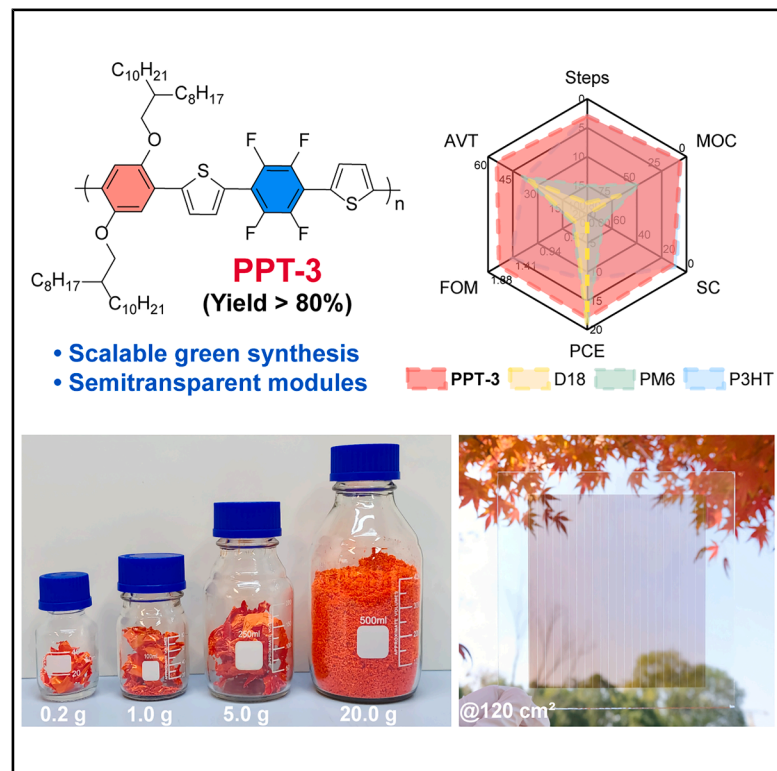


Scalable polymer for large-area semitransparent organic photovoltaics

Graphical abstract



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In brief

A scalable polymer donor (PPT-3) was designed for semitransparent solar windows. For the first time, we demonstrated the scalable synthesis of high-performance polymer donors (PCE >18%) at the 20-g scale. Ambient blade-coated semitransparent modules (120 cm²) attained a record efficiency of 6.69% with 40% transparency. This work provides a commercially viable approach to bridge the lab-to-fab gap for OSCs technology.

Highlights

- The first scalable synthesis of high-performance polymer donors with PCE >18%
- The material synthesis cost of the new polymer PPT-3 is comparable to that of P3HT
- A record-high efficiency of 18.03% is achieved using a DArP-derived polymer
- 120 cm² semitransparent modules attained a record 6.69% efficiency with 40% transparency

Article

Scalable polymer for large-area semitransparent organic photovoltaics

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CONTEXT & SCALE Although organic solar cells (OSCs) have achieved lab efficiencies exceeding 20%, their commercial application faces two key barriers: material scalability and suitable application scenarios. This study introduces PPT-3, a scalable polymer donor designed for semitransparent solar windows. Using simplified molecular design and tin-free synthesis, we achieved over 18% efficiency in opaque devices and a record 6.69% efficiency with 40% transparency in 120 cm² semitransparent modules. Crucially, we demonstrate the first scalable production of high-performance polymer donors at the 20-g scale, demonstrating a commercially viable approach to bridge the lab-to-fab gap for OSC technology.

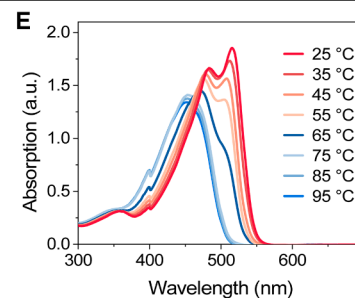
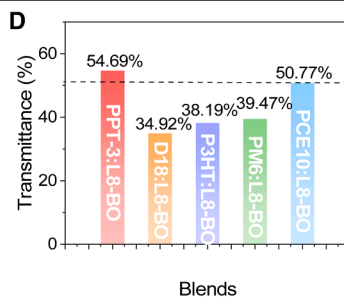
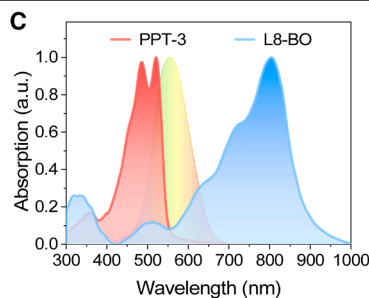
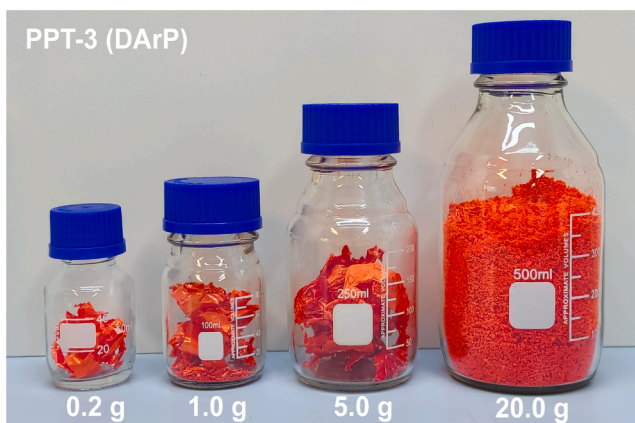
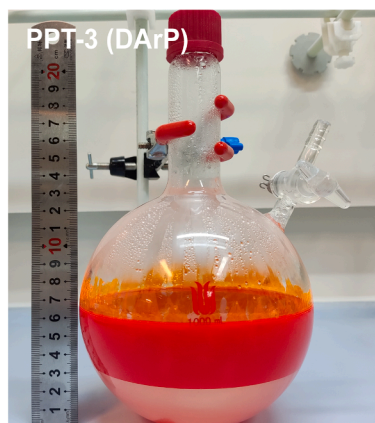
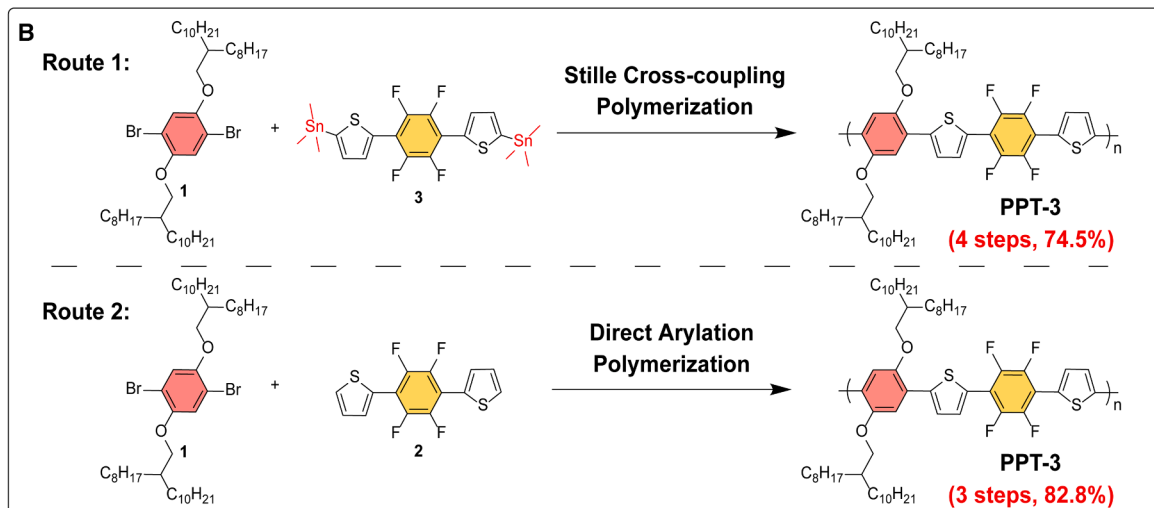
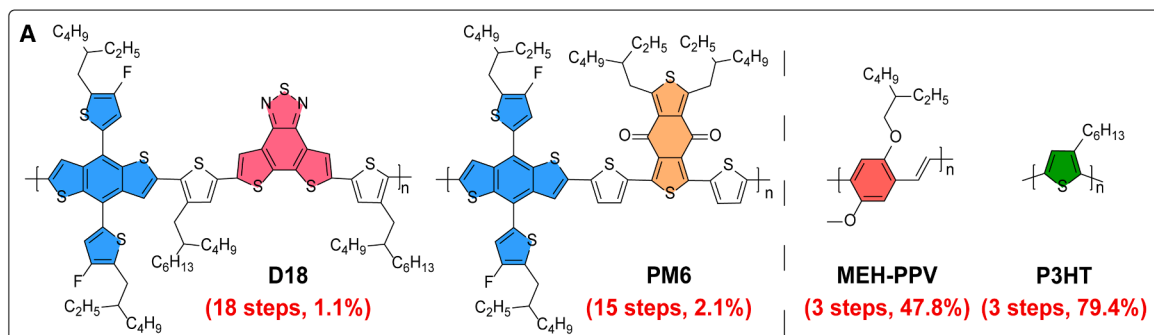
SUMMARY

