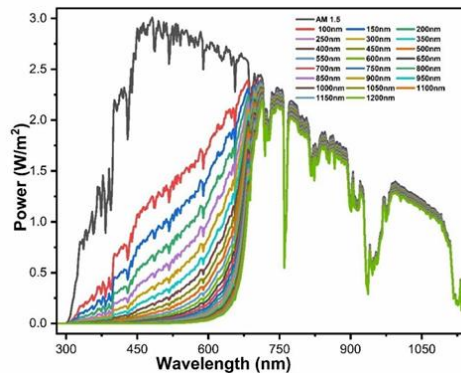


Figure 3: Filtered spectra with the original AM1.5G spectrum.

Figure 3. Current matching map as a function of the thickness of the absorber in Tandem 1, reconstructed from the set of thickness sweep of the absorbers. The design chosen consists of a 550 nm Cs₂AgBi_{0.75}Sb_{0.25}Br₆ top absorber and a 230 μm crystalline-silicon bottom absorber.

4. Results

4.1 Standalone sub-cell calibration

The calibrated wide-bandgap top cell achieved an efficiency of about 14.3% PCE in both tandem structures. For Tandem 1, the reported top-cell condition corresponding to the selected 550 nm absorber gave $JSC = 18.1068 \text{ mA cm}^{-2}$, $VOC = 1.12 \text{ V}$, $FF = 74.15\%$ and $PCE = 14.85\%$. The standalone silicon heterojunction reached $JSC =$

$36.152 \text{ mA cm}^{-2}$, $VOC = 0.731 \text{ V}$, $FF = 81.93\%$ and $PCE = 21.66\%$ at a reported calibrated thickness of 150 μm . The silicon current is much greater due to its lower silicon bandgap of 1.12 eV and wide NIR range of response.

For Tandem 2, the wide-bandgap top cell under AM1.5G generated $JSC = 16.92 \text{ mA cm}^{-2}$, $VOC = 1.137 \text{ V}$, $FF = 74.539\%$ and $PCE = 14.33\%$. The standalone CsSnI₃ cell generated $JSC = 33.31 \text{ mA cm}^{-2}$, a corrected VOC of approximately 0.704 V, $FF = 69.5\%$ and $PCE = 16.30\%$. These baselines show the expected asymmetry between the two cells: the wide bandgap has high voltage and low current, the narrow bandgap has high current under the un-filtered spectrum.

Table 4. Standalone performance used to establish the tandem baselines.

Sub-cell / condition	JSC (mA cm ⁻²)	VOC (V)	FF (%)	PCE (%)	Interpretation
T1 wide-gap top, selected thickness	18.1068	1.120	74.15	14.85	Current-limiting under standalone AM1.5G
Silicon bottom, standalone calibration	36.152	0.731	81.93	21.66	High-current near-infrared bottom cell
T2 wide-gap top, standalone	16.92	1.137	74.539	14.33	High-voltage top absorber
CsSnI ₃ bottom, standalone	33.31	≈ 0.704	69.50	16.30	Corrected voltage from PCE identity

4.2 Tandem 1: $\text{Cs}_2\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ /silicon

The photocurrent of the double-perovskite top absorber was increased while the irradiance reaching the silicon bottom cell was decreased as its thickness was increased. Optical calculations resulted in 23 thickness dependent filtered spectra

which were applied to silicon absorbers ranging about 50–300 μm in thicknesses. Several crossings of the upward trend of top-cell current and the downward family of silicon bottom-cell current were seen. The best common current was found at 550 nm top absorber and 230 μm Si wafer.

