Supporting Information

Butterfly-effect of Flexible Linker in Giant-molecule Acceptor: Optimized Crystallization and Aggregation for Enhancing Mechanical Durability and Approaching 19% Efficiency in Binary Organic Solar Cells

Qinrui Ye,^{a,b} Wei Song,^{a,b,*} Yongqi Bai,^{a,b} Zhenyu Chen,^{a,b,*} Pengfei Ding,^{a,b} Jinfeng Ge,^{a,b} Yuanyuan Meng,^{a,b} Bing Han,^c Xin Zhou,^d and Ziyi Ge^{a,b,*}

^a Zhejiang Engineering Research Center for Energy Optoelectronic Materials and Devices, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, China.

^b Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China.

^c The Hong Kong University of Science and Technology (Guangzhou), Sustainable Energy and Environment Thrust, Nansha, Guangzhou, Guangdong, China.

^d Beijing National Laboratory for Molecular Sciences, College of Chemistry and Molecular Engineering, Peking University, 100871 Beijing, China.

*E-mail: songwei@nimte.ac.cn; chenzhenyu@nimte.ac.cn; geziyi@nimte.ac.cn

Experimental section

Materials: compound 1, compound A, compound B, compound C, BTP-eC9, PM6(M_n =45000Da, M_w = 97000Da) and PY-IT (M_n =8500Da) were purchased from Solarmer Materials Inc. Toluene was purchased from Zhengzhou Alfa Chemical Co., Ltd., pyridine, chloroform were commercially available from China National Medicines Corporation Ltd. Pd₂(dba)₃ was obtained from Energy Chemical. All the solvents, materials were used without further purification. PEDOT: PSS (Clevios P VP 4083) was obtained from J&K Chemicals Inc. The indium-doped tin oxide (ITO)-coated glass (1.1 mm thick, \leq 15 Ω /square) were purchased from Wuhu Token Sciences Company.

Synthesis route:

The synthesis route of DSY-C4 exhibited as follows.



Scheme S1 The synthetic route of compound DSY-C4

Synthesis of Compound 2: compound 1 (0.3g, 0.386mmol) and NBS (0.207g, 1.16mmol) were added to a flask with three necks, then CF (10ml) and CH₃COOH (10ml) were injected. After stirring at 0°C for 6 hours, the reaction end. After removal of the solvent, the residue was purified by silica gel chromatography (dichloromethane: hexane=1:9) to get a red solid (0.3g, 83%).¹H NMR (400 MHz, Chloroform-*d*) δ 7.42 (s, 2H), 4.55 (d, *J* = 7.7 Hz, 4H), 2.09 – 1.94 (m, 2H), 1.18 – 0.76 (m, 30H), 0.65 (td, *J* = 15.7, 7.3 Hz, 16H).

Synthesis of Compound 3: compound 2 (0.445g, 0.636mmol) and THF (20ml) were added to a flask with three necks under nitrogen at -60°C, then LDA (1.27ml, 2.543mmol) was injected. The reaction was stirred at -60°C for 1 hour and room temperature for 6 hours. Then CH₃OH (2ml) was injected at -20°C to end the reaction. The mixture was quenched with water, extracted with dichloromethane. After removal of the solvent, the residue was purified by silica gel chromatography (dichloromethane: hexane=1:4) to get red liquid (0.3g, 67%). ¹H NMR (400 MHz,

Chloroform-*d*) δ 7.30 (s, 2H), 4.59 (d, *J* = 6.7 Hz, 4H), 2.10 – 1.99 (m, 2H), 1.14 – 0.76 (m, 30H), 0.74 – 0.56 (m, 16H).

