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Supporting Information

Achieving Small Non-Radiative Energy Loss Through Synergically Non-Fullerene Electron Acceptor Selection and Side Chain Engineering in Benzo[1,2-b:4,5-b']difuran Polymer-Based Organic Solar Cells

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Experimental Section

Device Fabrication: The BDF based devices were fabricated with the structure of ITO/ZnO/active layer/MoO_x/Ag in a N2-filled glovebox and characterized in the air without encapsulation. The ITO (150 nm)-coated glass substrates were cleaned with isopropyl alcohol, dishwashing liquid, deionized water, ethanol, acetone, and isopropanol for 30 min, respectively. After being dried, the ZnO precursor was spin-coated onto the cleaned ITO glass at 4000 rpm/min for 30s and annealed in the titanium plate under 250 ^{o}C for 30 mins. The resulting ZnO was 30 nm thick. The active layer was spin-coated in a N_2-filled glovebox from a solution of polymer:*m*-ITIC with 1:1.5 weight ratio in chlorobenzene. The resulting active layer was 100 nm thick. Then, we tried different annealing temperature of 100-150 ^{o}C for 2min and different additives, finally, we found 130 ^{o}C for 2min without additives achieved the highest PCEs. For BDFs: Y6 based devices, we refer to the literature that has reported that Y6 is used as the receptor and use similar optimization conditions to obtain the best efficiency. The active layer was spin-coated in a N_2-filled glovebox from a solution of BDFs:Y6 with 1: 1.2 weight ratio in chloroform and maintain a concentration of 14mg/ml with 0.5% 2-chloronaphthalene (CN) as solvent additives. Subsequently, the active layers were undergone thermal annealing treatment at 130 ^{o}C for 2 min. Finally, 1.5 nm MoO_x and100 nm Ag were sequentially evaporated on the active layer in the vacuum chamber under a pressure of ca. 2.5×10^{-4} Pa. The effective area of one cell is 4 mm^2.

Measurement: All the measurements were carried out under ambient condition. The current-voltage (J-V) characteristics of the devices were measured on a Keithley 2400

Source Measure Unit. The power conversion efficiency (PCE) was measured under an illumination of AM 1.5G (100 mW/cm^2) using a 7IS0503A (SOFN Instruments Co. Ltd.) solar simulator. The EQE was measured by 7-SCSpec (SOFN Instruments Co. Ltd.) which light intensity at each wavelength was calibrated with a standard single-crystal Si photovoltaic cell. The morphologies of the polymer/acceptor were investigated by AFM (Dimension Fastscan) in contacting under normal air conditions at room temperature with 2μm scanner.

Electroluminescence measurements were done using a source meter (Keithley 2400) to inject electric current, and the emitted photons were measured using a fluorescence spectrometer (KYMERA-3281-B2, Andor) with two sets of diffraction gratings, a Si EMCCD camera (DU970PBVF, Andor) for the wavelength range of 400~1000 nm, and an InGaAs camera (DU491A-1.7, Andor) for the wavelength range of 900~1600 nm. Photoluminescence measurements were done using a laser excitation (460 nm), and the emission spectra were obtained using the same setup used for recording electroluminescence spectra. The transient photovoltage decay measurements were done using two white LED lamps, driven by a Keithley 2450 source meter for different bias illumination intensities and an arbitrary function generator (AFG3022C, Tektronix) for the transient illumination. The peaks of the transient voltage signals, recorded by an oscilloscope (MDO4104C, Tektronix), were kept to approximated 5% of the DC bias photovoltage signal, by adjusting the driving voltage of the LED controlled by the function generator for each bias illumination intensity, and the record transient photovoltage decay signals were fitted using an exponential decay function for determining the decay time constants.

The GIWAXS measurements were carried out with a Xeuss 2.0 SAXS/WAXS laboratory beamline using a Cu Kα X-ray source (8.05 eV, 1.54 Å) and a Pilatus 3r 300 K detector. The incident angle was 0.2°.