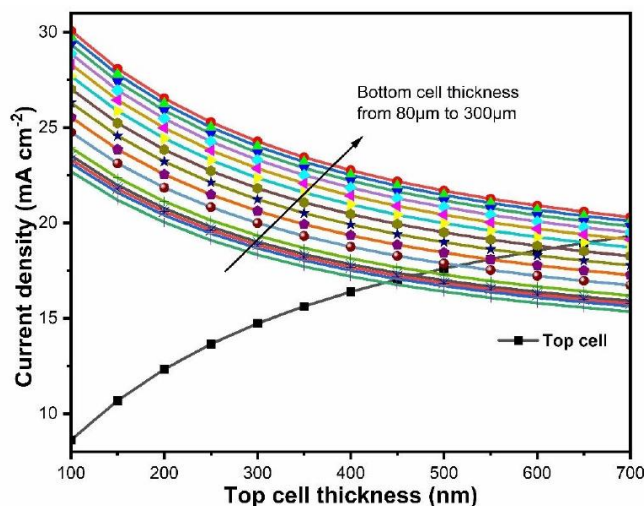


Figure 4: At thicknesses ranging from 50 to 700 nm for the top absorber layer, this curve was employed to establish current matching conditions for both the top and bottom cells.

Figure 4. Current-matching map for Tandem 2 as a function of the absorber thickness (reconstructed from the data available in the absorber-thickness sweep). The design selected has a 700nm

$\text{Cs}_2\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ absorber on top and a 65nm CsSnI_3 absorber on bottom.

Table 5. Current-matched photovoltaic parameters of Tandem 1.

Component condition	/	operating	Thickness	JSC (mA cm^{-2})	VOC (V)	FF (%)	PCE (%)
Wide-bandgap AM1.5G	top	under	550 nm	18.1068	1.120	74.15	14.85
Silicon bottom spectrum	bottom	under filtered	230 μm	18.085	0.7044	82.97	18.55
Reconstructed 2T tandem			550 nm / 230 μm	18.1068	1.812	79.18	25.95

The tandem efficiency is 19.81% better than the silicon tandem efficiency used for calibration, or 4.29 absolute percentage points (APPs) higher. The main source of the gain is from voltage addition where the tandem current is more or less half the standalone silicon current, and the operating voltage is more or less 2.48 times the standalone silicon VOC. The diagram shows the central tandem principle: the efficiency can be achieved without retaining the entire current of the low-bandgap cell, as long as the ratio between the

currents allocated by the spectrally allocated sub-cells is sufficiently high and the voltage losses of both sub-cells are kept under control.

4.3 Tandem 2: $\text{Cs}_2\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ / CsSnI_3

The thickness of the $\text{Cs}_2\text{AgBi}_{0.75}\text{Sb}_{0.25}\text{Br}_6$ top-absorber was varied from about 100 nm to 900-1000 nm for the all-perovskite tandem. The thickness of the CsSnI_3 bottom-absorber was changed from about 50 nm to 350 nm corresponding to each filtered spectrum. As the 'top

absorber' got thicker the 'current' coming to it began to move towards a saturation region, while the 'photon flux' and 'current' available to the 'bottom cell' decreased. A top absorber with 700 nm thickness and a bottom absorber with 65 nm thickness were used to obtain the desired current matching region.

Under the respective illumination conditions (15.57 mW cm⁻² and 15.51 mW cm⁻²), current densities of 16.81 mA cm⁻² and 17.10 mA cm⁻² were reported for the top and bottom cells, respectively,

and 16.91 mA cm⁻² was assigned to the reconstructed tandem. This is about 1.7% mismatch and more than the mismatch found at Tandem 1, and is considered to be a finite-step, rounding/transcription uncertainty, not a strict equality. However, the chosen family-thickness cross-point is in the crossing area of the two existing families. Based on Equation 5, the reported tandem VOC of 1.73 V and the FF of 83.72% and the PCE of 24.47% are consistent with each other.

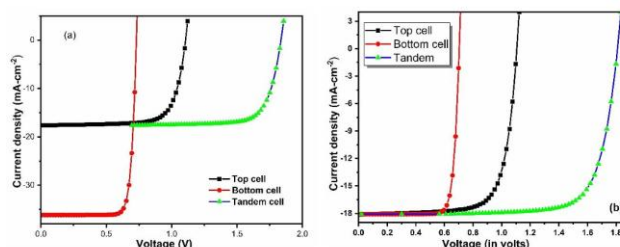


Figure 5: (a) Standalone J-V curves for the top, bottom, and tandem cells (b) J-V curves at the point of current matching.