Organic solar cells (OSCs) have achieved power conversion efficiencies (PCEs) exceeding 20%, yet the transition from lab to market remains challenging. This study proposes a systematic molecular engineering paradigm for developing cost-effective polymer donors, exemplified by PPT-3, specifically engineered for semitransparent solar windows. Using simple monocyclic aromatic units, we simplified the synthesis, enabling scalable production from milligrams to 20 g. Opaque OSCs based on PPT-3 achieved PCEs exceeding 18%, with excellent batch-to-batch reproducibility across multiple scales, including three Stille batches (0.2–5.0 g) and four direct-arylation batches (0.2–20.0 g). Moreover, ambient blade-coated semitransparent modules achieved a record PCE of 6.69%, an average visible transmittance of 40.30%, and a light-utilization efficiency of 2.70% over a 120 cm² active area. This work demonstrates the first scalable synthesis of high-performance polymers (PCE >18%) via a tin-free polymerization route, offering a transformative pathway for advancing OSCs from lab-scale research to commercial viability.

INTRODUCTION

Photovoltaic technology, as a sustainable and renewable energy solution, plays a pivotal role in mitigating global energy crises and environmental degradation.^{1–6} Among emerging photovoltaic technologies, organic solar cells (OSCs) have attracted considerable interest due to their unique advantages: light weight, mechanical flexibility, solution processability, and semitranspar-

ency.^{7–12} Driven by innovations in photovoltaic materials and device engineering, OSCs have achieved unprecedented progress, with certified power conversion efficiencies (PCEs) surpassing 20%.^{13–18} Nevertheless, the commercialization of OSCs has progressed more slowly than initially anticipated. Although historically recognized for the potential of OSCs in low-cost material systems and scalable roll-to-roll manufacturing, the rapid evolution of inorganic photovoltaics, particularly in efficiency



(legend on next page)

enhancement and cost reduction,^{19–21} has eroded the competitiveness of OSCs in conventional energy markets. This evolving landscape highlights the imperative to explore niche applications where OSCs can provide unique functionalities.

Semitransparent OSCs (ST-OSCs) exemplify such a strategic direction, combining energy generation with visible-light transmission for applications in building-integrated photovoltaics, vehicular glazing, and agrivoltaics. The exquisitely adjustable absorption characteristics of organic semiconductors endow ST-OSCs with the ability to function as power-generating windows featuring an extraordinarily high level of visible transmittance.^{22–33} This remarkable feature not only addresses the dual requirements of energy production and light penetration but also carves out a distinct market segment. However, most ST-OSCs still employ polymer donors optimized for opaque devices, where strong visible absorption by polymer donors such as PM6 and D18 disrupts the critical balance between PCEs and average visible transmittance (AVT).^{34–36} To address this limitation, we propose the rational design of high-mobility ultra-wide-band-gap polymer donors that synergistically pair with near-infrared electron acceptors.³⁷ This strategy circumvents visible absorption by the donor component, as ultrawide-band-gap polymers not only enable efficient hole transport but also exhibit selective near-ultraviolet absorption. This combination, supported by theoretical simulations,^{38,39} ensures optimal photovoltaic performance while providing visible-light transmission that aligns with the human eye photopic response curve, thereby maximizing both PCEs and AVT.

Beyond the identification of a distinctive application scenario, the advancement of industrial production in the photovoltaic realm is predominantly cost driven. These economic imperatives dictate that the production of organic photovoltaic materials must be economically viable. Specifically, organic photovoltaic materials should be of a relatively simple structure and obtainable through a minimal number of synthesis and purification steps.^{40–43} However, the pursuit of higher PCEs has led to a significant increase in the chemical complexity of organic photovoltaic materials, especially polymer donors.^{44–46} In contrast to structurally simple classical polymers, such as MEH-PPV and P3HT, state-of-the-art polymer donors, such as PM6 and D18, require multistep synthesis and rigorous purification protocols, resulting in elevated production costs, low overall yields, and extended production cycles (Figure 1A).^{14,22} These factors collectively undermine cost-effectiveness and hinder industrial scalability of OSCs. Economic analyses have revealed that photoactive materials account for the majority of OSC production costs, with polymer donors alone contributing over 50% of the total material expenses.^{47–51} Such high costs, coupled with the

associated risks, present formidable barriers to industrial adoption of OSCs. Moreover, the synthesis of state-of-the-art polymer donors often relies on hazardous reagents, including flammable and neurotoxic compounds, for monomer preparation and cross-coupling reactions.^{52–54} The use of organotin reagents, for instance, poses significant risks to human health (e.g., neurotoxicity and immune system damage) and environmental safety, rendering them unsuitable for sustainable large-scale production.^{52–54} To facilitate the transition of OSCs from laboratory research to industrial applications, it is imperative to develop polymer donors that balance cost-effectiveness with environmental sustainability, ensuring their long-term feasibility and scalability.

In this study, we demonstrate the large-scale synthesis of a novel wide-band-gap polymer donor (PPT-3) through a tin-free, direct-arylation polymerization (DAP) route, successfully scaling production from mg to 20 g while maintaining high synthetic quality. The optimized OSCs based on PPT-3 exhibited exceptional photovoltaic performance, achieving a PCE exceeding 18% and demonstrating outstanding batch-to-batch reproducibility across multiple synthesis scales, including three Stille polymerization batches (0.2–5.0 g) and four DAP batches (0.2–20.0 g). Notably, large-area semitransparent modules with an active area of 120 cm² based on PPT-3 achieved a PCE of 6.69% and an AVT of 40.30%, representing one of the highest performances reported to date for semitransparent modules. Moreover, a thorough cost-effectiveness assessment demonstrated a substantial decline in the cost burden linked to the production of polymer donors, thereby highlighting the economic viability of PPT-3. Through a meticulous and systematic approach to surmounting the core challenges associated with scalability, cost-effectiveness, and niche application scenarios, this study demonstrates a promising avenue for the prospective evolution and real-world utilization of OSC technology.

RESULTS

Polymer design, synthesis, and characterization

Starting with a monocyclic aromatic polymer design, we selected benzene and thiophene units as the foundational structures for constructing a poly(phenylene-*alt*-thiophene) skeleton (Figure 1B, Note S1, and Figures S1–S21). To facilitate a planar polymer structure, fluorine atoms and alkoxy hydrocarbon substituents were introduced to the benzene units,^{55–57} enabling multiple non-covalent interactions, such as F...S, S...O, and C–H...F. Furthermore, the presence of alkoxy groups endows the polymer with excellent solubility to dissolve in typical organic solvents, such as chloroform, chlorobenzene, and toluene. This

Figure 1. Design, synthesis, and characterization of PPT-3

- The chemical structures, synthesis steps, and total yield of D18, PM6, MEH-PPV, and P3HT.
- The chemical structure and synthesis routes of PPT-3. Route 1: Stille cross-coupling polymerization. Route 2: DAP. The reaction images and final products of PPT-3 (from 0.2 to 20.0 g) via the DAP route.
- The human eye photopic response curve and normalized absorption spectra of PPT-3 and L8-BO as thin films.
- The transmittance of blends based on different polymer donors (PPT-3, D18, P3HT, PM6, and PCE10) and L8-BO. The ratio of donor and acceptor is maintained at 1:1.2, with a thickness of 100 nm.
- The temperature-dependent absorption spectra of PPT-3 in chlorobenzene. When the temperature increased from 25 °C to 95 °C, the 0-0 absorption peak gradually decreased and disappeared at 75 °C.