Calculation of bandgaps of polymers and acceptors: The optical bandgap (E_g^{opt}) can be calculated by the intersection of the linear fitting curve of the absorption edge and the abscissa axis (or the tangent of absorption tail). The photovoltaic bandgap (Eg^{PV}) is the bandgap determined from the derivatives of the EQE^{PV} spectra. The edge bandgap (Eg^{edge}) is given by the crossing point between the EQE^{PV} spectra edge and the horizontal tangent of the EQE^{PV} peak.

Calculation of energy levels: The electrochemical properties of P-FT and P-FP are investigated by cyclic voltammetry (CV) measurements. According to the equation $E_{HOMO} = -e(V_{ox} + 4.80 - V_{ferro})$. According to the reduction onsets and calibration of the ferrocene/ferrocenium (Fc/Fc+) redox couple, the lowest unoccupied molecular orbitals (LUMOs) of these BDF based polymers are estimated to be -3.48 and -3.61eV, then the HOMO energy levels also can be calculated as -5.48 and -5.54eV.

Calculation of Voc^{SQ}:

$$qV_{oc}^{SQ} = \frac{kT}{q} ln \left\{ \frac{q.\int_{Eg}^{+\infty} \emptyset_{AM1.5G}(E).dE}{q.\int_{Eg}^{+\infty} \emptyset_{BB}(E).dE} \right\}$$

Calculation of interaction parameter: The estimation for $\chi_{D,A}$ can be derived using the empirical equation , where K is a constant while γ_D and γ_A are the surface energies of the polymer and acceptor films, respectively.

$$\chi_{DA} = K(\sqrt{\gamma_D} - \sqrt{\gamma_A})^2$$

Surface energies could be resolved into nonpolar (dispersion) and polar components.

$$\gamma=\gamma^d+\gamma^p$$

Furthermore, the dispersive and polar surface tension can be calculated based on contact angles obtained by the water and glycerol, as shown below

$$(1 + \cos\theta)\gamma_L = \frac{4\gamma_s^d\gamma_L^d}{\gamma_s^d + \gamma_L^d} + \frac{4\gamma_s^p\gamma_L^p}{\gamma_s^p + \gamma_L^p}$$

where θ is the contact angle of a specific solvent, γ_L represents the surface energy of the solvent, γ_s^d and γ_s^p refer to the dispersive and polar surface energy of the solid, respectively, and γ_L^d and γ_L^p represent dispersive and polar surface energy of the liquid solvent, respectively. Thus, the unknown γ_s^d and γ_s^p are obtained by determining contact angles of diiodomethane and water, respectively.