The respective illumination conditions of the top cell and the bottom cell in the source table give values of 16.81 mA/ cm² and 17.10 mA/ cm², respectively, and the value 16.91 mA/ cm² is assigned to the tandem cells reconstructed. This 1.7% spread is greater than the mismatch of the 'Tandem 1' and so should be considered as a

rounding or transcription error not a match. However, this selected region is definitely the crossing region of the two existing families. The reported tandem VOC of 1.73V, FF of 83.72% and PCE of 24.47% are consistent for each other, according to Equation 5.

Table 6. Current-matched photovoltaic parameters of Tandem 2.

Component condition	/ operating	Thickness	JSC (mA cm ⁻²)	VOC (V)	FF (%)	PCE (%)
Wide-bandgap AM1.5G	top under	700 nm	16.81	1.137	74.54	14.33
CsSnI3 bottom spectrum	bottom under filtered	65 nm	17.10	0.600	73.77	13.89
Reconstructed 2T tandem		700 nm / 65 nm	16.91	1.730	83.72	24.47

The tandem efficiency is 8.17 absolute percentage points (~ 50.12% in relative value) higher than the standalone CsSnI3 efficiency. The higher percentage increase compared to Tandem 1 is due to the lower voltage and fill factor of the standalone narrow-bandgap perovskite than the

calibrated silicon cell. The all-perovskite tandem is thus favourably impacted by the 1.137 V contribution of the top-cell, but the extremely thin 65 nm CsSnI3 absorber and the more streamlined interconnection are experimental items that will need to be tested.

4.4 Comparative performance

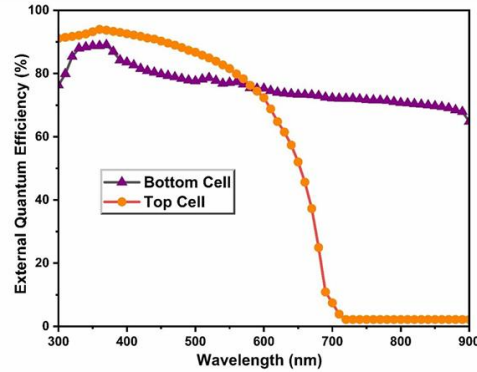


Figure 6. Efficiency of the standalone sub-cells and the corresponding tandem configurations in the supplied numerical dataset.

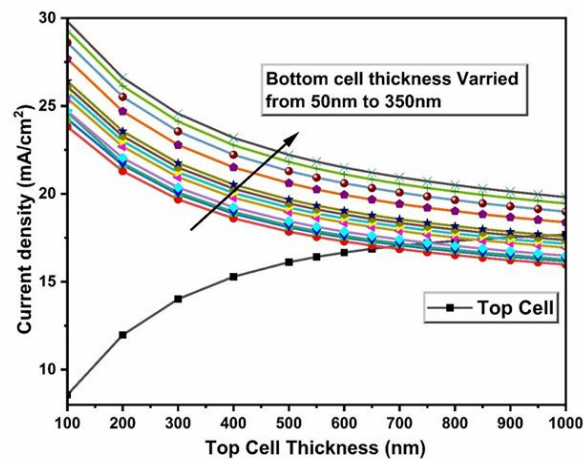


Figure 7. Comparison of the reported current-matched tandem metrics; bar heights are normalized within each metric and labels show the original values.

Table 7. Comparative interpretation of the two tandem architectures.

Comparison metric	Tandem 1: perovskite/silicon	Tandem 2: all-perovskite	Analytical implication
Top / bottom bandgap	1.8 / 1.12 eV	1.8 / 1.3 eV	Silicon provides broader near-infrared coverage
Matched JSC	18.1068 mA cm ⁻²	16.91 mA cm ⁻²	Tandem 1 retains the larger common current
Tandem VOC	1.812 V	1.73 V	Tandem 1 has the higher combined voltage
Fill factor	79.18%	83.72%	Tandem 2 reports the squarer reconstructed J–V curve
PCE	25.95%	24.47%	Tandem 1 leads by 1.48 percentage points
Relative PCE gain vs best standalone bottom	19.81%	50.12%	Tandem 2 derives the larger proportional benefit
Principal practical	Thick, high-quality silicon and	Sn oxidation,	Different scale-up constraints

risk	interconnection losses	ultrathin absorber control and all-perovskite stability	despite similar simulated PCE
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5. Discussion

