

Article

Calibrated Optoelectronic TCAD Optimization of All-Organic and Hybrid Organic/Crystalline Silicon Two-Terminal Tandem Solar Cells

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Abstract

The increasing demand for high-efficiency photovoltaic technologies has intensified research into tandem solar cells (TSCs) as a promising strategy to surpass the performance limits of single-junction devices. In this study, a comprehensive optoelectronic simulation using a Silvaco ATLAS TCAD simulator is employed to design, optimize, and compare two tandem architectures: all-organic and hybrid organic/silicon TSCs. The all-organic configuration consists of polymer donors comprising a PBDB-T:F-M top sub-cell stacked with a PTB7-Th:CO₂DFIC:PC₇₁BM bottom sub-cell, while the hybrid configuration integrates a PBDB-T:F-M organic top cell with a crystalline Si bottom cell. Calibrated device models, validated against experimental data of the individual sub-cells, are used to evaluate tandem performance and guide systematic optimization. The two tandem architectures employ the same PBDB-T:F-M wide-bandgap organic top absorber ($E_g \sim 1.60$ eV), while the rear sub-cell consists of either the PTB7-Th:CO₂DFIC:PC₇₁BM organic absorber ($E_g \sim 1.20$ eV) or crystalline silicon ($E_g \sim 1.12$ eV). The initial organic/organic tandem device accomplishes a power conversion efficiency (PCE) of 15.70% and J_{SC} of 10.96 mA/cm², whereas the organic/silicon tandem structure exhibits a higher initial PCE of 16.85% and J_{SC} of 12.26 mA/cm². Following absorber-thickness optimization, the all-organic and hybrid OSC/Si tandems achieve PCEs of 19.44% and 21.13%, respectively. Rather than constituting a simple efficiency ranking, the comparison reveals distinct optical, electrical, and technological trade-offs. The hybrid architecture benefits from the broader spectral utilization and efficient carrier collection of thin crystalline Si, whereas the all-organic configuration offers advantages associated with low-temperature solution processing, reduced material consumption, and potentially fully solution-processed photovoltaic fabrication. The calibrated comparative framework therefore provides design guidance for selecting and optimizing TSCs according to both performance and application requirements.



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1. Introduction

Solar energy stands as one of the most promising renewable energies [1,2], presenting a clean, abundant, and sustainable means of meeting the raising energy demands [3–5]. Among the various solar technologies currently in use, silicon-based solar cells have consistently dominated the market [6]. Crystalline silicon (c-Si) PV cells presently hold approximately 90% of the global PV market, driven by their high PCE, reliability, cost-effective manufacturing, and the well-established versatility of silicon in technologies including microelectronics and silicon photonics [7–9]. The greatest certified efficiency for c-Si cells has reached 26.7%, nearing the theoretical limit [10]. In addition to their market dominance, c-Si solar cells are considered ideal bottom-cell candidates in tandem architectures owing to their inherent material advantages. With a narrow bandgap of 1.12 eV, Si facilitates effective absorption of a large fragment of the solar spectrum.

Driven by the pursuit of higher efficiency and wider usability, researchers are exploring alternative materials and advanced designs like thin-film solar cells (TFSC), which use less material and offer lightweight, flexible applications [11]. To enhance the performance of TFSCs, researchers have explored new materials and device structures, including organic solar cells (OSCs), Copper Indium Gallium Selenide (CIGS), and perovskite solar cells (PSCs). In recent years, OSCs have appeared as promising alternatives because of their advantages such as lightweight structure, mechanical flexibility, and compatibility with low-cost solution-processing techniques. Organic photovoltaic devices typically utilize conjugated polymers or small molecules as active materials capable of absorbing sunlight and transporting charge carriers. The introduction of the bulk-heterojunction (BHJ) architecture significantly improved exciton dissociation and charge transport by creating nanoscale donor–acceptor interfaces [12]. Early OSCs demonstrated relatively low efficiencies below 10%, primarily due to limited absorption and charge-transport properties [13,14]. Nonetheless, the development of non-fullerene acceptor materials has dramatically improved device performance in recent years. For instance, some studies reported OSC efficiencies exceeding 17% through optimized polymer donor and acceptor materials [15], while other investigations demonstrated efficiencies reaching 18% using advanced morphology control in non-fullerene acceptor systems [16]. More recently, further improvements in molecular design and device engineering have enabled single-junction OSCs to achieve efficiencies approaching 20%, indicating significant progress toward commercialization [17]. In addition, state-of-the-art organic photovoltaic devices have even reported efficiencies of approximately 20.19% through the development of rigid dimer acceptor structures that reduce non-radiative recombination losses [18].

To surpass the efficiency limits of single-junction solar cells, researchers have increasingly explored tandem solar cell (TSC) architectures, which combine multiple photovoltaic materials with diverse bandgap energies to capture a wider portion of the solar spectrum. In tandem structures, the top cell absorbs high-energy photons while lower-energy photons are transmitted to the bottom cell, thereby reducing the thermalization losses inherent in single-junction solar cells, and overcome the theoretical Shockley–Queisser (S-Q) efficiency limit [19]. Numerous TSC architectures, including two-terminal (2-T) and four-terminal (4-T) arrangements, have been extensively investigated through both experimental and numerical studies. The 4-T configuration permits straightforward fabrication via mechanical stacking; however, it introduces system-level complexity due to the independent electrical outputs of each sub-cell. In contrast, the 2-T configuration, while requiring precise current matching and more advanced fabrication procedures, enables more efficient incorporation into PV modules. It employs a single transparent interlayer electrode, which reduces parasitic optical losses [19].

Various tandem architectures have been proposed, including all-organic and all-perovskite TSCs along with hybrid TSCs such as silicon-based tandem systems [20–22], and chalcopyrite-based systems [23–27]. In all-organic tandems, polymeric semiconductors are particularly attractive because they offer solution processability, mechanical flexibility, and tunable optical constants and bandgaps, features that also support their wide applicability in optoelectronic devices [28–31]. In all-organic [32,33] and all-perovskite [34–36] tandem configurations, both the front and bottom sub-cells are fabricated from structurally or chemically similar materials but are engineered to exhibit different bandgap energies.

Representative all-organic devices have reported efficiencies of 16.4% in a monolithic 2-T architecture [37], 20.6% (certified 20.3%) in a 2-T tandem [38], 19.9% in a 2-T tandem [39], and 16.94% in a 4-T configuration [40]. Theoretical modeling predicts that tandem configurations incorporating a silicon bottom cell and a top cell having a bandgap energy of approximately 1.7 eV could potentially achieve PCEs as high as 43% [41]. This efficiency represents an idealized theoretical limit under restrictive radiative/detailed-balance assumptions. For organic/Si tandems, both experimental and simulation studies have examined 2-T and 4-T arrangements [42–44]. Recent TCAD work has also extended calibrated optoelectronic optimization to other hybrid tandems, including monolithic lead-free perovskite/Si and perovskite/u-CIGS tandems [45,46]. These studies further emphasize the importance of coupled optical–electrical optimization and current management.

Despite these promising attributes, the integration of organic and Si cells in tandem architectures remains relatively unexplored compared with other tandem technologies, with only limited experimental and theoretical investigations reported to date [42–44]. Previous investigations have demonstrated the feasibility of combining organic absorbers with silicon; however, a systematic and experimentally calibrated comparison between all-organic (OSC/OSC) and hybrid organic/Si (OSC/Si) two-terminal tandems under an identical optoelectronic modeling framework remains limited. This highlights a significant research gap in the development and performance optimization of organic/silicon TSCs. In the present work, two monolithic two-terminal tandem architectures sharing the same PBDB-T:F-M organic top sub-cell are directly compared. In the first configuration, a PTB7-Th:CO₁8DFIC:PC₇₁BM organic absorber is employed as the bottom sub-cell, forming an all-organic tandem, whereas in the second configuration the organic bottom sub-cell is replaced by crystalline silicon to form a hybrid organic/Si tandem. The motivation for introducing Si as the rear absorber is based on its narrower bandgap, broader spectral response, and higher carrier mobilities, which are expected to enhance collection of the photons transmitted through the wide-bandgap organic top sub-cell. Thus, the working hypothesis is that the Si bottom junction can increase the tandem photocurrent and overall conversion efficiency, even though its smaller bandgap may introduce larger thermalization losses and may slightly reduce the total tandem V_{OC} . This hypothesis is evaluated through experimentally calibrated optoelectronic simulations.

The main contributions of this work are as follows. First, the individual organic top cell, organic bottom cell, and crystalline Si cell are calibrated against experimental J-V and EQE data before their incorporation into the tandem structures, providing a common experimentally anchored modeling framework. Second, simultaneous optimization of the top- and bottom-absorber thicknesses is performed. Finally, energy-loss decomposition, spatial current-density distributions, and energy-band profiles are analyzed to identify the physical mechanisms responsible for the different performances of the two tandem architectures. A distinctive aspect of the present work is the explicit distinction between the current-matching condition and the maximum-efficiency condition. In monolithic two-terminal tandem cells, current matching is commonly employed as an important optical design criterion. However, matching the generated photocurrents does not necessarily maximize

the electrical output power. Therefore, in this work, absorber-thickness variation is used to independently determine both the strict current-matching point and the maximum-PCE point through coupled optical–electrical simulations. The resulting comparison reveals that the two operating conditions do not necessarily coincide, providing insight into the trade-off between optical current balance and electrical device performance. This aspect is less frequently emphasized in previous optimization studies of organic-based two-terminal tandem solar cells. Through this combined approach, the study provides a physics-based explanation of why the hybrid organic/Si tandem achieves superior conversion efficiency.

2. TCAD Methodology and Device Structures

2.1. Silvaco ATLAS TCAD Simulation Framework

Relying on experimental trial and error to optimize TSCs is both costly and inefficient, frequently leading to unnecessary resource expenditure. As a result, simulations play a vital role in analyzing and refining TSCs before physical fabrication. Nevertheless, the accuracy and credibility of these simulation outcomes must be validated by comparing them to experimental data. In this work, both the all-organic and hybrid organic/Si TSCs are modeled using the ATLAS device simulator (version 5.32.1.R) within the Silvaco TCAD framework. Atlas numerically solves the fundamental semiconductor transport equations, namely, the carrier continuity equations for both electrons and holes, using the drift-diffusion model in conjunction with Poisson’s equation. To accurately represent physical processes, recombination mechanisms such as the Shockley–Read–Hall (SRH) and Auger mechanisms are activated, accounting for trap states within the energy gaps of different layers and the high carrier concentrations that may arise during device operation. All TCAD simulations are conducted under standard AM1.5G solar illumination conditions (1000 W/m^2) at a fixed temperature of 300 K. Figure S1 presents a schematic flow chart outlining the fundamental components of the Silvaco simulation process. Further details regarding the assumptions, limitations, and methodological constraints of the simulation framework are provided in the Supplementary Material (Supplementary Note S1). In addition, the assumptions and practical considerations associated with the ITO interconnecting layer, linking the top with the bottom cells, are addressed in Supplementary Note S2.

2.2. Calibration of Individual Organic Top and Bottom Cells and Si-Cell

The experimental data used for calibration in this study were obtained from previously published measurements. The top and bottom organic sub-cells are based on the experimental devices reported by Firdaus et al. [47]. That study also provides an experimentally demonstrated double-junction organic tandem based on the same material family, allowing an additional benchmark of the initial OSC/OSC tandem discussed later. The silicon calibration data were taken from the thin silicon heterojunction study in Ref. [48].

In this phase of our study, we calibrate the simulated solar cell models by benchmarking them against their experimentally fabricated counterparts to confirm the reliability of the simulation framework. This validation process involves a detailed comparison of key photovoltaic parameters, including PCE, J-V characteristic curves, and EQE spectra, between experimental measurements and simulation outputs. Table 1 summarizes the fundamental physical properties of all materials utilized in the simulations of the top OSC, bottom OSC, and Si solar cell. In addition to bulk recombination, which was represented through the carrier lifetimes listed in Table 1, interfacial recombination was also included in the simulations. Surface recombination velocities (SRVs) were assigned at the relevant organic absorber/transport-layer interfaces. Initial values on the order of 1 cm/s were adopted for the OSCs and slightly adjusted during calibration to reproduce the experimental characteristics. For the Si solar cell, no explicit interface recombination was introduced

because the simulated structure exhibits predominantly homojunction-like carrier transport, without a distinct heterointerface comparable to those present in the organic sub-cells. Accordingly, recombination in the Si device was primarily described through the bulk recombination mechanisms and carrier lifetimes included in the model.

Table 1. Key input parameters of the top OSC, bottom OSC and Si solar cell materials [47–51].