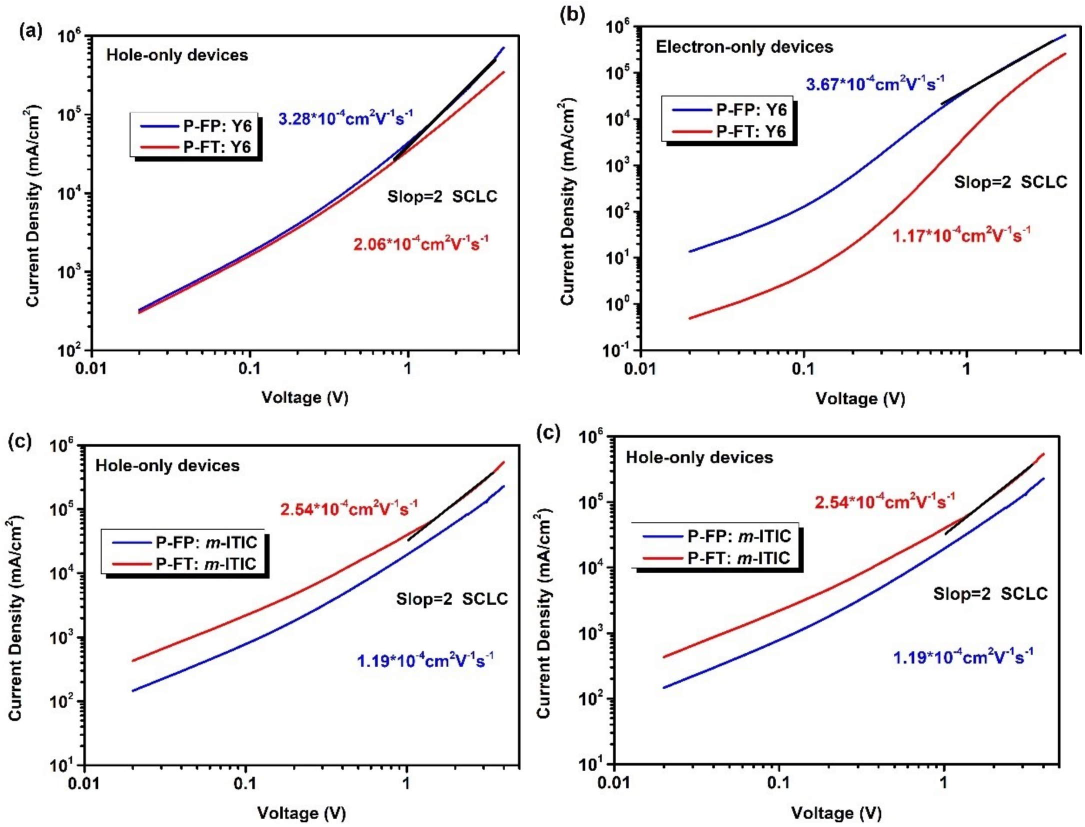

Figure S1. SCLC characteristics of the hole-only and electron-only devices.

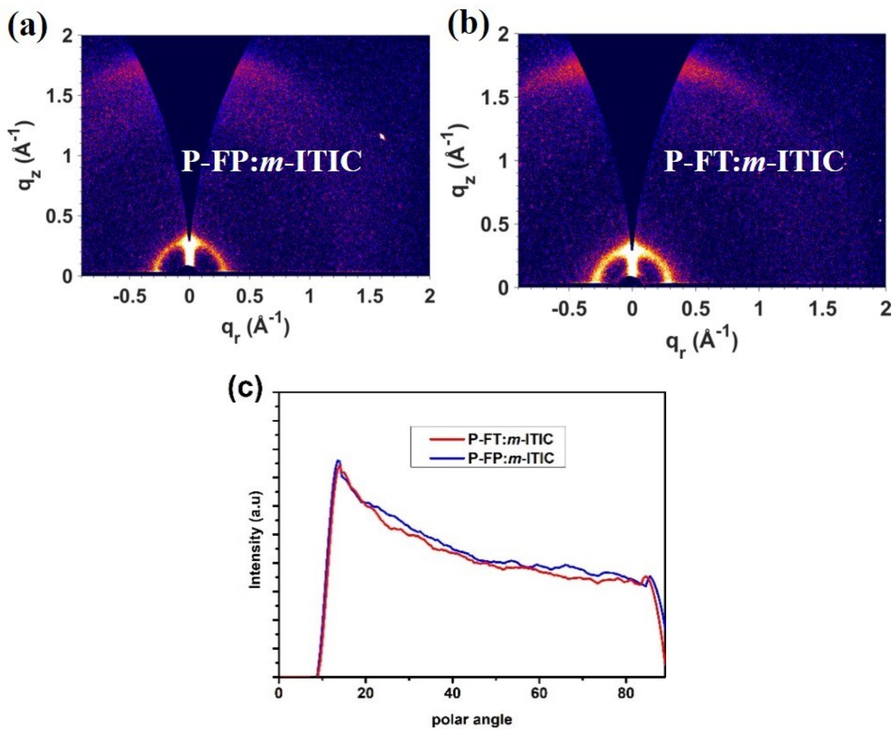

Figure S2. The 2D GIWAXS patterns of P-FT: *m*-ITIC and P-FP:*m*-ITIC (a, b), and corresponding linecuts of (010) diffraction peaks along polar angle(c).