5.1 Efficiency enhancement mechanism

From the results shown, you can see three different functions of the multinary top absorber. First, it absorbs a significant portion of high energy photons from the incident beam and absorbs the lower energy photons at a higher voltage than the bottom cell. Second, because of its finite thickness, it functions as an optical filter that dictates the current that is available to the back absorber. Third, due to its multinary composition, it provides a pathway to bandgap and defect tuning without lead. This theoretical optimum thickness is thus an electro-optical rather than an absorptive maximum thickness.

The higher PCE value of Tandem 1 is attributed to the silicon material which has a well-developed heterojunction contact system, good long-wavelength collection, and a high standalone fill factor. In the selected 230 μm wafer, the current under filtered-spectrum is kept around 18.1 mA/cm^2 and the additional voltage added to the top cell is about 0.70 V. Tandem 2 uses a lower matched current and voltage, but it's matched FF is higher than that of the other units, so it makes up for that loss. The all-perovskite architecture also provides for a thinner active stack and can also be processed at lower temperatures, although this is not assumed from a one-dimensional electrical model.

5.2 Position relative to recent tandem literature

The simulated efficiencies of 25.95% and 24.47% are reasonable but show efficiencies that are less than those of a few recent experimental devices that include perovskite/silicon combinations as cited in reviews and primary publications. The fully textured and nanotextured monolithic devices have been proven to allow optical management and interface quality to bring perovskite/silicon tandems from the mid 20% to surpass the 30% limit [7–12]. According to a 2026 review, the wider technology is at about 34.85% commercial readiness and durability, module integration and manufacturing cost are the remaining commercial barriers [6]. This new Tandem 1 result should thus be seen as a learning study of the absorber case, not as a record.

To date, experimental device and module performance with all-perovskite tandem devices has been improved due to improved grain surface passivation and module processing, as well as narrow-bandgap absorber control [13,14]. In recent years, SCAPS-1D calculations have shown efficiencies greater than 30% for materials with low amount of lead and alternative vacancy ordered perovskites [21]. That 24.47% corresponds to results from this paper, which are below results from best case simulations, yet has the benefit of being two of the reported absorber compositions that are lead-free. The value of the silicon-coupled version of the same WBP Top Absorber is therefore in the direct comparison with this.

Table 8. Contextual benchmark against selected tandem studies. Values are compared as reported and are not directly equivalent across area, certification and test protocol.

Study / year	Architecture	Type	Reported PCE	Relevance to the present results
Florent Sahli and co-authors, 2018 [7]	Perovskite/silicon, fully textured	Experimental, certified steady state	25.2%	Comparable to Tandem 1; demonstrates texture-enabled optical gain
Philipp Tockhorn and co-authors, 2022 [8]	Perovskite/silicon, nanotextured	Experimental, certified	29.80%	Shows the importance of optical texture and rear reflectors
Jiang Liu and co-	Perovskite/silicon	Experimental	>33%	Highlights interface

authors, 2024; reviewed in [4–6]	with bilayer passivation		class	passivation beyond the present simplified model
Renxing Lin and co-authors, 2022 [13]	All-perovskite tandem	Experimental	High-20% class	Demonstrates grain-surface passivation for all-perovskite tandems
Xuezeng Dai and co-authors, 2022 [14]	All-perovskite tandem module	Experimental module	>20% class	Addresses cell-to-module derating absent from SCAPS
Navdeep Kaur and co-authors, 2025 [21]	All-inorganic low- lead/lead-free simulated tandem	SCAPS-1D	31.93%	Recent simulation benchmark with different absorber chemistry
Present Tandem 1	Lead-free double- perovskite/silicon	SCAPS-1D	25.95%	Higher of the two supplied architectures
Present Tandem 2	Lead-free all- perovskite	SCAPS-1D	24.47%	Fully lead-free absorber pair