Synthesis of Compound 4: compound 3 (0.65g, 0.697mmol), compound A (0.282g, 0.581mmol), Pd₂(dba)₃ (5mg) and P(o-tol)₃ (15mg) were added to a flask with three necks under nitrogen, then toluene (20ml) was injected. After stirring at 120°C for 6 hours, the mixture was quenched with water, extracted with CF. After removal of the solvent, the residue was purified by silica gel chromatography (dichloromethane: hexane=1:3) to get yellow liquid (0.27g, 37%)..¹H NMR (400 MHz, Chloroform-*d*) δ 7.47 (s, 1H), 7.39 (s, 1H), 7.30 (s, 1H), 6.91 (s, 1H), 4.61 (t, *J* = 9.5 Hz, 4H), 2.63 (d, *J* = 6.9 Hz, 2H), 2.15 – 1.99 (m, 2H), 1.68 (t, *J* = 6.1 Hz, 1H), 1.36 (d, *J* = 14.9 Hz, 8H), 1.10 – 0.80 (m, 34H), 0.76 – 0.57 (m, 16H).

Synthesis of Compound 5: compound 4 (0.136g, 0.13mmol), compound B (0.036g, 0.065mmol), $Pd_2(dba)_3$ (5mg) and P(o-tol)₃ (15mg) were added to a flask with three necks under nitrogen, then toluene (20ml) was injected. After stirring at 120°C for 6 hours, the mixture was quenched with water, extracted with CF. After removal of the solvent, the residue was purified by silica gel chromatography (dichloromethane: hexane=1:2) to get red liquid (0.1g, 71%).. ¹H NMR (400 MHz, Chloroform-*d*) δ 7.46 (s, 2H), 7.43 (s, 2H), 7.40 (d, *J* = 3.4 Hz, 2H), 7.38 (s, 2H), 6.91 (s, 2H), 6.89 (d, *J* = 3.6 Hz, 2H), 4.64 (s, 8H), 2.97 (s, 4H), 2.63 (d, *J* = 6.8 Hz, 4H), 2.09 (t, *J* = 6.7 Hz, 4H), 1.91 (s, 4H), 1.71 – 1.63 (m, 2H), 1.35 (d, *J* = 13.4 Hz, 16H), 1.05 – 0.79 (m, 68H), 0.63 (dq, *J* = 15.7, 8.0, 7.3 Hz, 32H).

Synthesis of Compound 6: compound 5 (0.1g, 0.045mmol) and 1,2-C₂H₄Cl₂ were added to a flask with three necks under nitrogen at 0°C, then DMF (0.17ml, 1.81mmol), and POCl₃ (0.14ml, 1.81mmol) were injected. After stirring at 0°C for 2 hours, the flask was rising to 85°C and stirring for 8 hours. After that, the mixture was quenched with saturated sodium carbonate solution, extracted with CF. After removal of the solvent, the residue was purified by silica gel chromatography (dichloromethane: hexane=3:1) to get red solid (0.086g, 86%). ¹H NMR (400 MHz, Chloroform-*d*) δ 10.27 (s, 2H), 10.25 (s, 2H), 7.46 (d, *J* = 3.5 Hz, 2H), 7.41 (s, 2H), 7.20 (s, 2H), 7.02 (d, *J* = 3.6 Hz, 2H), 4.66 (s, 8H), 3.06 (s, 4H), 2.67 (d, *J* = 6.9 Hz, 4H), 2.06 (s, 4H), 1.98 (s, 4H), 1.66 (s, 2H), 1.40 – 1.29 (m, 16H), 0.94 (tq, *J* = 21.4, 13.8, 12.4 Hz, 68H), 0.64 (dt, *J* = 19.9, 7.4 Hz, 32H).

