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Citation for the original published paper (version of record):

Fan, Q., Xiao, Z., Wang, E. et al (2021). Polymer acceptors based on Y6 derivatives for all-polymer solar cells. *Science Bulletin*, 66(19): 1950-1953. <http://dx.doi.org/10.1016/j.scib.2021.07.002>

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Contents lists available at ScienceDirect

Science Bulletin

journal homepage: [www.elsevier.com/locate/scib](http://www.elsevier.com/locate/scib)

Perspective

## Polymer acceptors based on Y6 derivatives for all-polymer solar cells

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Significant progress has been witnessed in the development of organic solar cells (OSCs) with power conversion efficiency (PCE) over 18% due to the tremendous breakthrough of non-fullerene fused-ring small molecule acceptors (SMAs) in recent years [1–3]. Although OSCs comprising of a polymer donor and a SMA have achieved the highest PCE, all-polymer solar cells (all-PSCs) consisting of tightly entangled polymer donor and polymer acceptor are generally considered to be the most promising practical wearable power generators in the future due to their outstanding device stability under mechanical and thermal stresses [4,5]. Fan et al. [4] systematically compared the mechanical properties of the flexible devices based on all-polymer and polymer:SMA blends with the same polymer donor. The all-polymer blend shows excellent mechanical elongation and high toughness of 9.3 MJ m<sup>-3</sup>, which are 6 and 11 times higher than those of the polymer:SMA one, respectively. The related flexible all-PSCs maintained >90% of their initial PCE after bending and relaxing 1200 times at a bending radius of 4 mm, which is much superior to the SMA-based flexible OSCs. Lee et al. [5] developed an all-PSC that retained over 90% of its initial PCE even after 100 h continuous heating at 100 °C, better than the relevant SMA-based OSCs. Driven by the above-mentioned advantages, several polymer acceptors based on Y6-derivatives have been developed and boosted the PCEs of all-PSCs up to 17% in the past two years (Fig. 1 and Table 1).

Reviewing the road-map of all-PSCs, only few polymer acceptors demonstrated moderate PCEs before 2017 due to their clear shortcomings. For instance, the naphthalene/perylene diimide-based polymer acceptors have low extinction coefficient of ~10<sup>-4</sup> cm<sup>-1</sup> [6] and the B ← N-bridged ones suffer from narrow absorption spectra (300–700 nm) [7], resulting in PCEs below 12%. To overcome the shortcomings, Zhang et al. [8,9] developed a strategy of polymerizing small molecule acceptors (PSMAs) to make high-performance polymer acceptors in 2017. As the first milestone of this strategy, PZ1 achieved a promising PCE of over 9% in all-PSCs [8], for it inherits the advantages of its SMA precursor namely IDIC-C16, i.e., wide absorption spectrum (300–800 nm), high extinction coefficient (>10<sup>5</sup> cm<sup>-1</sup>), and high electron mobility. After that, plenty of PSMAs based on SMAs of IDIC or ITIC

derivatives have been developed and boosted the PCEs approaching 13% [9].

