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RECEIVED 29 September 2024

ACCEPTED 05 November 2024

PUBLISHED 21 November 2024

## CITATION

Wang M (2024) Significant improvement of organic solar cells with ternary additives. *Front. Phys.* 12:1503644. doi: 10.3389/fphy.2024.1503644

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# Significant improvement of organic solar cells with ternary additives

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Organic photovoltaics (OPVs) are gaining traction as a cost-effective and scalable alternative to conventional silicon-based solar cells, owing to their lightweight, flexibility, and tunable optical and electrical properties. Despite these advantages, the performance of OPVs is frequently hindered by non-optimal phase-separated morphologies that limit charge transport and extraction. A promising avenue to enhance the power conversion efficiency (PCE) of OPVs involves the incorporation of ternary additives, which have been shown to significantly refine the morphology of organic semiconductor blends. These additives promote the formation of a fibrillar-like nanoscale interpenetrating network, enhancing charge separation and transport. The dual-connected morphology achieved through ternary additives is crucial for improving the PCE. Additionally, the parallel bulk-heterojunction model formed by ternary additives has been instrumental in elucidating the photovoltaic parameters and understanding the effects of cascading heterojunction formation. Notably, block copolymer additives as one of ternary additives have demonstrated potential in stabilizing mixed donor-acceptor morphologies, leading to more efficient charge transfer. This review will explore the role of ternary additives in optimizing the morphology of OPVs and their impact on device performance, highlighting the mechanisms of block copolymer additives.

## KEYWORDS

morphology modulation, block copolymer, ternary additives, surface energy, open-circuit voltage

## 1 Introduction

OPVs are gaining global attention due to their unique characteristics, which include semi-transparency, non-toxicity, excellent flexibility, and low production costs [1–4]. Through systematic material design and meticulous processing optimization, significant progress has been made in enhancing the performance of OPVs. The power conversion efficiencies (PCEs) of single-junction OPV devices have notably reached up to 20%. However, a key challenge in the further development of these OPV devices lies in identifying reliable strategies for controlling the blend morphology [5–8]. A variety of strategies have been devised to manage film morphology and enhance device performance. Both materials and processing methods significantly influence the morphology, and thus, device performance can be optimized through a judicious design of material composition [4, 9–11].

The active layers of the heterojunction organic solar cells are composed of a blend of polymers or organic small molecules. As efficient acceptor materials, fullerene (C60) derivatives such as PC<sub>61</sub>BM or PC<sub>71</sub>BM are widely used in organic solar cells [12–15] and these types of organic solar cells are often referred to as fullerene-based organic solar

cells. Currently, the efficiency of fullerene-based organic solar cells, particularly those with ternary system heterojunction structures, can reach up to 20% [16–20]. However, fullerene acceptors have several disadvantages, including weak visible light absorption and limited energy levels, and expensive purification costs for fullerene derivative materials. Consequently, researchers have begun exploring non-fullerene acceptors as alternatives to fullerene derivatives, such as perylene diimide (PDI) and the small molecule material of ITIC. By carefully controlling the chemical structure and optoelectronic properties of the active layer material, researchers can achieve band alignment and finer phase separation, which can lead to efficiencies of 20% [16–20].

The use of ternary additives in the active layer represents an effective approach to controlling the morphology of donor and acceptor materials. This is because electron donor and acceptor materials form an interpenetrating network of nanoscale domains, creating interconnected pathways for charge transport and a large interfacial area for the dissociation of excitons into charge-separated states. Ternary additives can promote improved molecular ordering within the active layer, which can lead to more efficient charge transport and reduced energy losses, resulting in higher efficiency solar cells. Introducing a third component into the ternary system can effectively modulate the charge transfer state disorder, a key factor for exciton dissociation and charge transport, thereby suppressing charge recombination in the ternary device. Additionally, utilizing diverse materials in the ternary blend achieves complementary absorption spectra, which enhances light harvesting efficiency and results in increased short-circuit current density ( $J_{sc}$ ).