Synthesis of DSY-C4: compound 6 (0.1g, 0.044mmol), compound C (0.08g, 0.348mmol) and Pyridine(1ml) were added to a flask with three necks under nitrogen, then CF (20ml) was injected. After stirring at 60°C for 6 hours, the mixture was deposit in CH₃OH to wipe off pyridine. After removal of the solvent, the residue was purified by silica gel chromatography (CF: hexane=3:1) to get a black solid (0.1g, 72%). ¹H NMR (400 MHz, Chloroform-*d*) δ 9.15 (s, 2H), 9.12 (s, 2H), 8.54 (dd, *J* = 10.0, 6.4 Hz, 2H), 8.46 (dd, *J* = 10.0, 6.4 Hz, 2H), 7.73-7.66 (m, 4H), 7.43 (d, *J* = 3.6 Hz, 2H), 7.35 (s, 2H), 7.34 (s, 2H), 7.13 (d, *J* = 3.6 Hz, 2H), 4.86-4.66 (m, 8H), 3.11 (t, *J* = 7.6 Hz, 4H), 2.72 (d, *J* = 6.8 Hz, 4H), 2.20-2.08 (m, 4H), 2.06-1.96 (m, 4H), 1.73-1.63 (m, 2H), 1.46-1.28 (m, 16H), 1.23 – 0.88 (m, 64H), 0.88-0.55 (m, 36H).

The synthesis of DSY-C6, DSY-C8 and DSY-C10 are same as that of DSY-C4.

Synthesis of DSY-C6: ¹H NMR (400 MHz, Chloroform-*d*) δ 9.13 (s, 2H), 9.12 (s, 2H), 8.56-8.46 (m, 4H), 7.73-7.67 (m, 4H), 7.40 (d, *J* = 3.6 Hz, 2H), 7.35 (s, 2H), 7.34 (s, 2H), 7.09 (d, *J* = 3.6 Hz, 2H), 4.84-4.66 (m, 8H), 3.04 (t, *J* = 7.5 Hz, 4H), 2.71 (d, *J* = 6.8 Hz, 4H), 2.18-2.08 (m, 4H), 1.95-1.85 (m, 4H), 1.72-1.64 (m, 2H), 1.45-1.29 (m, 16H), 1.15 – 0.88 (m, 68H), 0.88 – 0.58 (m, 36H).

Synthesis of DSY-C8: ¹H NMR (400 MHz, Chloroform-*d*) δ 9.13 (s, 2H), 9.10 (s, 2H), 8.55 − 8.50 (m, 4H), 7.73 − 7.68 (m, 4H), 7.38 (d, *J* = 3.6 Hz, 2H), 7.36 (s, 2H), 7.34 (s, 2H), 7.07 (d, *J* = 3.6 Hz, 2H), 4.85-4.70 (m, 8H), 3.01 (t, *J* = 7.6 Hz, 4H), 2.72 (d, *J* = 6.8 Hz, 4H), 2.25-2.05 (m, 4H), 1.95-1.80 (m, 4H), 1.73-1.68 (m, 2H), 1.41 − 1.32 (m, 16H), 1.15 − 0.88 (m, 72H), 0.88-0.50 (m, 36H).

Synthesis of DSY-C10: ¹H NMR (400 MHz, Chloroform-*d*) δ 9.12 (s, 2H), 9.05 (s, 2H), 8.53 (dd, *J* = 10.0, 6.4 Hz, 2H), 8.43 (dd, *J* = 10.0, 6.4 Hz, 2H), 7.74 – 7.66 (m, 4H), 7.49 (s, 2H), 7.37 (d, *J* = 3.2 Hz, 2H), 7.36 (s, 2H), 7.06 (d, *J* = 3.2 Hz, 2H), 4.84-4.74 (m, 4H), 4.74-4.64 (m, 4H), 3.00 (t, *J* = 7.8 Hz, 4H), 2.81 – 2.69 (m, 4H), 2.39-2.27 (m, 2H), 2.13-2.03 (m, 2H), 1.92-1.82 (m, 4H), 1.78-1.70 (m, 2H), 1.52 – 0.90 (m, 92H), 0.87-0.50 (m, 36H).

Characterization and measurements

¹H NMR spectra were recorded on Bruker (AVANCE III 400MHz). High resolution time of flight mass spectrometer (HR-TOF-MS) was obtained from AB Sciex (Triple TOF 4600). Differential scanning calorimetry (DSC) measurement was performed on TG-DSC (STA 449F3) under Ar atmosphere at a heating rate of 10 °C min⁻¹. Spectrophotometer (Perkin-Elm er Lambda 950) was taken to test the UV-vis absorption spectrums.

