Soluble Poly(4-fluorostyrene): a High-performance Dielectric Electret for Organic Transistors and Memories

Supporting Information

Table S1. Dielectric polymers and their related OFET performances

Ref.	Dielectric/Semiconductor	Mobility ($cm^2 \cdot s^{-1} \cdot V^{-1}$)	I_{on}/I_{off}	Mem. W. (V)
[29]	SiO_2/pentacene	0.23	10^6	N/A
[29]	PVA/pentacene	0.07	10^5	N/A
[17]	PS/pentacene	0.71	5×10^4	4.3
[24]	a-PMMA/pentacene	0.38	$\sim10^7$	8
[S1]	Triple-layer PVP/pentacene	0.24	10^3	11
[29]	PVP/pentacene	0.21	10^5	17
[40]	PS/pentacene	0.02	N/A	19.4
[32]	PFPS-b-40/pentacene	0.28	10^6	20.9
[29]	PVPyr/pentacene	0.12	10^5	21
[29]	PS/pentacene	0.26	10^5	22.1
[29]	P4MS/pentacene	0.25	10^5	24.1
[29]	PaMS/pentacene	0.35	10^5	26
[29]	PVN/pentacene	0.61	10^6	27.8
[25]	PI(6FOH-ODPA)/1-aminopyrene/pentacene	0.09	10^5	29.9
[44]	PS-DPP-n-I/pentacene	0.22	3.2×10^5	32.7
[40]	P3TFMS/pentacene	0.03	N/A	33.1
[32]	PFBT-10/pentacene	0.35	10^6	34.8
[S2]	IPPA-CL/TIPS-pentacene	0.09	10^6	40.8
[22]	poly(4-vinylpyridine)-Phenol/pentacene	0.16	2.9×10^7	43
[41]	WG3/pentacene	0.33	10^6	45
[22]	poly(4-vinylpyridine)-2-Naphthol/pentacene	0.23	6.2×10^7	61
[S3]	PS/pentacene	0.6	10^5	70
[46]	P(St-Fl)3/pentacene	0.47	3×10^7	76
[22]	poly(4-vinylpyridine)-2-hydroxyanthracene/ pentacene	0.12	10^7	76
[39]	PI(BAPF-6FDA)/pentacene	0.73	10^6	77
[S4]	PI(6FDA-TPA-CN)/pentacene	5.0×10^{-4}	3×10^6	84
[3]	PaMS/pentacene	0.51	10^5	90
[26]	8-3-NTCDI/SiO_2	0.12	10^6	4.4
[73]	CYTOP/C_{60}	0.40	10^5	10
[S5]	PTCDI/PI-B	0.31	10^5	15.5
[25]	N(PTPMA)3/BPE-PTCDI	0.03	3.1×10^5	33.5
[23]	PI(APSP-6FDA)/BPE-PTCDI	3.6×10^{-3}	8.7×10^3	63
[42]	PVTT/BPE-PTCDI	7.1×10^{-5}	3.6×10^3	81

Table S2. Memory window, retention time, and charge density of electret in OFETs with various gate dielectric materials. No. of Ref. refers to Fig. 4e.