Parameter	ZnO (ETL)	Top OSC Absorber	ZnO NP (ETL)	Bottom OSC Absorber	MoO ₃ (HTL)	Poly-Si (Doped)	Si Absorber	Poly-Si (Doped)
Thickness	50 nm	130 nm	50 nm	110 nm	10 nm	100 nm	30 μ m	200 nm
Electrical bandgap (eV)	3.3	1.6	3.1	1.2	2.3	1.12	1.12	1.12
Electron affinity (eV)	4.1	3.7	4.1	4	3	4.05	4.05	4.05
Relative dielectric permittivity	9	3	9	10	3.5	11.7	11.7	11.7
CB effective DOS (1/cm ³)	2.2×10^{18}	1.0×10^{20}	1×10^{20}	1.0×10^{22}	1.0×10^{17}	2.8×10^{19}	2.8×10^{19}	2.8×10^{19}
VB effective DOS (1/cm ³)	1.8×10^{19}	1.0×10^{20}	1×10^{17}	1.0×10^{22}	1.0×10^{17}	1.04×10^{19}	1.04×10^{19}	1.04×10^{19}
Electron mobility (cm ² /Vs)	200	6.04×10^{-4}	200	4.9×10^{-4}	1.620×10^1	1000	1000	1000
Hole mobility (cm ² /Vs)	25	2.86×10^{-4}	25	4.9×10^{-4}	1.620×10^1	500	500	500
Donor density (1/cm ³)	1.0×10^{18}	0	1.0×10^{20}	1×10^{14}	0	4×10^{18}	1×10^{15}	4×10^{18}
Acceptor density (1/cm ³)	0	1×10^{14}	0	0	1×10^{18}	0	0	0
Carrier lifetime (s)	1×10^{-5}	1×10^{-3}	1×10^{-5}	1×10^{-3}	1×10^{-5}	5×10^{-6}	250×10^{-6}	5×10^{-6}

The parameters summarized in Table 1 have different origins. Device structures, absorber compositions, and experimentally reported layer thicknesses were retained from the corresponding fabricated reference cells [47,48]. Material properties available from experimental reports or the established literature were directly adopted and kept fixed during calibration. The material parameters corresponding to the ETL and HTL were adopted from Refs. [49–51]. Parameters that were not experimentally reported, particularly effective defect-related quantities and, where required, doping concentrations, were estimated within physically reasonable ranges and adjusted during calibration to reproduce the measured J-V and EQE responses. A detailed classification of the parameters according to their origin, experimental, literature-derived, estimated, or calibrated, is provided in Table S1 of the Supplementary Material.

These material parameters serve as input values in numerical modeling and significantly influence the optical absorption, charge transport, and PV performance of the simulated device structures. After incorporating all relevant material properties and physical modeling parameters, we perform simulations of the J-V and EQE characteristics for the top OSC, the bottom OSC and Si-bottom solar cell. The calibration was performed by simultaneously comparing the simulated and experimental J-V and EQE characteristics, as well as the extracted J_{SC} , V_{OC} , FF, and PCE. This multi-observable calibration reduces the degree of parameter ambiguity compared with fitting the J-V curve alone.

Figure 1a illustrates a schematic of the top OSC structure, highlighting the fundamental thin-film layers, whereas Figure 1b presents the band-edge levels of the various materials used, depicted prior to contact. The cell assembly comprises ITO/ZnO/PFN-Br/PBDB-T:F-M/Ag. In this cell, the absorber blend consists of PBDB-T and F-M. PBDB-T is a high-performance conjugated polymer donor known for its strong absorption coefficient,

deep-lying HOMO level, and high carrier mobility. F-M serves as the acceptor component, enabling effective exciton dissociation and charge transport when paired with the polymer donor PBDB-T. The effect of the ultrathin PFN-Br interlayer was modeled through an effective SRV assigned at the ZnO ETL/organic absorber interface. The fabrication details of this OSC are provided in Ref. [47]. This solar cell adopts a heterostructure design without an HTL, representing an HTL-free configuration. In this architecture, the organic thin-film blend PBDB-T:F-M functions as the photoactive absorber with a thickness of 130 nm. A 50 nm ZnO layer serves as the ETL, while the front indium tin oxide (ITO) and rear electrodes have work functions of 4.3 eV and 4.7 eV, respectively. The resulting simulated and experimental curves for our single-junction top cell are displayed in Figure 1c for EQE spectra and Figure 1d for J-V characteristics. Notably, the bulk and interface defect parameters of the OSC were fine-tuned to optimize the alignment between simulated results and empirical data, ensuring a high-fidelity model fit.

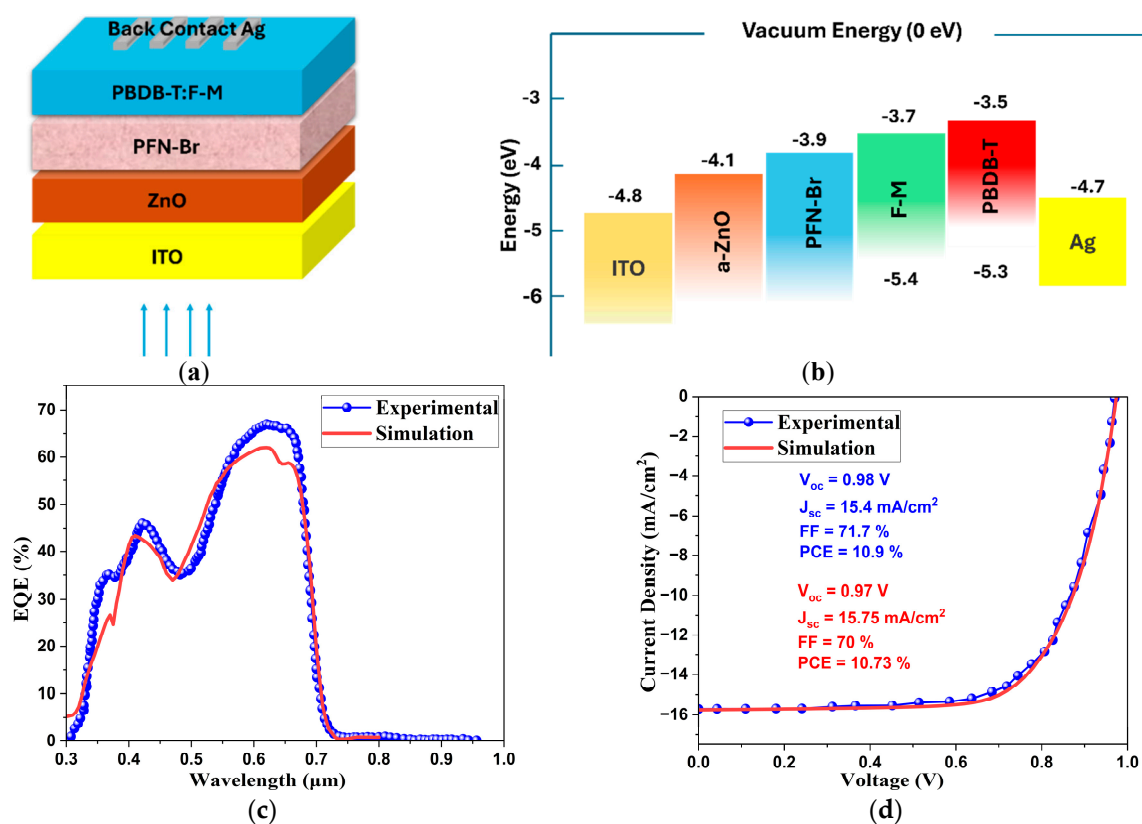


Figure 1. (a) Top OSC arrangement and (b) corresponding energy-band diagram, (c) EQE validation, and (d) J-V validation of the initial top OSC.

Additionally, for the bottom OSC [47], Figure 2a presents its schematic layout, illustrating the different layers, while Figure 2b depicts the band-edge levels of the materials prior to contact. The organic bottom-cell device architecture is composed of a multilayered structure starting with a 100 nm-thick Hsolar transparent conducting electrode. This is followed by a 50 nm thick electron transport layer of zinc oxide nanoparticles (ZnO NP). The active layer, a bulk heterojunction with a thickness of 110 nm, consists of a ternary blend of PTB7-Th, CO₈DFIC, and PC₇₁BM. PTB7-Th is the polymer donor in this ternary organic blend, widely used for its strong light absorption and effective hole-transport properties in bulk-heterojunction devices. CO₈DFIC and PC₇₁BM act as the acceptor components, with CO₈DFIC serving as the non-fullerene acceptor and PC₇₁BM as the fullerene acceptor that can help optimize charge transport and blend morphology. Above the organic blend, a 10 nm layer of molybdenum trioxide (MoO₃) serves as the HTL, and the structure is

completed with a silver (Ag) top electrode. The resulting simulated and experimental curves for our single-junction bottom cell are displayed in Figure 2c for EQE spectra and Figure 2d for J-V characteristics.

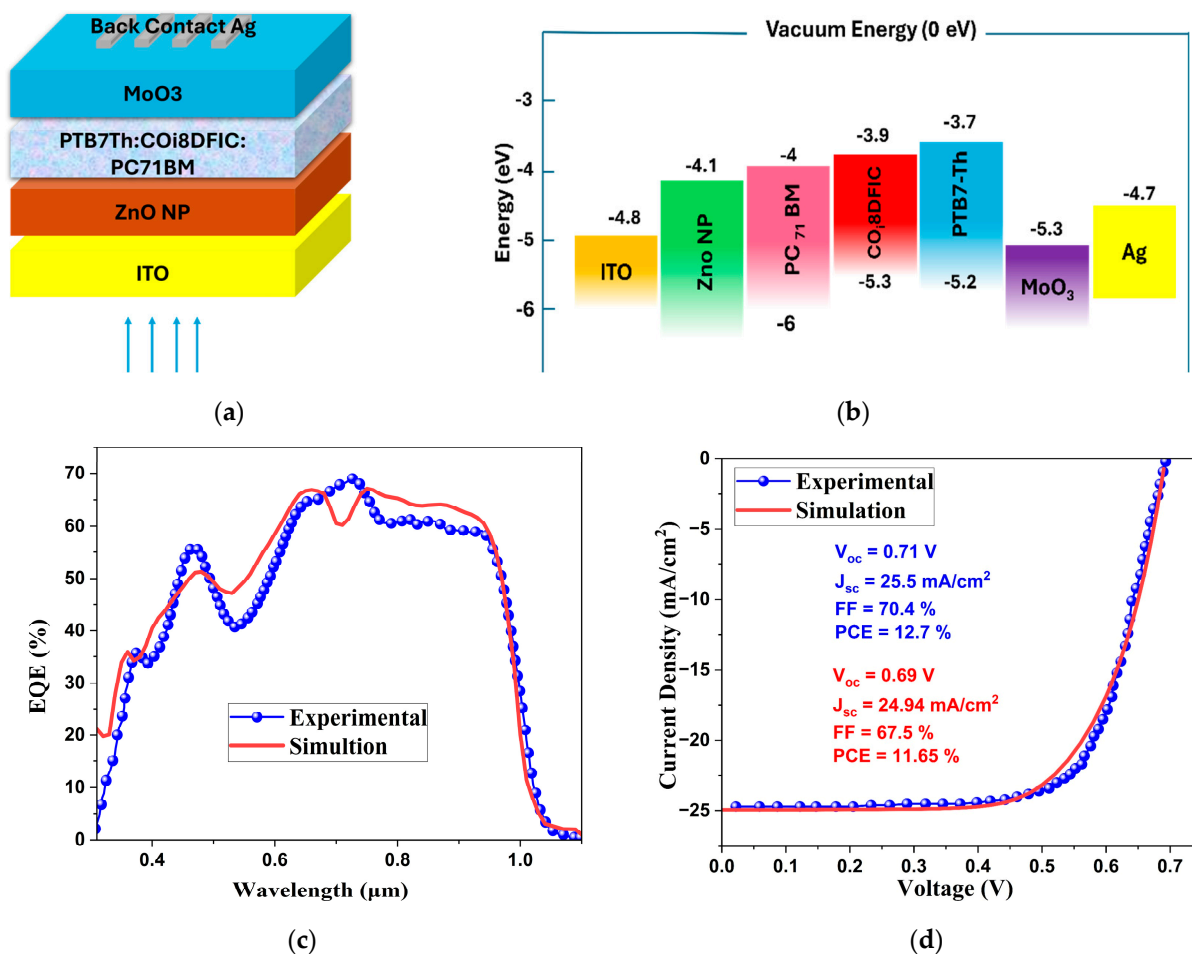


Figure 2. (a) Bottom OSC arrangement and (b) corresponding energy-band diagram, (c) EQE validation, and (d) J-V validation of the initial bottom OSC.

Finally, for the rear Si cell, Figure 3a presents a detailed schematic diagram illustrating the arrangement and composition of its various layers. Figure 3b further provides the corresponding band-edge energy levels, prior to electrical contact, of the Si substrate and the poly-Si layer, as well as the front and rear contact materials. The device is constructed on an n-type thin-film Si wafer with a thickness of 30 μm, which had been fabricated in earlier work, as described in [48]. While this thickness is comparatively larger than that of typical ultra-thin wafers, it still exhibits notable mechanical flexibility, a property confirmed by experimental results reported in [48]. This combination of mechanical robustness and flexibility makes it well-suited for integration into tandem structures. The resulting simulated and experimental curves for our silicon cell are displayed in Figure 3c for EQE spectra and Figure 3d for J-V characteristics.