Photovoltaic performances of the OSCs were performed via the solar simulator (Newport-Oriel® Sol3A 450W). The intensity of the AM 1.5G spectra was calibrated by a certified standard silicon solar cell. AFM images were obtained from peak force quantitation nanomechanical mapping (PFQNM) and tapping mode by a Veeco Dimension 3100V atomic force microscope. Grazing incidence wide-angle X-ray scattering (GIWAXS) was measured to research the molecular packing, crystallinity, and mode direction of the films.

Radius of gyration

All GMAs were tested in o-xylene configured as a 5mg/ml solution. SAXS analyses were measured using a Xeuss 3.0 SAXS/WAXS system (Xenocs SA, France). A Cu Ka X-ray source (GeniX3D Cu ULD), operating at 50 kV and 0.6 mA, produced radiation with a wavelength of 1.5418. An Eiger 2R Hybrid pixelphoton counting detector (500K model, vacuum compatible, windowless) with a silicon325sensor at a thickness of 450 μ m and a resolution of 512 x 1028 pixels (pixel size= 75 x 75 μ m²) was used to collect the scattered signals. Each SAXS pattern was collected after a 5-minute exposure. One-dimensional intensity profiles were integrated from background-corrected 2D SAXS patterns. The R_g fitting equation is guinier.

Density functional theory (DFT) simulations

We have employed the Vienna Ab Initio Simulation Package (VASP)^[1-2] to perform all the density functional theory (DFT) calculations within the generalized gradient approximation (GGA) using the PBE^[3] formulation. We have chosen the projected augmented wave (PAW) potentials^[4-5] to describe the ionic cores and take valence electrons into account using a plane wave basis set with a kinetic energy cutoff of 400 eV. Partial occupancies of the Kohn–Sham orbitals were allowed using the Gaussian smearing method and a width of 0.05 eV. The electronic energy was considered self-consistent when the energy change was smaller than 10–5 eV. A geometry optimization was considered convergent when the force change was smaller than 0.02 eV/Å. Grimme's DFT-D3 methodology was used to describe the dispersion interactions. The C4-dimer was in a tetragonal box of 35 Å×35 Å×30 Å and the C10-dimer was in a tetragonal box of 42 Å×42 Å×30 Å. During structural optimizations, the Γ point in the Brillouin zone was used for k-point sampling, and all atoms were allowed to relax. Electrostatic potential (ESP) maps are calculated with Gaussian software after fixing the conformation of GMAs.

Device fabrication and characterization

The device was fabricated with a structure of Glass/ITO/2PACZ/active layer/PDINN/Ag. The ITO glass was cleaned with sequential ultrasonication in a distilled water, acetone and isopropanol. Then, the ITO glass was treated in an ultraviolet ozone box for 20 minutes. Subsequently, 2PACZ in ethanol solution was spin-coated on the treated ITO glass at 3000 rpm for 30 s and annealed at 100 °C for 20 minutes. Then, the substrates were transferred into a nitrogen-filled glove box. A chloroform solution containing the PM6 and acceptors was prepared to spin coating for fabricating active layer. The total concentration was 15.4 mg/mL and the donor/acceptor ratio was kept as 1:1.2. And the active layer was annealed at 95 °C for 10 minutes. Subsequently, PDINN was spin-coated on the active layer at 2000 rpm for 30s. Finally, Ag was deposited about 150 nm under the pressure of 3×10⁻⁵ Pa. Photovoltaic properties of the device were measured under simulated solar light (100 mW/cm² AM 1.5G) provided by Newport-Oriel® Sol3A 450W solar simulator.

The flexible OSCs was fabricated with PET/ITO/PEDOT:PSS/active layer /PDINN/Ag. The flexible electrode (ITO/PET) was adhered onto polydimethylsiloxane (PDMS)-coated glass substrates, and then the normal spin coating process of the device is performed. The PEDOT:PSS (Clevios P VP 4083) solution was then spin-coated at 2500 rpm for 30 s and annealed on a hot stage at 115°C for 20 min. After that, the process conditions are the same as those of the rigid device.