No.	Ref.	Dielectric/Semiconductor	Mem.W. (V)	Retention (s)	Charge density of electret (10^{12} cm^{-2})
1	[25]	TIPS-pen:N(PTPMA)$_3$/BPE-PTCDI	30	5.0E3	2.05
2	[S2]	PPA/TIPS-pentacene	15	4.5E3	0.92
3	[S2]	PPA-Cl/TIPS-pentacene	11.1	6.2E3	0.62
4	[S2]	PPA-NO_2/TIPS-pentacene	14.6	5.2E3	0.88
5	[S2]	IPPA/TIPS-pentacene	33.4	1.2E4	1.12
6	[S2]	IPPA-Cl/TIPS-pentacene	40.8	4.0E3	2.66
7	[S2]	IPPA-NO_2/TIPS-pentacene	25.5	2.0E4	1.09
8	[29]	PS/pentacene	4.3	5.2E4	0.88
9	[29]	PaMS/pentacene	26	2.2E4	5.35
10	[29]	PVN/pentacene	27.8	3.4E4	5.73
11	[29]	PVPyr/pentacene	21	4.2E3	4.33
12	[29]	P4MS/pentacene	24.1	2.1E4	3.82
13	[29]	PVP/pentacene	17	2.4E3	2.69
14	[32]	PFBT-10/pentacene	34.9	1.0E4	4.81
15	[73]	PaMS/pentacene	90	3.6E5	2.81
16	[S4]	Phenol/pentacene	43	1.2E4	2.44
17	[28]	PMAA–GO1.5/TIPS-pentacene	5.9	2.0E4	1.36
18	[28]	PMAA–GO6/TIPS-pentacene	9.4	2.0E3	2.07
19	[S6]	PI(6FOH-ODPA)/pentacene	29.9	1.0E4	4.61
20	[46]	P(St-Fl)/pentacene	99	1.0E4	2.95
21	[45]	PPF/pentacene	70	8.0E2	4.81
22	[45]	PPF8/pentacene	55	1.2E3	3.77
23	[22]	PI(6FDA-TPA-CN)/pentacene	84	1.0E4	5.70
24	[39]	PI(BAPF-6FDA)/pentacene	77	1.0E4	5.30
25	[S7]	PVK/pentacene	40	1.0E3	2.74
26	[23]	PI(APSP-6FDA)/pentacene	63	3.0E3	4.33
27	[S8]	star-PTPMA/BPE-PTCDI	36.5	1.5E5	2.18
28	[S3]	PS with Au nanoparticles /pentacene	70	5.0E5	2.00
29	[44]	PS-DPP-n-I/pentacene	32.7	2.0E5	2.24
30	[42]	PVTT/BPE-PTCDI	81	1.0E5	4.21
31	[41]	WG3/pentacene	45	2.0E3	2.97

Table S3. Performance of C12-BTBT/FPS and C12-BTBT/PS OFET devices

	C12-BTBT/FPS	C12-BTBT/PS
μ_{max}	11.2	13.8
$\mu_{average}$	10.3	11.9
I_{on}/I_{off} (average)	~1×10^{7}	~3×10^{6}
$V_{th\text{-}average}$	-13	-24

Table S4. Performance of PCDTPT/FPS and PCDTPT/PS OFET devices

Device structure	PCDTPT/FPS	PCDTPT/PS
μ_{max}	0.36	0.39
$\mu_{average}$	0.33	0.31
I_{on}/I_{off} (average)	~5×10^{5}	~8×10^{5}
$V_{th\text{-}average}$	-12	-8

Table S5. Relaxation processes with activation energies of FPS and PS, according to Fig. 3a-3f

Relaxation	Mechanism	FPS	PS
α (high temp.)	Backbone relaxation	1.40 eV	1.79 eV
β (middle temp.)	Side chain relaxation	0.75 eV	0.39 eV
γ (low temp.)	Dipolar polarization	0.41 eV	N/A

