

Simultaneously Enhanced Efficiency and Mechanical Durability in Ternary Solar Cells Enabled by Low-Cost Incompletely Separated Fullerenes

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All-polymer solar cells (all-PSCs) are one of the most promising application-oriented organic photovoltaic technologies due to their excellent operational and mechanical stability. However, the power conversion efficiencies (PCEs) are mostly lower than 16%, restricting their core competitiveness. Furthermore, the improvement of mechanical durability is rarely paid attention to cutting-edge all-PSCs. This work deploys a low-cost “technical grade” PCBM (incompletely separated but pure mixtures containing $\geq 90\%$ [70]PCBM or [60]PCBM), into the efficient PM6:PY-IT all-polymer blend, successfully yielding a high-performance ternary device with 16.16% PCE, among the highest PCE values for all-PSCs. Meanwhile, an excellent mechanical property (i.e., crack onset strain = 11.1%) promoted from 9.5% for the ternary system is also demonstrated. The “technical grade” PCBM slightly disrupts the crystallization of polymers, and disperses well into the amorphous polymer regions of the all-PSC blends, thus facilitating charge transport and improving film ductility simultaneously. All these results confirm introducing low-cost “technical grade” PCBM with high electron mobility into all-polymer blends can improve carrier mobility, reduce charge recombination, and optimize morphology of the amorphous polymer regions, thus yielding more efficient and mechanically durable all-PSCs.

1. Introduction

Organic solar cells (OSCs) have attracted enormous attention thanks to the flexibility, semitransparency, and low-cost large-area fabrication.^[1–16] Benefitting from emergence of the non-fullerene small molecule acceptors (NF-SMAs), for example, Y6 and its derivatives, over 19% power conversion efficiencies (PCEs) have been achieved for OSCs with polymer donors and NF-SMAs.^[17–20] Unlike NF-SMAs-based polymer solar cells (PSCs), all-PSCs, consisting of polymer donor and acceptor, display unique features, including excellent thermal and morphological stability, and outstanding mechanical durability.^[21–36] However, the low PCE of all-PSCs restricts their core competitiveness against their PSC counterparts.^[25,29,33] Besides, rather limited works have been reported on improving the mechanical durability of all-polymer systems. Therefore, it is important to simultaneously enhance the PCE and mechanical robustness of all-PSCs for facilitating their applications in flexible electronic field.

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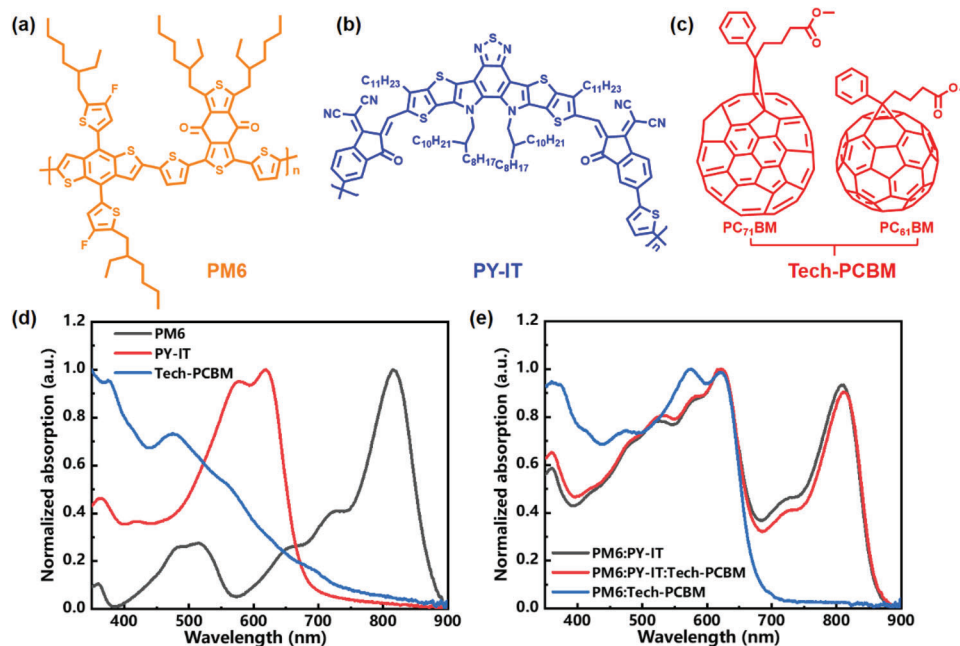