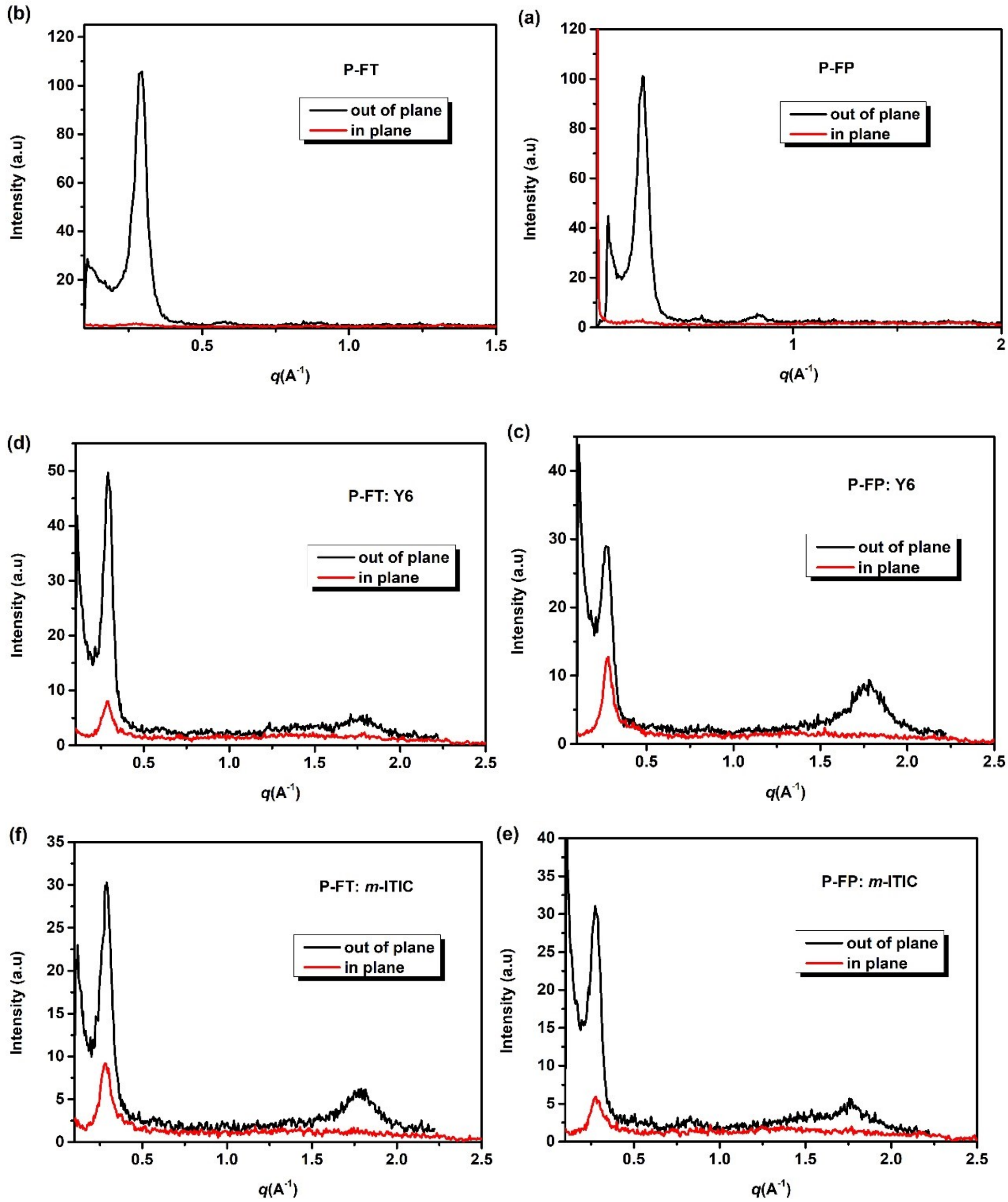

Figure S3. GIWAXS linecuts of the a) neat P-FT film, b) neat P-FP film, c) P-FT:Y6 blend film, d) P-FP:Y6 blend film, e) P-FT: *m*-ITIC and f) P-FP:*m*-ITIC blend film, along in-plane and out-of-plane directions, respectively.