To further enhance device performance, a ternary blend structure was designed to tune the energy levels of the donor and acceptor materials. More importantly, this structure provides a cascading effect that aids in charge separation [21]. Moreover, the highly ordered fibrillar morphology of the phase offers a sufficient electron donor/acceptor interface, which promotes photo-exciton dissociation. As a result, this increases both the short-circuit current ( $J_{sc}$ ) and the fill factor (FF) of the device. The ternary organic solar cell, which introduces another electron donor or acceptor into the traditional donor-acceptor binary system, can not only broaden the system's spectral response and reduce energy loss but also induce the formation of a fibrillar morphology network. Based on theoretical simulations, researchers can rationally design bicontinuous donor and acceptor ratios by inputting related parameters, including surface energy values and Flory-Huggins interaction parameters. Through the combination of simulations and experiments, it is likely to form a bicontinuous morphology using a ternary blend of small molecules and an all-conjugated block copolymer. For example, the block copolymer (BCP) acts as a compatibilizer in a ternary blend active layer, supported by the alloy model and the parallel bulk-heterojunction model at different BCP compositions. Block copolymers can tune the optical and electronic properties of organic solar cells by influencing molecular ordering and domain characteristics. Molecular ordering can lead to better control over the morphology of the active layer, which is critical for achieving a balance between phase separation and intermixing of donor and acceptor materials. After designing appropriate photovoltaic materials, the next step

is to further implement the overall device properties and propose their widespread application in large-scale production [4].

## 2 High-performance ternary additives for OSCs

### 2.1 Small molecules additives

Figure 1 show the structure, development and efficiency, and GIWAXS (Grazing Incidence Wide Angle X-ray Scattering) and AFM (Atomic Force Microscope) characteristics of small molecules based ternary organic solar cells. Under the appropriate balance of compatibility, crystallinity, and surface energy between the host and guest acceptors, as well as the control of interfacial interactions between the donor and dual acceptors, some small molecules additives can penetrate into the interior of the host acceptor phase, forming embedded host/guest alloy aggregates. This feature greatly optimizes the morphology, maximizes energy transfer, and enhances exciton/charge behavior. In the ternary active layer of solar cells, a “guest” alloy component is embedded, which can broaden the spectrum of absorbed light, thereby improving the overall absorption rate of the cell to sunlight. This means that a light-sensitive component is introduced into the raw material, allowing the original solar cell to receive a broader spectrum of light, thereby improving the PCE. Research has confirmed the superiority of the LA1 alloy structure, which is a small molecule-based system with Y6 family receptors as the primary component. This has resulted in an impressive power conversion efficiency of 19.17% [16]. This research highlights the importance of phase distribution in ternary systems and reveals the controlling factors of phase distribution in ternary organic solar cells.

The innovation of organic photovoltaic acceptor materials, especially the use of acceptor-donor-acceptor (A-D-A) and acceptor-(donor-acceptor1-donor)-acceptor type small molecule acceptors (SMAs), has driven significant progress in OSCs. Typically, some SMAs have a banana-shaped molecular conformation, which helps multi-modal stacking and forms a dense 3D network structure, and increases the charge transport channels. The mainstream strategy for developing highly efficient non-halogen solvent-processable SMAs is focused on side chain engineering, such as extending side chains, changing the position of alkyl chain branching, and introducing polyethylene glycol chains. However, this strategy mainly focuses on modifying the alkyl chains on the central nucleus of SMAs, while not modifying the central nucleus itself, which limits the development of new acceptors. Research has shown that the A1 unit with a deficiency of electrons in the central nucleus plays a crucial role in constructing high-performance structure SMAs. Fine-tuning this unit can optimize the molecular energy level and luminescent properties, reduce the energy loss of devices, and improve the charge transport and exciton diffusion process. SMAs featuring benzo [1, 2, 5]thiadiazole (BP) or mono chlorinated BP as the electron deficient unit A1 group, including non chlorinated NA1, 10 chlorine substituted NA2, 8-chlorine substituted NA3, and 7-chlorine substituted NA4. The naphthalene ring is in the central nucleus of A-DA1D-A-type SMAs, which is different from common quinoline derived electron deficient units. The dual component

