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Recent Advances in PSMA-Type Polymer Acceptors for All-Polymer Solar Cells

Haoran Cai*

College of Polymer Science and Engineering, Sichuan University, Chengdu, Sichuan, China

**Corresponding author: Haoran Cai.*

Abstract

All-polymer solar cells (all-PSCs) offer intrinsic advantages in mechanical flexibility, film toughness, and morphological stability, making them promising candidates for flexible and scalable organic photovoltaics. Polymerized small-molecule acceptors (PSMAs) inherit the broad spectral absorption and tunable electronic structures of high-performance small-molecule acceptors, effectively overcoming the insufficient near-infrared absorption of conventional polymer acceptors and driving the power conversion efficiencies of all-PSCs into the 17–19% range. However, once high-performance PSMAs provide suitable absorption and energy levels, further improvements can no longer be achieved by independently optimizing a single intrinsic parameter, such as optical absorption, energy-level alignment, or charge-carrier mobility. Instead, polymer-chain entanglement, molecular and energetic disorder, suboptimal donor–acceptor matching, and the difficulty in simultaneously controlling crystallization, phase separation, and vertical composition distribution have become the major barriers to further efficiency gains. Recent efficiencies exceeding 20% demonstrate that high-performance all-PSCs rely on the concerted optimization of PSMA molecular structures, donor–acceptor interactions, active-layer morphology, and processing kinetics. This review, therefore, focuses on comparing the strategies that have enabled breakthroughs beyond 20% efficiency. On this basis, the remaining challenges associated with long-term stability, green-solvent processing, batch-to-batch consistency, and scalable manufacturing are further discussed, and the common mechanisms underlying the crossing of the 20% efficiency threshold, together with future development directions, are summarized.

Keywords

all-polymer solar cells, polymerized small-molecule acceptors, donor–acceptor matching, morphology control

1. Introduction

Compared with conventional crystalline silicon solar cells, organic solar cells (OSCs) offer several advantages, including low weight, mechanical flexibility, solution processability, and compatibility with large-area coating and printing. These characteristics make OSCs particularly attractive for applications such as wearable electronics and building-integrated photovoltaics [1]. Among the various OSC configurations, all-polymer solar cells (all-PSCs) employ an active layer composed entirely of a polymer donor and an acceptor polymer. Entanglement and kinetic constraints among long polymer chains can enhance film cohesion while suppressing long-range diffusion of components and domain coarsening, giving all-PSCs potential advantages in mechanical flexibility, crack resistance, and long-term morphological stability [2, 3].

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The performance of polymer acceptors mainly limited the early development of all-PSCs. Conventional acceptors, such as N2200, exhibit relatively high electron mobility and good ambient stability, but their weak near-infrared absorption limits photon harvesting further improvement in short-circuit current density [4]. By contrast, high-performance small-molecule acceptors such as Y6 feature strong absorption, low band gaps, tunable energy levels, and relatively low energy losses [3]. However, their molecular diffusion and crystallization can compromise the mechanical and morphological stability of the active layer. Polymerized small-molecule acceptors (PSMAs), which incorporate high-performance small-molecule acceptor units into conjugated polymer backbones, combine the favorable optoelectronic properties of small molecules with the film-forming ability, mechanical flexibility, and morphological stability of polymers, and have therefore become a key acceptor platform for all-PSCs [3, 5].

The rapid development of Y-series PSMAs has substantially improved light harvesting and charge generation, pushing all-PSC efficiencies into the 17–19% range [5, 6]. progress, however, remains constrained by intrinsic features of all-polymer blends. Entanglement between donor and acceptor chains limits segmental diffusion and rearrangement, while competing crystallization further complicates control of morphology. Variations in molecular conformation and packing can increase energetic disorder, promote nonradiative recombination, and reduce charge-transport efficiency [4]. Excessive donor–acceptor miscibility may produce mixed phases with insufficient phase purity, whereas insufficient miscibility can lead to overly coarse phase separation [5]. Unfavorable vertical composition distributions and imbalanced electron–hole transport can also hinder charge extraction and reduce the fill factor [7]. Consequently, further breakthroughs can no longer be achieved simply by enhancing PSMA absorption or tuning energy levels.

Recent efficiencies exceeding the 20% mark signal a shift from optimizing individual material parameters to multilevel cooperative regulation. These advances arise from the combined effects of donor–acceptor co-design, control of acceptor conformation and molecular packing, multicomponent morphology engineering, selective post-treatment, and regulation of film-formation kinetics and vertical composition distribution [1, 7-9].