Table 1. The scalable polymerization condition of PPT-3

Batch	Catalyst	Ligand	Additive	Base	M_n^c (kg mol ⁻¹)	D_M^d	Yield (%) ^e
Stille-0.2 ^a	Pd ₂ (dba) ₃	P(o-MePh) ₃	–	–	60	1.80	96
Stille-1.0 ^a	Pd ₂ (dba) ₃	P(o-MePh) ₃	–	–	69	1.96	94
Stille-5.0 ^a	Pd ₂ (dba) ₃	P(o-MePh) ₃	–	–	68	2.15	96
DARp-0.2 ^b	Pd ₂ (dba) ₃	P(o-OMePh) ₃	NDA	Cs ₂ CO ₃	56	1.96	95
DARp-1.0 ^b	Pd ₂ (dba) ₃	P(o-OMePh) ₃	NDA	Cs ₂ CO ₃	65	2.15	95
DARp-5.0 ^b	Pd ₂ (dba) ₃	P(o-OMePh) ₃	NDA	Cs ₂ CO ₃	59	2.14	96
DARp-20.0 ^b	Pd ₂ (dba) ₃	P(o-OMePh) ₃	NDA	Cs ₂ CO ₃	58	1.88	93

^aStille cross-coupling polymerization conditions: monomer 1 (1 equiv), monomer 3 (1 equiv), Pd₂(dba)₃ (0.02 equiv), P(o-MePh)₃ (0.16 equiv), solvent (o-xylene), monomer concentration (0.15 mmol mL⁻¹), 110°C, 48 h. The number after Stille represents the weight of each batch of products.

^bDirect-arylation polymerization conditions: monomer 1 (1 equiv), monomer 2 (1 equiv), Pd₂(dba)₃ (0.01 equiv), P(o-OMePh)₃ (0.04 equiv), solvent (o-xylene), additive (NDA, neodecanoic acid, 1 equiv), base (Cs₂CO₃, 3 equiv), monomer concentration (0.15 mmol mL⁻¹), 110°C, 48 h. The number after DARp represents the weight of each batch of products.

^c M_n : number average molecular weight.

^d D_M : molar-mass dispersity (M_w/M_n).

^eIsolated yields after Soxhlet extraction.

systematic molecular design strategy was developed to simultaneously reduce synthetic complexity and minimize visible-light absorption. We initiated the synthesis of PPT-3 using classical Stille cross-coupling polymerization, with the detailed polymerization conditions summarized in Table 1. Based on prior studies, Pd₂(dba)₃ and P(o-MePh)₃ were selected as the catalyst combination, resulting in batch Stille-0.2 with a molecular weight (M_n) of 60 kg mol⁻¹ and a high yield of 96%. The product quantity for Stille-0.2 was approximately 0.2 g, which aligns with typical polymer preparation quantities reported in the literature.^{14,22} Building upon this result, we scaled up the polymerization, conducting batch reactions Stille-1.0 (1 g) and Stille-5.0 (5 g), yielding M_n values of 69 and 68 kg mol⁻¹, respectively (Table 1). These successful gram-scale polymerizations demonstrate the feasibility of large-scale production of PPT-3 via the Stille method.

However, for future large-scale production, synthesizing 100 g of PPT-3 via the Stille method would require approximately 15.41 g of *n*-BuLi and 49.26 g of Me₃SnCl, as detailed in Note S2. The inherent flammability and pronounced neurotoxicity of these compounds render the large-scale synthesis of polymer donors an unsustainable endeavor. Consequently, we attempted to synthesize PPT-3 via DARp, an economically efficient and ecologically green method.^{58–62} Taking the synthesis of 100 g of PPT-3 using DARp as an example, this approach not only reduces the use of hazardous reagents but also lowers material costs by approximately 40% while increasing the total yield from 74.5% to 82.8% (Table S1). Drawing on the palladium-catalyzed (aromatic) C–H bond metalation-deprotonation mechanism,^{63,64} the varied reactivity of hydrogen atoms in the thiophene unit enables the DARp polymerization of PPT-3 (Note S2, Figures S22–S24). Although numerous DARp polymerization conditions have been developed, most high-performance polymer donors with PCEs exceeding 10% in OSCs were synthesized using Ozawa's protocol,⁶⁵ as demonstrated in prior studies.^{66–68} Thus, we also optimized the DARp conditions of PPT-3 based on Ozawa's protocol—including catalyst, ligand, monomer concentration, additives,^{69,70} and solvent—and successfully scaled up the DARp synthesis of PPT-3 by 100-fold,

increasing the batch size from mg to 20 g in the laboratory (Table 1; Figures S25–S28; Tables S2 and S3; Video S1). The M_n of PPT-3 synthesized via DARp ranged from 50 to 70 kg mol⁻¹, comparable to that achieved through Stille polymerization (Figure S29). Furthermore, ¹H and ¹⁹F nuclear magnetic resonance (NMR) analyses confirmed the consistent polymer structure of PPT-3 synthesized via both Stille and DARp methods. The characteristic proton signals were unambiguously assigned to the polymer backbone and side chain (Figures S30–S32). The ¹⁹F NMR spectra of PPT-3 showed a consistent signal at about –139.83 ppm, matching the fluorine signal of monomer 2 (–139.92 ppm) (Figures S33–S35). These results demonstrate molecular-level consistency in material quality and the feasibility of large-scale PPT-3 production using tin-free synthetic methods.

The ultraviolet-visible (UV-vis) absorption spectrum of the PPT-3 neat film exhibits an absorption edge at 550 nm, corresponding to an optical band gap of 2.26 eV (Figure 2C). The absorption coefficients of PPT-3 neat films were calculated to be 9.87×10^4 cm⁻¹ (Stille-1.0 batch) and 9.82×10^4 cm⁻¹ (DARp-1.0 batch), respectively (Figure S36). The nearly identical absorption coefficients demonstrate excellent material consistency between the two synthetic methods. Distinct 0-0 and 0-1 absorption peaks at 485 and 521 nm, respectively, indicate strong π - π interactions between polymer chains in the solid state. Notably, the blend of PPT-3 with the state-of-the-art electron acceptor L8-BO demonstrates a transmittance of 54.69% (Figures 1D and S37), attributed to minimal spectral overlap with the human eye photopic response, highlighting its potential for ST-OSCs. Temperature-dependent absorption spectra in chlorobenzene reveal consistent aggregation behavior across different PPT-3 batches, with deaggregation temperatures (defined by the disappearance of the 0-0 peak) of 75°C (Figures 1E and S38). This consistent temperature-dependent aggregation behavior simplifies morphology control and ensures reproducibility during device fabrication. The energy levels of PPT-3 were determined by cyclic voltammetry (CV) (Figure S39). The highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels