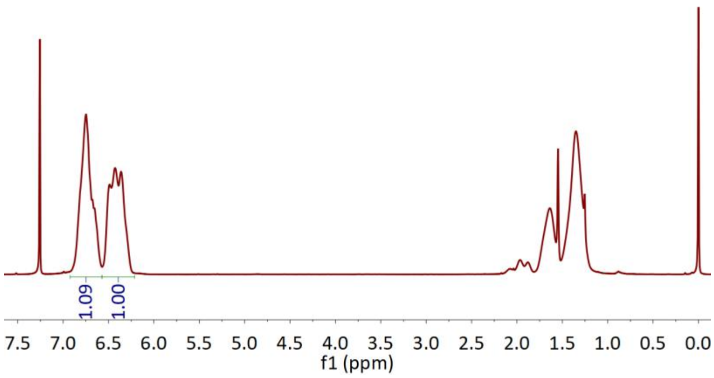
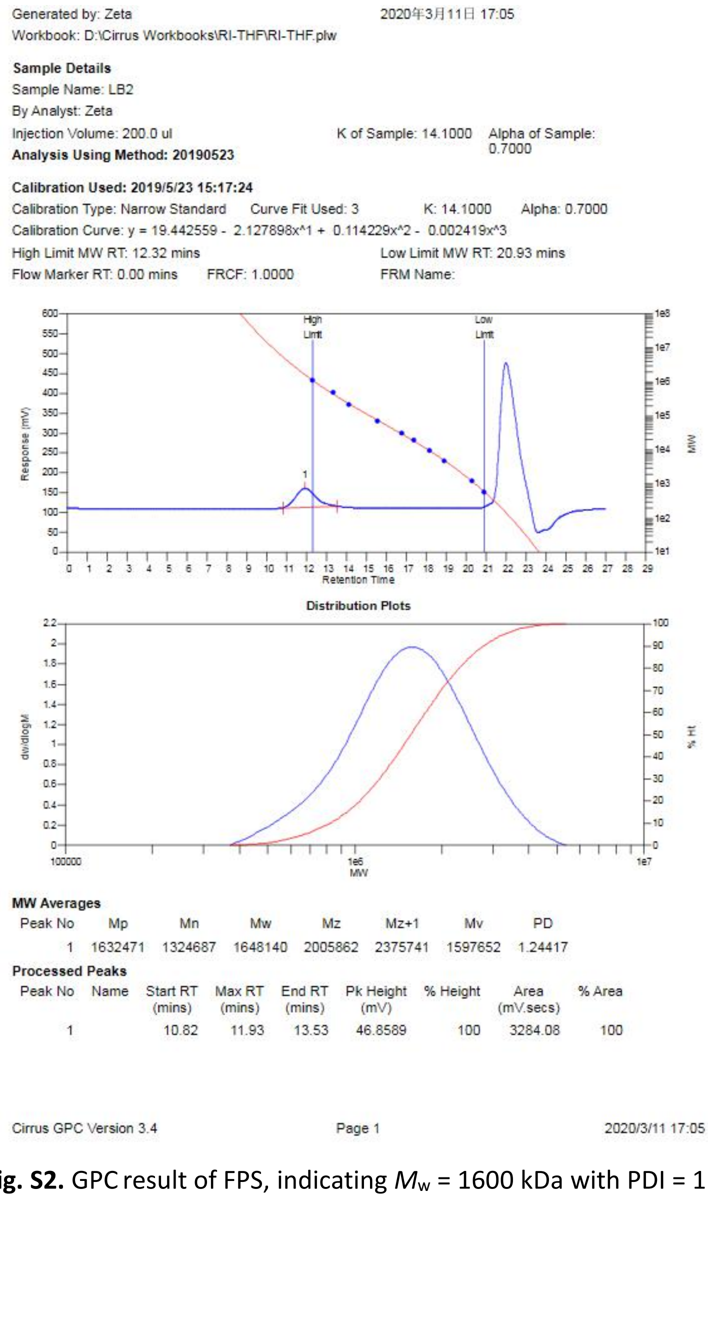


Fig. S1. The 1H NMR spectrum of poly(4-fluorostyrene). The multi peaks at *δ* 6.93 – 6.58 (a) and *δ* 6.40 – 6.57 (b) assigned to hydrogens (2H) at the benzene near and away from the fluorine respectively.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Sample Details
Sample Name: LB2
By Analyst: Zeta
Injection Volume: 200.0 ul K of Sample: 14.1000 Alpha of Sample: 0.7000
Analysis Using Method: 20190523

Calibration Used: 2019/5/23 15:17:24
Calibration Type: Narrow Standard Curve Fit Used: 3 K: 14.1000 Alpha: 0.7000
Calibration Curve: y = 19.442559 - 2.127898x^1 + 0.114229x^2 - 0.002419x^3
High Limit MW RT: 12.32 mins Low Limit MW RT: 20.93 mins
Flow Marker RT: 0.00 mins FRCF: 1.0000 FRM Name:

MW Averages

Peak No	Mp	Mn	Mw	Mz	Mz+1	Mv	PD
1	1632471	1324687	1648140	2005862	2375741	1597652	1.24417

Processed Peaks

Peak No	Name	Start RT (mins)	Max RT (mins)	End RT (mins)	Pk Height (mV)	% Height	Area (mV.secs)	% Area
1		10.82	11.93	13.53	46.8589	100	3284.08	100

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Fig. S2. GPC result of FPS, indicating M_w = 1600 kDa with PDI = 1.25

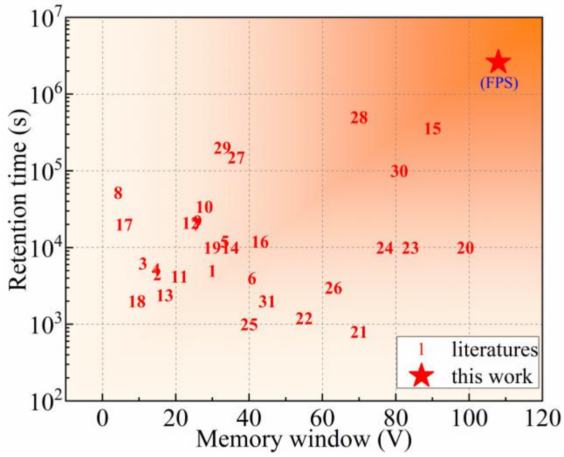

Fig. S3. Memory window and retention time of OFET performances for different polymeric gate dielectric materials. The reference numbers are provided in Table S2.