Cyclic voltammetry (CV)

The energy levels for DSY-C4, DSY-C6, DSY-C8 and DSY-C10 were measured by cyclic voltammetry (CV) using a PC controlled 604E electrochemical workstation, which was performed by using Ag/AgCl as reference electrode in 0.1 mol L⁻¹ tetra-*n*-butylammonium hexafluorophosphate (n-Bu4NPF₆) solution, and ferrocene/ferrocenium (Fc/Fc⁺) (-0.1 eV versus Ag/AgCl) was used as internal reference. The CV system was constructed using a Pt disk as the working electrode, a Pt wire as the counter electrode, and an Ag/AgNO₃ (0.1 mol L⁻¹ in acetonitrile) electrode as the reference electrode. The HOMO and LUMO were calculated according to the following equations:

$$E_{\rm HOMO/LUMO} = -(\varphi_{\rm ox/red} - \varphi_{\rm Fc/Fc^+} + 4.8) \, \rm eV$$

Where φ_{ox} is the onset of oxidation and the φ_{red} relates to the reduction potential, respectively.





Figure S1 ¹H NMR spectra of Compound 2 in CDCl₃.



Figure S2 ¹H NMR spectra of Compound 3 in CDCl₃



Figure S3 ¹H NMR spectra of Compound 4 in CDCl₃



Figure S4 ¹H NMR spectra of Compound 5 in CDCl₃



Figure S5 ¹H NMR spectra of Compound 6 in CDCl₃



Figure S6 ¹H NMR spectra of DSY-C4 in CDCl₃



Figure S7 NOESY spectra of DSY-C4 in CDCl₃



Figure S8 Mass spectrum of DSY-C4 in CDCl3



Figure S9 ¹H NMR spectra of DSY-C6 in CDCl₃



Figure S10 NOESY spectra of DSY-C6 in CDCl₃.



Figure S11 Mass spectrum of DSY-C6 in CDCl3



Figure S12 ¹H NMR spectra of DSY-C8 in CDCl₃



Figure S13 NOESY spectra of DSY-C8 in CDCl₃.



Figure S14 Mass spectrum of DSY-C8 in CDCl₃



Figure S15 ¹H NMR spectra of DSY-C10 in CDCl₃



Figure S16 NOESY spectra of DSY-C10 in CDCl₃.



Figure S17 Mass spectrum of DSY-C10 in CDCl3



Figure S18 DFT simulation calculation results of DSY-C6.



Figure S19 DFT simulation calculation results of DSY-C8.



Figure S20 Standard concentration solution fitting curve of GMAs.



Figure S21 Absorption spectra of blend solutions (in CF) under different temperatures.



Figure S22 Absorption spectra of DSY-C4 (in CF) under different temperatures.





Figure S24 (a) The films absorption and fluorescence emission spectra of GMAs. (b) The solution absorption and fluorescence emission spectra of GMAs.



Figure S25 Electrochemical cyclic voltammograms of GMAs.





BTP-eC9



Figure S26 Materials used in this article.



Figure S27 *J*–*V* characteristics of the ternary device.

PHOTOVOLTAIC AND WIND POWER SYSTEMS QUALITY TEST CENTER, IEE, CHINESE ACADEMY OF SCIENCES

Report No: PWQC-WT-P25022621-1R



End of Report –



Figure S28. PCE certified at Institute of Electrical Engineering, Chinese Academy of Sciences, showing an efficiency of 19.39% (Report No: PWQC-WT-P25022621-1R).



Figure S29 Electron mobility curves measured by SCLC method.



Figure \$30 Hole mobility curves measured by SCLC method.



Figure S31 P dependence on J_{SC} of devices.



Figure S32 P dependence on V_{OC} of devices.



Figure S33 EQE curves of the flexible device.



Figure S34 PCEs, COS_{FOW} and EDF of flexible devices (Efficiency Deformation Factor (EDF), defined as the product of PCE and the crack-onset strain (COS_{FOW}) value. The PCE is derived from the blend film-based flexible device, while COS_{FOW} is obtained using the films-on-water (FOW) method. EDF=PCE×COS_{FOW}).