Accordingly, this review focuses on the breakthrough beyond 20% efficiency and examines the synergy among PSMA molecular design, donor–acceptor matching, active-layer morphology, and device processing. Different routes toward high efficiency are compared, followed by a discussion of the remaining challenges in long-term stability, green-solvent processing, batch-to-batch reproducibility, and large-area manufacturing.

2. Theoretical Background

2.1 Photophysical Processes and Photovoltaic Parameters

All-polymer solar cells (all-PSCs) are excitonic photovoltaic devices. Because organic semiconductors have relatively low dielectric constants, photon absorption in the active layer initially generates bound excitons in either the polymer donor or acceptor rather than free charge carriers. These excitons subsequently diffuse to the donor–acceptor interface, where charge transfer occurs under the influence of interfacial energy-level offsets and local electrostatic interactions, leading to the formation of charge-transfer (CT) states. Further dissociation of the CT states generates free electrons and holes. Electrons are transported through the polymer acceptor phase, whereas holes move through the polymer donor phase and are finally collected by the corresponding electrodes. Efficient photovoltaic conversion, therefore, requires both sufficient donor–acceptor interfacial area for exciton dissociation and relatively pure, continuous transport pathways for efficient carrier extraction [5].

The power conversion efficiency (PCE) is expressed as:

$$\mathrm{PCE} = \frac{V_{\mathrm{OC}} J_{\mathrm{SC}} \mathrm{FF}}{P_{\mathrm{in}}}$$

where the short-circuit current density J_{sc} is mainly governed by light absorption, exciton diffusion and dissociation, and charge collection. The open-circuit voltage V_{oc} depends on donor–acceptor energy-level alignment, CT-state energy, and radiative and nonradiative energy losses. The fill factor (FF) reflects carrier mobility, electron–hole transport balance, trap states, recombination, and active-layer morphology. In PSMA-

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based all-PSCs, chain entanglement, energetic disorder, and unfavorable vertical composition distributions can simultaneously limit several photovoltaic parameters.

2.2 Structure–Interaction–Morphology–Performance Relationship

Polymerized small-molecule acceptors (PSMAs) are conjugated polymer acceptors formed by covalently linking high-performance small-molecule acceptor building blocks through π-bridge units. Their repeating units are typically derived from A–D–A or A–DA′D–A structures, while side chains, polymerization sites, molecular weight, and dispersity further regulate solubility, backbone rigidity, intermolecular packing, and film formation. These structural parameters determine not only the intrinsic absorption and frontier molecular orbital levels of PSMAs, but also chain conformation and the degree of conformational and energetic disorder [10]. In particular, modification of the central A′ unit, π-bridge, or side-chain topology can simultaneously influence electron affinity, backbone planarity, aggregation strength, and compatibility with a specific polymer donor [3, 5].

The performance of PSMA-based all-PSCs can therefore be understood through a structure–interaction–morphology–performance relationship. The PSMA molecular structure first governs donor-acceptor interactions and solution-state pre-aggregation, which subsequently affect crystallization, phase separation, fibrillar network formation, and vertical composition distribution during film formation. Donor–acceptor miscibility must remain within an appropriate range: excessive miscibility produces mixed phases with low phase purity, whereas insufficient miscibility can lead to coarse phase separation and hinder exciton diffusion to the interface [5]. Likewise, continuous bicontinuous fibrillar networks provide efficient pathways for electron and hole transport, while unfavorable vertical distributions impede carrier extraction. Energetic disorder arising from local variations in conformation and packing broadens the density of states, increases trapping and nonradiative recombination, and reduces carrier mobility [4]. Consequently, active-layer morphology governs charge generation, transport, and recombination, and is ultimately reflected in J_{sc}, V_{oc}, FF, and operational stability. This framework highlights that PSMA optimization must integrate molecular design, donor matching, and processing control rather than maximize any single material parameter in isolation.

3. Literature Review

3.1 Molecular Design toward High-Performance PSMAs

With the introduction of Y-series building blocks, PSMA design shifted from extending absorption to controlling aggregation and solid-state organization. Du et al. compared two A–DA′D–A-type PSMAs, PS-Se and PN-Se, and found that changing the central A′ unit affected not only absorption and energy levels, but also solution-state pre-aggregation and blend morphology. Compared with PS-Se, the PBDB-T:PN-Se precursor solution formed more uniform nanoscale aggregates and developed into a bicontinuous interpenetrating network of about 10–20 nm in the film, enabling a PCE of 16.16% [3]. Thus, PSMA building blocks should be judged not only by their optical and electronic properties, but also by their effects on pre-aggregation and morphology.