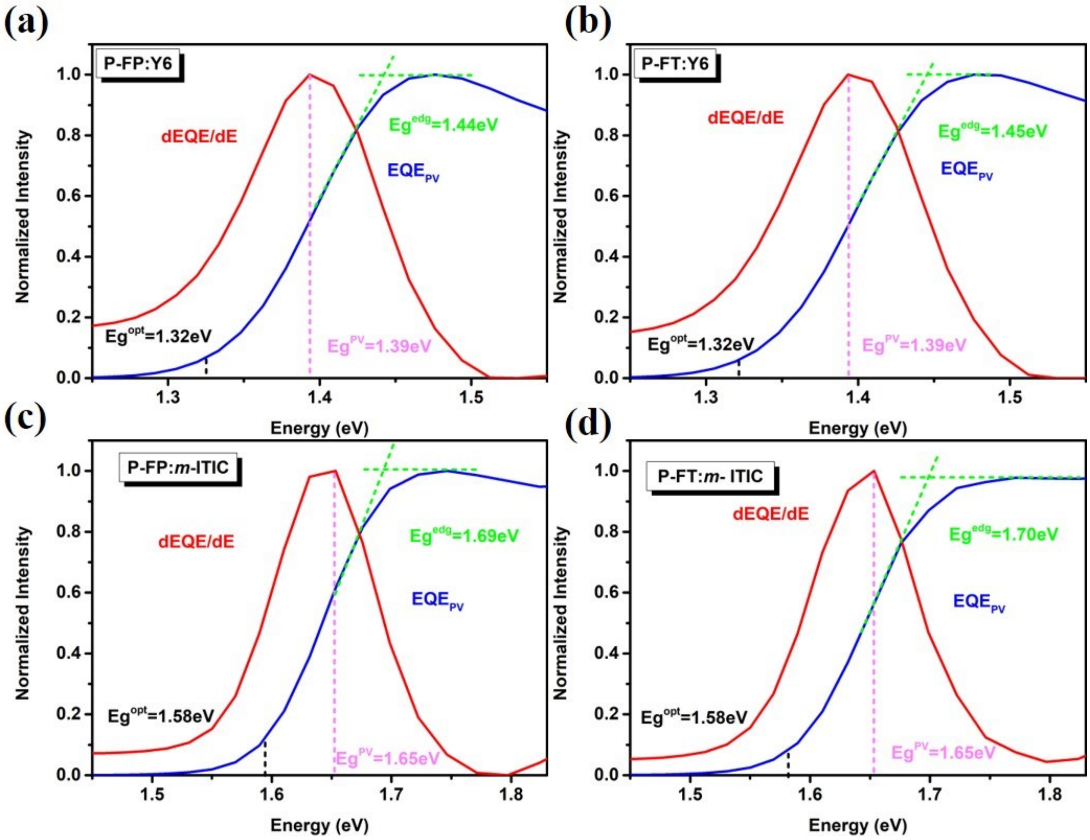

Figure S4. E_g determination for photoactive layers prepared under different condition using different methods. (a-c). P-FP, P-FT:Y6; d-f) P-FP, P-FT :*m*-ITIC. onset E_g is the optical gap determined from the absorption onset, while E_g^{PV} is the optical gap determined from the derivatives of the EQE spectra and edge E_g is given by the crossing point between the EQE spectra edge and horizontal tangent of EQE peak.

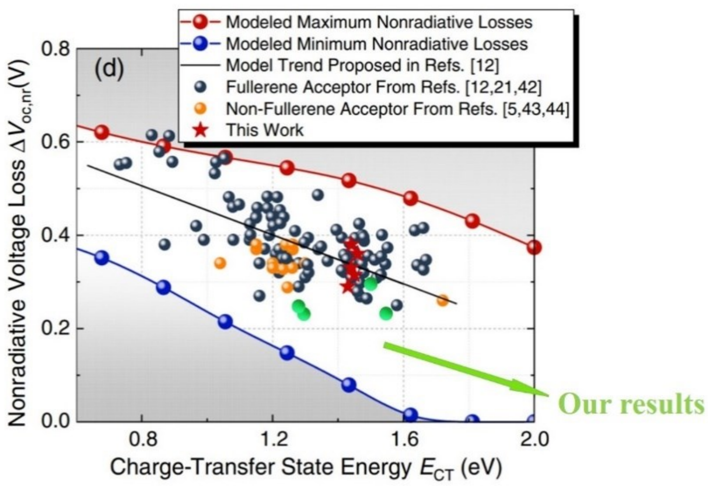

Figure S5. The results of our study in comparison with the predicted nonradiative voltage losses obtained using the proposed model by Nelson and coworkers. The figure is duplicated from the literature (*Phys. Rev. X.* **2018,** *8*, 031055.) with our results marked by green circles.