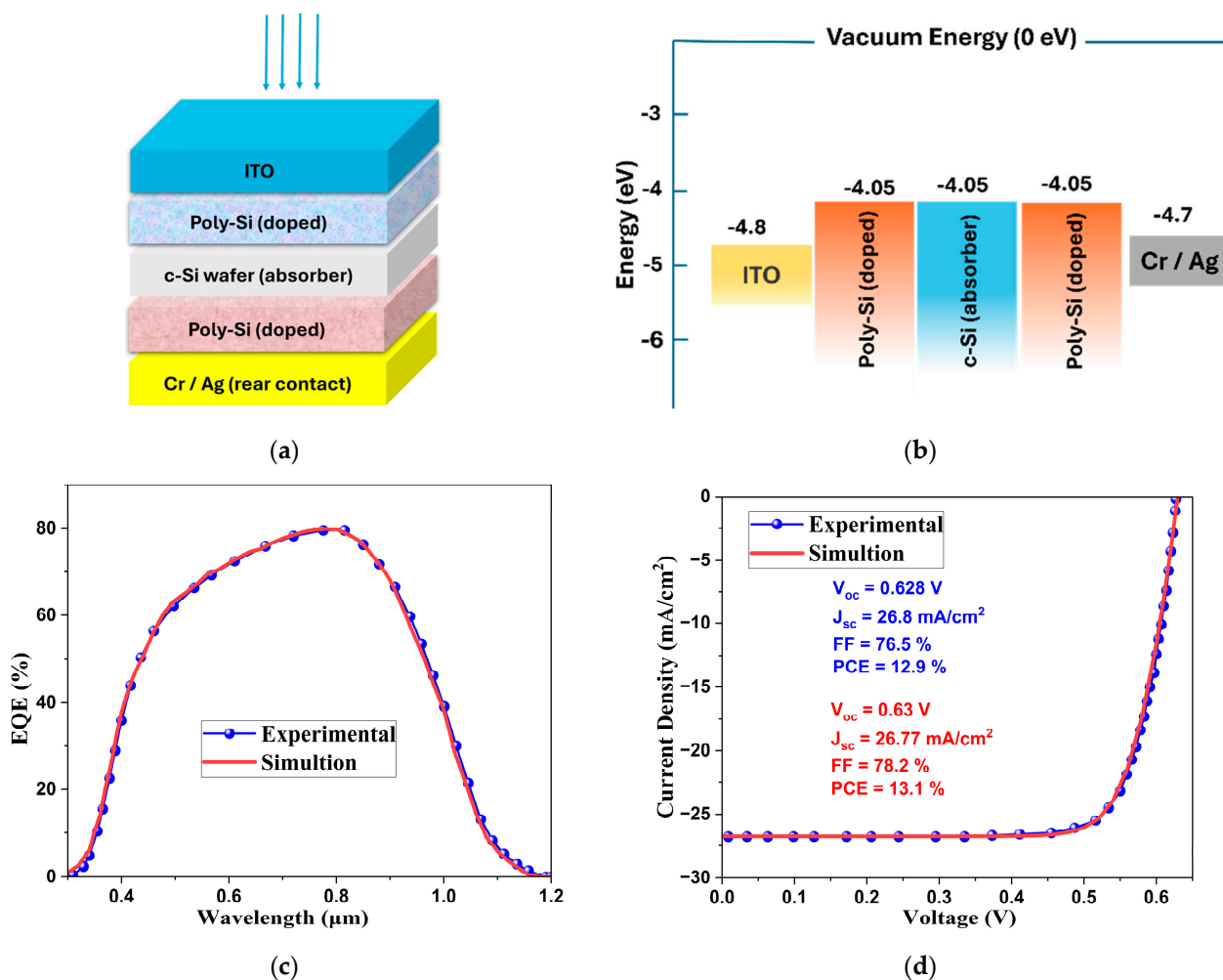


Figure 3. (a) Si-bottom-cell arrangement, (b) corresponding energy-band diagram cell, (c) EQE validation, and (d) J-V validation of the initial Si cell.

The results illustrated in the J-V and EQE curves that compare the experimental vs simulation results detailed in Table 2 clearly highlight the success of the calibration process, as indicated by the strong correlation between the simulated outcomes and experimental measurements. To further provide a quantitative assessment of the J-V calibration quality, the root-mean-square error (RMSE) was calculated between the simulated and experimental current densities at corresponding voltage points according to

$$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^N [J_{\text{sim}}(V_i) - J_{\text{exp}}(V_i)]^2}$$

where J_{sim} and J_{exp} are the simulated and experimental current densities calculated at voltage V_i , and N is the number of samples. The resulting RMSE values were 0.357, 0.886, and 0.0941 mA/cm² for the top OSC, bottom OSC, and Si cell, respectively. These relatively small deviations, together with the agreement in the extracted photovoltaic parameters and EQE spectra, support the use of the calibrated models for the subsequent tandem-cell analysis.

Table 2. PV parameters of both experimental and simulated organic and Si cells.

PV Parameters		J_{SC} (mA/cm ²)	V_{OC} (V)	FF (%)	PCE (%)
Top OSC	Experimental	15.40	0.98	71.70	10.90
	This work	15.75	0.97	70.00	10.73
Bottom OSC	Experimental	25.50	0.71	70.40	12.70
	This work	24.94	0.69	67.50	11.65
Bottom Si cell	Experimental	26.80	0.63	76.50	12.90
	This work	26.77	0.63	78.20	13.10

3. Results and Discussion

This section presents a comparative performance and physics-based analysis of the proposed OSC/OSC and hybrid OSC/Si TSCs, developed using experimentally calibrated TCAD models. First, the initial configurations of both tandem architectures are established and their baseline J-V and EQE characteristics are evaluated to provide a common reference for the subsequent optimization. Next, an energy-loss analysis of the individual constituent cells is presented to identify the dominant optical and electrical loss mechanisms and to clarify their influence. The effects of the top- and bottom-absorber thicknesses are then systematically investigated to determine the thickness combinations that maximize the PCE of each architecture. The analysis is subsequently extended to examine the relationship between strict current matching and maximum-efficiency operation. Although ideal tandem performance requires matched photocurrents, slightly mismatched conditions may yield higher overall efficiency. Therefore, both operating regimes are evaluated and compared to quantify efficiency limits and current-balance constraints in OSC/OSC and OSC/Si TSCs. Next, the internal device physics is examined through spatial current-density distributions and energy-band alignment profiles. These results provide insight into voltage addition, carrier transport, recombination behavior, and charge extraction mechanisms. Finally, the intrinsic challenges associated with each architecture are discussed, highlighting material limitations in OSC/OSC tandems and optical–electrical coupling and interface constraints in OSC/Si devices. This comparative analysis establishes clear design considerations for optimizing fully organic and hybrid TSCs.

3.1. Initial Configuration and Baseline Performance of Organic/Organic and Organic/Si Tandem Cells

The schematics of the proposed organic/organic and organic/Si tandem solar cell configurations are presented in Figures 4a and 5a, while the complete energy-band diagrams are illustrated in Figures 4b and 5b, respectively. These designs strategically combine a wide-bandgap organic top cell with a narrow bandgap organic or Si bottom cell to optimize light absorption. In the subsequent simulations, the top and rear cells are interconnected utilizing an ITO interlayer, which serves as a shared electrode to facilitate seamless current flow between the two sub-cells and to ensure proper boundary conditions are maintained.

Figure 6a,b present the simulated J-V characteristics of the two investigated tandem configurations, while Figure S2 provides a direct comparison between their performances. In addition, Figure 7a,b illustrate the EQE spectra of both devices. The extracted photovoltaic parameters are listed in Table 3. As shown in the table, the organic/organic TSC exhibits a V_{OC} of approximately 1.7 V, a J_{SC} of 10.96 mA/cm² and PCE of 15.7%. In contrast, the organic/Si tandem configuration demonstrates a slightly lower V_{OC} of 1.63 V but achieves a higher J_{SC} of 12.26 mA/cm² resulting in a boosted PCE of 16.85%. The higher V_{OC} observed in the organic/organic TSC can be attributed to the larger combined bandgap of the stacked organic sub-cells, which increases the built-in potential of the device.

However, the organic/silicon tandem structure benefits from the superior charge-transport properties and broader spectral absorption of silicon, leading to improved photocurrent generation and overall device efficiency.

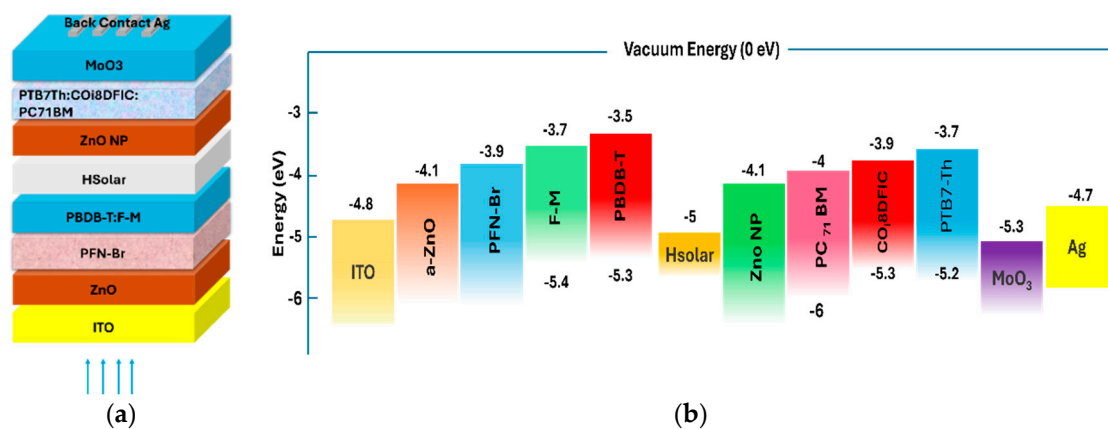


Figure 4. (a) Basic organic/organic tandem arrangement and (b) corresponding energy-band diagram.

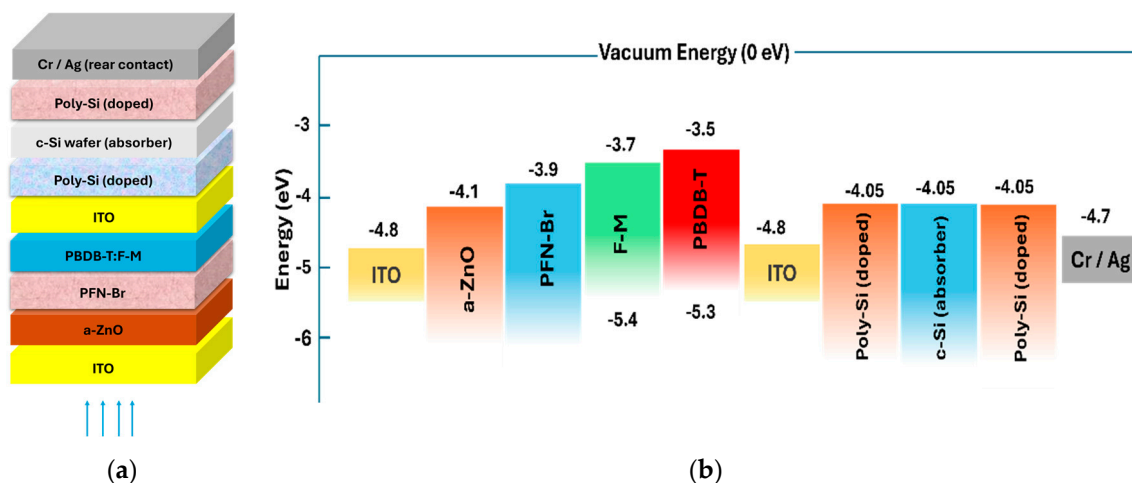


Figure 5. (a) Basic organic/Si tandem arrangement and (b) corresponding energy-band diagram.