Figure S35 Bending test of flexible devices.



Figure S36 Crack-onset strain (COS) on the film on water (FOW) test of PM6:DSY-C4 and PM6:DSY-C10..



Figure S37 Contact angle tests of PM6 and GMAs.



Figure S38 2D spectra of in-situ PL for blend films.



Figure S39 1D spectra of in-situ PL for blend films.



Figure S40 Mechanical properties of PM6:BTP-eC9:DSY-C10.

	λ _{peak,s} [nm]	λ _{onset,s} [nm]	λ _{peak,f} [nm]	λ _{onset,} f [nm]	E ^{opt} [eV]	Е _{номо} [eV]	E _{LUMO} [eV]	E ^{cV} [eV]	Rg [Å]	PL _f [nm]	PL, [nm]
DSY-C4	734	788	797	892	1.57	-5.54	-3.95	1.59	40.36±1.38	972	779
JSY-C6	734	788	804	893	1.57	-5.58	-3.98	1.60	44.12±1.68	960	779
82-YS	733	786	805	894	1.58	-5.59	-3.97	1.62	58.55±5.93	947	775
DSY- C10	735	796	812	895	1.56	-5.54	-3.97	1.57	70.81±4.69	952	775

Supplementary Tables

Absorption	0	5mg/ml	10mg/ml	15mg/ml	Saturated solution	Solubility (mg/ml)
DSY-C4	9.54×10 ⁻⁴	0.286	0.523	0.793	0.554	10.421
DSY-C6	9.54×10 ⁻⁴	0.258	0.492	0.774	0.521	10.237
DSY-C8	9.54×10 ⁻⁴	0.198	0.430	0.621	1.171	12.258
DSY-C10	9.54×10 ⁻⁴	0.220	0.481	0.753	0.958	19.303

Table S2 Absorption of GMAs of different concentration.

Table S3 Normalized absorption of GMAs under different temperatures.

Normalized Absorption	20°C	30°C	40°C	50°C	60°C
DSY-C4	1	0.975	0.952	0.926	0.898
DSY-C6	1	0.976	0.951	0.923	0.898
DSY-C8	1	0.986	0.968	0.946	0.914
DSY-C10	1	1.001	1.011	1.005	0.993

Table S4 Detailed GIWAXs parameters of neat films and blend films.

	π - π stacking (OOP)							
Films	qz	d	FWHM	CCL				
	(Å-1)	(Å)	(Å-1)	(Å)				
DSY-C4	1.590	3.952	0.6043	9.67				
DSY-C6	1.592	3.947	0.5989	9.76				
DSY-C8	1.619	3.881	0.5334	10.95				
DSY-C10	1.625	3.867	0.5167	11.31				
PM6: DSY-C4	1.636	3.841	0.4738	12.33				
PM6: DSY-C6	1.643	3.824	0.4724	12.37				
PM6: DSY-C8	1.656	3.794	0.4658	12.54				
PM6: DSY-C10	1.669	3.765	0.4417	13.23				

Tuble Se culler mounty of obes.							
Active layer	$\mu_{\rm h}~({\rm cm}^2{\rm V}^{-1}{\rm s}^{-1})$	$\mu_{\rm e}~({\rm cm}^2{\rm V}^{-1}{\rm s}^{-1})$	$\mu_{ m e}/\mu_{ m h}$				
PM6: DSY-C4	6.19×10 ⁻⁴	5.45×10 ⁻⁴	0.88				
PM6: DSY-C6	6.01×10 ⁻⁴	5.78×10 ⁻⁴	0.96				
PM6: DSY-C8	5.96×10 ⁻⁴	5.78×10 ⁻⁴	0.97				
PM6: DSY-C10	6.03×10 ⁻⁴	6.17×10 ⁻⁴	1.02				

Table S5 Carrier mobility of OSCs.

 Table S6 Photovoltaic parameters of flexible OSCs PCEs based on different active layer systems.