Figure 1. Chemical structures of a) PM6, b) PY-IT, and c) Tech-PCBM. d) Absorption spectra of PM6, PY-IT, and Tech-PCBM neat films; e) PM6:PY-IT, PM6:PY-IT:Tech-PCBM (1:0.8:0.2), and PM6:Tech-PCBM blend films.

Improving mechanical and photovoltaic performances can be enabled by designing and synthesizing new polymer materials, such as those reported via introducing flexible conjugation-break spacer units into polymer acceptors (PAs).^[37] However, material innovation often required additional chemical inputs. In contrast, the ternary tactic has been proved as an advantageous and simple strategy to uplift device performances.^[38,39] By adding a third component, for example, polymer donor, SM donor, fullerene derivative, NFAs, et. al., the film absorption, energy level, and morphology can be finely tuned.^[40–42] Notably, fullerenes and their derivatives, as excellent electron acceptors, play a non-negligible role in the field of OSCs. By introducing fullerene derivative, the PCEs of PSCs has exceeded 17%, even 18% in several cases, and other performances could be promoted, too.^[43] To date, fullerene derivatives as the third component in all-PSCs have been rarely reported, raising the confusion of whether fullerene derivative can be applicable in all-PSCs. Moreover, pure fullerenes have high energies of production, resulting in a high cost.^[44] Hence, deploying cost-effective fullerenes into cutting-edge all-PSCs for building highly efficient and mechanically robust ternary systems is urgently needed.

In this work, “technical grades” fullerenes (Tech-PCBM, **Figure 1**) were selected to construct a ternary blend with the previously reported efficient PM6:PY-IT system.^[21,45] “Technical grades” fullerenes are incompletely separated blends of the [60]PCBM and [70]PCBM, which are less processed than the pure fullerenes, leading to a reduced cost.^[44] With these, a ternary system achieving both enhanced efficiency and mechanical durability was demonstrated via combining the merits of all-polymer and incompletely separated fullerenes. With the introduction of 20% Tech-PCBM, a 16.16% PCE of the ternary device was achieved, significantly higher than PM6:PY-IT system (15.47%). The PCE enhancement is enabled by the greater open-circuit voltage (V_{oc}),

short-circuit current density (J_{sc}), and fill factor (FF), mainly ascribing to complementary film absorption, improved carrier mobility, and suppressed charge recombination corporately. Morphology studies reveal that Tech-PCBM slightly disrupts the crystallization of polymers, and disperses well into the amorphous polymer regions of the all-PSC blends, thus facilitating charge transport and improving film ductility simultaneously. These results suggest that incorporating the Tech-PCBM into all-polymer systems offers a simple and economical example for realizing mechanically robust and highly efficient all-PSCs.

2. Results and Discussion

The molecular structures of PM6, PY-IT, and Tech-PCBM are depicted in Figure 1a–c. Tech-PCBM shown in Figure 1c consists of $\geq 90\%$ [70]PCBM and the remainder [60]PCBM, which are quasi-spherical and spherical, respectively, allowing electron transfer from any direction and the possibility of morphology modulation. Figure 1d,e display the complementary absorption spectra of the three components, fully covering visible region and extending to near-infrared (NIR) region. As shown in Figure 1e, after incorporating Tech-PCBM into the all-polymer blend, the ternary films exhibited higher absorption than PM6:PY-IT blend films in the range of 350–650 nm. The complementary strong absorption of the ternary system contributes to boosting the J_{sc} of ternary device.