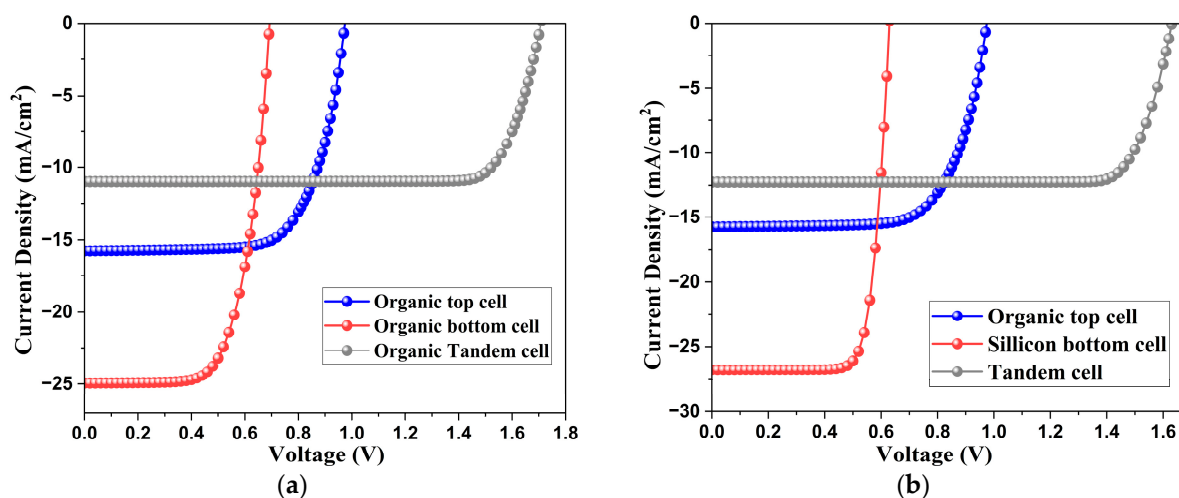


Figure 6. (a) OSC/OSC J-V curve for front vs rear vs tandem. (b) OSC/Si J-V curve for top vs bottom vs tandem.

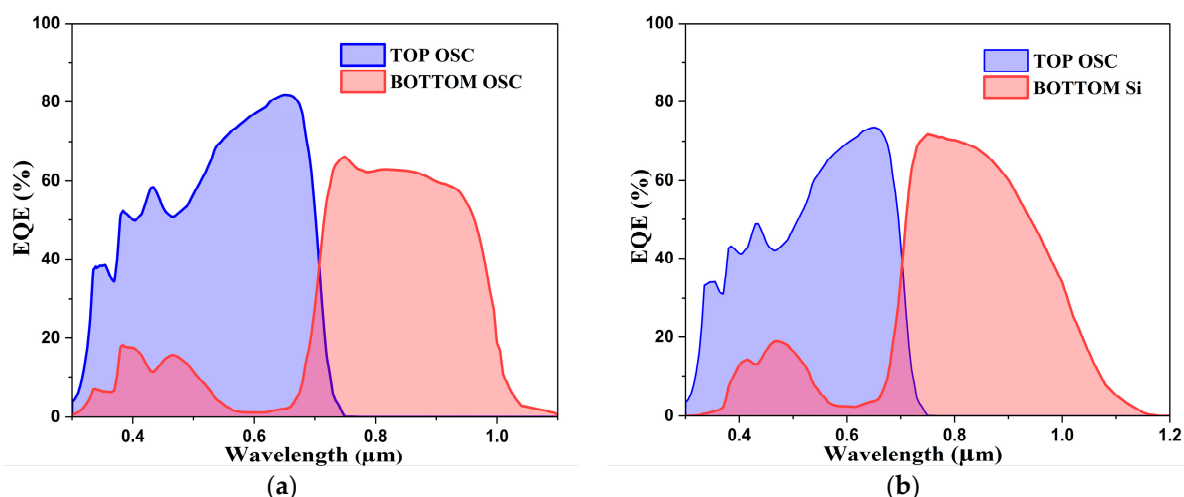


Figure 7. (a) OSC/OSC EQE curve for top and bottom. (b) OSC/Si EQE curve for top and bottom.

Table 3. TSC parameters of OSC/OSC and OSC/Si.

PV Parameters	J_{SC} (mA/cm ²)	V_{OC} (V)	FF (%)	PCE (%)
Organic/organic tandem	10.96	1.70	83.76	15.70
Organic/Si tandem	12.26	1.63	84.31	16.85

It is worth noting that additional experimental support is available for the initial all-organic tandem configuration. The organic sub-cell structures employed in the present model were adopted from the same experimental study in which the same organic tandem device was demonstrated. The reported experimental tandem exhibited an average V_{OC} of about 1.66 V, and PCE of about 16.3%. The corresponding initial OSC/OSC tandem simulated in the present work yields a PCE of 15.7% and V_{OC} of approximately 1.70 V, showing reasonably close agreement with the experimentally demonstrated tandem and providing additional confidence in the adopted modeling framework. In contrast, the OSC/Si tandem investigated here represents a proposed material combination and device architecture for which no directly corresponding experimental tandem has been reported.

3.2. Energy-Loss Analysis

To evaluate the energy losses in the analyzed standalone cells, we categorize the various loss mechanisms in accordance with Refs. [52,53]. Notably, a quantitative analysis of energy losses is a critical preliminary step in the design of the proposed tandem structure, as it enables the identification of the dominant mechanisms limiting device performance. The total energy losses can be split into different components, as addressed in the Supplementary Materials (Note S2).

Figure 8 shows energy-loss components for the three cells, while Table 4 compares all the distributed energy components of the top OSC, bottom OSC and bottom Si cell. For the top OSC, the transmission loss was determined to be 43.052 mW/cm², compared to 22.86 mW/cm² for the bottom OSC and 18.42 mW/cm² for the silicon cell. This reduction in transmission losses for the bottom cells can be attributed to their broader spectral absorption range. In terms of absorption losses, the top OSC exhibited a value of 31.486 mW/cm², while the bottom OSC and silicon cells showed slightly higher values of 34.93 mW/cm² and 31.20 mW/cm², respectively.

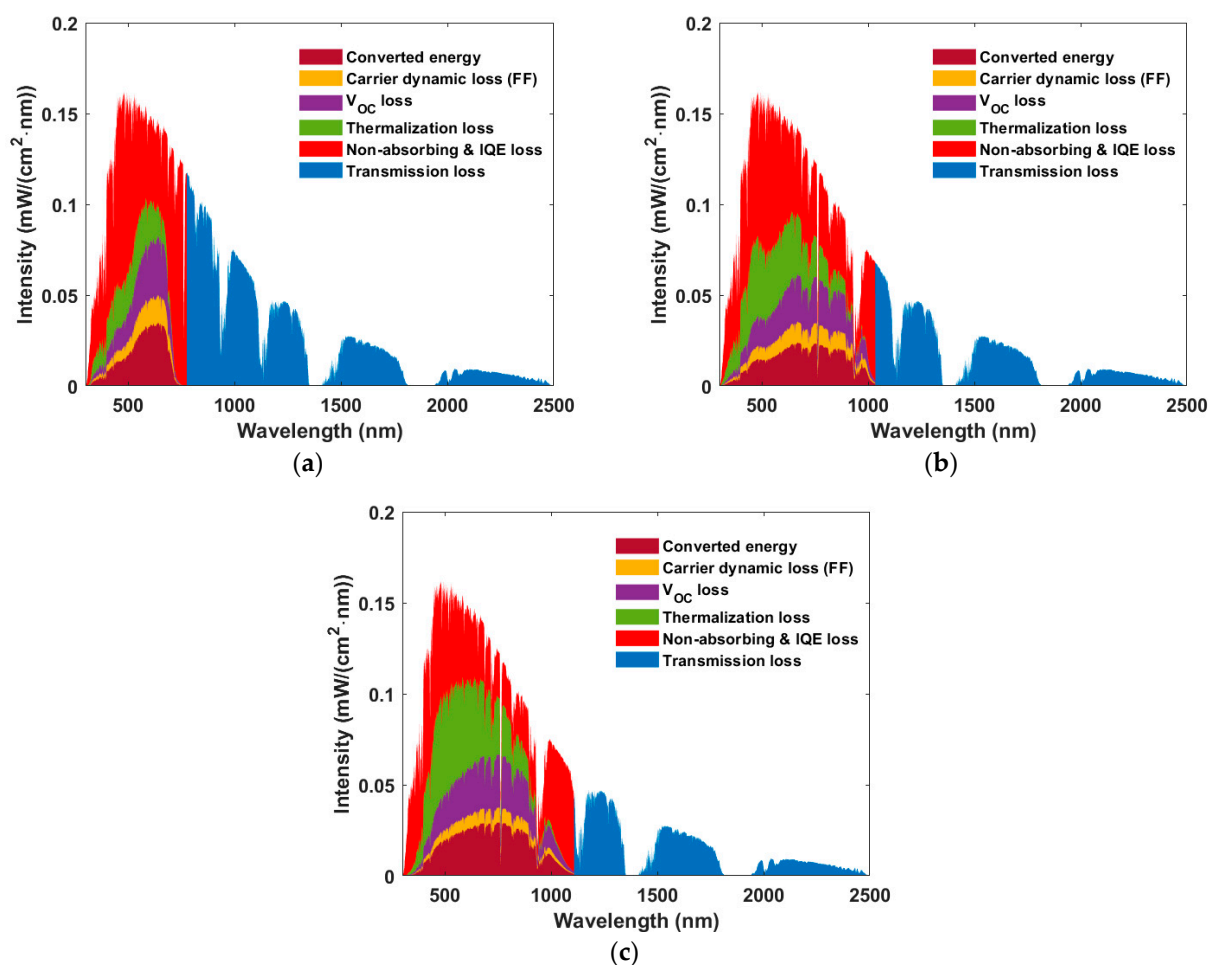


Figure 8. Spectral power density distribution illustrating the energy-loss elements in (a) the initial top OSC cell structure, (b) the initial bottom OSC cell structure and (c) the initial bottom Si cell structure.

Table 4. Comparison between the distributed energy components of the top OSC, bottom OSC and bottom Si cell.

W/cm^2	E_{trans}	E_{abs}	E_{th}	E_{VOC}	E_{FF}	E_{conv}
Top OSC	43.052	31.486	6.823	6.937	3.253	7.430
Bottom OSC	22.86	34.93	14.93	11.16	4.91	10.20
Bottom Si	18.42	31.20	19.50	13.07	3.66	13.14

Thermalization losses are significantly lower in the top OSC (6.823 mW/cm^2) compared to the bottom OSC (14.93 mW/cm^2) and the silicon cell (19.50 mW/cm^2), which can be primarily explained by the wider bandgap of the top sub-cell. Furthermore, the three devices demonstrated comparable values for E_{VOC} and E_{FF} , with the top OSC exhibiting marginally reduced losses. The converted energy (E_{conv}) is found to be 7.43 mW/cm^2 for the top OSC, 10.2 mW/cm^2 for the bottom OSC, and 13.14 mW/cm^2 for the silicon cell.

3.3. Thicknesses of Both Organic/Organic and Organic/Silicon Absorber Layers

Notably, when the front absorber layer is excessively thin, insufficient absorption of high-energy photons occurs, leading to a limitation in J_{SC} . In contrast, an overly thick front absorber increases series resistance, thereby decreasing the fill factor. Moreover, excessive front-layer thickness diminishes the spectral irradiance transmitted to the rear sub-cell, consequently reducing its J_{SC} . In addition, a thicker organic layer increases the

carrier-transport distance and, because of the comparatively low effective mobilities of the bulk-heterojunction absorber, increases the sensitivity to recombination and incomplete carrier collection. This interpretation is specific to the organic absorber. The thickness of the rear absorber also exerts a significant influence on device performance. An increased rear absorber thickness enhances the absorption of low-energy photons, contributing to higher J_{SC} and V_{OC} . However, when the rear absorber becomes too thick, carrier recombination losses intensify, and FF degradation can occur, resulting in a saturation or decline in overall device performance.

In our analysis of thickness variations, the thickness of the top PBDB-T:F-M absorber layer (t_{TA}) in the OSC/OSC TSC was varied from 50 to 350 nm in increments of 25 nm, while the thickness of the bottom PTB7-Th:CO₂8DFIC:PC₇₁BM absorber layer (t_{BA}) was adjusted between 200 and 400 nm using the same step size. Figure 9a presents the dependence of tandem PCE on t_{TA} for multiple values of t_{BA} . On the other hand, for the organic/silicon TSC, Figure 9b shows an analogous analysis in which the thickness of the top PBDB-T:F-M organic absorber (t_{TA}) was similarly varied from 50 to 400 nm with 25 nm increments, whereas the thickness of the Si bottom absorber (t_{BA}) was varied from 30 μm to 100 μm in steps of 10 μm .