In 2019, a milestone A-DA'D-A type SMA, namely Y6, was developed by Yuan et al. [10], and its single-junction OSCs obtained a record-breaking PCE of 15.7%. Unlike previous A-D-A type SMAs, Y6 has benzothiadiazole-fused central core (dithienothiophen [3,2-*b*]pyrrolobenzothiadiazole (BPT)) with a DA'D fused structure, which results in strong quinoidal character for broadening molecular absorption to near-infrared region. Moreover, the twisted central core conjugated with two end-groups on the same side offers the mixed intermolecular interactions from BPT-core in the perpendicular direction and from end-groups in the horizontal direction, forming a three-dimensional interpenetrating network for efficient intermolecular charge transport [11]. So far, the OSCs based on Y6 derivatives have achieved PCE over 18% [2,12]. Inspired by the successes of the PSMA strategy and Y6-derivatives, rapid development in polymer acceptors based on Y6-derivatives has occurred for more efficient all-PSCs (Fig. 1 and Table 1). Jia et al. [13] reported a polymer acceptor PJ1 with a narrow bandgap and a high absorbance by polymerizing a Y6-derivative with a thiophene spacer, and the resulting all-PSCs achieved an impressive PCE of 14.4%. At the same time, Wang et al. [14] developed another similar polymer acceptor PYT with different side-chain on BPT core and obtained a PCE of 13.44% by finely controlling molecular weight. Fan et al. [15] developed a polymer acceptor PF5-Y5 by replacing thiophene spacer with benzodithiophene. PF5-Y5 shows strong absorption (onset at 880 nm and extinction coefficient >10<sup>5</sup> cm<sup>-1</sup> in film), excellent electron mobility (>10<sup>3</sup> cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>), and high-lying lowest unoccupied molecular orbital (LUMO) level (-3.84 eV), which is conducive to both high open-circuit voltage ( $V_{oc}$ ) and high short-circuit current density ( $J_{sc}$ ) in all-PSCs. Moreover, the PF5-Y5-based all-PSCs possess smaller energy loss ( $E_{loss}$ ) (0.57 eV) and more efficient charge separation and transport in comparison with the OSCs based on its SMA precursor Y5. As a result, the PF5-Y5-based all-PSCs achieved a high PCE of 14.45%. Different from the above polymer acceptors with an electron-donating  $\pi$ -spacer, Sun et al. [16] reported an A-A type polymer acceptor L14 with narrow bandgap and low-lying energy levels by copolymerizing a Y6-derivative with a bithiophene-fused imide as  $\pi$ -spacer. Such low-lying energy levels offer higher electron transfer character without sacrificing  $V_{oc}$ , being attributed to a

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<https://doi.org/10.1016/j.scib.2021.07.002>

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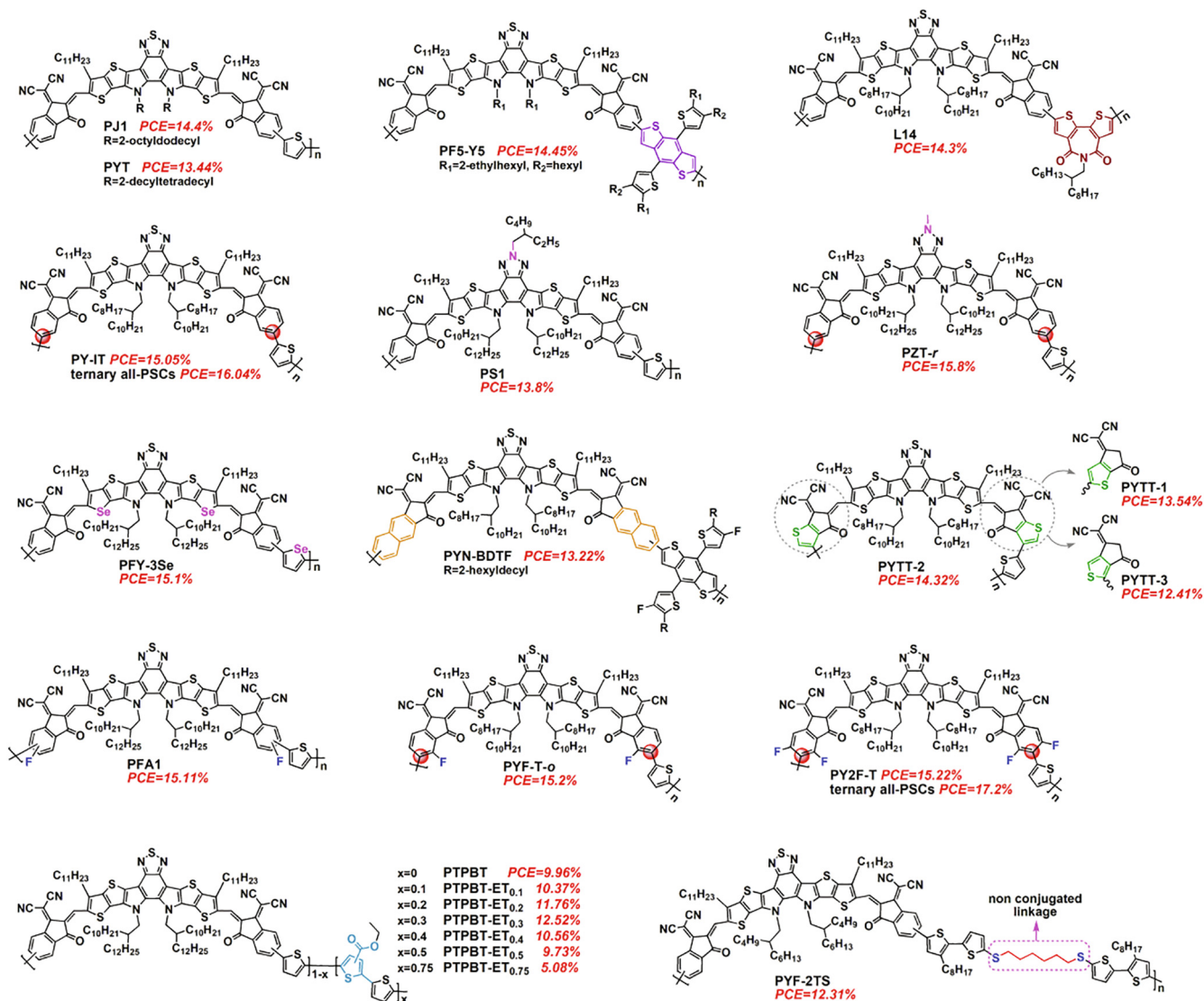