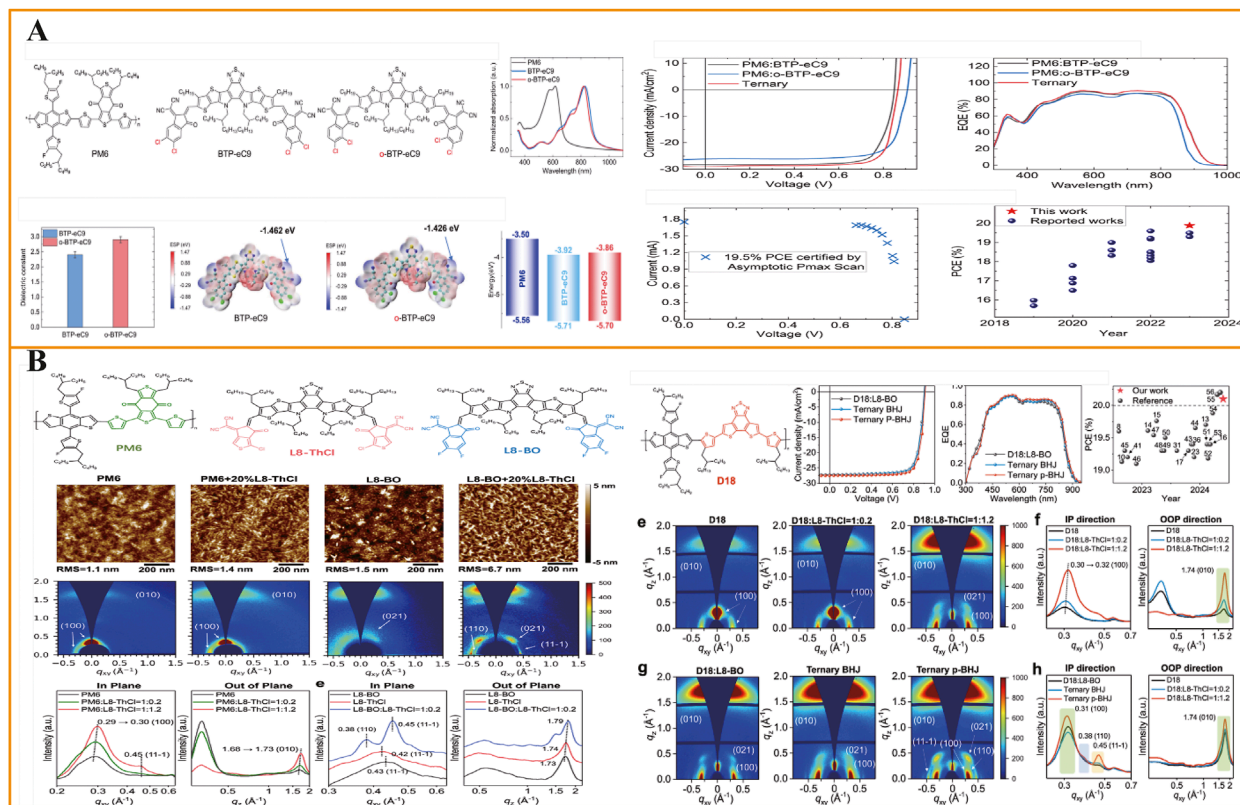


FIGURE 1 (A) Structure, development and efficiency, and characteristics of ternary organic solar cells, reproduced with permission from [18]. (B) Characteristics, development, efficiency and structure of ternary organic solar cells, reproduced with permission from [19].

composed of PM6 and NA3 achieved the highest photoelectric conversion efficiency in non halogen solvent treated OSCs, reaching 18.94%. By incorporating D18-Cl into the PM6 and NA3 binary system, the efficiency of the device was further improved to 19.75% [17].

A small molecules acceptor, BTP-eC9, has been synthesized. The researchers synthesized the unsaturated fatty acids of BTP-eC9 (CAS: 2598965-39-8, C<sub>86</sub>H<sub>94</sub>Cl<sub>4</sub>N<sub>8</sub>O<sub>2</sub>S<sub>5</sub>). BTP-eC9, with its excellent solubility and superior electron transport capabilities, outperforms its counterpart. The incorporation of BTP-eC9 into organic solar cells leads to a significant enhancement in both the short-circuit current and the fill factor, thereby boosting the overall power conversion efficiency. This improvement can be attributed to the material's ability to optimize the active layer's morphology, facilitate efficient charge transport, and suppress recombination losses. Owing to the enhancement of charge-transfer states, BTP-eC9 significantly reduces the energy loss in organic solar cells. Moreover, the ternary additives exhibit excellent solubility, crystallization, and energy-level compatibility with the host acceptor BTP-eC9 and o- BTP-eC9, enabling an efficiency of 19.9% for a ternary system [18]. The phase clusters in the ternary film are smaller and more uniform acting as recombination sites to capture charge carriers, while on the other hand, the band bending at the grain boundary-grain interface is beneficial for charge separation. By the way, the inclusion of BTP-OS, a similar and

important small molecule material, is crucial for enhancing the efficiency of OSCs [19].