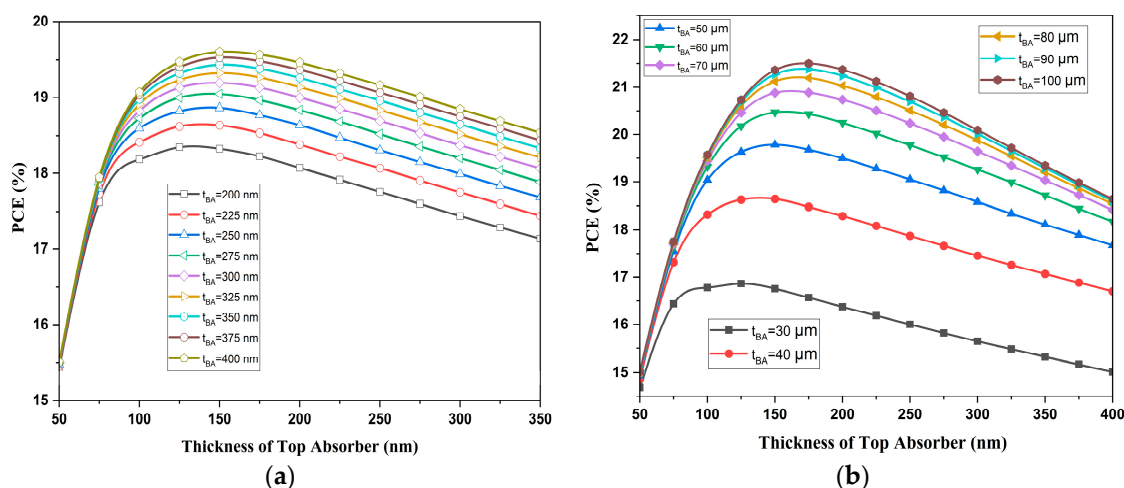


Figure 9. (a) Impact of thickness of both PBDB-T:F-M/PTB7-Th:CO₂8DFIC:PC₇₁BM layers on the PCE of OSC/OSC TSC, and (b) impact of thickness of both PBDB-T:F-M/Silicon layers on the PCE of OSC/Si TSC.

For the OSC/OSC TSC, the results clearly demonstrate that the PCE exhibits a strong dependence on the thickness of the PBDB-T:F-M top absorber for each selected bottom-absorber thickness. In all cases, the PCE increases with t_{TA} up to an optimal value, beyond which it decreases. Moreover, the maximum achievable PCE increases as t_{BA} becomes larger. For $t_{BA} = 200$ nm, the maximum PCE slightly exceeds 18%, reaching its optimum of approximately 18.2% at a top-absorber thickness of around 125 nm. When t_{BA} is increased to 400 nm, the PCE attains a higher peak of about 19.5% at a t_{TA} of roughly 150 nm. These results indicate that thicker bottom-absorber layers shift the optimal top-absorber thickness toward higher values.

In the organic/silicon TSC, the PCE strongly depends on the thickness of the PBDB-T:F-M top absorber. For all silicon bottom-absorber thicknesses, the PCE rises with t_{TA} until reaching an optimum and then decreases. As the silicon bottom absorber becomes thicker (from 30 μm to 100 μm), the maximum PCE increases. At 30 μm , the device achieves ~16.75% PCE at ~125 nm top thickness, whereas at 100 μm , the peak improves to ~21.5% at ~175 nm. Thus, increasing the silicon absorber thickness shifts the optimal top-absorber thickness to higher values.

The selection of absorber thicknesses for the maximum-efficiency configuration was based on identifying a practical optimum that provides high PCE while avoiding unnecessary increases in material consumption. As the bottom-absorber thickness increases, the PCE initially improves; however, beyond a certain thickness, the additional efficiency gain becomes marginal. Therefore, a bottom-organic absorber thickness of 350 nm was selected for the OSC/OSC tandem, since further increasing the thickness to 400 nm results in only a very small PCE enhancement. Similarly, an 80 μm Si bottom absorber was selected for the OSC/Si tandem because further increases in Si thickness provide only limited improvement in PCE while requiring additional absorber material. Thus, the selected thicknesses represent practical near-optimal design points that balance photovoltaic performance and material utilization.

3.4. Current Matching vs Maximum Efficiencies for Both TSCs

To achieve current matching in both TSC configurations, a series of optical–electrical simulations were performed in which the absorber-layer thicknesses were systematically varied. For the organic/organic TSC, the thickness of the top-absorber layer (t_{TA}) was varied from 50 nm to 250 nm in increments of 25 nm. The bottom-absorber layer (t_{BA}) was scanned from 100 nm to 400 nm with a step size of 50 nm. For the organic/silicon TSC, t_{TA} was likewise varied from 50 nm to 250 nm, while the silicon bottom absorber was varied from 30 μm to 100 μm in increments of 10 μm .

The resulting J_{SC} of both the front and rear sub-cells were evaluated, as illustrated in Figure 10a for the organic/organic TSC and Figure 10b for the organic/silicon TSC. In the organic/organic TSC, increasing t_{TA} leads to an increase in the top-cell J_{SC} and a corresponding decrease in the bottom-cell J_{SC} . This behavior arises because a thicker top absorber captures a larger fraction of incident photons, thereby decreasing the transmitted spectrum available to the bottom sub-cell. Furthermore, increasing t_{BA} improves the J_{SC} of the bottom sub-cell as expected for thicker absorber layers. Current matching is achieved at the intersection point where the J_{SC} values of the two sub-cells become equal.

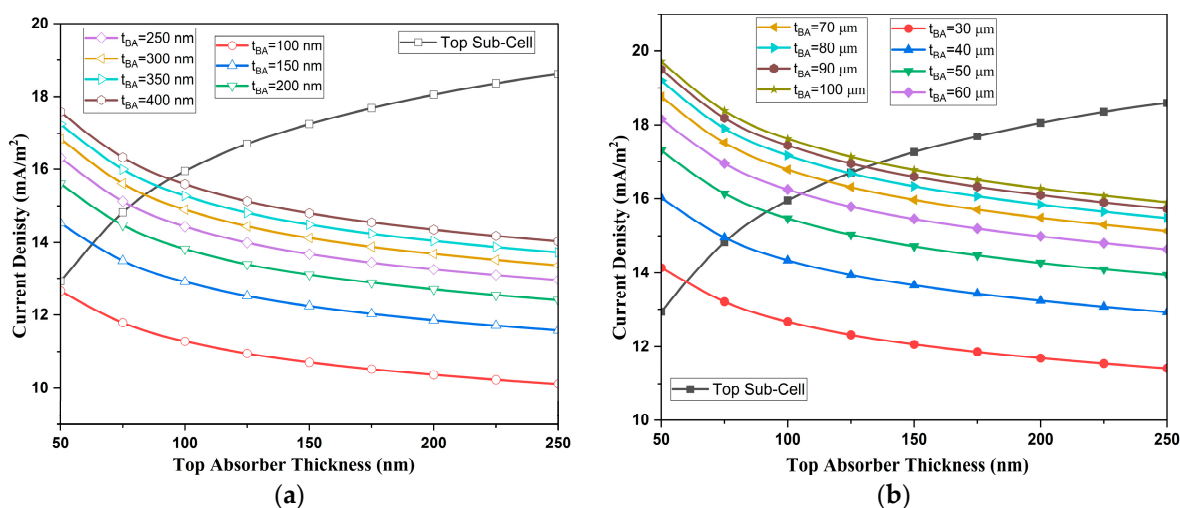


Figure 10. (a) Illustrating current-matching conditions for organic/organic TSC; (b) illustrating current-matching conditions for organic/silicon TSC.

A similar trend is observed for the OSC/Si tandem, where increasing the top-absorber thickness enhances the top-cell current while reducing the bottom-cell current. Meanwhile, increasing the Si thickness improves the bottom-cell current owing to enhanced absorption. Analysis of the data presented in Figure 10 indicates that, for the organic/organic TSC, the optimal current-matching condition is obtained at a top-absorber thickness of $t_{\text{TA}} = 89.5$ nm

and a bottom-absorber thickness of $t_{BA} = 350$ nm, resulting in a matched J_{SC} of 15.4 mA/cm^2 . For the OSC/Si TSC achieves optimal current matching at $t_{TA} = 124.5$ nm and $t_{BA} = 80 \text{ }\mu\text{m}$, corresponding to a matched J_{SC} of 16.69 mA/cm^2 .

To provide further physical insight into the absorber-thickness optimization, additional spatial photogeneration profiles are presented in Figure S3 of the Supplementary Material for the OSC/OSC tandem. The results show that increasing the top-absorber thickness from 89.5 to 150 and 250 nm extends the photogeneration region within the top sub-cell, allowing a larger fraction of the incident photons to be absorbed before reaching the rear sub-cell. Consequently, the photogeneration rate in the bottom organic absorber progressively decreases as the top absorber becomes thicker. The 89.5 nm configuration, corresponding to the current-matching condition, allows greater photon transmission to the bottom sub-cell, whereas the 250 nm top absorber produces stronger optical attenuation and noticeably suppresses rear-cell photogeneration. The 150 nm configuration provides a more favorable balance between photon harvesting in the front cell and sufficient photogeneration in the rear cell, consistent with the maximum-PCE condition obtained from the coupled optoelectronic optimization.

Figure 11 displays the J-V characteristics under maximum-efficiency conditions for the two device architectures. Specifically, Figure 11a corresponds to the all-organic TSC with an active layer thickness of $t_{TA} = 150$ nm for the top-absorber and $t_{BA} = 350$ nm for the bottom absorber. In contrast, Figure 11b represents the hybrid organic/silicon TSC, where the top absorber thickness is maintained at $t_{TA} = 150$ nm, while the silicon bottom absorber has a significantly larger thickness of $t_{BA} = 80 \text{ }\mu\text{m}$. These results highlight the effect of absorber material and thickness on charge transport and overall device performance under optimized efficiency conditions.

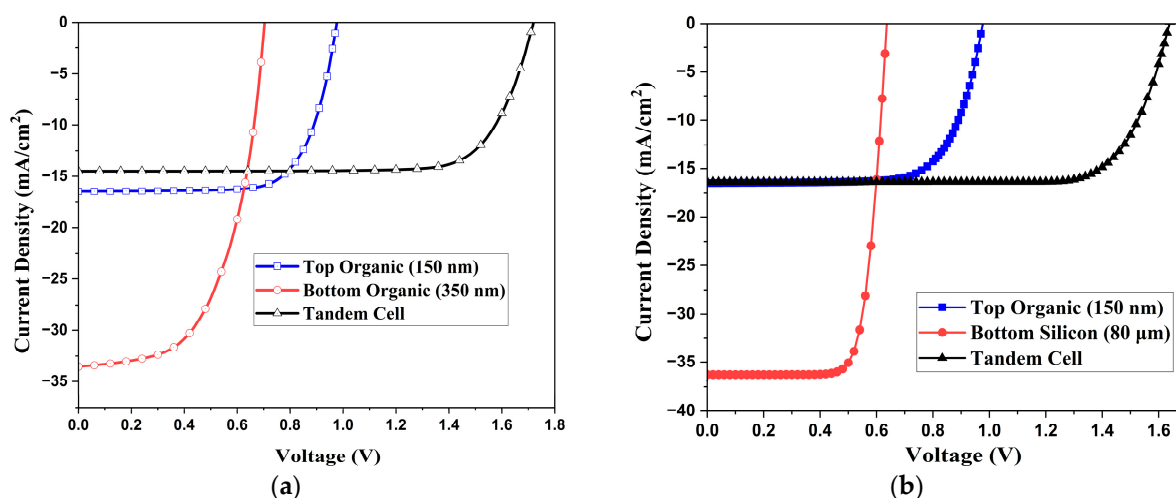


Figure 11. J-V curves at maximum efficiency for (a) optimized OSC/OSC TSC, and (b) optimized OSC/Si TSC.

Figure 12 depicts the J-V characteristics under current-matching conditions, which are critical for tandem device optimization due to the series electrical configuration of sub-cells. For the all-organic TSC (Figure 12a), current matching is achieved at $t_{TA} = 89.5$ nm and $t_{BA} = 350$ nm. Meanwhile, the organic/silicon TSC (Figure 12b) requires a slightly thicker top-absorber layer of $t_{TA} = 124.5$ nm, with the silicon bottom layer thickness remaining at $t_{BA} = 80 \text{ }\mu\text{m}$. The shift in optimal thickness values reflects the differences in absorption coefficients and carrier generation profiles between organic and inorganic materials.

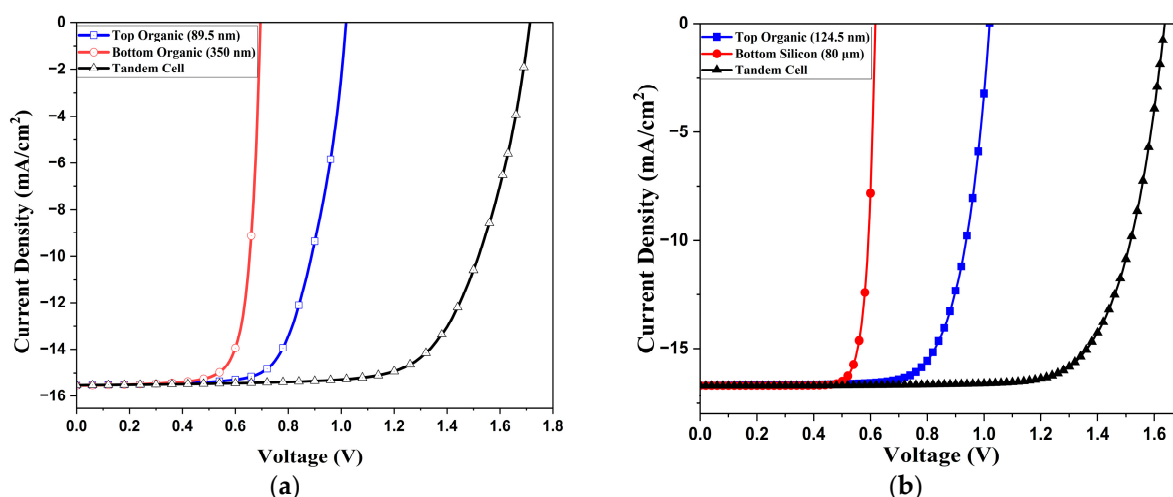


Figure 12. J-V curves at current matching for (a) optimized OSC/OSC TSC, and (b) optimized OSC/Si TSC.