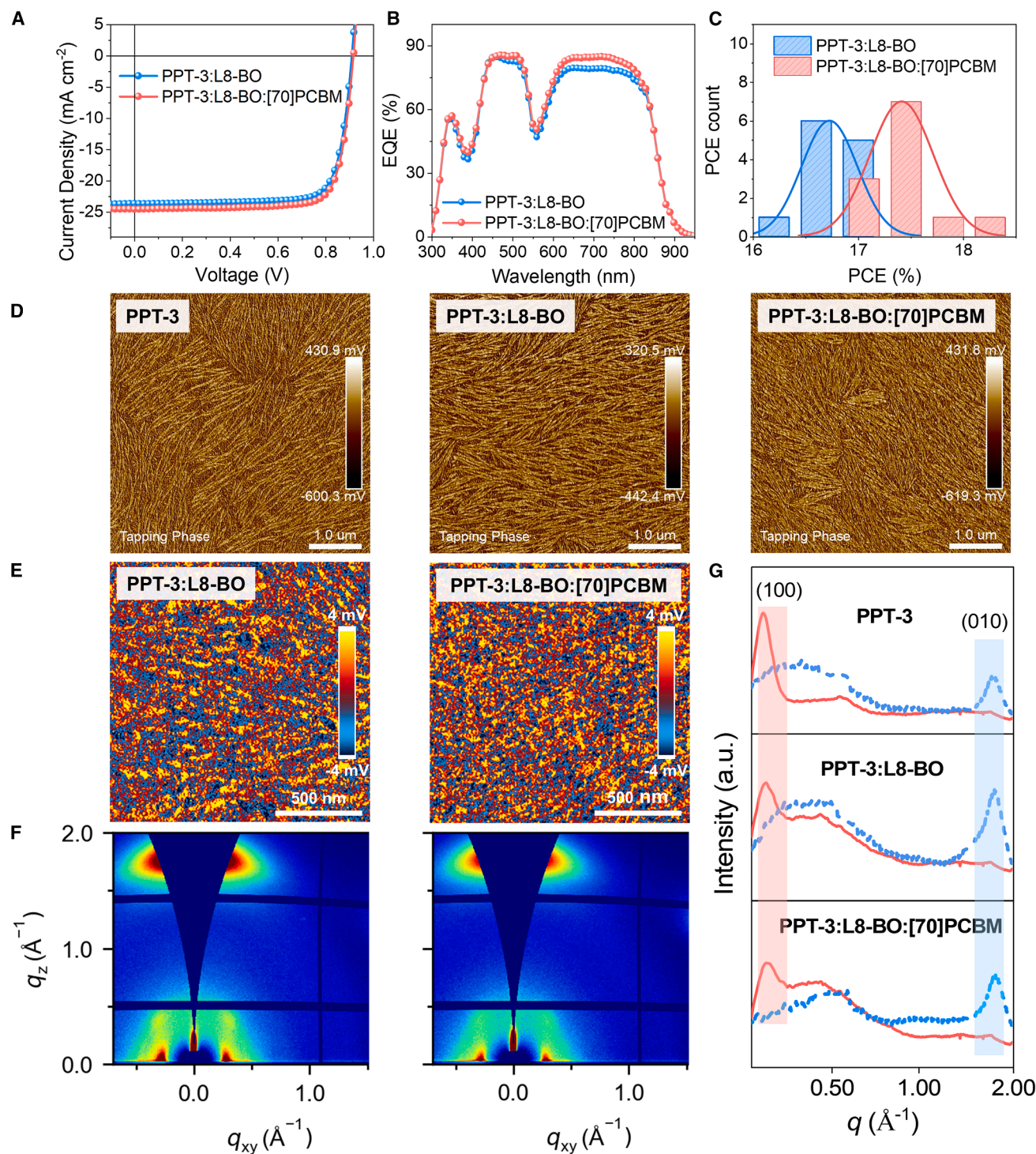


Figure 2. Photovoltaic properties and morphology investigation

(A–C) (A) The J–V curves, (B) EQE spectra, and (C) statistical PCE distribution of the opaque OSCs based on PPT-3. The integrated J_{sc} calculated from EQE data has less than a 5% mismatch with J–V measurements under a solar simulator.

(D) The AFM phase images of PPT-3 neat film as well as binary and ternary blends.

(E) The AFM-IR images of binary and ternary blends at a wave number of $1,430 \text{ cm}^{-1}$ (representing L8-BO).

(F) The two-dimensional GIWAXS patterns of binary and ternary blends.

(G) The one-dimensional GIWAXS line-cut profiles of PPT-3 neat film as well as binary and ternary blends.

Table 2. Device parameters of the opaque OSCs based on PPT-3 under AM1.5G irradiation (100 mW cm⁻²)

Active layer	V_{oc} (V)	J_{sc} (mA cm ⁻²)	J_{sc} (mA cm ⁻²) ^a	FF (%)	PCE (%) ^b
PPT-3:L8-BO = 1:1.2	0.91	23.69	22.64	79.13	17.04 (16.72 ± 0.26)
PPT-3:L8-BO:[70]PCBM = 1:1.2:0.1	0.91	24.58	23.85	79.71	18.13 (17.42 ± 0.30)

^aThe current densities were integrated from the EQE curves of the solar cells.

^bThe statistics were obtained from over 12 devices.

of PPT-3 were calculated to be -5.54 and -3.28 eV, respectively, which match well with those of L8-BO. This harmonious match of energy levels not only facilitates highly efficient exciton dissociation but also favors a high open-circuit voltage (V_{oc}). Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were employed to assess the thermal properties of PPT-3 (Figures S40 and S41). The decomposition temperature at 5% weight loss (T_d) is 376°C , indicating sufficient thermal stability for OSC fabrication. Additionally, PPT-3 exhibits distinct melting and crystallization peaks, with corresponding melting (T_m) and crystallization (T_c) temperatures of 260°C and 234°C , respectively. Grazing-incidence wide-angle X-ray scattering (GIWAXS) was used to investigate the crystalline ordering and orientation of PPT-3 (Figure S42). Neat PPT-3 films demonstrate favorable face-on orientation, with clear (010) diffraction in the out-of-plane (OOP) direction and (100) diffraction in the in-plane (IP) direction. The characteristic distances for π - π stacking and lamellar stacking are 3.62 and 23.00 Å, respectively, with corresponding coherence lengths (L_c) of 28.26 and 104.66 Å. The strong interchain interactions and polymer crystallinity of PPT-3 could be a result of the rigid polymer skeleton with multiple non-covalent interactions. These features synergistically enhance charge transport, yielding a hole mobility of $4.41 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for the PPT-3 neat film (Figure S43). Moreover, the neat PPT-3 film displays a distinct nanofiber network structure, which will be discussed in detail below. Density functional theory (DFT) calculations corroborate the coplanar conjugated skeleton of PPT-3 (Figure S44), stabilized by non-covalent interactions as observed in analogous small-molecule single crystals.^{71,72} To confirm the presence of non-covalent interactions in the PPT-3 skeleton, we further measured the interatomic distances. The observed distances of 2.73 – 2.74 Å (O...S), 2.78 – 2.79 Å (F...S), and 2.24 – 2.25 Å (F...H) are significantly shorter than the sum of their corresponding van der Waals radii (3.31 Å for O...S, 3.27 Å for F...S, and 2.67 Å for F...H), which provides definitive evidence for the existence of these non-covalent interactions (Figure S45).⁷³ This coplanar conjugated skeleton promotes intermolecular interactions, thereby enhancing crystallinity and charge carrier mobility.

Photovoltaic properties and morphology investigation

To assess the photovoltaic performance of PPT-3, the Stille-1.0 batch was selected for systematic device optimization, morphology characterization, and electrical property analysis. The current density-voltage (J - V) characteristics, external quantum efficiency (EQE) spectra, and PCE distribution of the optimal devices are shown in Figures 2A–2C. The device parameters are listed in Table 2 (Tables S4–S6). The binary OSCs based on PPT-3:L8-BO achieved a PCE of 17.04% with a V_{oc} of 0.91 V, a J_{sc} of 23.69 mA cm^{-2} , and an FF of 79.13% . By incorporating

[70]PCBM as a ternary component, the PPT-3:L8-BO:[70]PCBM-based devices exhibited enhanced performance, delivering a PCE of 18.13% with a V_{oc} of 0.91 V, a J_{sc} of 24.58 mA cm^{-2} , and an FF of 79.71% . This represents the highest PCE for OSCs based on monocyclic aromatic polymers to date (Figures S46 and S47; Table S7). The enhanced J_{sc} and FF of ternary OSCs suggests that both carrier generation and transport processes have been optimized. In the EQE spectra, the response in the 400 – 550 nm range is mainly attributed to the absorption of PPT-3, while the response in the 630 – 900 nm range stems from L8-BO. Due to the blue-shifted absorption of PPT-3, the EQE spectrum shows a remarkable decline in the 500 – 630 nm region. This spectral window aligns well with the human eye photopic response, highlighting the potential of PPT-3 for semitransparent applications. Furthermore, the enhanced EQE values of ternary OSCs in the 650 – 900 nm range indicate the improved photon-utilization efficiency, consistent with the enhanced J_{sc} . The photostability of binary and ternary OSCs based on PPT-3 under maximum power point tracking (MPPT) were measured, as shown in Figure S48. The PPT-3:L8-BO:[70]PCBM-based devices demonstrated better stability, retaining approximately 80% of the initial efficiency after 200 h of continuous illumination.