The small molecule of L8-ThCl has been introduced as a third component into the PM6:L8-BO and D18:L8-BO systems. At the same time, fibrillar morphology of donor and acceptor phases is realized, and photovoltaic conversion efficiency is greatly improved. In the PM6:L8-BO system achieved 19.4% PCE, in the D18:L8-BO system reached a breakthrough 20.1%, and in the national photovoltaic product quality testing Center certification of 20.0%, is the world's first certification efficiency of more than 20% single-junction organic solar cell device [20]. Through atomic force microscopy (AFM) characterization, researchers found that the introduction of L8-ThCl can effectively induce the growth of polymer acceptor PM6 fibrillar morphology. Meanwhile, the L8-BO thin film, which could not form fibrillar morphology previously, also showed a dense fibrillar morphology-like morphology after the introduction of L8-ThCl. Through grazing incidence wide-angle X-ray diffraction (GIWAXS) characterization, researchers further found that after the introduction of L8-ThCl, the side-chain stacking (edge-on) in the donor film changed to the main-chain stacking (face-on), accompanied by an enhancement of p-p stacking.

An asymmetric phenyl-containing non-fullerene acceptor, Z8, has been utilized to create alloy acceptors in ternary organic solar cells (OSCs). The alkyl side chains substituted with phenyl

groups intermolecular interactions, improve the morphology of the film, effectively dissociate exciton, and reduce charge recombination. Asymmetric non fullerene receptor NFA (Z8) with phenyl substituted side chains exhibits ideal optoelectronic properties, such as high photoluminescence quantum yield (PLQY) and delocalized exciton, increasing the possibility of achieving low non radiative energy loss and efficient charge generation simultaneously. The phenyl group in Z8 can participate in intermolecular interactions, affecting receptor acceptor and donor receptor intermolecular interactions. Two types of NFAs, Z8 and L8-BO, can form alloy acceptors, and blending with polymer donor D18 helps to form good morphology, including appropriate phase separation, good vertical component distribution, and enhanced crystallinity. Organic solar cells based on the D18:Z8:L8-BO ternary blend have demonstrated an impressive efficiency of 20.2% [22].

## 2.2 Block copolymer additives

Figure 2 show the material structure, GIWAXS and TEM results, single chain in mean field (SCMF) results, and stretchability characteristics of block copolymer based ternary organic solar cells. The new material of block-copolymerized conjugated polymer of D18<sub>0.8</sub>-s-PEHDT<sub>0.2</sub> show great photovoltaic and mechanical performances. This new material combines highly stretchable polymers with a conductive polymer that has excellent electrical properties through chemical bonding. The efficiency of this flexible solar cell can reach up to 19%, and its stretching ability is ten times that of existing similar products [8]. In fact, the organic solar cell is capable of stretching its length by 40% while maintaining normal functionality. This attribute underscores its flexibility and potential for applications in wearable electronics and other flexible devices that require durable performance under deformation. This new type of conductive polymer has high photovoltaic characteristics and can be stretched like rubber. The potential of this newly engineered polymer is vast, with expectations that it will serve as a power source for the next-generation of wearable electronic devices.

The high open-circuit voltage ( $V_{OC}$ ) observed in this system reason can be elucidated by compatibility of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of the ternary blend materials as the cascade structure. For a simplified case, in ternary blend solar cells, the active layer typically has two donor polymers paired with an acceptor molecule. When light strikes the donor polymers, an electron is excited from the HOMO to the LUMO, thereby creating an exciton. The electron travels through the electron transport layer to the cathode while the holes travel in the opposite direction through the hole transport layer to the anode. This process occurs for both donor polymers, and the relative LUMO energy levels of the three materials provides a cascading effect that aids in charge separation as represented by Mok et al [14]. Cascading structure as energy payback element explains the increase of  $V_{OC}$ . The changes are on basis that an average HOMO and an average LUMO energy level exist in these BHJ active layers and that these energy levels are composition dependent. Therefore, constituent

miscibilities in such ternary blends must play an important role in determining their electronic structures. According to the parallel bulk heterojunction model, the  $V_{OC}$  is determined by the blend composition.