Fig. 1. (Color online) The chemical structures of representative polymer acceptors based on Y6-derivatives.

Table 1

Performance data for binary all-PSCs with polymer acceptors based on Y6-derivatives.

Acceptors	Donors	$E_g$ (eV) <sup>a</sup>	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Ref.
PJ1	PBDB-T	1.41	0.90	22.3	70	14.4	[13]
PYT	PM6	1.41	0.93	21.78	66.33	13.44	[14]
PF5-Y5	PBDB-T	1.41	0.946	20.65	74	14.45	[15]
L14	PM6	1.41	0.96	20.6	72.1	14.3	[16]
PY-IT	PM6	1.39	0.933	22.3	72.3	15.05	[17]
PY-IT:N2200	PM6	1.39	0.947	22.60	74.9	16.04	[18]
PS1	PTzBI-oF	1.39	0.92	22.47	66.7	13.8	[19]
PZT-r	PBDB-T	1.36	0.896	24.7	71.3	15.8	[20]
PFY-3Se	PBDB-T	1.35	0.871	23.6	73.7	15.1	[21]
PYN-BDTF	PBDB-T	1.38	0.86	22.28	69	13.22	[22]
PYTT-1	PBDB-T	1.49	0.93	20.66	70.35	13.54	[23]
PYTT-2	PBDB-T	1.49	0.91	22.00	71.53	14.32	[23]
PYTT-3	PBDB-T	1.44	0.82	21.99	68.47	12.41	[23]
PFA1	PTzBI-oF	1.41	0.87	23.96	72.67	15.11	[24]
PYF-T-o	PM6	1.38	0.901	23.3	72.4	15.2	[25]
PY2F-T	PM6	1.37	0.86	24.27	72.62	15.22	[26]
PY2F-T:PYT	PM6	1.34	0.90	25.2	76.0	17.2	[27]
PTPBT	PBDB-T	1.42	0.849	19.82	59.2	9.96	[28]
PTPBT-ET <sub>0.3</sub>	PBDB-T	1.42	0.899	21.33	65.3	12.52	[28]
PYF-2TS	PBDB-T	1.40	0.906	20.47	66.3	12.31	[29]

<sup>a</sup> Calculated from absorption onsets of polymer acceptors.

small nonradiative energy loss of 0.22 eV. Blending with PM6, L14 yielded a PCE of 14.3% in all-PSCs.