Finally, Figure S4a and Figure S4b present the EQE spectra under current-matching conditions for the all-organic and organic/silicon TSCs, respectively. These EQE profiles provide insight into the wavelength-dependent photo response of each sub-cell and confirm the spectral complementarity required to achieve effective current matching in tandem configurations.

Table 5 summarizes the photovoltaic parameters of the organic/organic TSC under two operating conditions: maximum efficiency and current matching. At the maximum-efficiency point, the TSC exhibits a J_{SC} of 14.5 mA/cm² and a V_{OC} of 1.72 V. Under current-matching conditions, the device achieves a higher J_{SC} of 15.4 mA/cm², while the V_{OC} slightly decreases to 1.70 V. Although current matching does not correspond to the absolute maximum efficiency, the associated performance loss is minimal. Current matching occurs at a top-absorber thickness (t_{TA}) of 89.5 nm, yielding a tandem PCE of 18.68%. By comparison, the highest PCE of 19.44% is obtained at a larger t_{TA} of 150 nm. These results indicate that the tandem device maintains nearly equivalent performance under current-matching conditions at reduced absorber thickness, with only a minor efficiency trade-off.

Table 5. PV metrics of the 2-T Organic/Organic TSC at maximum-efficiency and at current-matching conditions.

	Cell	Thickness	J_{SC} (mA/cm ²)	V_{OC} (V)	FF (%)	PCE (%)
Maximum Efficiency Organic/organic	Top	150 nm	16.46	0.98	71.76	11.53
	Bottom	350 nm	33.57	0.70	56.78	13.41
	Tandem		14.50	1.72	77.92	19.44
Current Matching Organic/organic	Top	89.5 nm	15.40	1.02	68.82	10.88
	Bottom	350 nm	15.40	0.69	77.75	8.38
	Tandem		15.40	1.70	70.27	18.68

Table 6 presents the corresponding photovoltaic metrics for the OSC/Si TSC under maximum efficiency and current-matching conditions. At maximum efficiency, the tandem cell delivers a J_{SC} of 16.34 mA/cm² and a V_{OC} of 1.64 V. Under current-matching conditions, the J_{SC} increases to 16.69 mA/cm², while the V_{OC} remains unchanged at 1.64 V. Similar to the organic/organic configuration, operation under current-matching conditions leads to only a slight reduction in efficiency. Current matching is achieved at a t_{TA} of 124.5 nm, resulting in a tandem PCE of 20.95%. The maximum PCE of 21.13% is attained at a thicker

front absorber layer of 150 nm. This demonstrates that the organic/silicon TSC device can operate close to its peak efficiency under current-matching conditions while employing thinner absorber layers, incurring only a marginal efficiency penalty.

Table 6. PV performance metrics of the monolithic organic/silicon TSC at maximum-efficiency and at current-matching conditions.

	Cell	Thickness	J_{SC} (mA/cm ²)	V_{OC} (V)	FF (%)	PCE (%)
Maximum Efficiency Organic/silicon	Top	150 nm	16.46	0.98	71.76	11.53
	Bottom	80 μ m	36.30	0.636	76.20	17.58
	Tandem		16.34	1.64	78.97	21.13
Current Matching Organic/silicon	Top	124.5 nm	16.69	1.02	73.10	12.44
	Bottom	80 μ m	16.69	0.62	82.48	8.48
	Tandem		16.69	1.64	75.44	20.59

3.5. Current Densities and Band Energy Profiles for Both TSCs

To better understand the operation and charge carrier dynamics of the OSC/OSC and OSC/Si TSC, the current-density and energy-band profiles are presented in Figures 13 and 14. These results provide insight into charge transport and energy alignment within the devices. Figures 13a and 14a illustrate the current-density profiles across the thickness of both TSC structures, organic/organic and organic/silicon, under CM conditions. These figures provide a detailed visualization of the current flow within the devices, emphasizing the interaction between the sub-cells and the recombination interlayer junction. In these plots, three current components are presented: the electron current density (J_n), the hole current density (J_p), and the total current density (J).

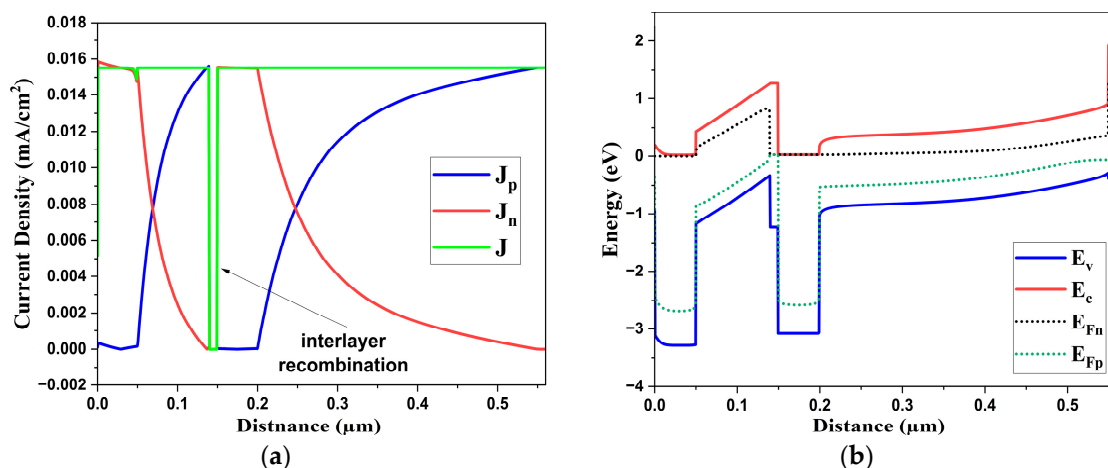


Figure 13. (a) Current density across the thickness of organic/organic TSC at current-match condition and (b) energy-band profile of the OSC/OSC TSC.

For the front organic sub-cell, J_n gradually declines toward the recombination junction due to electron–hole recombination and charge transfer processes. The sharp fall beneath the interlayer indicates an active recombination region, facilitating effective inter-band charge transfer between the two sub-cells. Similarly, in the bottom sub-cell, J_p decreases toward the recombination interlayer, reflecting analogous recombination behavior. The trends observed in J_n and J_p collectively demonstrate a balanced recombination and charge transfer process between the sub-cells. The distinct discontinuity at the recombination interlayer signifies a region of significant carrier interaction, confirming the efficient coupling of the sub-cells in both tandem configurations.

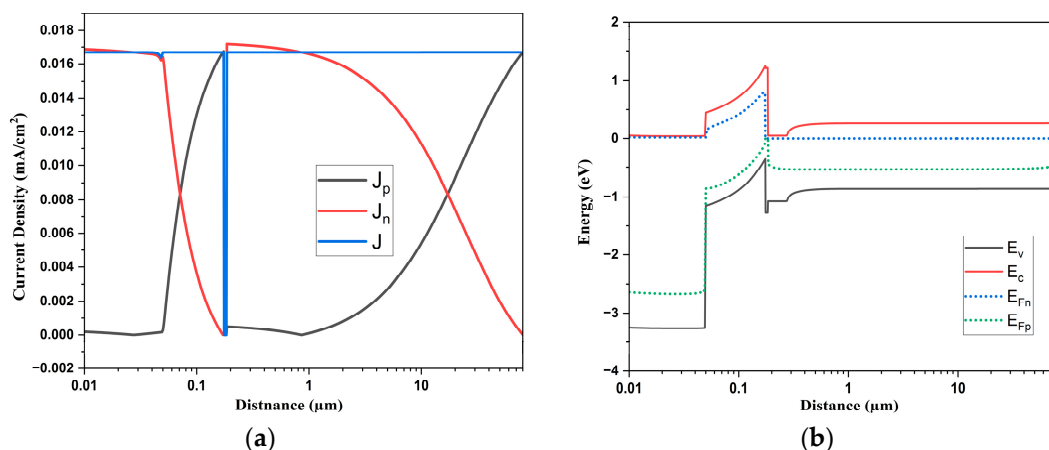


Figure 14. (a) Current density across the thickness of organic/silicon TSC at current-match condition, and (b) energy-band profile of the OSC/Si TSC.

Figures 13b and 14b present the band energy profiles of both TSCs under the same conditions. These profiles provide an in-depth visualization of the band alignment, charge-transport barriers, and potential gradients across the various layers of the tandem structures. The noticeable band bending within the photoactive layers indicates the existence of built-in electric fields, which serve as the primary driving forces for charge separation and transport. Furthermore, the band diagrams reveal the lack of a significant voltage drop at the recombination junction, as evidenced by the close alignment of the electron and hole quasi-Fermi levels (E_{Fn} and E_{Fp}). This alignment implies that the recombination interlayer facilitates efficient charge transfer between sub-cells with minimal energy loss, thereby contributing to the overall high performance and low interfacial resistance of both tandem configurations.

3.6. Organic/Organic Versus Organic/Si Tandem Solar Cells

From a device-physics standpoint, the high performance of the hybrid OSC/Si tandem is mainly a consequence of stronger spectral complementarity and more efficient carrier collection in the bottom sub-cell. In both tandems, the PBDB-T:F-M top absorber acts as a wide-bandgap front junction. The decisive difference appears in the rear junction. When the bottom cell is the ternary organic blend, the tandem benefits from low-temperature processability and reasonable spectral extension into longer wavelengths, but the rear current remains limited by the intrinsically lower carrier mobilities, lower dielectric screening, and stronger sensitivity to trap-assisted recombination that is typical of organic semiconductors. By contrast, replacing the organic rear junction with silicon introduces a bottom absorber with a broader effective absorption window, substantially higher carrier mobilities and more robust collection of photogenerated carriers. This is why the OSC/Si tandem delivers higher J_{SC} and PCE even though its total V_{OC} is slightly lower than that of the OSC/OSC structure.

This comparison is consistent with the calibrated energy-loss analysis and with the thickness-dependent trends observed in this work. The organic top cell exhibits relatively large transmission loss. In an OSC/OSC tandem, the bottom organic cell can recover only part of the transmitted flux, since its optical absorption and electrical extraction remain constrained by finite thickness, moderate mobility, and recombination losses. However, in the OSC/Si tandem, the Si cell converts a larger fraction of the transmitted red and near-infrared photons, thereby reducing the effective spectral loss of the tandem as a whole. Although silicon experiences larger thermalization and voltage losses because of its smaller bandgap, these penalties are overcompensated by the much larger usable

photocurrent and more efficient charge extraction. Therefore, the hybrid tandem achieves a more favorable balance between transmission loss, carrier collection, and series-connected current generation.

The current-matching behavior further reflects the underlying transport mechanisms. In a monolithic two-terminal tandem, the current is limited by the weaker sub-cell, and therefore thickness optimization is fundamentally important to control the optical distribution while preserving electrical extraction. In the OSC/OSC tandem, increasing the top-organic thickness rapidly increases the front-cell photocurrent but simultaneously deprives the bottom organic cell of photons. Since the rear organic cell is already transport-limited, the current-matching window remains relatively narrow. This explains the stronger trade-off between optical gain in the top cell and electrical penalty in the bottom cell. In the OSC/Si tandem, the silicon bottom junction acts as a much stronger current reservoir. Once sufficient photons are transmitted through the front cell, silicon can sustain a higher rear current because of its superior absorption depth and collection efficiency. As a result, the current-matched and maximum-efficiency operating points remain closer to each other, and the efficiency penalty associated with current matching is smaller in the hybrid structure than in the all-organic device.

From the fabrication perspective, the all-organic tandem offers an important technological advantage because both sub-cells can be produced through low-temperature solution-based processing on lightweight and flexible substrates. Nevertheless, the fabrication window is narrow. Since both photoactive junctions are organic, the deposition of upper layers can partially dissolve or restructure previously deposited layers [33,37–40]. The hybrid OSC/Si tandem follows a fundamentally different integration mechanism. In this case, the silicon bottom cell is usually fabricated first using a mature wafer-based or thin silicon process, after which the organic top sub-cell is integrated by direct low-temperature deposition or by lamination approaches. This route benefits from the technological maturity of silicon photovoltaics. However, the integration challenge becomes more severe at the organic/inorganic interface. Surface textures that are beneficial for single-junction silicon light trapping are often unfavorable for very thin organic layers, because excessive roughness can lead to thickness nonuniformity [43,48]. Therefore, OSC/Si tandems require especially careful optimization of refractive-index matching and low-damage transparent contacts.