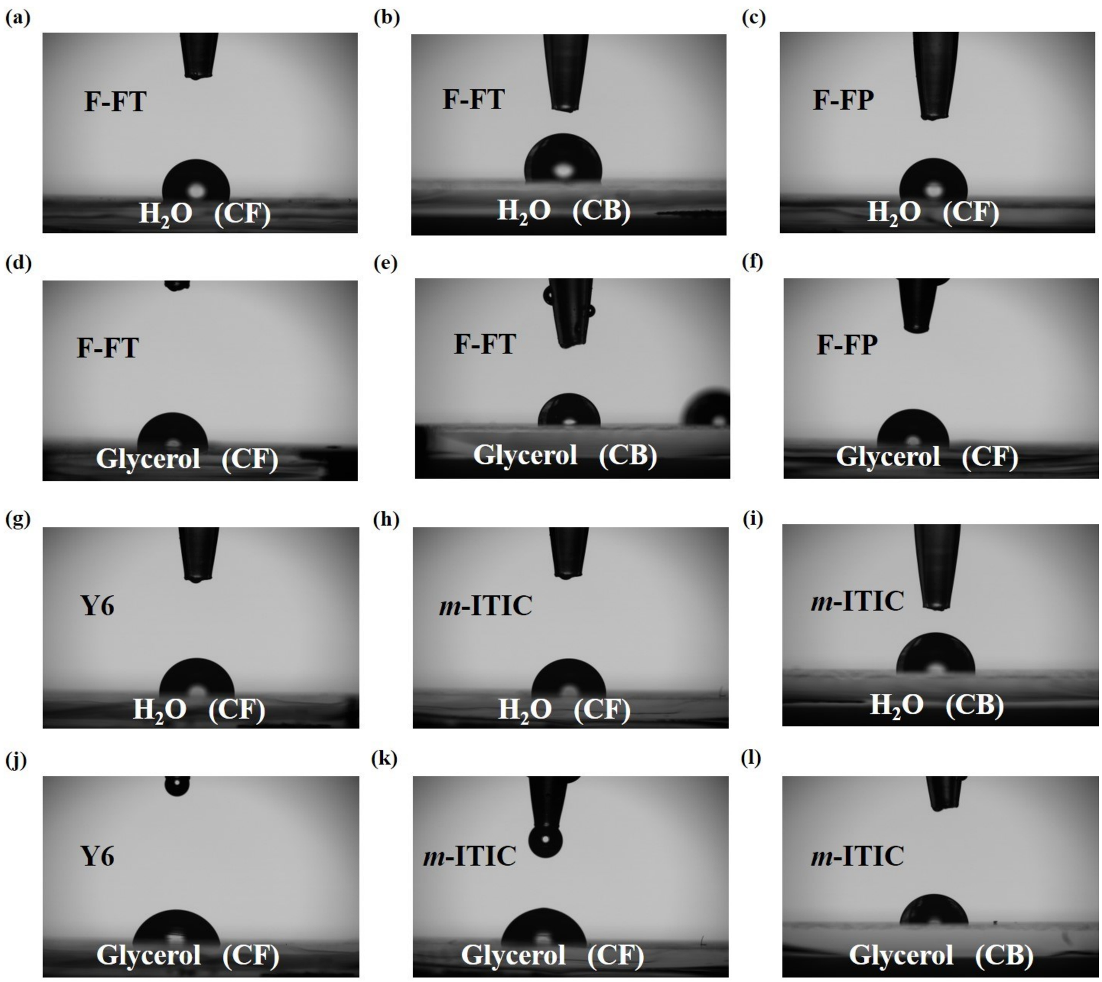

Figure S6. Water and glycerol contact angle images of (a,d) P-FT, (c,f) P-FP, (g,j) Y6 and (h,k) *m*-ITICand (b,e) P-FT, (i,l) m-ITIC casting from CB, respectively.

Table S1. Summary of the E_g determination.