Considering the previously reported Y6-derived monomers containing three isomers with bromines at different positions, Luo et al. [17] separated these isomeric monomers and synthesized a regio-regular polymer acceptor PY-IT. Compared to its random analog PY-IOT (PYT), PY-IT shows red-shifted absorption and improved electron mobility, yielding an excellent PCE of 15.05% in PM6:PY-IT-based all-PSCs. Afterwards, Ma et al. [18] made PM6:PY-IT:N2200-based ternary all-PSCs with an optimized morphology by introducing a crystalline polymer acceptor N2200 as the third component, and the corresponding devices achieved a higher PCE over 16%. Recently, by replacing BPT central core of PJ1 with dithienothiophen[3,2-*b*]pyrrolobenzotriazole (BPTz), Zhu et al. [19] developed a polymer acceptor PS1. Owing to the additional solubilizing alkyl side-chain in BPTz unit, PS1 can be readily dissolved in non-halogenated 2-methyltetrahydrofuran (THF-Me) solvent, and the all-PSCs gave a promising PCE of 13.8%. Fu et al. [20] reported another BPTz-containing polymer acceptor PZT- $\gamma$  by combining side-chain engineering and regio-specific end-group. Because of less electron-deficient property of benzotriazole unit, PZT- $\gamma$  shows significantly red-shifted absorption and up-shifted LUMO level in comparison with BPT-containing PYT, leading to an improved  $J_{sc}$  of 24.7 mA cm<sup>-2</sup> and a lower  $E_{loss}$  of 0.51 eV in all-PSCs. As a result, a record-high PCE of 15.8% was demonstrated. In addition to wide absorption and high LUMO level, it is also important to note that PSMA has good batch-to-batch consistency and excellent reproducibility in device performance. By combining selenophene-fused Y6-derived framework and selenophene  $\pi$ -spacer, Fan et al. [21] reported a multi-selenophene-containing narrow-bandgap polymer acceptor PFY-3Se with a high PCE of 15.1% in all-PSCs. Owing to the strong intermolecular interaction of selenophene units, three batches of PFY-3Se with different number-average molecular weights ( $M_n$ ) show similar temperature-dependent aggregation property, helping the formation of consistent blend morphology. In all-PSCs, PFY-3Se shows excellent batch-to-batch reproducibility in device performance, and all the devices based on PFY-3Se with different  $M_n$  (25.8–42.4 kDa) gave high PCEs of 14.5% – 15.1%. Recently, the end-group modification strategies of SMA building blocks have been also applied in designing high-performance Y6-derived polymer acceptors. Su et al. [22] reported a polymer acceptor, PYN-BDTE, by introducing  $\pi$ -extended naphthalene-based end-groups. Compared to its analogous polymers with benzene-based end groups, PYN-BDTE shows a red-shifted absorption onset extending to ~900 nm. Blended with PBDB-T, PYN-BDTE-based all-PSCs exhibited a PCE of 13.22% with a high  $J_{sc}$  of 22.28 mA cm<sup>-2</sup>. Wang et al. [23] developed three polymer acceptors (PYTT-1, PYTT-2, and PYTT-3) with different isomeric thiophene-fused end-groups. The isomeric molecular geometry significantly affects the optoelectronic, packing, and charge-transport properties. PYTT-2 offered the highest PCE of 14.32% in all-PSCs due to the improved charge transport, less non-radiative loss, faster charge extraction, and optimized morphology.