PSMA design then progressed toward coordinated regulation of π-bridges and side chains. Sun et al. introduced a fluorinated thiophene π-bridge into PY-IT to improve backbone planarity and electron transport. Although PY-IT2F exhibited higher electron mobility, its excessive miscibility with PM6 led to severe recombination, thereby reducing device performance. After introducing branched side chains, PG-IT2F achieved a better balance between electron transport and donor–acceptor compatibility, allowing PM6:PG-IT2F devices to reach 17.24% [5]. This shows that improving one intrinsic property is insufficient; aggregation, miscibility, and charge transport must be considered together.

More recently, PSMA design has expanded from local functional-group modification to polymerization mode and overall molecular configuration. Yu et al. proposed a core-to-core polymerization strategy, distinct from conventional end-to-end linkage, to form PffBQx-T with a “double-decker” configuration featuring intramolecular packing. This architecture reduces conformational freedom, promotes terminal-group interactions, and enhances molecular packing and charge delocalization, leading to an 18.7% PCE in ternary devices [10]. Before the 20% regime, PSMA design had therefore evolved toward integrated control of

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backbone conformation, aggregation, packing, and three-dimensional configuration. At the same time further progress required better donor–acceptor matching, lower energy loss, and more precise control of morphology.

3.2 The Pre-20% Efficiency Regime: Emerging Limitations Beyond Molecular Improvement

As PSMA properties improved, the main limitation shifted from insufficient acceptor performance to the challenge of converting these properties into low-loss devices. Zhang et al. designed PQB terpolymer donors and paired them with PY-IT. PQB-2 showed a stronger face-on orientation and lower energetic disorder than PBDB-TF and PBQx-TF. In the PQB-2:PY-IT blend, hole energetic disorder was further reduced, while the DTQx unit provided an efficient hole-transfer pathway, lowering nonradiative energy loss and increasing V_{oc}. The device reached 18.1% PCE [4], indicating that PSMA performance strongly depends on the energetic landscape and interfacial charge-transfer processes with the paired donor.

Morphology optimization also moved beyond simple nanoscale phase separation toward multiscale regulation. Zeng et al. combined a 1,4-diiodobenzene solid additive with thermal annealing and solvent-vapor annealing in PM6:PY-IT. By controlling additive removal and polymer crystallization, they formed a highly crystalline bicontinuous nanofibrillar network together with a microscale surface structure. The nanoscale network promoted exciton dissociation and carrier transport, while the microscale topography enhanced light trapping. The device reached 19.06% PCE, with a certified efficiency of 18.59% [6]. Thus, at the 18–19% level, the focus shifted from finding a "better PSMA" to enabling an existing high-performance PSMA to operate optimally within a specific donor and processing environment.

3.3 Integrated Strategies for Crossing the 20% Efficiency Threshold

Recent studies have pushed PSMA-based all-PSCs beyond 20% efficiency. These advances show that the performance ceiling can be raised through acceptor structure, molecular environment, and film-formation pathway. Rather than pointing to one universal strategy, different routes can succeed when molecular organization, interfacial structure, and charge transport are mutually compatible.

3.3.1 Intrinsic Structural and Ordering Control of PSMAs

One direct route is to reduce structural disorder within the PSMA itself. Sequence-regulated copolymer acceptors show that the arrangement of building blocks along the backbone strongly affects chain conformation and intermolecular packing. In RC10, the regular arrangement of benzothiadiazole and benzoquinoxaline units produced a more defined backbone configuration and more favorable aggregation than in the random copolymer UC10. When paired with PM6, RC10 achieved a PCE of 20.03% [8]. This extends PSMA design from selecting building blocks to controlling their sequence along the backbone. Even at similar compositions, improved sequence regularity can reduce local structural fluctuations and promote more favorable solid-state organization.

Side-chain and terminal-group engineering provide another route to regulate ordering. PQx-FT and PQxO-IT exhibit distinct aggregation strengths and characteristics due to their distinct substitution patterns. PQxO-IT exhibits stronger intermolecular interactions and tighter π–π stacking, whereas PQx-FT shows a different aggregation tendency. Combining them allows these tendencies to complement each other and avoids both insufficient and excessive aggregation. The PM6:PQx-FT:PQxO-IT device reached 20.29% PCE, with a certified efficiency of 20.03% [8]. The goal is therefore not simply to maximize molecular order, but to achieve a controllable degree of order that supports efficient transport and favorable phase organization.