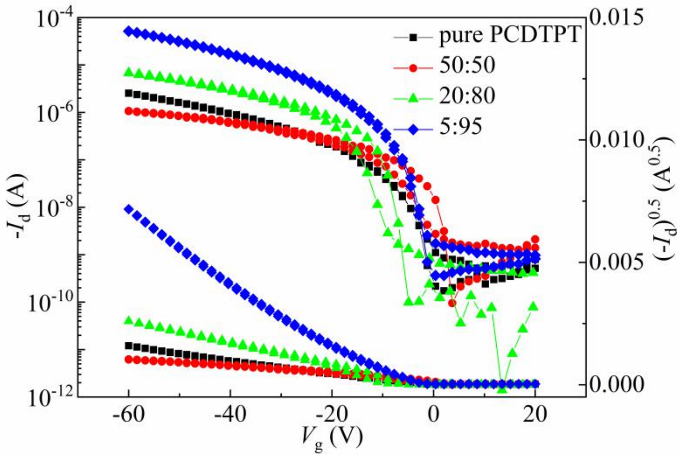

Fig. S4. The transfer characteristics of PCDTPT/PS OFETs with the content ratios (PCDTPT:PS wt%) of a) pure PCDTPT b) 50:50 c) 20:80 and d) 5:95.

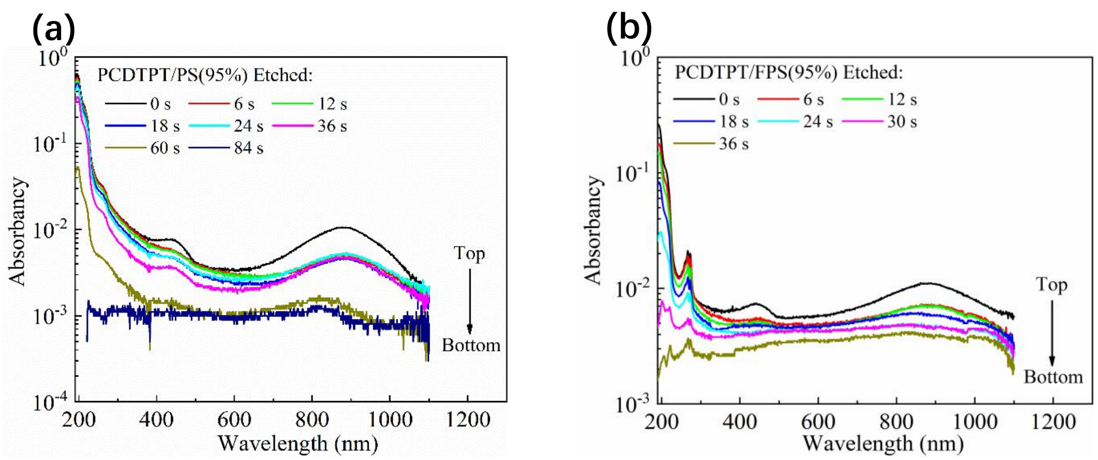

Fig. S5. Light absorbance of (a) PCDTPT/PS (95%) (b) PCDTPT/FPS (95%) blend films during oxygen plasma etching (~ 30 Pa). After oxygen plasma treatment (≥ 6 s), the evolution of the absorption peaks of PCDTPT (450 nm and 900 nm) decrease with substantially unchanged absorption peaks of PS and FPS (~190 nm), which implies that PCDTPT is partly enriched at the top sublayers of the blend film. However, there are still some minor PCDTPT molecules in the PS matrix.