The block copolymer (BCP) PTB7-b-PNDI has been shown to enhance miscibility between donor and acceptor phases, thereby effectively controlling the interface between these domains. The coexistence of BCP droplets with a PTB7:PCBM continuous phase indicates the formation of a bicontinuous microemulsion. This system undergoes a microphase-to-macrophase transition, transitioning directly into a lamellar phase. A combination of simulations and experiments is investigated the equilibrium-like morphology formed by a ternary blend of this system. Continuous pathways promote transport of charges between electrodes and the block copolymer can be used as a polymer-compatibilizer to create co-continuous donor and acceptor domains and high interfacial overlap between the polymer domains. The single chain in mean field (SCMF) is applied to study the phase behavior of this system [4]. The blend compositions corresponding to the various phase transitions were found to depend more sensitively on the composition of the BCP and on the values of the Flory-Huggins interaction parameters. The mixing property, resulting in the active layer morphology, is determined by the surface energy values. The surface energies were previously measured through contact angle measurements and can be used to estimate the enthalpic interactions between different components. A significant improvement of PCE was conducted with amounts of 3 wt% BCP additives. Most interesting is the great increase in the open-circuit voltage ( $V_{OC}$ ) with the addition of BCP additives, from a value of 0.72 V to 0.82 V. Importantly, the tendency of  $V_{OC}$  is distinct from that typically observed for ternary blend systems, where a number of studies have reported either a linear change in  $V_{OC}$  for blends with good compatibility or pinning of  $V_{OC}$  to a low value for blends with poor compatibility since pinning of  $V_{OC}$  to a higher value is observed. The short circuit current ( $J_{SC}$ ) and Fill factor (FF) increase with the added BCP and then quickly degrade for BCP contents above 10 wt%. The results indicate that the PTB7-b-PNDI block copolymer significantly enhances the performance of PTB7:PCBM solar cells. This enhancement was achieved at relatively low loadings of the compatibilizer (2 wt%), and the increase in solar cell performance is primarily attributed to the enhancement of the open-circuit voltage ( $V_{OC}$ ) from 0.68 V to 0.78 V [4].

Upon absorption of light by the organic solar cell, an exciton is generated within the electron donor phase, where the exciton consists of bound positive and negative charges. These excitons must be dissociated into free charge carriers, which implies that the layer thickness should be optimized for exciton movement. It is essential to establish percolating pathways to facilitate the transfer of charge carriers to the electrodes. Transmission electron microscopy (TEM) images reveal a distinct mixing trend, which corroborates the PCE results from device testing, as well as data from single-chain in mean field (SCMF) and grazing incidence small-angle X-ray scattering (GISAXS) studies. By incorporating the block copolymer P3HT-b-PFTBT into the active layer, the chain segments link and interpenetrate, enhancing the aggregation of the active layer and, consequently, the device's stretchability.