Active layer	Voc [V]	Jsc [mA cm ⁻²]	FF	PCE _{max} [%]	COS _{FOW} [%]
PM6: BTP-eC9	0.841	27.24	0.750	17.20	5.30
PM6: DSY-C10	0.865	25.74	0.765	17.04	9.95
PM6: PY-IT	0.934	24.69	0.708	16.34	10.79

 Table S7 Contact angles of PM6 and GMAs.

Surface	θ _{Water}	θ Diiodomethane	γ ^d	γ ^p	γ	D-A a) X	D-Ab) γ
	ľ	[^o]	[mN/m]	[mN/m]	[mN/m]	[K]	[mN/m]
PM6	102.47	59.99	28,78	0.24	29.02		
DSY-C4	94.86	45.00	36.91	0.48	37.39	0.53	1.058
DSY-C6	93.33	44.97	36.51	0.72	37.23	0.51	1.018
DSY-C8	92.58	44.99	36.29	0.86	37.16	0.50	1.001
DSY-C10	90.01	45.02	35.58	1.43	37.01	0.49	0.965

Year	Active layer	PCE	COS _{FOW} ^{a)}	EDF	Refer.
		(%)	(%)	(%)	
2020	PM6:Y6	12.69	5.75	0.73	[6]
	PM6:Y6 (30% PAEF)	12.28	25.07	3.08	
2021	PBDB-T:PYTS-0.0	10.12	18.84	1.90	[7]
	PBDB-T:PYTS-0.3	11.04	21.6	2.38	
2021	PM6-C0:Y7	10.29	2.33	0.24	[8]
	PM6-C5:Y7	11.64	12.09	1.42	
	PBDB-T:PYFS-Reg	10.64	22.4	2.38	
2022	PM6:BTP-eC9	10.59	2.99	0.32	[9]
	PM6-OEG5:BTP-eC9	12.05	10.50	1.27	
2022	PhAm5:Y7	12.73	13.8	1.76	[10]
2023	PM7:L8-BO	11.28	2.6	0.29	[11]
	PM7-Thy10:L8-BO	13.69	13.7	1.88	
2023	PM6:L8-BO	11.21	2.29	0.26	[12]
	PM6- <i>b</i> -PDMS _{5k} :L8-BO	12.01	4.30	0.52	
	PM6-b-PDMS _{12k} :L8-BO	11.48	8.13	0.93	
	PM6- <i>b</i> -PDMS _{19k} :L8-BO	11.34	18.14	2.06	
2023	PM6:PY-IT	12.86	9.7	1.25	[13]
	PM6-A:PYTCl-A	13.81	20.1	2.78	
2023	PM6:L8-BO	15.54	5.67	0.88	[14]
	PM6:L8-BO: 5wt% SEPS	15.09	13.5	2.04	
	PM6:L8-BO: 10wt% SEPS	14.53	16.1	2.34	
	PM6:L8-BO:BTP-eC9:5wt% SEPS	15.71	14.0	2.20	
	PM6:L8-BO:BTP-eC9:10wt% SEPS	15.26	16.0	2.44	
2024	D18:MYT	12.19	1.3	0.16	[15]
	D18:TYT-L	13.10	6.4	0.84	
	D18:TYT-S	14.37	21.6	3.10	
2024	P1 _{0.8} /P2 _{0.2} : Y6	15.66	8.01	1.25	[16]
	P1 _{0.8} /P2 _{0.2} -TCl: Y6	17.03	9.09	1.55	
2024	D18 _{0.8} - <i>r</i> -PEHDT _{0.2} : L8-BO	10.97	10.4	1.14	[17]
	D180.8-s-PEHDT0.2: L8-BO	14.31	17.2	2.46	
2024	PM6:BTP-eC9	17.20	5.30	0.91	This
	PM6:DSY-C10	17.04	9.95	1.70	work
	PM6:PY-IT	16.34	10.79	1.76	

Table S8 Comparisons of PCE and cos values of plastic substrate-based flexible binary devices in the recent years.