3.3.2 Donor–Acceptor Matching and Multicomponent Regulation

A second route reshapes the host PSMA environment through donor design or multicomponent regulation. Qiu et al. incorporated benzo[1,2-d:4,5-d′]bisthiazole units into a PM6-type donor backbone to form PBZ terpolymers. Optimized PBZ-10 exhibited greater compatibility with PY-IT and a favorable fibrillar morphology, yielding a binary PCE of about 19.1%. Adding PBQx-TF as a second polymer donor further improved absorption, charge transport, and active-layer organization, increasing the PCE to 20.04% [9]. Because PY-IT itself was unchanged, this result shows that the performance limit of a PSMA cannot be defined independently of its donor environment.

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A useful boundary case introduces the small-molecule acceptor AQI4 into a polymer-donor/PSMA host system. The device reached 20.1% PCE and 19.5% certified efficiency, together with finer mixed regions and a fibrillar network [11]. Because AQI4 is not a polymer, this system should not be included in a strict all-polymer efficiency benchmark; it is retained only to illustrate how a third component can regulate interfacial organization.

A more representative fully polymeric example is steric locking. In PM6:PYF-T-o, the guest polymer acceptor PY-IDT mainly modifies the crystallization environment of the host acceptor rather than providing additional absorption. It suppresses excessive self-aggregation of PYF-T-o, lowers energetic disorder and nonradiative loss, and preserves favorable transport pathways. With o-xylene processing, the device reached 20.53% PCE and a certified efficiency of 19.79% [12]. Thus, ternary components can regulate host-PSMA organization rather than merely provide spectral complementarity.

3.3.3 Processing Kinetics and Vertical Morphology Control

A third route achieves efficiencies above 20% without major changes to the host donor and acceptor structures. Ma et al. developed wet-assisted annealing (WAA) for PM6:PY-DT by introducing assist solvents during thermal treatment. Compared with conventional thermal annealing, WAA temporarily increases the rearrangement ability of PY-DT and its lower-molecular-weight fractions, enhancing acceptor packing, optimizing phase separation, and producing a more favorable vertical composition distribution. The binary-device PCE increased from 19.02% to 20.04% [7]. The strategy also improved batch-to-batch reproducibility, suggesting that kinetic control can partially reduce performance variation across polymer batches.

Layer-by-layer (LBL) processing intervenes earlier in the formation of morphology. Sequential deposition partially decouples donor and acceptor aggregation and crystallization, allowing different additives to regulate each layer more independently. Subsequent controlled interdiffusion then creates the required donor–acceptor contacts and vertical concentration gradient. The optimized binary device achieved a PCE of 20.03%, with a certified efficiency of 19.60% [1]. Unlike post-treatment, LBL changes the morphology-evolution pathway before the final structure becomes kinetically fixed.

Overall, recent breakthroughs have not established a single dominant design rule. Instead, the remaining performance losses increasingly arise from coupling among molecular structure, the blend environment, and morphology formation. Further progress will likely depend on controlling interactions across these levels rather than optimizing any one of them in isolation.

4. Discussion

4.1 Common Principles behind the Breakthrough beyond 20% Efficiency

The recent achievement of PCEs exceeding 20% in PSMA-based all-polymer solar cells indicates that this field has entered a new stage of development beyond previous optimization strategies. At this efficiency level, continuously improving a single material parameter is unlikely to yield substantial sustained benefits. More importantly, once a specific loss pathway is suppressed, the dominant limitation within the device system may shift to other processes. For example, enhanced molecular ordering may initially improve charge transport; however, when transport is no longer the primary limitation, phase purity, interfacial charge generation, or vertical transport may become the new bottlenecks. Therefore, the dominant factors limiting device performance are not necessarily fixed. As one loss mechanism is reduced, other processes may become relatively more important, with the specific evolution depending on the material combination and processing history. From this perspective, further improvement beyond 20% efficiency should be regarded as a continuous rebalancing of multiple coupled loss pathways rather than the elimination of a single universal limitation.

This understanding also suggests that a “high-performance PSMA” should no longer be considered merely an intrinsic material label. Previous studies have demonstrated that the same polymer acceptor can exhibit significantly different device performances depending on the paired donor, third components, and processing conditions. Therefore, attention should be paid not only to the optical and electronic properties of the PSMA itself, but also to whether its molecular organization can respond controllably to the surrounding molecular environment and processing conditions. Based on these considerations, morphological tunability can be regarded as an important design criterion. An ideal PSMA should maintain sufficient structural order to support

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efficient charge transport while allowing its molecular packing and phase organization to be adjusted within a practically accessible processing window.