5.3 Interpretation of the multinary-material role

The findings suggest a larger take (apart from replacing toxic or rare elements): The potential of multinary semiconductors is not just in replacing other elements in tandem photovoltaics. Composition (A Design Variable): It is a variable that relates materials chemistry to device-level spectral management. In the high energy absorber, such as Bi/Sb substitution, the electronic structure changes and consequently the gap value is approximately 1.8 eV which is suitable for high energy photon conversion. The lower gap provided by the tin iodide framework will enable the bottom absorber to collect near infrared. Similar composition–bandgap concepts are employed in mixed-halide perovskites and CIGS and CZTSSe, but each class of material has different defect and phase-stability limitations [20,22–25].

The comparison also shows that not all the limitations can be overcome with the tunable absorber. VOC can be increased by a high bandgap, while low mobility and indirect transition will reduce the current and fill factor. A low band gap can increase the spectral coverage, while high self-doping and deep recombination centres can restrict the voltage. Thus, composition, phase purity, interfaces and optical thickness needs to be managed simultaneously to be successful with tandem optimisation. In the present model, the thickness and the size of the defect are only poorly described and the model cannot solve the problem of phase segregation or ion motion depending on the composition.

5.4 Model limitations and uncertainty

The major constraint is the sequential sub-cell method. It is helpful for two terminal tandem analysis, but does not solve the recombination junction, interlayer band bending, tunnel transport and luminescent coupling in one coupled domain explicitly. When there is information from interconnection resistive or recombination loss, the voltage-addition procedure may overestimate the performance. Similarly, Equation 4 fails to consider front surface reflections, multiple reflections within the medium, coherent interference, texture-dependent scattering and angle-dependent absorption. For silicon tandems, the condition of light trapping and parasitic absorption in the transparent contacts is significant for current matching and the above mentioned omissions are particularly relevant in this context.

The calculation of the SCAPS is a one-dimensional model and assumes that each layer is laterally uniform. It cannot accommodate pinholes, networks of grain-boundaries, rough interfaces, local shunts, or inadequate coverage and/or thickness variations. It is also not a model that includes dynamic ion migration, redistribution of halides, oxidation of Sn²⁺, moisture ingress, thermal cycling or mechanical stress. The reported output should therefore be viewed as an upper-bound device design screen and not as a foretaste of stabilised module output.

There are also indications of numerical uncertainty in the provided data. The top and bottom currents difference for Tandem 1 is small and the summed

sub-cell voltages differ from the reported tandem voltage by 12 mV. Tandem 2 has a larger spread among the top, bottom and tandem current values. The inconsistencies do not affect the qualitative ranking, but precise curve reconstruction, uncertainty propagation and independent replication would require raw output files from SCAPS.

A number of values included in the simulation records provided have some uncertainty. The top

and bottom cell currents of Tandem 1 differ slightly, and the voltage at the bottom cell is 12 mV different from the reported tandem cell voltage. The range of the top-cell, bottom-cell and reconstructed tandem currents is relatively large in tandem 2. They do not change the qualitative ranking of the two architectures, but it would need to have access to the original SCAPS project files and the exported J–V data sets to be able to exactly reconstruct the curves and replicate independently.

Table 9. Data-integrity corrections and interpretation of inconsistent numerical values.

Numerical issue	Inconsistent value	Value adopted in the analysis	Basis for correction
Silicon filtered-spectrum VOC	7044 V	0.7044 V	Physically realistic value and consistency with voltage addition
Standalone CsSnI ₃ VOC	0.073 V	≈0.704 V	Required by the reported JSC, FF and 16.30% PCE
Tandem 1 selected top thickness	500 nm in one summary record	550 nm	Current-matching curve and final output consistently identify 550 nm
Tandem 2 matched currents	16.81, 17.10 and 16.91 mA cm ⁻²	Values retained separately; 16.91 mA cm ⁻² used for the tandem	Preserves the numerical outputs while acknowledging finite mismatch
Silicon absorber thickness	250 μm preliminary value and 150 μm calibration value	230 μm in the tandem	Distinguishes preliminary design, standalone calibration and tandem optimum

A comprehensive study on SCAPS-1D analysis of multinary compound semiconductors for next generation tandem PV is presented. The wide-bandgap top cell was a Cs₂AgBi_{0.75}Sb_{0.25}Br₆ cell and the bottom cell was a crystalline-silicon heterojunction or narrow-bandgap CsSnI₃ perovskite. For each architecture the region of series-current matching was identified by thickness-specific filtered-spectrum coupling, and by absorber-thickness sweeps.