Fluorination of polymer acceptors based on Y6-derivatives was also carried out to improve photovoltaic performance. Peng et al. [24] developed a polymer acceptor PFA1 based on a Y6-derivative with monofluorinated 1,1-dicyanomethylene-3-indanone (IC) end group. Compared with its fluorine-free analog PY5T (PJ1) with a low PCE of 4.01% in all-PSCs based on a wide-bandgap polymer donor PTzBI-oF, PFA1 achieved an outstanding PCE of 15.11% due to broadened absorption spectrum, higher extinction coefficient, increased electron mobility, and better compatibility in PTzBI-oF:PFA1 blend. Unlike random PFA1 (PYF-T), the regio-regular polymer acceptor PYF-T-o with fluorine on the specific position showed more ordered inter-chain packing and suitable phase separation in active layer [25]. Blended with a polymer donor PM6, the PYF-T-o-

based all-PSCs offered an increased PCE of 15.2% than PFA1-based ones (14.0%). Further, Yu et al. [26] developed a polymer acceptor PY2F-T based on a difluorinated end group. Compared with PYF-T-o with a monofluorinated end group, PY2F-T presented broadened absorption and the corresponding all-PSCs gave a higher  $J_{sc}$  of 24.27 mA cm<sup>-2</sup>. Then, PYT as the third component was introduced into PM6:PY2F-T host system [27]. Thanks to the complementary absorption and fine-tuned microstructures of the ternary blend, the ternary all-PSCs achieved a record-high PCE of 17.2%, which is comparable to the polymer donor:SMA system.

On the other hand, Du et al. [28] synthesized a series of polymer acceptors namely PTPBT-ETx via a random ternary copolymerization strategy with a Y6-derived building block, a thiophene  $\pi$ -spacer, and fine controlled amount of 3-ethylesterthiophene (ET) units. With increasing ET content in PTPBT-ETx, the related all-PSCs showed gradually increased  $V_{oc}$ . Among the devices with PTPBT-ETx, the PTPBT-ET<sub>0.3</sub>-based all-PSCs yielded the highest PCE of 12.52% with a long-term photostability over 300 h. Interestingly, Fan et al. [29] reported a non-fully conjugated polymer acceptor PFY-2TS via polymerizing a Y6-derived unit (namely YBO) with a non-conjugated thioalkyl linkage. Compared with YBO-based fully-conjugated polymer acceptor PFY-DTC, PFY-2TS showed similar absorption spectrum and electron mobility, but quite different crystallinity and aggregation properties, leading to optimal active layer morphology and better device physical processes in all-PSCs. As a result, PFY-2TS-based all-PSCs gave a PCE of 12.31% with a small non-radiative energy loss (0.24 eV), better than PFY-DTC-based ones (11.08%).

Viewing the development of Y6-derived polymer acceptors, the various molecular design strategies focus on side-chain engineering on Y6-derived building blocks for optimizing solubility,  $\pi$ -spacer modification for regulating intermolecular interaction, fluorination and regio-regulation on the end group for broadening absorption, and screening of Y6-derived units and introduction of the third component for optimizing crystallinity and energy levels. Thanks to the rapid development of Y6-derived polymer acceptors, the highest PCE over 17% has been achieved in all-PSCs [27], which significantly narrows the PCE gap between all-PSCs and SMA-based OSCs. With fine-tuned polymer structures and optimized active-layer morphologies, all-PSCs achieve high  $J_{sc}$  enabled by broadening absorption spectra (300–920 nm), increased  $V_{oc}$  due to well-matched energy levels and decreased energy loss (0.50–0.55 eV), and improved fill factor, leading to the state-of-the-art all-PSCs performing quite close to the most efficient SMAs-based devices. To this end, one can confidently conclude that the photovoltaic performance of all-PSCs does not necessarily fall behind SMAs-based devices. Apart from further improvement of PCEs for all-PSCs through the development of novel polymer donors and acceptors, optimization of morphologies and innovation of device architectures, further attention should be paid to the device stability, batch-to-batch variations, and large-area processing techniques in near future.

## Conflict of interest

The authors declare that they have no conflict of interest.

## Acknowledgments

E. Wang thanks the Swedish Research Council, Formas and the Wallenberg Foundation. L. Ding thanks the National Key Research and Development Program of China (2017YFA0206600) and the National Natural Science Foundation of China (51773045, 21772030, 51922032, 21961160720) for financial support.

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