To investigate the effect of Tech-PCBM on photovoltaic performance, OSCs were constructed with a conventional architecture of ITO/PEDOT:PSS-PA/PM6:PY-IT:Tech-PCBM/PNDIT-F3N/Ag, where PNDIT-F3N was selected as the electron transport material to transport and collect electrons. The relevant molecular structure is presented in Figure S1, Supporting Information. After a systematic optimization, the weight ratio of

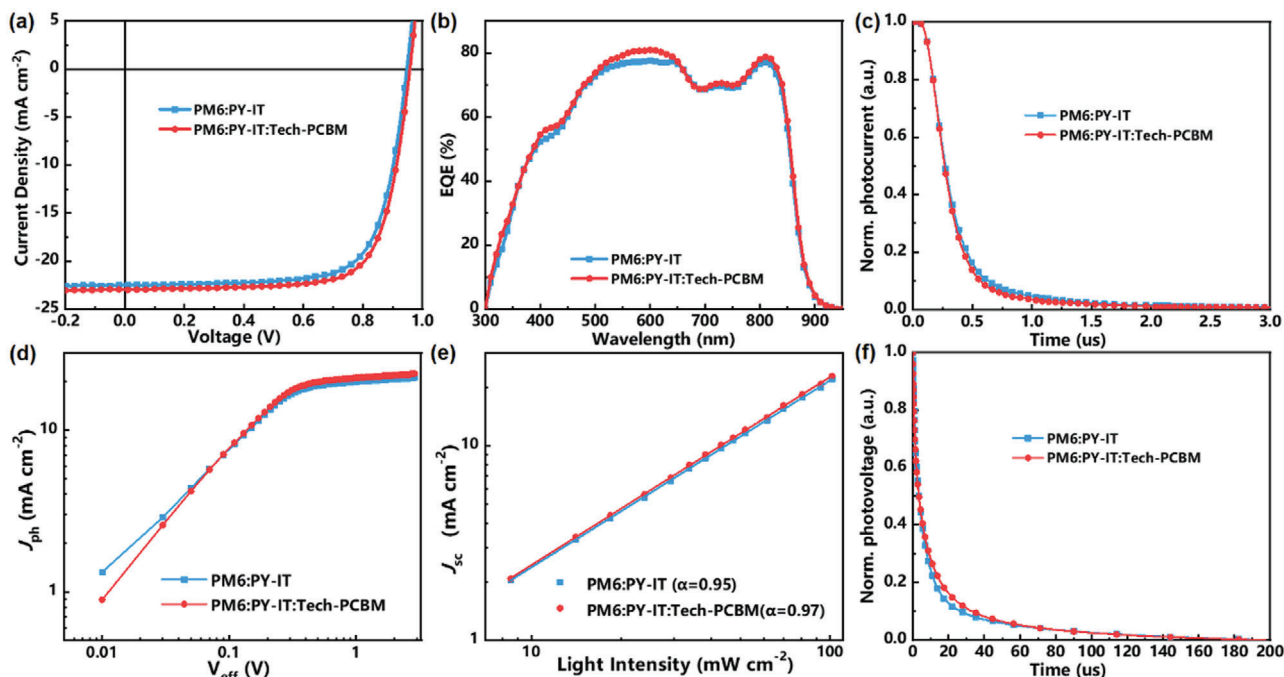


Figure 2. a) J - V curves; b) EQE spectra; c) TPC; d) J_{ph} versus V_{eff} ; e) light-intensity dependence of J_{sc} , and f) TPV of the optimal ternary and binary systems.

Table 1. Device performances of PM6:PY-IT:Tech-PCBM-based solar cells.

PM6:PY-IT:Tech-PCBM	V_{oc} [V]	J_{sc} [$\text{mA}\cdot\text{cm}^{-2}$]	FF [%]	PCE [%]
1:1:0	0.947	22.48/22.20 ^{a)}	72.6	(15.28 \pm 0.27) ^{b)} /15.47
1:0.9:0.1	0.952	22.53/22.28	73.2	(15.56 \pm 0.23)/15.72
1:0.8:0.2	0.956	22.93/22.72	73.6	(15.92 \pm 0.36)/16.16
1:0.7:0.3	0.959	22.60/22.36	72.9	(15.59 \pm 0.31)/15.83
1:0.5:0.5	0.961	18.19/17.93	69.5	(11.95 \pm 0.28)/12.16
1:0:1	0.967	13.81/13.40	66.8	(8.71 \pm 0.15)/8.93

^{a)} J_{sc} values integrated from EQE spectra. ^{b)} Average values in brackets are based on 20 devices.