Although flexibility is frequently regarded as an inherent advantage of all-organic photovoltaics, it should not be considered an exclusive differentiator in the present comparison because the crystalline Si absorber adopted here is only 30 μm thick, a thickness regime compatible with mechanically flexible Si photovoltaic devices. The more distinctive advantage of the OSC/OSC architecture therefore lies in its compatibility with low-temperature and potentially fully solution-processed fabrication.

Overall, the comparison reveals a clear physical trade-off between the two tandem concepts. The OSC/OSC architecture is more compatible with fully solution-processed, lightweight, and mechanically flexible photovoltaics. However, its performance remains more sensitive to interfacial recombination and current-matching constraints. The OSC/Si tandem, on the other hand, sacrifices some voltage advantage but gains substantially in rear-cell current generation, charge-transport robustness, and total conversion efficiency. In other words, the all-organic tandem is more attractive when processing simplicity, low weight, and flexibility are the primary targets, whereas the hybrid organic/silicon tandem is the more promising route when the goal is to maximize efficiency. These considerations explain the simulated superiority of the OSC/Si design in the present study and provide practical guidance for future device engineering in both tandem platforms.

3.7. Comparison with Previously Reported Organic-Based Tandem Solar Cells

To further assess the performance and distinctive features of the proposed TSCs, Table 7 compares the present results with representative previously reported all-organic and hybrid organic/Si tandems. The comparison considers the absorber materials, tandem configuration, investigation approach and connection, as well as principal PV parameters.

Table 7. Comparison of the proposed TSCs with representative previously reported all-organic and hybrid organic/silicon tandems.

Ref.	TSC	Top Absorber	Bottom Absorber	Method	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	PCE (%)
[37]	OSC/OSC	PM6:m-DTC-2F	PTB7-Th:BTPV-4F:PC ₇₁ BM	Exp., 2-T	14.5	1.65	68.5	16.40
[38]	OSC/OSC	PBDB-TCl:AiTC:BTP-eC9	PFBCPZ:AiTC	Exp., 2-T	13.3	2.02	76.6	20.6
[39]	OSC/OSC	PM6:BTP-eC9	PM6:BTP-eC9	Exp., 2-T	—	—	—	19.9
[40]	OSC/OSC	PM6:L8-BO	PTB7-Th:O6T-4F	Exp., 4-T	—	—	—	16.94
[42]	OSC/a-Si	PDPP3T:PC ₆₁ BM	a-Si	Exp., 2-T	7.59	1.45	76.0	8.32
[43]	OSC/c-Si	PBDB-T-2F:Y6	c-Si TOPCon	Exp., 2-T	15.81	1.08	55.57	8.32
[43]	OSC/c-Si	PBDB-T:ITIC	c-Si	Exp., 4-T	—	—	—	15.25
[44]	OSC/c-Si	PBDB-TCl:AiTC	c-Si	TCAD, 2-T	19.28	1.81	79.31	27.60
This work	OSC/OSC	PBDB-T:F-M	PTB7-Th:CO _i 8DFIC:PC ₇₁ BM	TCAD, 2-T	14.50	1.72	77.92	19.44
This work	OSC/c-Si	PBDB-T:F-M	c-Si	TCAD, 2-T	16.34	1.64	78.97	21.13

As summarized in Table 7, the reported performance of organic-based tandems varies considerably depending on the absorber combination, terminal configuration, interconnection strategy, and degree of device optimization. Several experimental all-organic TSCs have demonstrated PCEs approaching or exceeding 20%, whereas reported hybrid organic/Si devices show a wider performance range depending on the organic absorber, silicon structure, and integration approach. The present work is distinguished not by targeting a record simulated PCE, but by providing a direct comparison of all-organic and hybrid organic/c-Si two-terminal tandem architectures within experimentally calibrated TCAD framework.

4. Conclusions

In this work, two different TSC architectures were proposed, designed, and systematically compared using comprehensive optoelectronic TCAD numerical simulations. The first configuration was an all-organic tandem solar cell composed of PBDB-T:F-M as the top sub-cell and PTB7-Th:CO_i8DFIC:PC₇₁BM as the bottom sub-cell, while the second configuration combined an organic top sub-cell (PBDB-T:F-M) with a crystalline silicon bottom sub-cell. The initial organic/organic TSC structure, based on conventional and experimentally calibrated layer configurations, achieved a PCE of 15.7%, whereas the organic/silicon tandem device exhibited a significantly higher initial PCE of 16.85%. Further optimization of absorber thicknesses in both structures led to substantial efficiency improvements. For the organic/organic tandem cell, a top-cell thickness of 150 nm and a bottom-cell thickness of 350 nm resulted in a maximum PCE of 19.44%. In contrast, the organic/silicon tandem configuration reached a higher maximum PCE of 21.13% with a 150 nm organic top absorber and an 80 μ m silicon bottom cell.

Under current-matching conditions, a slight reduction in efficiency was observed compared to the maximum-efficiency scenario. The organic/organic tandem cell achieved a PCE of 18.68% at a top-cell thickness of 89.5 nm, only slightly lower than its peak efficiency, demonstrating that near-optimal performance can be maintained with thinner absorber layers. Similarly, the organic/silicon tandem cell reached a PCE of 20.59% at a current-matched top-cell thickness of 124.5 nm, which is only slightly lower than its maximum efficiency obtained at 150 nm.

Overall, the results clearly indicate that organic/silicon tandem solar cells consistently outperform all-organic tandem configurations in terms of efficiency. Nevertheless, both architectures demonstrate strong potential, particularly due to the use of low-cost, earth-abundant, and environmentally benign materials. These findings highlight the viability of tandem solar cell designs as a promising pathway toward high-efficiency and sustainable photovoltaic technologies.

Beyond the numerical efficiency improvement, the present results provide several practical design implications for the experimental development of organic-based TSCs. In particular, the analysis demonstrates that strict current matching should not necessarily be treated as the sole optimization criterion, since the maximum-PCE condition results from the combined optical and electrical response of the tandem and may occur at a different absorber-thickness combination. Therefore, experimentally selecting absorber thicknesses should consider not only photocurrent balance but also carrier transport and recombination. The identified thickness ranges can consequently serve as useful starting points for experimental fabrication while avoiding unnecessary increases in absorber material once the efficiency gain becomes marginal.

Several limitations should nevertheless be recognized. Although the individual organic and Si sub-cells were calibrated, the complete tandem structures remain primarily simulation-based, particularly the proposed OSC/Si architecture. In addition, the organic bulk-heterojunction layers were described using an effective drift-diffusion framework, while detailed morphology-dependent exciton transport, field-dependent mobility, explicit interface-recombination mechanisms, and fully coherent optical-interference effects were not independently resolved. The ITO interconnecting layer was also treated under a low-loss assumption, whereas practical devices may exhibit additional parasitic absorption. Future work should therefore focus on experimental fabrication and validation of the proposed tandems, detailed optimization of the interconnecting layer and interfaces, and incorporation of more advanced organic-transport and optical models. Such developments would provide an important bridge between the present calibrated TCAD predictions and experimentally realizable high-efficiency TSC devices.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/cryst16080545/s1>, Figure S1: Flow chart of the Silvaco simulation process, illustrating the fundamental steps and governing equations used in the modeling workflow; Figure S2: J-V curve for OSC/OSC vs OSC/Si tandems; Figure S3: Spatial photogeneration profiles across the OSC/OSC tandem solar cell for top PBDB-T:F-M absorber thicknesses of (a) 89.5 nm, corresponding to the current-matching condition, (b) 150 nm, corresponding to the maximum-PCE condition, and (c) 250 nm, representing a comparatively thick top absorber; Figure S4: EQE curves of front and rear sub cells at current matching for (a) optimized OSC/OSC TSC, and (b) optimized OSC/Si TSC; Table S1: Classification, and justification of the material and device parameters used in the calibrated top OSC, bottom OSC, and Si solar-cell models.

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Writing—review and editing, M.F., H.M.H., M.A. and A.S.; Supervision, H.M.H. and A.S.; Software, M.F., M.O. and A.S. All authors have read and agreed to the published version of the manuscript.

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References

1. Pourasl, H.H.; Barenji, R.V.; Khojastehnezhad, V.M. Solar Energy Status in the World: A Comprehensive Review. *Energy Rep.* **2023**, *10*, 3474–3493. [\[CrossRef\]](#)
2. Allouhi, A.; Rehman, S.; Buker, M.S.; Said, Z. Up-to-Date Literature Review on Solar PV Systems: Technology Progress, Market Status and R&D. *J. Clean. Prod.* **2022**, *362*, 132339. [\[CrossRef\]](#)
3. Ibrahim, T.; Durillon, B.; Faraj, J.; Ali, S.; Saudemont, C.; Harion, J.; Khaled, M. A Comprehensive Review of Solar Energy Systems: Technical, Economic, and Environmental Perspectives for Sustainable Development. *Int. Commun. Heat Mass Transf.* **2025**, *165*, 109095. [\[CrossRef\]](#)
4. Ali, A.O.; Elgohr, A.T.; El-Mahdy, M.H.; Zohir, H.M.; Emam, A.Z.; Mostafa, M.G.; Al-Razgan, M.; Kasem, H.M.; Elhadidy, M.S. Advancements in Photovoltaic Technology: A Comprehensive Review of Recent Advances and Future Prospects. *Energy Convers. Manag. X* **2025**, *26*, 100952. [\[CrossRef\]](#)
5. El-Ghonemy, A.M.K. Photovoltaic Solar Energy: Review. *Int. J. Sci. Eng. Res.* **2012**, *3*, 1–43. [\[CrossRef\]](#)
6. Al-Ali, S.; Olabi, A.G.; Mahmoud, M. A Review of Solar Photovoltaic Technologies: Developments, Challenges, and Future Perspectives. *Energy Convers. Manag. X* **2025**, *27*, 101057. [\[CrossRef\]](#)
7. Mahrous, H.; Fedawy, M.; El Sabbagh, M.; Fikry, W.; Gad, M. A Compact 120 GHz Monolithic Silicon-on-Silica Electro-Optic Modulator. *Opt. Quantum Electron.* **2020**, *52*, 111. [\[CrossRef\]](#)
8. Mahrous, H.; Fedawy, M.; Abboud, M.; Shaker, A.; Fikry, W.; Gad, M. A Multi-Objective Genetic Algorithm Approach for Silicon Photonics Design. *Photonics* **2024**, *11*, 80. [\[CrossRef\]](#)
9. Wan, Y.; He, W.; Jaussi, J.; Liao, L.; Pan, D.Z.; Bowers, J.E.; Rong, H. Integrating Silicon Photonics with Complementary Metal–Oxide–Semiconductor Technologies. *Nat. Rev. Electr. Eng.* **2026**, *3*, 15–31. [\[CrossRef\]](#)
10. Green, M.A.; Dunlop, E.D.; Hohl-Ebinger, J.; Yoshita, M.; Kopidakis, N.; Hao, X. Solar Cell Efficiency Tables (Version 59). *Prog. Photovolt. Res. Appl.* **2022**, *30*, 3–12. [\[CrossRef\]](#)
11. Farooq, W.; Alshahrani, T.; Kazmi, S.A.A.; Iqbal, J.; Khan, H.A.; Khan, M.; Raja, A.A.; Rehman, A.U. Materials Optimization for Thin-Film Copper Indium Gallium Selenide (CIGS) Solar Cell Based on Distributed Bragg's Reflector. *Optik* **2021**, *227*, 165987. [\[CrossRef\]](#)
12. Vogelbaum, H.S.; Sauvé, G. Recently Developed High-Efficiency Organic Photoactive Materials for Printable Photovoltaic Cells: A Mini Review. *Synth. Met.* **2017**, *223*, 107–121. [\[CrossRef\]](#)
13. Santhoshi Kiran, K.S.; Preethi, V.; Kumar, S. A Brief Review of Organic Solar Cells and Materials Involved in Its Fabrication. *Mater. Today Proc.* **2022**, *56*, 3826–3829. [\[CrossRef\]](#)
14. Casademont-Viñas, M.; Capolat, D.; Quesada-Ramírez, A.; Reinfelds, M.; Trimmel, G.; Sanviti, M.; Martín, J.; Goñi, A.R.; Kirchartz, T.; Campoy-Quiles, M. Combinatorial Screening of Wide Band-Gap Organic Solar Cell Materials with Open-Circuit Voltage between 1.1 and 1.4 V. *J. Mater. Chem. A* **2024**, *12*, 16716–16728. [\[CrossRef\]](#)
15. Jin, J.; Wang, Q.; Ma, K.; Shen, W.; Belfiore, L.A.; Bao, X.; Tang, J. Recent Developments of Polymer Solar Cells with Photovoltaic Performance over 17%. *Adv. Funct. Mater.* **2023**, *33*, 2213324. [\[CrossRef\]](#)
16. Zhu, L.; Zhang, M.; Zhong, W.; Leng, S.; Zhou, G. Progress and prospects of the morphology of non-fullerene acceptor based high-efficiency organic solar cells. *Energy Environ. Sci.* **2021**, *14*, 4341–4357. [\[CrossRef\]](#)
17. Mohamed El Amine, B.; Zhou, Y.; Li, H.; Wang, Q.; Xi, J.; Zhao, C. Latest Updates of Single-Junction Organic Solar Cells up to 20% Efficiency. *Energies* **2023**, *16*, 3895. [\[CrossRef\]](#)
18. Shi, W.; Han, Q.; Zhao, W.; Wang, R.; Li, L.; Song, G.; Chen, X.; Long, G.; Yao, Z.; Lu, Y.; et al. A Large Conjugated Rigid Dimer Acceptor Enables 20.19% Efficiency in Organic Solar Cells. *Energy Environ. Sci.* **2025**, *18*, 5356–5364. [\[CrossRef\]](#)
19. Zhang, M.; Lin, Z. Efficient Interconnecting Layers in Monolithic All-Perovskite Tandem Solar Cells. *Energy Environ. Sci.* **2022**, *15*, 3152–3170. [\[CrossRef\]](#)
20. Alanazi, T.I.; Eid, O.I.; Okil, M. Numerical Study of Flexible Perovskite/Si Tandem Solar Cell Using TCAD Simulation. *Opt. Quantum Electron.* **2023**, *55*, 1152. [\[CrossRef\]](#)