The charge-transport characteristics were investigated using the space-charge-limited current (SCLC) method in single-carrier devices.⁷⁴ We observed that the introduction of [70]PCBM led to an increase in both hole and electron mobilities, implying the formation of a more favorable active layer morphology (Figures S49 and S50). The exciton dissociation efficiency (η_{diss}) can be calculated by the equation $\eta_{diss} = J_{ph}/J_{sat}$, where J_{ph} denotes the photocurrent density, and J_{sat} represents the saturated photocurrent density.⁷⁵ The η_{diss} values of the binary and ternary OSCs were 97.0% and 98.3% (Figure S51), respectively, indicating that the ternary device has a higher exciton dissociation efficiency. The correlation between V_{oc} and P_{light} was exploited to examine trap-assisted recombination in OSCs (Figure S52).⁷⁶ The parameter n was estimated to be 1.29 and 1.27 for the binary and ternary devices, indicating that the trap-assisted recombination was further mitigated in the ternary OSCs. Overall, the well-balanced charge transport, efficient exciton dissociation, and reduced recombination jointly contributed to the high performance of the ternary OSCs based on PPT-3.

To clarify the underlying reasons for the improved photovoltaic performance of ternary devices, we conducted a more in-depth investigation into the film microstructures. Atomic force microscopy (AFM) images reveal that the PPT-3 neat film exhibits a well-defined fibrillar network with an average fibril width of 19.61 nm (Figures 2D and S53). These nanoscale fibrillar structures underscore the strong crystallinity of PPT-3, consistent with the results of DSC and GIWAXS. Notably, the fibrillary morphology is preserved in both the PPT-3:L8-BO binary blend

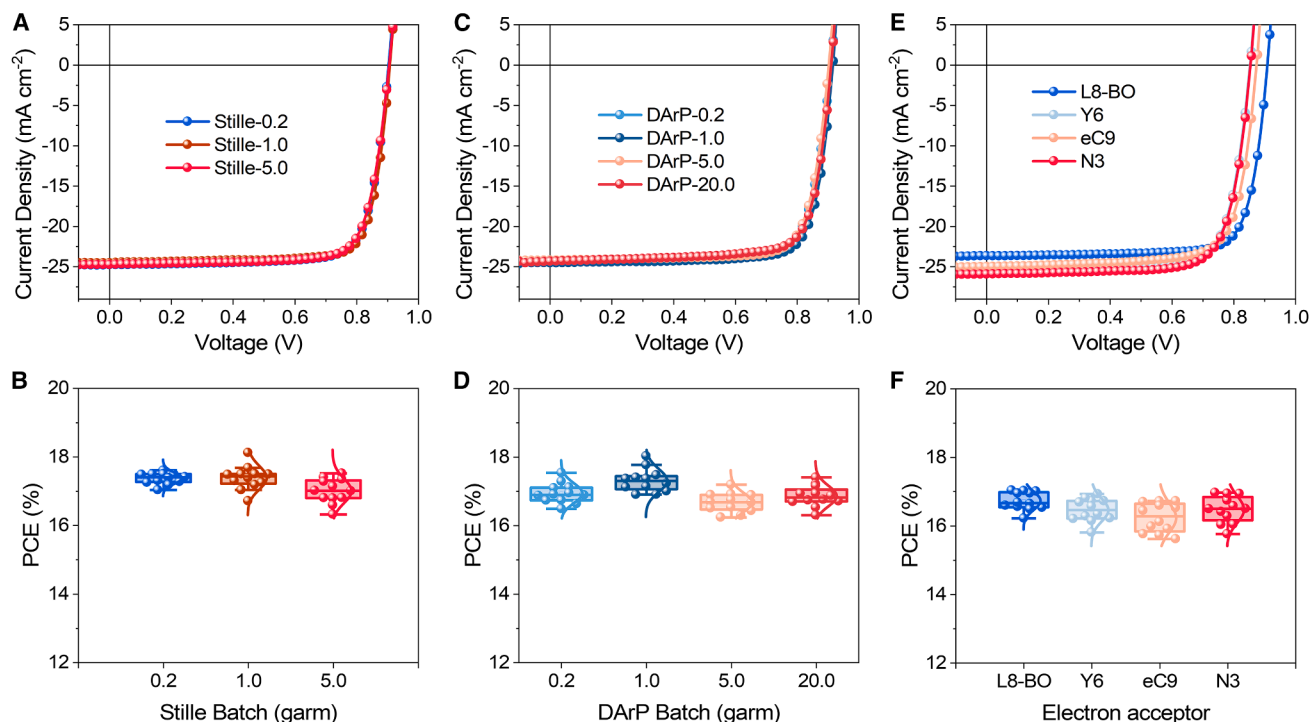


Figure 3. Photovoltaic properties of the OSCs

(A and B) (A) The J - V curves and (B) statistical PCE distribution of ternary OSCs based on Stille batches of PPT-3.

(C and D) (C) The J - V curves and (D) statistical PCE distribution of ternary OSCs based on DArP batches of PPT-3.

(E and F) (E) The J - V curves and (F) statistical PCE distribution of binary OSCs based on PPT-3 and different electron acceptors (L8-BO, Y6, eC9, and N3).

and the PPT-3:L8-BO:[70]PCBM ternary blend, providing efficient pathways for charge carrier transport.⁷⁷ The fibril texture in the PPT-3:L8-BO binary blend is roughly defined with a larger fibril diameter and phase distribution. With the introduction of [70]PCBM, the ternary blend exhibited a slightly smaller fibril diameter and reduced phase separation. The average fibril width of binary and ternary blends is 22.67 and 18.23 nm, respectively, which is a suitable length scale of phase separation for efficient exciton diffusion and balanced charge carrier transport.⁷⁷ Further morphological characterization via AFM-infrared spectroscopy (AFM-IR) and transmission electron microscopy (TEM) confirms continuous fibril networks in both blends (Figures 2E, S54, and S55). The reduced phase separation in the ternary blend increases the donor/acceptor interfacial area, rationalizing the improved EQE response in the 600–900 nm range.

GIWAXS measurements were performed to further investigate the crystallization behavior, intermolecular packing, and molecular orientation in blend films.⁷⁸ The two-dimensional GIWAXS patterns and one-dimensional GIWAXS line-cut profiles are presented in Figures 2F, 2G, and S56. The results reveal that both binary and ternary blends maintain a preferential face-on orientation, which is advantageous for efficient charge transport. Detailed numerical parameters are summarized in Table S8. For the PPT-3:L8-BO binary blend, the (010) diffraction peak in the OOP direction appears at $1.76 \text{ \AA}^{-1}$, corresponding to a π - π stacking distance of $3.57 \text{ \AA}$. The IP (100) peak is observed at $0.29 \text{ \AA}^{-1}$,

indicating a lamellar stacking distance of $21.96 \text{ \AA}$ with a coherence length (L_c) of $122.87 \text{ \AA}$. In the PPT-3:L8-BO:[70]PCBM ternary blend, a slight reduction in the π - π stacking distance ($3.56 \text{ \AA}$) and lamellar stacking distance ($21.81 \text{ \AA}$) is observed. These findings suggest that the incorporation of [70]PCBM refines the crystalline structure of the active layer, consistent with the enhanced charge carrier mobility and improved device performance.