Polymer	Acceptor	E_g^{onset} (eV)	E_g^{PV} (eV)	E_g^{edge} (eV)
P-FP	Y6	1.32	1.39	1.44
	m-ITIC	1.58	1.65	1.69
P-FT	Y6	1.32	1.39	1.45

m-ITIC	1.58	1.65	1.70

Table S2. Contact angles and surface energy parameters of the polymer and acceptor films.

surface	θ_{water}	$\theta_{glycerol}$	γ_s^d	γ_s^p	γ
P-FT(CF)	106.40	106.02	2.82	13.32	16.14
P-FT(CB)	110.08	96.00	18.40	2.05	20.45
P-FP	108.00	97.01	13.95	4.42	18.37
Y6	93.10	79.40	18.79	8.28	27.07
m-ITIC(CF)	99.60	79.99	27.38	3.22	30.6
m-ITIC(CB)	97.13	82.65	19.44	6.37	25.81

Table S3. The interaction parameter of blend films.

Polymer	Acceptor	χ_{DA}
P-FP	Y6	0.84
	m-ITIC	1.55
P-FT	Y6	1.41
	m-ITIC	0.31

Table S4. Summary for E_{loss}, PCE and EQE_{max} of BDT and other polymers based OSCs.

Number	E_{loss}(eV)	PCE(%)	EQE_{max}(%)	Reference
1	0.52	10.6	68	1
2	0.68	11.2	75	2
3	0.65	12.1	78	3
4	0.64	11.3	75	3
5	0.64	11.4	75	4
6	0.7	10.4	75	5
7	0.68	10.4	74	6
8	0.62	12.1	74	7
9	0.65	11.2	72	8
10	0.68	12.1	77	9
11	0.67	11.8	79	10
12	0.66	11.6	79	11
13	0.64	12.1	77	11

14	0.62	10.2	72	11
15	0.61	10.7	72	12
16	0.62	11.1	78	12
17	0.67	10.1	66	12
18	0.71	10.1	74	13
19	0.63	11	77	14
20	0.59	11.2	79	15
21	0.62	10.1	70	16
22	0.65	10.8	76	16
23	0.68	11.5	77	16
24	0.65	11.9	79	17
25	0.66	10.8	78	17
26	0.63	11.3	81	18
27	0.63	12.2	74	19
28	0.58	11.9	81	20
29	0.57	10	73	21
30	0.58	10.1	74	22
31	0.55	10.3	70	22
32	0.66	13.1	83	23
33	0.68	10.1	72	24
34	0.54	10.8	78	25
35	0.54	11.7	73	26
36	0.57	10.5	62	27
37	0.63	13.7	75	27
38	0.74	10.74	79	28
39	0.8	10.41	72	28
40	0.74	11.2	79	28
41	0.63	10.9	80	29
42	0.52	10.2	66	29
43	0.75	13.2	79	30
44	0.64	14.4	83	30
45	0.6	14.2	81	31
46	0.45	10.2	69	32
47	0.53	10.46	67	33
48	0.58	12.16	81	34
49	0.56	11.7	79	35
50	0.47	12.8	80	35
51	0.53	10	76	36
52	0.47	11.6	71	37
53	0.67	10.1	78	38
54	0.67	10.08	77	39
55	0.63	11.8	72	40
56	0.87	10.3	78	41

57	0.88	10.8	87	42
58	0.66	11.47	80	43
59	0.65	12.1	86	44
60	0.55	13.63	79	45
61	0.5	13.04	76	46
62	0.52	12.5	78	47
63	0.67	10.5	75.8	48
64	0.54	10.1	71	48
65	0.62	10.3	71	49
66	0.63	11.6	80	50
67	0.5	12.1	78	51
68	0.57	12.2	75.7	52
69	0.53	11.3	73	52
70	0.56	11.2	70	52
71	0.59	12.7	73	53
72	0.59	12.6	72.2	54
73	0.57	10.6	63.7	54
74	0.52	10.3	53.7	54
75	0.59	10.4	68	55
76	0.52	11.1	75	56
77	0.69	12	80	57
78	0.71	11.5	80	57
79	0.71	10.9	80	57
80	0.67	11	75	58
81	0.64	11.7	72	59
82	0.54	12.8	81.7	60
83	0.55	11.7	76	60
84	0.67	10.08	76	61
85	0.62	10.26	77	61
86	0.76	12.5	79	62
87	0.7	12.1	80	62
88	0.67	10.7	72.5	63
89	0.7	12.1	80	64
90	0.54	10.5	81	65
91	0.63	12.5	83.5	66
92	0.61	12.5	81	66
93	0.59	11.5	66	67
94	0.61	13.1	80	68
95	0.74	10.6	80	69
96	0.59	11.9	83	70
97	0.71	10.2	80	70
98	0.64	13	80	71
99	0.55	14.1	79	72

