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### Constructing a new polymer acceptor enabled non-halogenated solvent-processed all-polymer solar cell with an efficiency of 13.8%<sup>†</sup>

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A new polymer acceptor, PS1, was developed by connecting the nonfullerene acceptor building block of dithienothiophen[3,2-b]pyrrolobenzotriazole capped with 3-(dicyanomethylidene)-indan-1-one through a thiophene spacer. The solubilizing alkyl side groups in the central unit enabled PS1 to be readily dissolved in non-chlorinated solvents. By using 2-methyltetrahydrofuran as the processing solvent, the all-polymer solar cell (all-PSC) containing PS1 and a polymer donor PTzBI-oF in the light-harvesting layer exhibited an impressively high power conversion efficiency of 13.8%.

All-polymer solar cells (all-PSCs) present great potential for practical applications because of their remarkable thermal and photochemical stability, robust mechanical properties, and compatibility for large-scale roll-to-roll processing.1-6 The primary concern regarding the development of all-PSCs remains the photovoltaic performance, as the power conversion efficiency (PCE) lags behind those obtained from small-molecule non-fullerene acceptors (NFAs) that have achieved a remarkable value of 18%.<sup>7-12</sup> In principle, the achievement of high photovoltaic performances for all-PSCs requires light-harvesting materials with efficient light absorption, high electron mobility, and excellent donor and acceptor miscibility to obtain a favorable film morphology.<sup>13-15</sup> To realize highperformance all-PSCs, we developed a series of polymer donors based on an imide-functionalized benzotriazole (PTzBI) unit by fine-tuning the molecular structures.<sup>16-20</sup> These copolymers could be easily dissolved in non-chlorinated solvents, for instance, 2-methyltetrahydrofuran (2-MeTHF), cyclopentyl methyl ether, d-limonene, and so forth.<sup>18-20</sup> When combined with one of the most extensively used n-type polymer acceptors N2200, the resulting all-PSCs containing PTzBI derivatives presented a PCE exceeding 11% after delicately optimizing the device

architecture and processing conditions.<sup>21</sup> However, the low extinction coefficient of N2200 in the near-infrared region significantly limits the light-harvest of the bulk-heterojunction layer. In this regard, the short-circuit current densities  $(I_{SC})$  of all-PSCs utilizing the polymer acceptor of N2200 are typically below 20 mA  $cm^{-2}$ .

An effective strategy for overcoming such limitations is the development of new polymer acceptors, which have high extinction coefficient in the NIR region. This can be achieved by copolymerizing the recently emerged building blocks of high-performance NFAs through a  $\pi$ -conjugated spacer.<sup>22–29</sup> This strategy has been proven very effective, as a range of polymer acceptors containing fused rings with well-extended conjugated lengths have been developed and showed impressive PCEs of 13-15%.<sup>30-35</sup> However, these all-PSCs are typically processed with chlorinated solvents (Table S1 in the ESI<sup>+</sup>) because of their large and coplanar structure. In pursuance of highly efficient all-PSCs that can be processed with non-chlorinated solvents to reduce the harmfulness to the environment,36-39 here we designed a narrow bandgap polymer acceptor, PS1 (Scheme 1). This copolymer contains a building block of dithienothiophen[3,2-b]pyrrolobenzotriazole end-capped with 3-(dicyanomethylidene)-indan-1-one. The long solubilizing side-chain in the central benzotriazole moiety imparts solubility in the non-chlorinated solvent of 2-MeTHF. Moreover, this copolymer presents a high absorption coefficient in the NIR region.

The synthesis of the target polymer acceptor PS1 was conducted through multiple steps, as shown in Scheme 1. By treating the dialdehyde compound TTPTAZ-CHO with a mono-brominated 2-(3-oxo-2,3-dihydro-1H-inden-1-ylidene) malononitrile, the dibromo monomer TTPTAZ-ICBr was obtained with a high yield of 80%. Synthesis of the target polymer PS1 was carried out via the palladium-catalyzed Stille polymerization of TTPTAZ-ICBr with 2,5-bis(trimethylstannyl)thiophene. The obtained copolymer PS1 was purified by Soxhlet extraction and extracted using chloroform (details are shown in the ESI<sup>†</sup>). The number-average molecular weight of PS1 was 10.9 kDa with a dispersity of 2.52, which was

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evaluated by high-temperature gel permeation chromatography, utilizing 1,2,4-trichlorobenzene as the eluent (measured at 150  $^{\circ}$ C, Fig. S1, ESI†).