Following the systematic investigation of the photovoltaic properties and morphological characteristics, we further evaluated the batch-to-batch reproducibility of PPT-3. The J - V characteristics and PCEs for devices fabricated from multiple synthesis batches are summarized in Figures 3A–3D (Table S9). Notably, three Stille batches (0.2–5.0 g) and four DArP batches (0.2–20.0 g) consistently achieved high PCEs in the range of 17.20% to 18.13%. There were only minimal variations in the key parameters: the V_{oc} was in the range of 0.91–0.92 V, the J_{sc} was between 24.24 and 24.82 mA cm^{-2} , and the FF ranged from 77.36% to 79.71%. These results demonstrate the exceptional reproducibility of PPT-3 throughout the scale-up processes. Remarkably, devices utilizing DArP-synthesized PPT-3 achieved a maximum PCE of 18.03%, which is the highest value for DArP-derived polymers to date (Figure S57; Table S10). Photostability tests confirmed that devices from both Stille and DArP batches devices showed comparable degradation trends, retaining $\sim 80\%$ of their initial PCE after 200 h continuous illumination, demonstrating excellent reproducibility in device performance (Figure S58). To further elucidate the origin of

device performance retention, TEM and AFM analyses were conducted on ternary blends based on different batches of PPT-3 (Figures S59 and S60). All blends exhibited a well-defined fibrillar interpenetrating network morphology with consistent phase separation scales and nanofibril structures. These findings corroborate that the resemblance in microstructure and morphology stands as the primary determinant for the exceptional reproducibility of PPT-3-based OSCs. This reproducibility can be further ascribed to the rigorous quality control of materials and the consistent pre-aggregation behavior across different batches. To evaluate the universality of PPT-3, we fabricated binary OSCs using several state-of-the-art electron acceptors, including Y6,¹³ eC9,⁷⁹ and N3.⁸⁰ The *J*-*V* characteristics and PCEs of these devices are presented in Figures 3E and 3F and Table S11. Notably, binary OSCs based on PPT-3 achieved PCEs nearing 17% when combined with diverse electron acceptors. This result demonstrates the outstanding compatibility of PPT-3 with various acceptors, a feature that allows for the selection of cost-effective acceptors in the large-scale production of OSCs.

ST-OSCs application

Having successfully addressed the challenges associated with large-scale synthesis and established a fundamental understanding of the photovoltaic properties of PPT-3, we further investigated its application in ST-OSCs (Figure S61). The ST-OSCs were fabricated with a device architecture of ITO/PEDOT:PSS/PPT-3:L8-BO:[70]PCBM (0.6:1.2:0.1, 80 nm)/PFN-Br/Ag (12 nm), which was defined as the control device for subsequent comparative analyses (Figure 4A). The control device achieved a PCE of 12.38%, with an AVT of 27.66% and an LUE of 3.42% (Tables 3, S12, and S13). The corresponding *J*-*V* curves, EQE, and transmittance spectra are presented in Figures 4F and 4G. To further boost the performance of ST-OSCs, a bilayer optical coupling layer (OCL) was applied atop the semitransparent electrode by using a high-throughput optical screening method,^{36,81} and the detailed simulation methods are described in supplemental methods. An optimal OCL structure was identified as ($n_1 = 2.4$, $d_1 = 10$ nm, ZnS)/($n_2 = 1.4$, $d_2 = 50$ nm, LiF), which can deliver the highest LUE value, close to the theoretical limit (Figures 4A, 4B, and S62). As a result, ST-OSCs containing an OCL obtained a PCE of 11.46%, an AVT of 41.04%, and a LUE of 4.72% (Tables S14 and S15). The bilayer OCL effectively reduces the reflection from the metal electrode by improving impedance matching, thereby redirecting more photons toward transmission. As evidenced by the optical field distribution (Figures 4C and 4D), the transmitted region (wavelength: 400–700 nm, position >327 nm) demonstrated higher field intensity, directly correlating with enhanced light transmission. Conversely, the reflected region (wavelength: 400–700 nm, position <150 nm) exhibited diminished field intensity, corresponding to suppressed reflection losses across the device stack. Such optimized light management resulted in a significantly enhanced AVT (from 27.66% to 41.04%). Critically, the exciton generation rate in the active layer remained largely unaffected in the visible spectrum, ensuring minimal compromise to device efficiency (Figure 4E). Furthermore, 100 nm MgF₂ was selected as an antireflective coating (ARC) on the bot-

tom glass surface to reduce the reflection (*R*) (Figure S63). This design leverages intermediate refractive index ($n \approx 1.38$) of MgF₂, which minimizes the optical mismatch between glass ($n \approx 1.5$) and air ($n \approx 1.0$), thereby suppressing Fresnel reflection through destructive interference.⁸² The optimal ST-OSCs integrating the OCL and ARC achieved a PCE of 12.07%, an AVT of 41.76%, and a LUE of 5.04%, representing one of the best-performing ST-OSCs with AVT over 40% (Table 3; Figures 4F and 4G). The optical analysis of corresponding ST-OSCs was also performed to validate the accuracy and validity of *J*-*V* measurements, and all the EQE + AVT + *R* values were below 100%, confirming the reliability of the results (Figure S64). We further investigated the photostability of control and optimal ST-OSCs. Compared with the control devices, the optimal ST-OSCs with an OCL demonstrated superior photostability, indicating that the implementation of an OCL can effectively mitigate device degradation (Figure S65). To conclude this section, the overall performance of the ST-OSCs based on PPT-3 can be optimized through an OCL and ARC to achieve a delicate balance between AVT and PCE, thereby leading to improved LUE. These results underscore the exceptional benefits of PPT-3 a key material, in the development of ST-OSCs, and it is expected to achieve a further breakthrough in the LUE value when integrated with state-of-the-art ST-OSCs manufacturing processes.^{34–36}

Inspired by the outstanding performance of PPT-3 in small-area ST-OSCs, we further fabricated ST-OSC modules to identify their viability as power-generating windows. Small-area devices fabricated under ambient conditions exhibited over 95% performance retention compared with those prepared in a nitrogen-filled glovebox (Table S16). Then, the large-area semitransparent module was fabricated via a blade-coating process under ambient conditions, with a device structure of ITO/PEDOT:PSS/PPT-3:Y6/ZnO/AgNWs (Tables S17–S19). The module area and active layer area of the large-area semitransparent module is 132 and 120 cm², respectively, which consists of 22 sub-cells in series, and each sub-cell is 6 cm² (12 cm × 0.5 cm) (Figure S66). The *J*-*V* curves and transmittance spectra are provided in Figures 4H and 4I, respectively. The large-area semitransparent module exhibits a PCE of 6.69%, an AVT of 40.30%, and an LUE value of 2.70%, which represents, to the best of our knowledge, the best-performing ST-OSC module achieved thus far (Table 3; Figures 4J and S67; Table S20). Figure 4K presents an image of a large-area semitransparent module based on PPT-3, which exhibits high visible-light transparency, enabling clear visibility of background elements, such as the sky, white clouds, and maple leaves, achieving a visually stunning aesthetic appearance. In addition, we have integrated the large-area semitransparent module with a green light-emitting diode (LED) lamp. This integration showcases that the ST-OSCs can effectively power the LED, as shown in Video S2 (Figure S68). Collectively, these results highlight the successful integration of large-scale material synthesis and modular device fabrication, which lays a solid foundation for the practical implementation of organic photovoltaics.