Given the dependence of PSMA system on their environment and the importance of morphological tunability, the evaluation of post-20% systems should extend beyond the highest reported efficiency. Achieving a record efficiency within a narrow processing window demonstrates a high performance potential, but does not necessarily indicate sufficient robustness of the material platform. For practical applications, a more rigorous criterion is whether the device can maintain near-optimal performance under reasonable variations in composition ratio, film thickness, and processing conditions.

4.2 Beyond Efficiency: Toward Practical PSMA-Based All-Polymer Photovoltaic Systems

After surpassing the 20% efficiency threshold, material reproducibility is one of the first challenges to address. The performance of PSMA-based blends can be sensitive to molecular weight, dispersity, sequence distribution, and low-molecular-weight fractions. Recent processing studies suggest that appropriate post-treatment strategies can partially mitigate the effects of polymer batch variations on device performance; However, this tolerance remains strongly dependent on the specific material system. Therefore, future PSMA development should consider not only intrinsic optoelectronic properties, but also tolerance toward molecular-weight variations, synthetic reproducibility, and the complexity of polymer preparation.

Another important aspect is processing tolerance and scalable manufacturing. The optimized morphology obtained by laboratory-scale spin coating cannot be assumed to remain unchanged during large-area deposition, where drying kinetics, shear effects, and temperature gradients differ significantly. Therefore, scaling up high-efficiency devices should not simply be regarded as increasing device area, but rather as a challenge of morphology transfer. Future efforts should focus on expanding the processing window, reducing reliance on complex post-treatments, and improving tolerance to thicker active layers and fluctuations during coating processes.

Environmental sustainability should also be considered together with processing performance. Existing studies indicate that the use of green or non-halogenated solvents does not necessarily require sacrificing morphology control. However, future evaluation should not simply focus on replacing one solvent with another, but should consider solvent toxicity, volatility, waste generation, and compatibility with large-area manufacturing. Therefore, green processing should gradually evolve from an additional advantage into an essential design constraint for high-performance PSMA systems.

Long-term operational stability is another critical issue. Polymerization can suppress large-scale molecular diffusion and potentially improve thermal and morphological stability in some systems, but it cannot eliminate degradation caused by photochemical reactions, thermal stress, interfacial instability, and mechanical deformation. Therefore, stability evaluation should be conducted under more standardized conditions that better reflect practical operating environments.

Ultimately, progress beyond 20% efficiency should no longer be evaluated solely by the highest PCE value. Other factors, including certified and average efficiencies, device-to-device variation, batch reproducibility, processing tolerance, large-area device performance, and operational lifetime, should also be considered. In other words, the next stage of development is not simply to achieve higher efficiency records, but to translate laboratory-scale record efficiencies into high-performance photovoltaic systems that are reproducible, environmentally sustainable, and compatible with scalable manufacturing.

5. Conclusion

PSMA-type polymer acceptors have played a decisive role in advancing all-polymer solar cells by combining the favorable optoelectronic characteristics of high-performance small-molecule acceptors with the structural advantages of polymeric materials. Their development has progressively overcome the optical and electronic limitations of earlier polymer acceptors,enabling champion PCEs above 20%, with certified efficiencies beyond 20% also emerging in selected systems. These advances demonstrate that the recent efficiency breakthrough cannot be attributed to a single molecular modification or processing strategy, but reflects progress across multiple levels of active-layer design.

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More importantly, the 20% milestone marks a change in the design objective of PSMA-based all-polymer solar cells. The central task is no longer to create an acceptor with increasingly favorable intrinsic properties, but to construct material systems in which those properties can be translated reliably into high photovoltaic performance. In this sense, the value of a PSMA should ultimately be judged not only by the highest efficiency it enables, but also by whether its high-performance state can be consistently reproduced.

Future progress should therefore focus on translating record efficiencies into robust practically accessible performance. Improvements in stability, reproducibility, environmentally compatible processing, and scalable fabrication will be essential if the advantages of polymer-based active layers are to extend beyond laboratory-scale devices. The next meaningful step for PSMA-based all-polymer solar cells may thus be defined less by another marginal increase in champion PCE than by the reliable realization of near-record efficiencies under realistic fabrication and operating conditions.

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Conflicts of Interest

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