The resulting polymer PS1 exhibited a broad absorption profile in the 600-1000 nm range, which was correlated with a strong charge-transfer effect and maximum absorption at approximately 810 nm (Fig. 1a). The absorption peak of PS1 in 2-MeTHF solution was 810 nm, which was bathochromically shifted by approximately 15 nm relative to that in chloroform (CF) solution (796 nm) (Fig. S4, ESI<sup>+</sup>). The PS1 thin film processed with 2-MeTHF had a high absorption coefficient of  $7.0 \times 10^4$  cm<sup>-1</sup>, which was slightly higher than that of the film processed with chloroform (5.1  $\times$  10<sup>4</sup> cm<sup>-1</sup>). The absorption profile of PS1 complemented the wide-bandgap conjugated donor polymer PTzBI-oF (Fig. 1a). The optical bandgap value of PS1 as estimated based on the onset of film absorption was 1.39 eV. Based on the cyclic voltammetry (CV) measurement (ferrocene/ferrocenium, Fc/Fc<sup>+</sup> potential: 0.39 V versus Hg/ Hg<sub>2</sub>Cl<sub>2</sub> electrode), the highest occupied molecular orbital energy level  $(E_{HOMO})$  and the lowest unoccupied molecular orbital ( $E_{LUMO}$ ) energy level of PS1 were -5.56 eV and -3.70 eV, respectively (Fig. 1b), on the basis of the equation:  $E_{HOMO}/E_{LUMO} =$  $-[e(E_{ox}/E_{red} + 4.8 - E_{Fc/Fc}^{+})]$  (eV), where  $E_{ox}/E_{red}$  is the onset oxidation/reduction potential versus the Hg/Hg<sub>2</sub>Cl<sub>2</sub> electrode. We note that the bandgap calculated from the CV measurement is 1.86 eV, which is obviously higher than the optical bandgap. This can be understood as the optical bandgap correlating to the exciton of the electron-hole pair has a relatively high Coulomb force interaction (binding energy), while such binding energy is not involved in the CV measurement.



Fig. 1 (a) Normalized absorption spectra of PTzBI-oF and **PS1** in thin films processed by 2-MeTHF. (b) Cyclic voltammetry curve of **PS1**.

To evaluate the performance of **PS1** as a polymer acceptor, all-PSCs were fabricated (structure: ITO/PEDOT:PSS/PTzBIoF:PS1/PFN-Br/Ag), with the current density-voltage (J-V) curves shown in Fig. 2a. Here the PTzBI-oF:PS1 layer was processed with chloroform or non-chlorinated 2-MeTHF under the optimized conditions (Tables S3-S5, ESI<sup>+</sup>). Note that the device processed with chloroform or 2-MeTHF presented an identical open-circuit voltage  $(V_{OC})$  of 0.92 V. The device processed with 2-MeTHF delivered a higher PCE of 13.8% compared to 12.1% for the chloroform-processed device, which was primarily due to the concurrently increased short-circuit current density  $(J_{\rm SC})$  of 22.47 mA cm<sup>-2</sup> and a fill factor of 66.70% (Table 1), and it is among the highest values achieved so far for all-PSCs that were processed with non-chlorinated solvents (Table S8, ESI<sup>+</sup>). These J<sub>SC</sub> values were verified from the external quantum efficiency spectra (Fig. 2b). The obtained device presented good thermal stability, with the PCE remaining at about 95% with respect to the initial value after thermal annealing at 80 °C for 60 h (Fig. S9, ESI<sup> $\dagger$ </sup>). It is also worth pointing out that the large-area (1 cm<sup>2</sup>) device based on PTzBI-oF:PS1 presented an impressively high PCE of 12.4% (Fig. S9, ESI<sup>†</sup>), suggesting its great potential for fabricating module devices.

The photoluminescence spectra of the pure PTzBI-oF films and PTzBI-oF:**PS1** films processed with either chloroform or 2-MeTHF showed that the emission quenching of the 2-MeTHF-processed film was more pronounced than that of the chloroform-processed film (Fig. 3a). Furthermore, the device processed with 2-MeTHF presented hole and electron mobilities of  $2.45 \times 10^{-3}$  and  $3.57 \times 10^{-4}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>, respectively, both of which were slightly higher than those of the device processed with chloroform ( $1.29 \times 10^{-3}$  and  $1.56 \times 10^{-4}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>, respectively), with the



Fig. 2 (a) Current density–voltage curves. (b) External quantum efficiency (EQE) and integrated  $J_{sc}$  curves for all-polymer solar cells based on PTzBI-oF:**PS1** processed by chloroform (CF) or 2-MeTHF.

Table 1 Photovoltaic parameters of PTzBI-oF: PS1-based devices

Solvent	$V_{\rm OC}$ (V)	$J_{ m SC}~({ m mA~cm^{-2}})$	FF (%)	$PCE^{a}$ (%) (best/avg.)
CF	0.92	21.45	61.36	12.1/11.7
2-MeTHF	0.92	22.47	66.70	13.8/13.5

 $^{a}$  Power conversion efficiency (PCE) values were obtained from 12 separate devices.

corresponding curves shown in Fig. S5 and Table S6 (ESI<sup> $\dagger$ </sup>). These observations were consistent with a slightly higher  $J_{SC}$  of the 2-MeTHF-processed device.