Economic scalability analysis

Following the success of scale-up synthesis and fabrication of large-area semitransparent modules, we assessed the

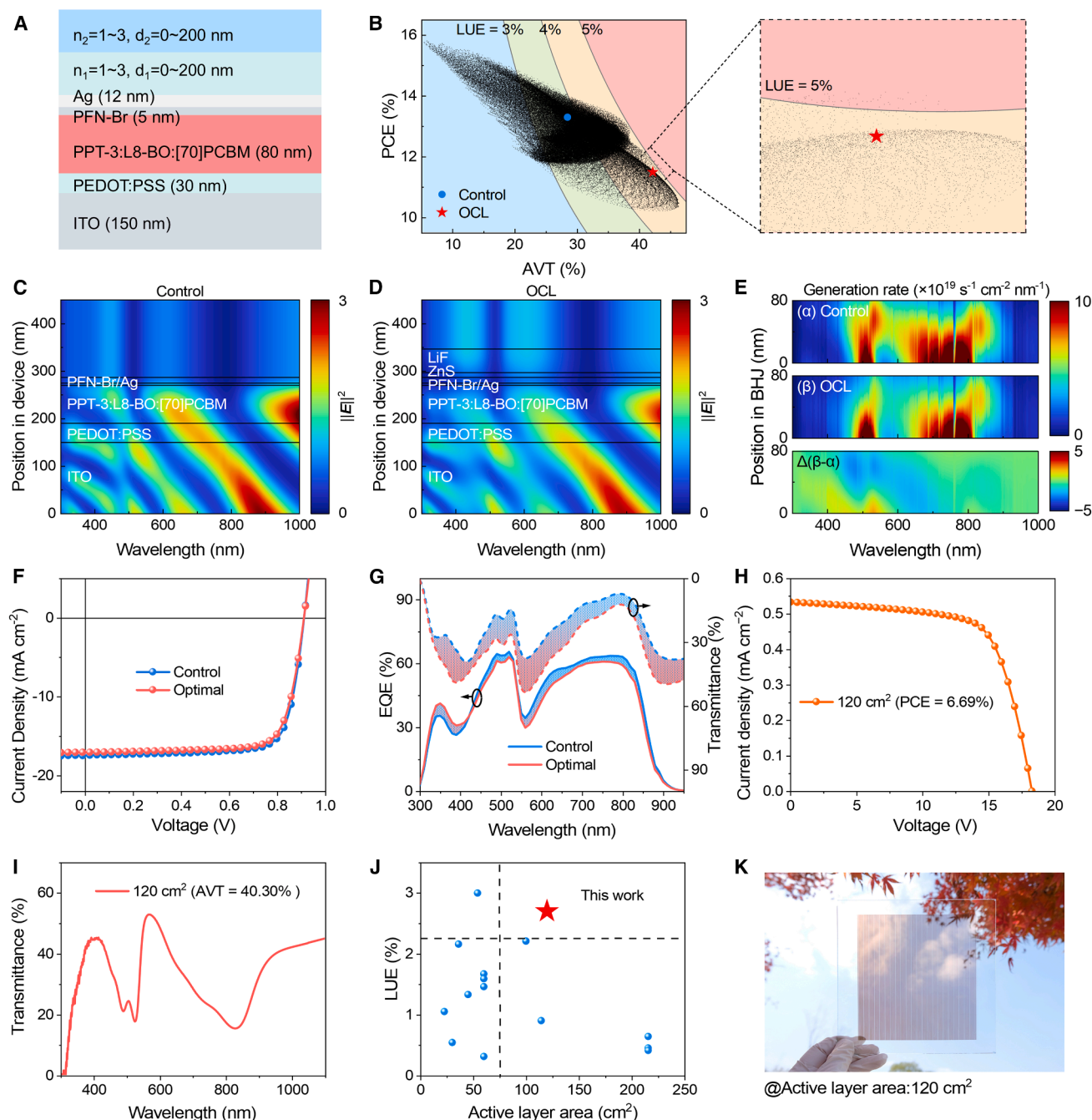


Figure 4. ST-OSCs application

(A) Device structure of the ST-OSCs with OCLs.

(B) Contour map of simulated PCE-AVT as a function of the refractive index and layer thickness of the OCL. The black dots, the blue circle, and the red star denote the high-throughput screened results, the simulated result of the control device without an OCL, and the simulated result of the device with an OCL of ZnS (10 nm)/LiF (50 nm), respectively. The blue curves represent contour lines for the LUE values. Right: magnification of the part that gives the maximal LUE values.

(C and D) Profiles of the electrical field distribution were calculated for the control device without an OCL (C) and the ST-OSC with an OCL (D).

(E) Exciton generation rate in the BHJ active layer for the control device (labeled α), the ST-OSC with an OCL (labeled β), and the difference in exciton generation rate in the BHJ active layer between the control device and the ST-OPV with an OCL (labeled $\beta-\alpha$).

(F and G) (F) The J-V curves and (G) EQE and transmittance spectrum of the ST-OSCs based on PPT-3:L8-BO:[70]PCBM.

(H and I) (H) The J-V curves and (I) transmittance spectrum of the large-area semitransparent module based on PPT-3:Y6.

(J) The summary of recently reported large-area ST-OSCs with active areas of over 20 cm².

(K) The figure of the large-area semitransparent module based on PPT-3:Y6.

Table 3. Device parameters of the ST-OSCs based on PPT-3 under AM 1.5G irradiation (100 mW cm^{-2})

Device ^a	V_{oc} (V)	J_{sc} (mA cm^{-2})	J_{sc} (mA cm^{-2}) ^b	FF (%)	PCE (%)	AVT (%)	LUE (%)
Control	0.91	17.49	17.13	77.24	12.38	27.66	3.42
Optimal	0.91	17.07	16.43	77.03	12.07	41.76	5.04
120 cm^2	18.30	0.54	–	68.00	6.69	40.30	2.70

^aThe structure of control device: ITO/PEDOT:PSS/active layer/PFN-Br/Ag (12 nm); the structure of optimal device: ARC/ITO/PEDOT:PSS/active layer/PFN-Br/Ag (12 nm)/OCL. The device structure of large-area modules: ITO/PEDOT:PSS/active layer/ZnO/AgNWs.

^bThe current densities were integrated from the EQE curves of the solar cells.

economic scalability of PPT-3 using multiple metrics, including synthesis complexity (SC),⁸³ figure of merit (FOM, $\text{FOM} = \text{PCE}/\text{SC}$),⁸⁴ scalability factor (SF),^{85,86} and material-only costs (MOCs).⁸⁷ The following analysis is based on laboratory-scale procedures, which may differ from optimized industrial processes, and the results are intended as preliminary benchmarks rather than absolute metrics.

Previous economic analyses have demonstrated that photoactive materials constitute a significant portion of the total material costs in OSCs, with polymer donors alone accounting for over 50%.^{47–51} Given the strong correlation between the cost of organic photovoltaic materials and their SC, optimizing SC while maintaining high PCE is essential for enhancing the commercial viability of OSCs. The SC of widely used polymer donors, such as PM6 and D18, exceeds 50% due to their multistep synthesis procedures (Figure 5A; Table S21). In contrast, polythiophenes and their derivatives exhibit lower SC values, typically ranging from 20% to 50%. Notably, the SC of PPT-3 has been reduced to 10.67%, comparable to that of P3HT, a benchmark low-cost polymer. Figure 5B summarizes the SC versus PCE for OSCs based on representative polymer donors. Most state-of-the-art polymer donors, including D18 and PM6, exhibit FOM values far below 1 due to their extremely high SC. Significantly, PPT-3 achieves an optimal FOM of 1.69, making it one of only two materials (alongside P3HT) with an FOM greater than 1. This exceptional balance between cost-effectiveness and performance underscores the potential of PPT-3 for practical applications. The SF, a metric based solely on synthetic step count and reaction yield, was employed to evaluate scale-up potential (Table S22). The DArP-synthesized PPT-3 exhibited an SF of 4.1, which is significantly lower than that based on the Stille method ($\text{SF} = 5.2$). These results demonstrate the superior scalability of the DArP route for industrial production. To further quantify the economic advantages of PPT-3, we calculated its MOC. The MOC of PPT-3 (DArP) amounts to 6.1 USD g^{-1} , representing one-tenth that of D18 (67.3 USD g^{-1}) and even slightly lower than that of P3HT (6.9 USD g^{-1}). By selecting alternative raw chemical suppliers, the MOC of PPT-3 (DArP) can be further reduced to 3.3 USD g^{-1} (Figure 5C; Tables S23–S35). We further estimated the production cost for 100 g of PPT-3 synthesized via direct-arylation polymerization (DArP), accounting for both synthesis and purification reagents. The substantial increase in solvent expenses elevated the total production cost to 860 USD per 100 g, suggesting that solvent recycling must be further optimized in scaled-up manufacturing to mitigate costs.