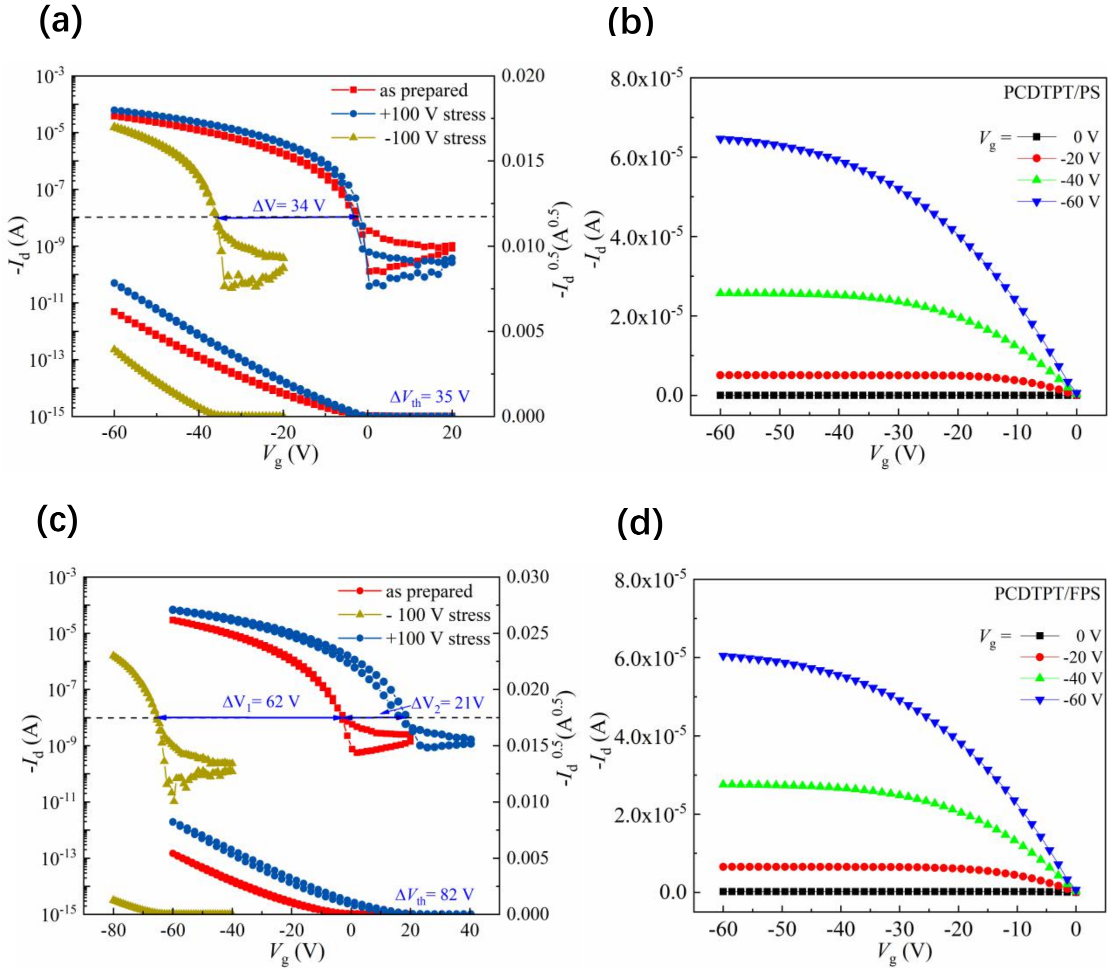

Fig. S6. OFET performances of PCDTPT/PS and PCDTPT/FPS (a) Transfer characteristics of PCDTPT/PS OFET with $\pm$100 V and without gate stresses, (b) Output characteristics of an as-prepared PCDTPT/PS OFET. (c) Transfer characteristics of PCDTPT/FPS OFET with $\pm$100 V and without gate stresses. (d) Output characteristics of an as-prepared PCDTPT/FPS OFET.

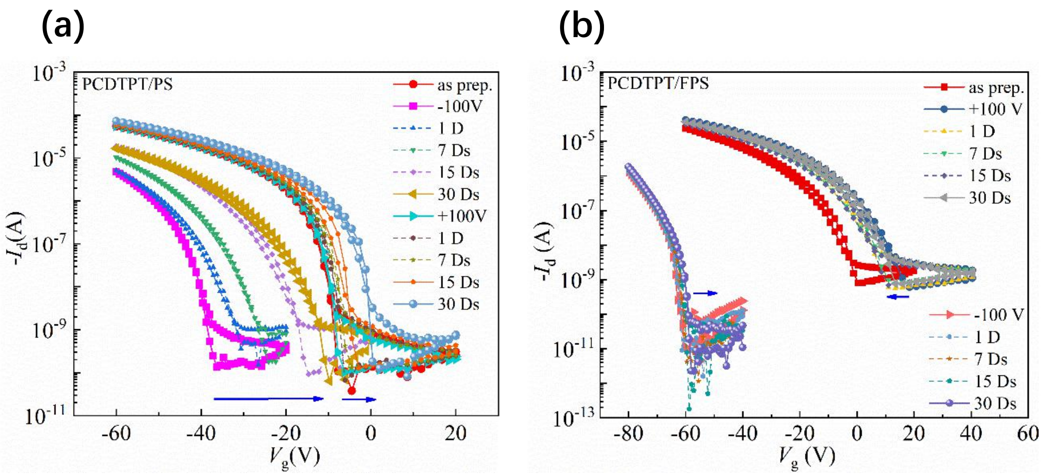

Fig. S7. Ambient memory stabilities of (a) PCDTPT/PS and (b) PCDTPT/FPS OFET devices with positive and negative electrets in one month.