5.5 Experimental validation pathway

A credible experimental programme starts with independent films/dices. The composition and phase purity of Cs₂AgBi_{0.75}Sb_{0.25}Br₆ should be confirmed by X-ray diffraction and elemental analysis, and the absorption, photoluminescence, carrier lifetime and mobility measurements ought to confirm if the assumed optical and transport parameters are indeed realistic for the compound. Thickness-dependent top-cell devices should then be fabricated to assess the ability of the 550–700 nm film thickness range to be free of pinholes and

to compensate for the current gain due to bulk recombination.

The silicon tandem should be deposited on planar and textured silicon heterojunction substrates, and sub-cells resolved EQE, EL, and calibrated tandem J–V measurements should be performed. The main difficulties associated with the all-perovskite design are the preparation of a uniform 65 nm CsSnI₃ absorber, excluding oxygen from the process and device, and ensuring the low loss recombination layer can be used in both halves. Stability tests should feature maximum power point tracking, thermal exposure, humidity, and repeated lighting and post-ageing structural analysis.

6. Conclusions

The integrated SCAPS-1D study of multinary compound semiconductors application in next generation Tandem PV Devices is presented. The top cell CSFW was first combined with a crystalline-silicon heterojunction bottom cell, and then with a narrow-bandgap CsSnI₃ perovskite bottom cell. Each architecture's series current-

matching region was found by applying both thickness-specific filtered-spectrum coupling and absorber-thickness sweeps. The perovskite/silicon tandem achieved the highest numerical results, with $JSC = 18.1068 \text{ mA cm}^{-2}$, $VOC = 1.812 \text{ V}$, $FF = 79.18\%$ and $PCE = 25.95\%$ for thicknesses of the top-absorber (TA) of 550 nm and the bottom-absorber (BA) of 230 μm . The all-perovskite tandem reached $JSC = 16.91 \text{ mA cm}^{-2}$, $VOC = 1.73 \text{ V}$, $FF = 83.72\%$ and $PCE = 24.47\%$ using a 700 nm top absorber and a 65 nm bottom absorber. The efficiency of the silicon based tandem and all-perovskite tandem cells were 19.81% and 50.12% higher respectively when compared with the best stand-alone bottom cells. The results show that multinary semiconductors can improve the performance of the tandem in three interrelated ways: (1) the bandgap allocation, (2) the complementary utilization of the spectrum, and (3) the additive generation of voltage. The front-cell bandgap engineering feature enabled by Bi/Sb substitution in the wide-bandgap double perovskite, combined with the lower-bandgap CsSnI_3 absorber feature that allows for photon collection in the near-infrared region, creates a lead-free sunlight absorption pathway. This comparison also demonstrates that a multinary absorber is more effective than just nominal band gap. Optimization of carrier mobility, defect density, phase stability, and interface recombination and optical thickness must be optimised simultaneously. Technological readiness for immediate use should not be assumed from the simulated efficiencies. Sequential SCAPS-1D does not explicitly address recombination-junction transport, luminescent coupling, optical interference, texture-dependent scattering or ion migration and/or environmentally induced degradation. For future work, coupled modelling between optical and electrical parts, losses on interconnections, and experimental validation under maximum-power-point conditions, which takes into account the composition of the defects, are suggested. In this scope the two architectures studied offer a convenient template for analyzing sustainable tandem PV designs. It has been found that the results set the stage for making significant contribution to the current matching, spectral complementarity and voltage enhancement of lead-free multinary compound materials and

devices and also displayed the difficulty encountered at material level and device level for realizing high efficiency tandem cells in experiment.

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