PM6:PY-IT:Tech-PCBM was optimized to be 1:0.8:0.2 in the ternary system. The current density-voltage (J - V) curves of the optimum ternary and binary devices are presented in Figure 2a, and the corresponding photovoltaic parameters are also summarized in Table 1. The PM6:PY-IT system presented a PCE of 15.47%, which is concordant with previous reports.^[21,45] After adding Tech-PCBM, a significantly improved PCE of 16.16% with synergistically enhanced V_{oc} (0.956 V), J_{sc} (22.93 $\text{mA}\cdot\text{cm}^{-2}$), and FF (73.6%) was achieved for the optimal ternary system. To our knowledge, this impressive PCE of 16.16% is among the highest values for all-PSCs to date. Furthermore, we explored the performance of ambient atmosphere fabricated devices. Consequently, the ternary devices with the optimal weight ratio of 1:0.8:0.2 showed an excellent 15.76% PCE under this condition, while the PM6:PY-IT binary solar cells only provided 15.00% efficiency (Table S1, Supporting Information).

To verify the higher J_{sc} value of the ternary device, external quantum efficiency (EQE) measurement was conducted as displayed in Figure 2b. The ternary device displayed enhancement across the range of 400–620 nm compared to that PM6:PY-IT-based device, leading to a slightly higher J_{sc} . The enhanced parts were ascribed to the contribution of Tech-PCBM by a careful comparison of UV-vis absorption. The integrated J_{sc} values for the PM6:PY-IT and ternary devices are calculated to be 22.20 and 22.72 $\text{mA}\cdot\text{cm}^{-2}$, respectively, which agree well with the J_{sc} values derived from the J - V curves.

To further understand how Tech-PCBM affects device performance, the relevant photophysical processes were studied. The charge transport and extraction behavior were firstly investigated according to previously reported methods. The charge carrier mobility was evaluated via space-charge-limited current (SCLC) analysis. (Figure S2 and Table S2, Supporting Information). After adding molecule Tech-PCBM, the hole mobility gave an enhancement from 1.73×10^{-4} to $1.85 \times 10^{-4} \text{ cm}^{-2} \text{ V}^{-1} \text{ s}^{-1}$. While the ternary blend exhibited a higher electron mobility ($1.70 \times 10^{-4} \text{ cm}^{-2} \text{ V}^{-1} \text{ s}^{-1}$) than that of the PM6:PY-IT system ($1.55 \times 10^{-4} \text{ cm}^{-2} \text{ V}^{-1} \text{ s}^{-1}$). A possible reason for enhanced electron mobility is that the quasi-spherical and spherical shapes of Tech-PCBM allow electron transfer from any direction. Moreover, the ternary blends displayed more balanced charge mobilities, which played a crucial role in the improvement of J_{sc} and FF. Furthermore, transient photocurrent (TPC) decay results confirm the promoted charge collection in the ternary system. As presented in Figure 2c, the charge extraction time of 0.33 μs for the optimal ternary blend was slightly shorter compared to the PM6:PY-IT binary system (0.38 μs). The facilitated charge extraction of the ternary system well explains the improvement in J_{sc} and FF.