^{a)} The COS value tested obtained by film on water (FOW) method;

^{b)} EDF (efficiency deformation factor) is defined as PCE×COS_{FOW}, where the PCE is obtained from the corresponding blend film-based flexible device and COS_{FOW} is obtained from the FOW method.

Supplemental References

- [1] G. Kresse, J. Furthmüller, *Computational Materials Science* **1996**, *6*, 15-50.
- [2] G. Kresse, J. Furthmüller, *Physical Review B* 1996, 54, 11169-11186.
- [3] J. P. Perdew, K. Burke, M. Ernzerhof, *Physical Review Letters* **1996**, 77, 3865-3868.
- [4] G. Kresse, D. Joubert, *Physical Review B* **1999**, *59*, 1758-1775.
- [5] P. E. Blöchl, *Physical Review B* **1994**, *50*, 17953-17979.
- [6] J. Han, F. Bao, D. Huang, X. Wang, C. Yang, R. Yang, X. Jian, J. Wang, X. Bao, J. Chu, Adv. Funct. Mater. 2020, 30, 2003654.
- [7] Z. Genene, J. W. Lee, S. W. Lee, Q. Chen, Z. Tan, B. A. Abdulahi, D. Yu, T. S. Kim, B. J. Kim, E. Wang, *Adv. Mater.* **2022**, *34*, e2107361.
- [8] J.-W. Lee, D. Jeong, D. J. Kim, T. N.-L. Phan, J. S. Park, T.-S. Kim, B. J. Kim, *Energy Environ. Sci.* 2021, 14, 4067-4076.
- [9] J. W. Lee, C. Lim, S. W. Lee, Y. Jeon, S. Lee, T. S. Kim, J. Y. Lee, B. J. Kim, *Adv. Energy Mater.* 2022, 12, 2202224.
- [10] J. W. Lee, S. Seo, S. W. Lee, G. U. Kim, S. Han, T. N. Phan, S. Lee, S. Li, T. S. Kim, J. Y. Lee, B. J. Kim, *Adv. Mater.* **2022**, *34*, e2207544.
- [11] Q. Wan, S. Seo, S. W. Lee, J. Lee, H. Jeon, T. S. Kim, B. J. Kim, B. C. Thompson, J. Am. Chem. Soc. 2023, 145, 11914-11920.
- [12] S. Seo, J. W. Lee, D. J. Kim, D. Lee, T. N. Phan, J. Park, Z. Tan, S. Cho, T. S. Kim, B. J. Kim, Adv. Mater. 2023, 35, e2300230.
- J. Liu, J. Deng, Y. Zhu, X. Geng, L. Zhang, S. Y. Jeong, D. Zhou, H. Y. Woo,
 D. Chen, F. Wu, L. Chen, *Adv. Mater.* 2023, *35*, e2208008.
- [14] X. Zheng, X. Wu, Q. Wu, Y. Han, G. Ding, Y. Wang, Y. Kong, T. Chen, M. Wang, Y. Zhang, J. Xue, W. Fu, Q. Luo, C. Ma, W. Ma, L. Zuo, M. Shi, H. Chen, *Adv. Mater.* **2023**, *36*, e2307280.
- [15] J. W. Lee, C. Sun, J. Lee, D. J. Kim, W. J. Kang, S. Lee, D. Kim, J. Park, T. N. L. Phan, Z. Tan, F. S. Kim, J. Y. Lee, X. Bao, T. S. Kim, Y. H. Kim, B. J. Kim, *Adv. Energy Mater.* 2024, *14*, 2303872.
- [16] C. Lin, R. Peng, W. Song, Z. Chen, T. Feng, D. Sun, Y. Bai, Z. Ge, Angew. Chem. Int. Ed. Engl. 2024, 63, e202407040.
- [17] J.-W. Lee, H.-G. Lee, E. S. Oh, S.-W. Lee, T. N.-L. Phan, S. Li, T.-S. Kim, B. J. Kim, *Joule* 2024, 8, 204-223.