To further elaborate the dependence of the photovoltaic performances of all-PSCs on the processing solvents, we recorded the photocurrent density  $(J_{\rm ph})$ -effective voltage  $(V_{\rm eff})$ characteristics (Fig. 3b). The  $J_{\rm ph}$  of the PTzBI-oF:PS1 devices processed with chloroform gradually approached saturated values  $(J_{sat})$  of 23.04 mA cm<sup>-2</sup>, slightly lower than the 24.27 mA cm<sup>-2</sup> for the 2-MeTHF processed device at  $V_{\rm eff}$ exceeding 1 V, respectively (Table S7, ESI<sup>†</sup>). On the basis of the ratio of  $J_{\rm ph}/J_{\rm sat}$ , the charge dissociation probability parameter P(E,T) was estimated to be 90.7% and 93.9% for the devices processed with chloroform and 2-MeTHF, respectively. This finding implied that the exciton dissociation and charge collection efficiency of the 2-MeTHF-processed device is slightly more efficient than the counterpart device processed with chloroform, which is consistent with the higher EQE and  $J_{SC}$ of the former. To explore the carrier recombination behaviors, we studied the correlation of  $J_{SC}$  and the light intensity ( $P_{light}$ ), following the characteristics of  $J_{sc} \propto (P_{\text{light}})^{S}$ . The estimated exponential factor was 0.987 and 0.992 for the devices based on PTzBI-oF:PS1 processed with chloroform and 2-MeTHF, respectively, thereby suggesting a slightly lower bimolecular recombination in 2-MeTHF-processed devices. For the  $P_{\text{light}}$ - $V_{\text{OC}}$ characteristics, the devices processed with chloroform and 2-MeTHF presented slopes of 1.31 kT  $q^{-1}$  and 1.29 kT  $q^{-1}$ , respectively (Fig. S6b, ESI<sup>†</sup>). These observations demonstrated that both bimolecular recombination and Shockley-Read-Hall recombination were negligible in both devices.

Two-dimensional grazing incidence wide-angle X-ray scattering was employed to reveal the molecular stacking inside the neat and blended films. The neat **PS1** processed with chloroform and 2-MeTHF showed an intense  $\pi$ - $\pi$  stacking peak at *q* of 1.68 Å<sup>-1</sup>



Fig. 3 (a) Photoluminescence spectra of PTzBI-oF and PTzBI:**PS1** films processed by chloroform (CF) or 2-MeTHF excited at 530 nm. (b)  $J_{ph}$  *versus*  $V_{eff}$  characteristics of polymer solar cells based on PTzBI-oF:**PS1** processed by CF or 2-MeTHF.

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Fig. 4 Two-dimensional grazing incidence wide-angle X-ray scattering patterns of PTzBI-oF:**PS1** blend films processed by (a) chloroform (CF) and (b) 2-MeTHF under optimum conditions. Transmission electron microscopy images of PTzBI-oF:**PS1** blend films processed by (c) chloroform (CF) and (d) 2-MeTHF.

in the out-of-plane (OOP) direction, which was indicative of the preferential face-on polymer packing arrangement (Fig. S7, ESI<sup>+</sup>). The PTzBI-oF:PS1 blends processed with chloroform and 2-MeTHF displayed comparable scattering signals with a strong  $\pi$ - $\pi$  stacking peak ( $q = 1.76 \text{ Å}^{-1}$ ) in the OOP direction, associated with lamellar peaks ( $q = 0.35 \text{ Å}^{-1}$ ) in both OOP and in-plane directions. The peaks generated by the 2-MeTHF-prepared blend showed enhanced intensities when compared with those from the chloroform-prepared blend (Fig. 4a and b). The mesoscale morphology of the PTzBI-oF:PS1 blend was also investigated through transmission electron microscopy (TEM). Delicate and inter-continuous bright and dark regions were visible in both blends in the TEM images, but these features were finer in the 2-MeTHF-processed blend (Fig. 4c and d). From the atomic force microscopy images, one can note that both blends presented uniform and smooth surfaces, with a relatively small root-meansquare roughness of approximately 1.5 nm (Fig. S8, ESI<sup>+</sup>). These observations demonstrated the improved crystallinity and reduced-size domains in the 2-MeTHF-processed blend, which can assist in charge transfer and transportation and eventually increase the  $J_{SC}$  and fill factor of all-PSCs.

In summary, we have developed a new fused-aromatic-ringconstructed polymer acceptor, **PS1**, *via* a universal design strategy. The bulky and solubilizing alkyl chain on the key building block enabled the superior solubility of the polymer. **PS1** possessed excellent light harvesting capabilities, suitable energy levels, and a more favorable film morphology when blended with an electron-donating copolymer PTzBI-oF. The photovoltaic performance of all-PSCs highly depends on the processing solvent, where the non-chlorinated solvent 2-MeTHF resulted in a more favorable film morphology than the halogenated solvent chloroform. The optimized all-PSCs based on 2-MeTHF-processed PTzBI-oF:**PS1** delivered a high PCE of 13.8%. Our results suggest that this new polymer acceptor **PS1** has great potential for use in high-performance and non-chlorinated solvent-processed organic photovoltaic devices in future practical applications.

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#### Conflicts of interest

There are no conflicts to declare.

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