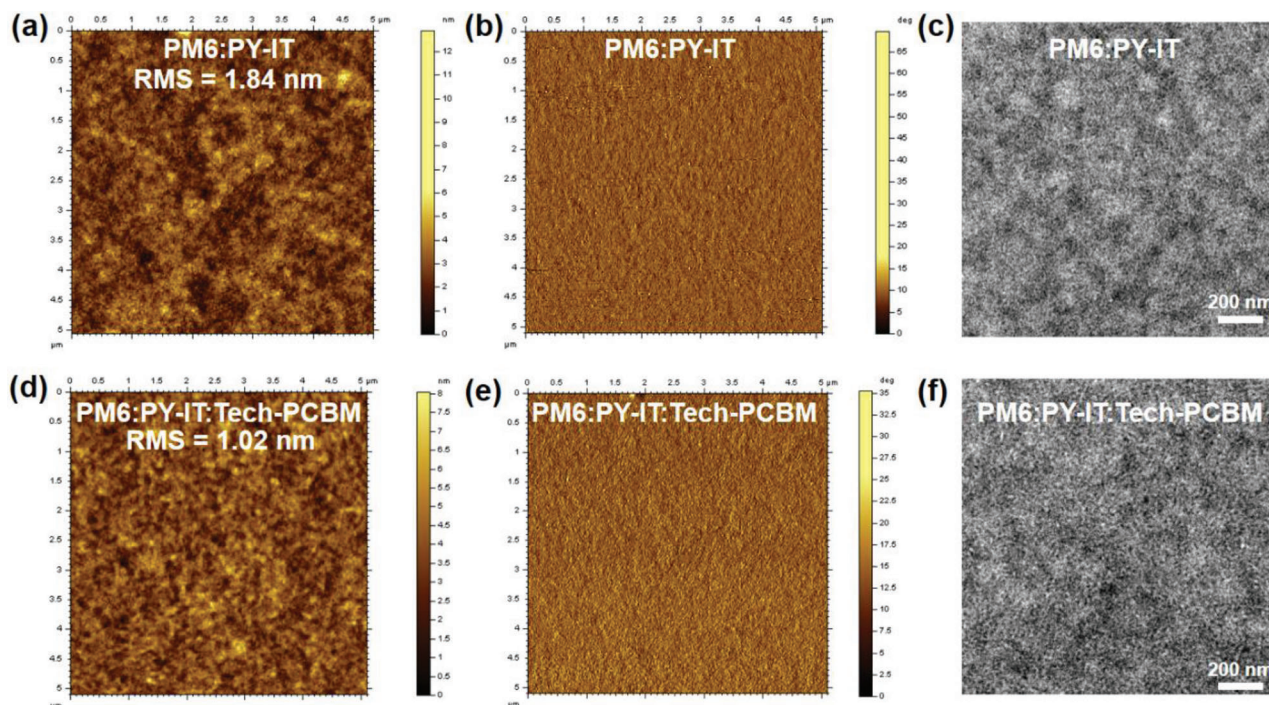


Figure 3. AFM height, phase and TEM images for a–c) PM6:PY-IT and d–f) PM6:PY-IT:Tech-PCBM (1:0.8:0.2) films.

The photocurrent density (J_{ph}) versus effective applied voltage (V_{eff}) was further characterized to investigate the charge generation and recombination behavior (Figure 2d). when V_{eff} exceeds 1.5 V, J_{ph} reaches saturation current (J_{sat}) for both devices indicating a small amount of charge recombination at high voltage.^[46] The total charge dissociation probability is estimated from the ratio of J_{ph}/J_{sat} . The ternary blend gave a higher value of 95.3% compared to the PM6:PY-IT binary system (91.5%) under short-circuit conditions, demonstrating improved charge dissociation and extraction in the ternary system. Moreover, the α values from light- from intensity (P) dependence of J_{sc} values were then analyzed. As shown in Figure 2e, the optimal ternary and all-polymer binary devices showed α values of 0.97 and 0.95, respectively, indicating the introduction of Tech-PCBM effectively decrease the bimolecular recombination. Furthermore, the charge recombination dynamics of the devices were investigated by the transient photovoltage (TPV) measurements (Figure 2f). The ternary blend exhibited a longer charge carrier lifetime (41 μ s) compared to the PM6:PY-IT binary blend (37 μ s), indicating the efficacy of Tech-PCBM in suppressing bimolecular recombination. All these results confirmed that the introduction of Tech-PCBM can synthetically promote charge extraction, suppress recombination, increase and balance charge mobility, thereby contributing to the high J_{sc} and FF.

The blend morphology plays an important role in determining the device performance.^[47] The atomic force microscope (AFM) and transmission electron microscope (TEM) measurements were conducted to gain more information on morphological changes (Figure 3). AFM results confirmed the lack of coarse phase separation in the binary and ternary blends, indicating nanoscale grain-like structures. Compared with the binary system (root-mean-square (RMS) = 1.84 nm), the ternary film ex-

hibits a smoother surface RMS of 1.02 nm, which may be due to a decreased crystallinity for the ternary blend. TEM images demonstrate that the introduction of Tech-PCBM can further refine the phase separation of PM6:PY-IT blend and boost charge separation at the donor/acceptor interface.