100	0.7	13.3	83.1	72
101	0.71	10.9	76	73
102	0.68	14.04	83.5	74
103	0.57	13.8	80	75
104	0.49	13.7	80	76
105	0.78	11.3	79	77
106	0.65	11.3	72	78
107	0.81	11.7	85	79
108	0.62	10.1	72.5	80
109	0.79	11.5	73	81
110	0.67	10.88	72	82
111	0.72	11.03	81	83
112	0.65	10.16	72	84
113	0.6	11.63	77	85
114	0.67	11.47	77	86
115	0.7	11.12	80	87
116	0.55	12.17	76	88
117	0.82	10.33	80	89
118	0.6	11	85	90
119	0.55	10.21	68	91
120	0.81	10.4	76	92
121	0.55	10.1	65	93
122	0.6	8.6	70	94
123	0.752	13.17	80	95
124	0.59	14.3	81	96
125	0.47	7.66	58	97
126	0.721	13.03	81	98
127	0.49	15.43	80	99
128	0.89	8.77	73	100
129	0.57	11.48	73	101
130	0.689	14.16	77	102
131	0.662	13.68	72	102
132	0.51	13.9	77	103
133	0.75	12.88	79	104
134	0.72	13.54	79	104
135	0.552	16.2	86	105
136	0.537	17.52	86	105
137	0.56	13.15	82	106
138	0.55	13.75	75	106
139	0.52	12.06	72	107
140	0.758	7.66	78	108
141	0.53	16.52	81.5	109
142	0.6	14.2	81.5	110

143	0.542	17.53	82	[111]
144	0.49	10.05	65	[112]
145	0.789	11.26	80	[113]
146	0.715	12.1	80	[113]
147	0.55	10.2	72	[114]
148	0.62	13.4	82	[115]
149	0.76	11.47	81	[116]
150	0.526	13.6	79	[117]
151	0.575	12.86	72	[117]
152	0.52	10.12	65	[118]
153	0.67	10.4	79	[119]
154	0.63	13.1	78	120
155	0.64	11.1	77	[120]
156	0.57	13.42	71	[121]
157	0.57	13.4	74	[121]
158	0.564	16.5	82	[122]
159	0.559	15.5	82	[122]
160	0.78	10.2	76	[123]
161	0.67	10.48	76	[124]
162	0.7	10.4	77	[125]
164	0.58	10.97	78	[126]
165	0.6	11	83	[127]
166	0.82	11.1	82	[128]
167	0.49	11.1	80	[129]
168	0.81	10.16	82	[130]
169	0.63	10.48	75	[131]
170	0.83	10.5	78	[132]
171	0.78	10.1	82	[133]
172	0.7	10.05	71	[134]
173	0.81	10.72	80	[135]
174	0.55	16.5	84	[136]
175	0.549	16.21	81	[137]
176	0.56	15.7	86	[138]
177	0.53	16.4	81.5	[139]
178	0.55	16.67	82	[140]
179	0.67	14.7	86	[141]
180	0.55	13.44	78	[142]
181	0.42	15.37	78	[143]
182	0.56	13.1	79	[144]
183	0.63	10.28	74	[145]
184	0.713	14	82	[146]
185	0.721	12.72	81	[146]
186	0.57	12.93	83.01	[147]

187	0.907	11.13	76	[148]
188	0.822	10.57	74	[148]
189	0.815	10.23	74	[148]
190	0.69	12.42	81.6	[149]

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