21. Tahir, S.; Mushtaq, S.; McQueen, J.; Iqbal, J.; Almufarij, R.S.; Alqurashi, R.S.; Bonilla, R.S.; Ashfaq, A. Computational Insights into Wide Bandgap Lead Free Perovskite Solar Cells for Silicon Based Tandem Configurations. *Sci. Rep.* **2025**, *15*, 20020. [[CrossRef](#)] [[PubMed](#)]
22. Sharma, A.K.; Chourasia, R.K.; Chourasia, N.K.; Sangani, K. 2T-Si/Sb2S3 Tandem Solar Cell: Impedance Spectroscopic and Electrical Circuit Analysis. *J. Electroanal. Chem.* **2026**, *1006*, 119895. [[CrossRef](#)]
23. Jošt, M.; Köhnen, E.; Al-Ashouri, A.; Bertram, T.; Tomšič, Š.; Magomedov, A.; Kasparavicius, E.; Kodalle, T.; Lipovšek, B.; Getautis, V.; et al. Perovskite/CIGS Tandem Solar Cells: From Certified 24.2% toward 30% and Beyond. *ACS Energy Lett.* **2022**, *7*, 1298–1307. [[CrossRef](#)]
24. Priyanka, E.; Muchahary, D. Performance Improvement of Perovskite/CIGS Tandem Solar Cell Using Barium Stannate Charge Transport Layer and Achieving PCE of 39% Numerically. *Sol. Energy* **2024**, *267*, 112218. [[CrossRef](#)]
25. Alanazi, T.I.; El Sabbagh, M. Proposal and Design of Flexible All-Polymer/CIGS Tandem Solar Cell. *Polymers* **2023**, *15*, 1823. [[CrossRef](#)] [[PubMed](#)]
26. Mailoa, J.P.; Lee, M.; Peters, I.M.; Buonassisi, T.; Panchula, A.; Weiss, D.N. Energy-Yield Prediction for II-VI-Based Thin-Film Tandem Solar Cells. *Energy Environ. Sci.* **2016**, *9*, 2644–2653. [[CrossRef](#)]
27. Zein, W.; Alanazi, T.I.; Saeed, A.; Salah, M.M.; Mousa, M. Proposal and Design of Organic/CIGS Tandem Solar Cell: Unveiling Optoelectronic Approaches for Enhanced Photovoltaic Performance. *Optik* **2024**, *302*, 171719. [[CrossRef](#)]
28. Elkattan, M.; Gad, M. Investigation of the Absorption Edge and the Optical Bandgap of PVA/PVP-Based Thin Films. *J. Electron. Mater.* **2025**, *54*, 831–847. [[CrossRef](#)]
29. Gad, M.; Elkattan, M. Tailoring the Optical Properties of Polyvinyl Alcohol-Polyvinyl Pyrrolidone Based Polymers. *Opt. Quantum Electron.* **2023**, *55*, 985. [[CrossRef](#)]
30. Lim, J.W. Polymer Materials for Optoelectronics and Energy Applications. *Materials* **2024**, *17*, 3698. [[CrossRef](#)] [[PubMed](#)]
31. R. Murad, A.; Iraqi, A.; Aziz, S.B.; N. Abdullah, S.; Brza, M.A. Conducting Polymers for Optoelectronic Devices and Organic Solar Cells: A Review. *Polymers* **2020**, *12*, 2627. [[CrossRef](#)] [[PubMed](#)]
32. Li, M.; Gao, K.; Wan, X.; Zhang, Q.; Kan, B.; Xia, R.; Liu, F.; Yang, X.; Feng, H.; Ni, W.; et al. Solution-Processed Organic Tandem Solar Cells with Power Conversion Efficiencies > 12%. *Nat. Photonics* **2017**, *11*, 85–90. [[CrossRef](#)]
33. Ullah, F.; Chen, C.C.; Choy, W.C.H. Recent Developments in Organic Tandem Solar Cells toward High Efficiency. *Adv. Energy Sustain. Res.* **2021**, *2*, 2000050. [[CrossRef](#)]
34. He, R.; Ren, S.; Chen, C.; Yi, Z.; Luo, Y.; Lai, H.; Wang, W.; Zeng, G.; Hao, X.; Wang, Y.; et al. Wide-Bandgap Organic-Inorganic Hybrid and All-Inorganic Perovskite Solar Cells and Their Application in All-Perovskite Tandem Solar Cells. *Energy Environ. Sci.* **2021**, *14*, 5723–5759. [[CrossRef](#)]
35. Lim, J.; Park, N.G.; Il Seok, S.; Saliba, M. All-Perovskite Tandem Solar Cells: From Fundamentals to Technological Progress. *Energy Environ. Sci.* **2024**, *17*, 4390–4425. [[CrossRef](#)]
36. Zhou, J.; Fu, S.; Zhou, S.; Huang, L.; Wang, C.; Guan, H.; Pu, D.; Cui, H.; Wang, C.; Wang, T.; et al. Mixed Tin-Lead Perovskites with Balanced Crystallization and Oxidation Barrier for All-Perovskite Tandem Solar Cells. *Nat. Commun.* **2024**, *15*, 2324. [[CrossRef](#)] [[PubMed](#)]
37. Jia, Z.; Qin, S.; Meng, L.; Ma, Q.; Angunawela, I.; Zhang, J.; Li, X.; He, Y.; Lai, W.; Li, N.; et al. High Performance Tandem Organic Solar Cells via a Strongly Infrared-Absorbing Narrow Bandgap Acceptor. *Nat. Commun.* **2021**, *12*, 178. [[CrossRef](#)] [[PubMed](#)]
38. Wang, J.; Zheng, Z.; Bi, P.; Chen, Z.; Wang, Y.; Liu, X.; Zhang, S.; Hao, X.; Zhang, M.; Li, Y.; et al. Tandem Organic Solar Cells with 20.6% Efficiency Enabled by Reduced Voltage Losses. *Natl. Sci. Rev.* **2023**, *10*, nwad085. [[CrossRef](#)] [[PubMed](#)]
39. Yu, J.; Hao, L.; Zhang, K.; Zheng, J.; Zhang, J.; Xu, Y.; Dong, M.; Xie, J.; Li, H.; Luo, X.; et al. High Efficiency Homojunction Tandem Organic Solar Cells with All Solution Processable Interconnect Layer. *Chem. Eng. J.* **2024**, *500*, 156887. [[CrossRef](#)]
40. Bernal-texca, F.; Martorell, J. Four-Terminal Tandem Based on a PM6: L8-BO Transparent Solar Cell and a 7 Nm Ag Layer Intermediate Electrode. *Sol. RRL* **2024**, *8*, 2300728. [[CrossRef](#)]
41. Yu, Z.J.; Carpenter, J.V.; Holman, Z.C. Techno-Economic Viability of Silicon-Based Tandem Photovoltaic Modules in the United States. *Nat. Energy* **2018**, *3*, 747–753. [[CrossRef](#)]
42. Liu, X.; Wu, S.M.; Liang, Y.; Xu, W.; Liu, H. Organic/Amorphous Silicon Hybrid Tandem Solar Cell with PDPP3T as Organic Active Layer. *Microelectron. J.* **2021**, *113*, 105098. [[CrossRef](#)]
43. Park, H.J.; Park, S.H.; Lee, S.W.; Kang, Y.; Kim, D.; Son, H.J.; Lee, H.S. Novel Polymer-Based Organic/c-Si Monolithic Tandem Solar Cell: Enhanced Efficiency Using Interlayer and Transparent Top Electrode Engineering. *Macromol. Rapid Commun.* **2021**, *42*, 2100305. [[CrossRef](#)] [[PubMed](#)]
44. Salem, M.S.; Okil, M.; Shaker, A.; Abouelatta, M.; Salah, M.M.; Al-Dhlan, K.A.; Gad, M. TCAD-Based Design and Optimization of Flexible Organic/Si Tandem Solar Cells. *Crystals* **2024**, *14*, 584. [[CrossRef](#)]
45. Wang, S.; Li, W.; Yu, C.; Shi, W.; Kang, Q.; Cao, F.; Gao, K.; Yang, L.; Yang, B.; Zhou, J.; et al. Flexible Perovskite/Silicon Tandem Solar Cells with 33.6% Efficiency. *Nature* **2026**, *649*, 59–64. [[CrossRef](#)] [[PubMed](#)]

46. Mosalanezhad, R.; Shayesteh, M.R.; Pourahmadi, M. Silvaco TCAD Modeling, Optical Simulation, and Optimization for High-Current Perovskite and u-CIGS Tandem Solar Cells with Efficiencies above 30%. *Sci. Rep.* **2026**, *16*, 8611. [[CrossRef](#)] [[PubMed](#)]
47. Firdaus, Y.; Ho, C.H.Y.; Lin, Y.; Yengel, E.; Le Corre, V.M.; Nugraha, M.I.; Yarali, E.; So, F.; Anthopoulos, T.D. Efficient Double- and Triple-Junction Nonfullerene Organic Photovoltaics and Design Guidelines for Optimal Cell Performance. *ACS Energy Lett.* **2020**, *5*, 3692–3701. [[CrossRef](#)]
48. Wang, X.; Zheng, J.; Ying, Z.; Li, X.; Zhang, M.; Guo, X.; Su, S.; Sun, J.; Yang, X.; Ye, J. Ultrathin (~30 Mm) Flexible Monolithic Perovskite/Silicon Tandem Solar Cell. *Sci. Bull.* **2024**, *69*, 1887–1894. [[CrossRef](#)] [[PubMed](#)]
49. Suchikova, Y.; Nazarovets, S.; Konuhova, M.; Popov, A.I. Binary Oxide Ceramics (TiO₂, ZnO, Al₂O₃, SiO₂, CeO₂, Fe₂O₃, and WO₃) for Solar Cell Applications: A Comparative and Bibliometric Analysis. *Ceramics* **2025**, *8*, 119. [[CrossRef](#)]
50. Sani, S.; Usman, A.; Bhatranand, A.; Jiraraksopakun, Y.; Muhammad, K.S.; Yahaya, U. A Study on Defect, Doping, and Performance of ETLs (ZnO, TiO₂, and IGZO) for the Lead-Free CsSnCl₃ Perovskite Solar Cell by SCAPS-1D Framework. *Mater. Today Commun.* **2024**, *38*, 107575. [[CrossRef](#)]
51. Soro, D.; Sylla, A.; Gbané, A.; Franck, B.A.; Guaita, J.M.; Bouich, A.; Toure, S.; Marí, B. Modeling and Simulation of an Organic Photovoltaic Cell: ITO/MoO₃/CARAPA/PCBM/Alq₃/Al with SCAPS. *Model. Numer. Simul. Mater. Sci.* **2024**, *14*, 79–96. [[CrossRef](#)]
52. Zuo, L.; Shi, X.; Jo, S.B.; Liu, Y.; Lin, F.; Jen, A.K.-Y. Tackling Energy Loss for High-Efficiency Organic Solar Cells with Integrated Multiple Strategies. *Adv. Mater.* **2018**, *30*, 1706816. [[CrossRef](#)] [[PubMed](#)]
53. Zhang, Q.; Chen, X.; Lim, E.L.; Shi, L.; Wei, Z. Advancing All-Perovskite Two-Terminal Tandem Solar Cells: Optimization of Wide- and Narrow-Bandgap Perovskites and Interconnecting Layers. *Energy Environ. Sci.* **2025**, *18*, 3060–3084. [[CrossRef](#)]

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