To further explore the role of Tech-PCBM in molecular packing, 2D grazing-incidence wide-angle X-ray scattering (GIWAXS) were characterized. 2D patterns and the corresponding line-profiles are presented in Figure 4, with calculated parameters in Tables S3 and S4, Supporting Information. In the out-of-plane (OOP) profile, the (010) peak shows increased d -spacing values from 3.85 to 3.90 Å after incorporating Tech-PCBM. Besides, the crystal coherence lengths (CCLs) for the π - π stacking peaks exhibited a decrease from 24.5 to 22.1 Å, indicating that Tech-PCBM slightly disrupted the crystallization of polymers. However, this decreased crystallinity does not negatively affect electron mobility in ternary films (as shown in SCLC above), implying that it could be compensated by the inherent high electron mobility of Tech-PCBM. Furthermore, by replacing 20 wt % PY-IT with Tech-PCBM, there is no Tech-PCBM peak observed (Figure 4e), implying no aggregation of Tech-PCBM in the ternary blend, which is mainly due to the fraction of Tech-PCBM below the threshold concentration. Thus, most of the Tech-PCBM molecules are dispersed in the amorphous polymer matrix instead of the formation of brittle Tech-PCBM aggregates,^[48] conducive to the improvement of mechanical properties.

Aside from PCE, mechanical durability is of great significance for evaluating the comprehensive performance of OSCs, but only a few reports have focused on this issue in recent years.^[37,43,49] To date, very few works have provided mechanical performance in high-performance all-PSCs, and work to further improve their mechanical durability is also in shortage. Under the status quo,

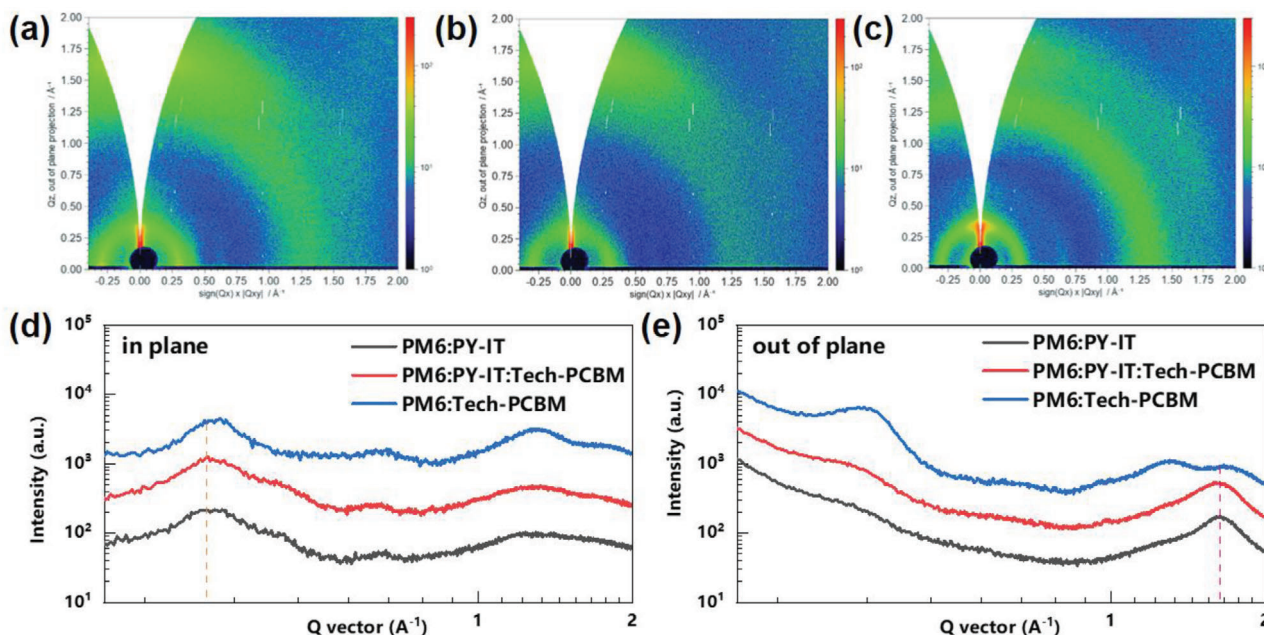


Figure 4. a–c) 2D-GIWAXS patterns, d) 1D OOP, and e) in-plane (IP) GIWAXS line profiles of PM6:PY-IT, PM6:PY-IT:Tech-PCBM (1:0.8:0.2), and PM6:Tech-PCBM blends.