To assess the economic impact of low-cost polymer donors on the future OSCs industry, we conducted a cost-effectiveness

analysis comparing PPT-3 with two widely used polymer donors, D18 and PM6, using an established economic model from the literature.^{47–51} This analysis is based on a bottom-up manufacturing cost model, with detailed assessment methods and assumptions provided in Note S3. The cost structure diagrams in an industrial scenario reveal that the total cost of active layer materials (donor and acceptor) in the PPT-3 system constitutes only 36%, with the donor contributing 9% (Figure 5D). In contrast, the active layer materials account for 65% and 57% of the total cost in the D18 and PM6 systems, respectively, with donor materials alone representing 50% and 39% (Figures S69–S71). These results underscore the dominant role of photoactive material costs in the overall expense of OSCs production and highlight the potential of PPT-3 to significantly reduce manufacturing costs. Further financial parameters and assumptions used to calculate the minimum sustainable price (MSP) in USD W^{-1} for the industrial scenario are detailed in Tables S36–S45. For a target MSP of 0.25 USD W^{-1} , the module efficiencies required for PPT-3-, D18-, and PM6-based devices are 15.4%, 19.0%, and 20.8%, respectively (Figure 5E). These results even surpass those of some other photovoltaic technologies (Figure S72).⁵¹ Owing to its low-cost advantage, PPT-3 demonstrates the highest feasibility for achieving the target MSP, indicating that the development of cost-effective polymer donors can alleviate the financial barriers impeding OSC industrialization. A comprehensive comparison of PPT-3, P3HT, PM6, and D18 is presented in Figure 5F, encompassing synthesis steps, MOC, SC, PCE, FOM, and AVT. PPT-3 exhibits exceptional performance across all metrics, achieving a PCE comparable to PM6 and D18 while maintaining an MOC on par with P3HT. Given its scalable synthesis, unique suitability for semitransparent applications, and significant economic advantages, PPT-3 represents a compelling candidate for accelerating the transition of OSCs from laboratory research to industrial deployment.

DISCUSSION

In summary, this study aims to overcome the vital challenges in the scalable synthesis of polymer donors and the construction of large-area semitransparent devices, thereby facilitating the practical application of organic photovoltaics. We successfully achieved a 100-fold scale-up synthesis of polymer donor PPT-3 via the DArP method, increasing batch sizes from mg to 20 g. The optimized opaque OSCs based on PPT-3 exhibited exceptional photovoltaic performance, achieving a PCE exceeding 18% and demonstrating outstanding batch-to-batch reproducibility across multiple synthesis scales. Notably, large-area

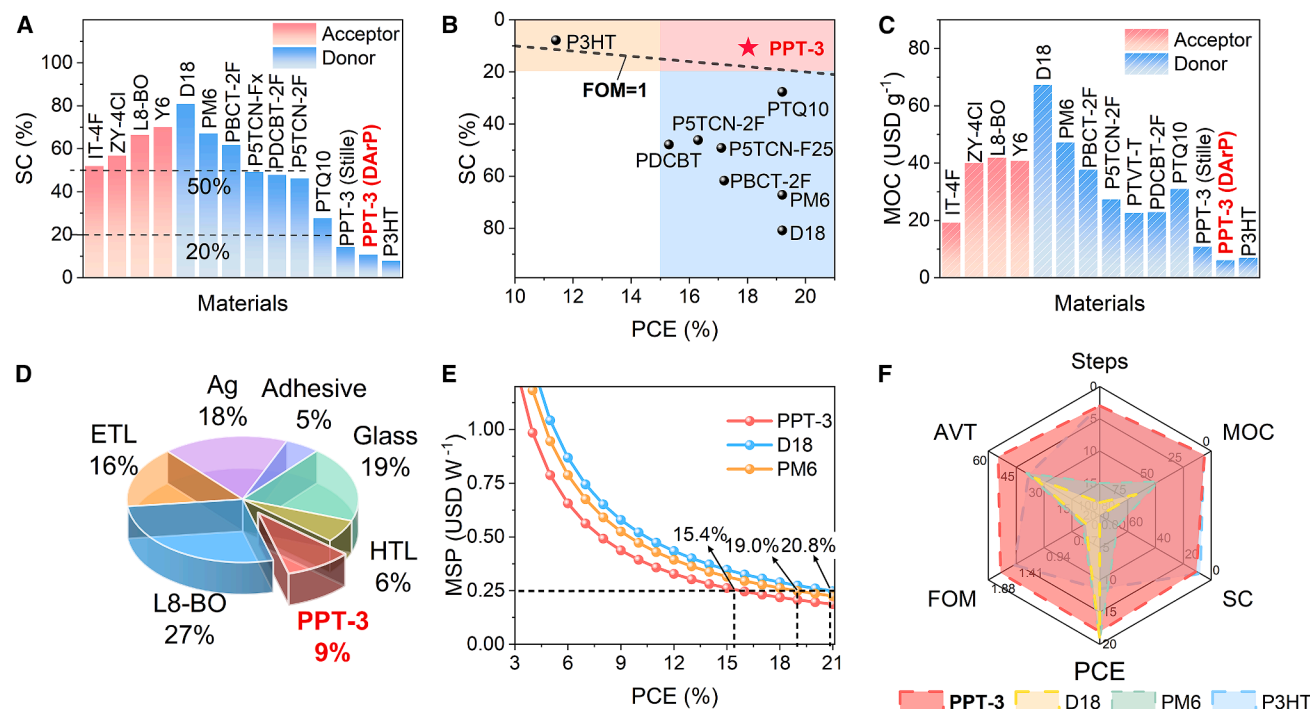


Figure 5. Economic scalability analysis

(A) The SC value of representative organic photovoltaic materials.
 (B) The SC versus PCE for OSCs based on representative polymer donors. The dotted line represents FOM = 1 (FOM = PCE/SC).
 (C) The MOC of representative organic photovoltaic materials.
 (D) The cost structure of the industrial model of solar modules based on PPT-3.
 (E) MSP-PCE function of the OSCs based on D18, PM6, and PPT-3.
 (F) A multifactorial evaluation of PPT-3, P3HT, PM6, and D18.

semitransparent modules with an active area of 120 cm², fully printed under open-air conditions, achieved a record LUE of 2.70% alongside a PCE of 6.69% and an AVT of over 40%. A comprehensive cost-benefit analysis demonstrates that the development of low-cost polymer donors effectively alleviates the economic barriers hindering the industrialization of OSCs. The remarkable cost-performance balance positions PPT-3 as a highly promising candidate for practical applications. This study not only provides critical insights into the design and scalable synthesis of economically viable polymers but also establishes a transformative pathway for transitioning OSCs from laboratory-scale research to commercial viability.

METHODS

Further details regarding the methods can be found in the [supplemental methods](#).

RESOURCE AVAILABILITY

Lead contact

Further information and requests for resources should be directed to, and will be fulfilled by, the lead contact, Chunhui Duan (duanchunhui@scut.edu.cn).

Materials availability

All unique reagents generated in this study are available from the [lead contact](#) without restriction.

Data and code availability

The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

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AUTHOR CONTRIBUTIONS

C.D. conceived the idea and supervised the project. D.X. carried out the material synthesis, device fabrication, and characterization. B.W., Z.C., L.P., and K.L. contributed to the monomer synthesis. Q.W., Q.J., and D.L. carried

out the large-area semitransparent device fabrication. B.D. and T.X. performed the optical simulation. J.L., Q.J., and H.F. contributed to the AFM, AFM-IR, and TEM characterization. J.Z. and H.L. performed the GIWAXS measurements. R. X., X.Y., and Y.Z. participated in data analysis. F.H. and Y.C. participated in project administration. D.X. and C.D. prepared the manuscript. All authors discussed the results and commented on the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

SUPPLEMENTAL INFORMATION

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