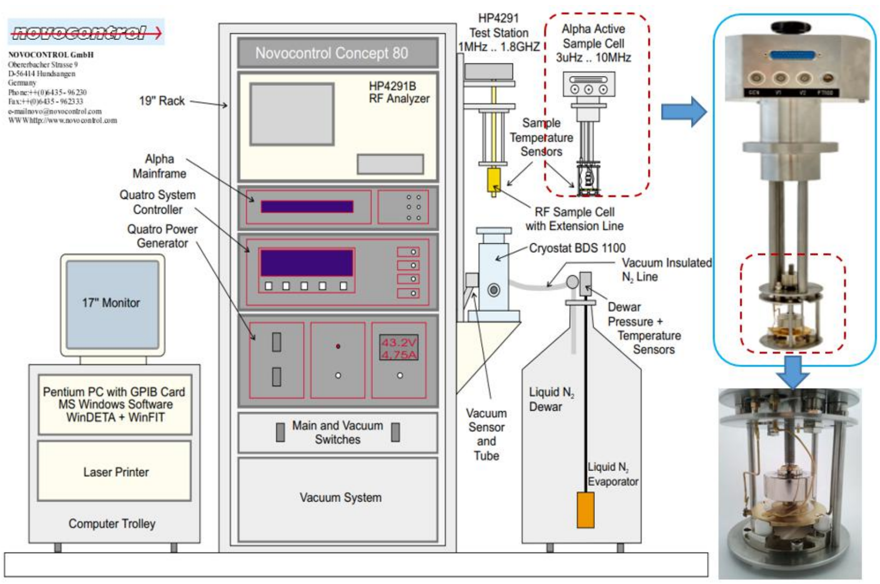

Fig. S8. Schematic of Novo control Concept 80 testing system and Alpha –A sample cell for dielectric characterization

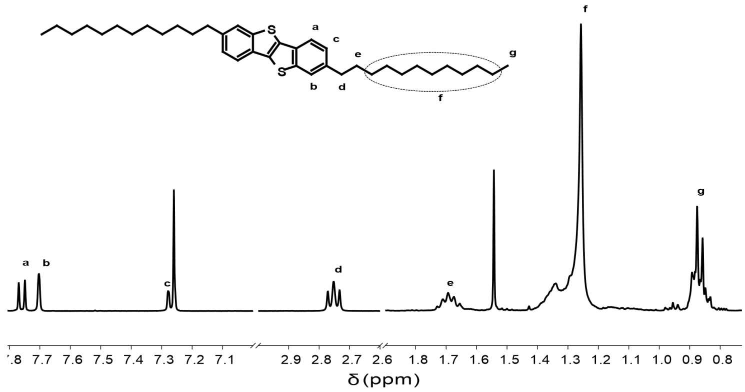

Fig. S9. ^{1}H NMR spectrum of C12-BTBT

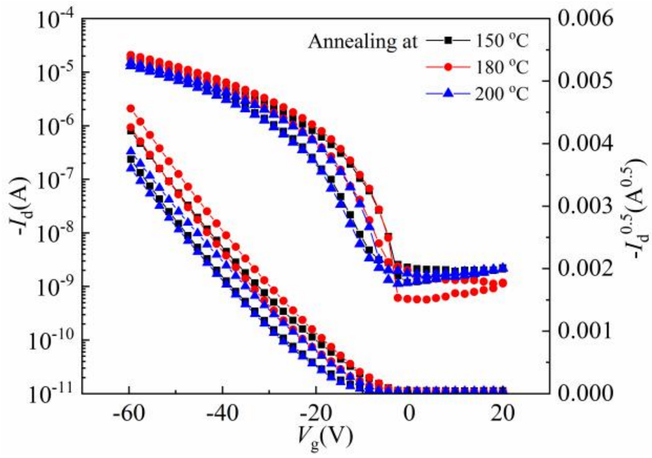

Fig. S10. Transfer characteristics of PCDTPT/PS (95%) OFETs with annealing temperatures of 150 °C, 180 °C, and 200 °C respectively, showing 180 °C is the best selected annealing temperature with improved device performance.

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