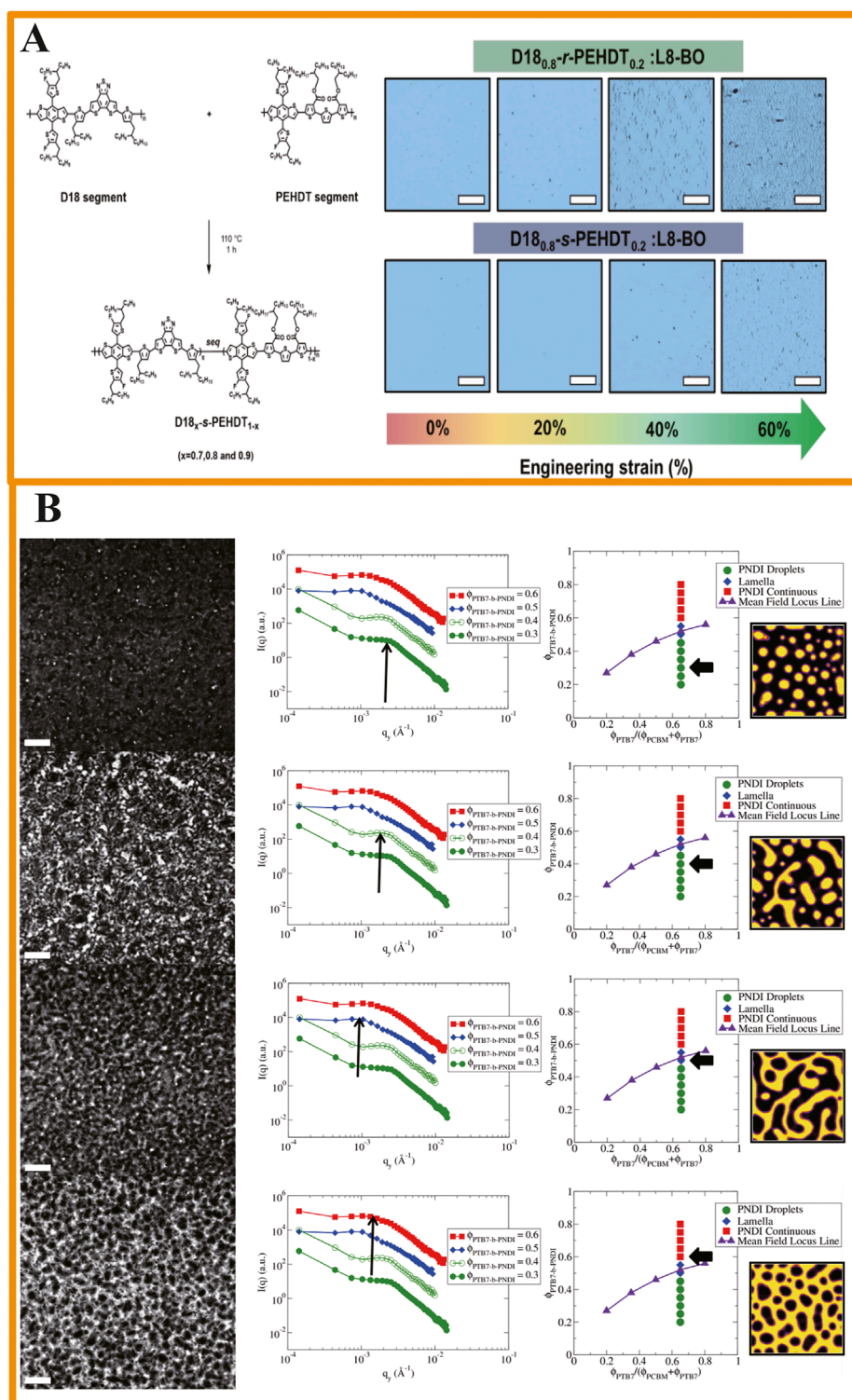


FIGURE 2

(A) Structure, and stretchability characteristics of ternary organic solar cells, reproduced with permission from [8]. (B) TEM, GIWAXS and SCMF results of ternary organic solar cells, reproduced with permission from [4].

Moreover, adjustments in material composition have led to the discovery that specific conditions can induce the formation of fiber-like structures, which significantly enhance the device's stretchability [4, 23].

### 3 Conclusion

The exceptional performance of organic photovoltaics with ternary additives shows the critical role of the rational design of

the donor/acceptor morphology. This design precision is achieved through the strategic blending of the ternary small organic molecules and block copolymers, leveraging a combination of computational simulations and empirical experiments to optimize the morphology. The results indicate that the ternary blend systems embody the characteristics of a parallel bulk-heterojunction model, where the third additive forms a distinct acceptor or donor phase. The compatibilizer within these systems facilitates the formation of a parallel-type solar cell, enhancing the overall performance. The results highlight the novel effects of ternary additives as compatibilizer, suggesting a promising direction for the development of ternary organic solar cells. The ternary additives can help tune electronic properties and significantly enhance the performance of polymer-based solar cells. These materials not only demonstrate exceptional photovoltaic performance but also exhibit outstanding mechanical properties. They efficiently convert sunlight into electricity and maintain strong durability under various mechanical stresses. The ternary additive-based organic solar cells, with their unique combination of high efficiency in solar energy conversion and the capability to maintain structural integrity, stand out as a promising candidate for a broad spectrum of applications.

## Author contributions

MW: Conceptualization, Methodology, Writing—original draft.

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## Funding

The author(s) declare that no financial support was received for the research, authorship, and/or publication of this article.

## Conflict of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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