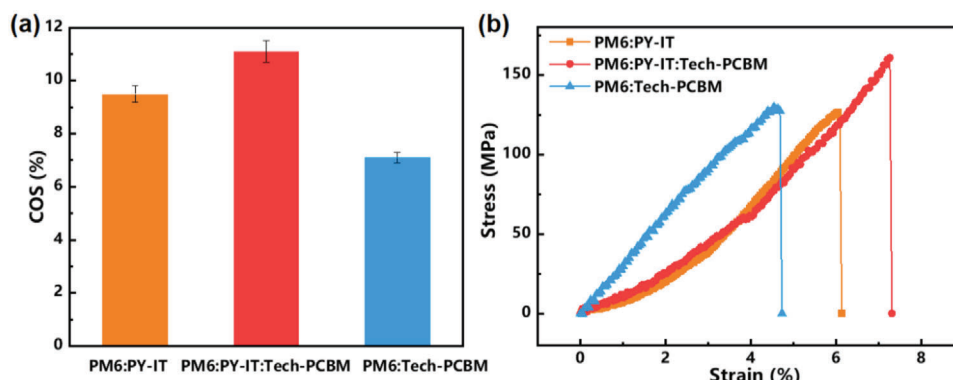


Figure 5. a) Plots of COS of the blend films via FOE method. b) Stress–strain curves of the blend films via FOW method.

the mechanical properties were studied with two well-established methods.^[43] As a figure of merit for the film stretchability under tension, the crack-onset-strain (COS) was firstly conducted with the film-on-elastomer (FOE) method. The corresponding crack optical images are presented in Figure S3, Supporting Information. As shown in **Figure 5a**, the COS is $9.5\% \pm 0.3\%$, $11.1\% \pm 0.4\%$, and $7.1\% \pm 0.2\%$ for PM6:PY-IT, PM6:PY-IT:Tech-PCBM, and PM6:Tech-PCBM blend films, respectively. Furthermore, a similar trend was observed for the stress-strain curves acquired from film-on-water (FOW) (Figure 5b). We note that the mechanical performances of PM6:Tech-PCBM-based devices were inferior to PM6:PY-IT-based devices because polymer acceptor is more ductile and demonstrates improved molecular packing interactions with the polymer donor. Interestingly, the addition of a small amount of Tech-PCBM slightly disrupts the crystallization of polymers and avoids forming large crystallites and Tech-PCBM aggregates, thereby leading to enhanced mechanical per-

formance (with 16.8% COS incensement). Thus, improved mechanical durability was achieved for all-PSCs with low-cost Tech-PCBM, together with high efficiency, resulting in mechanically robust and efficient all-PSCs.

3. Conclusion

We demonstrate a ternary solar cell via incorporating low-cost Tech-PCBM to the efficient PM6:PT-IT all-polymer system, achieving both improvements in PCE and mechanical properties. A PCE of 16.16% for the ternary device was obtained, significantly higher than that of the PM6:PT-IT binary system (15.47%). The efficiency improvement is attributed to complementary film absorption, improved charge extraction, suppressed charge recombination, enhanced charge mobility, and optimized phase separation. In addition, morphology studies indicate Tech-PCBM slightly disrupts the crystallization of polymers, and disperses

well into the amorphous polymer phases of the all-PSC blends, supporting the improved mechanical durability of the ternary films. Our results suggest that combining the advantages of all-PSCs and low-cost Tech-PCBM can realize the “best of both worlds” of the photovoltaic and mechanical performance, might open up a new road to achieve highly efficient and mechanically robust flexible OSCs.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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After initial online publication, the spelling of the name of author C.Y. was corrected on May 10, 2022, due to a previous error.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Keywords

all-polymer solar cells, fullerene, low cost, mechanical ductility, ternary solar cells

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