Supporting Information

A Facile Membraneless Method for Detecting Alkali-Metal Cations Using Organic Electrochemical Transistors

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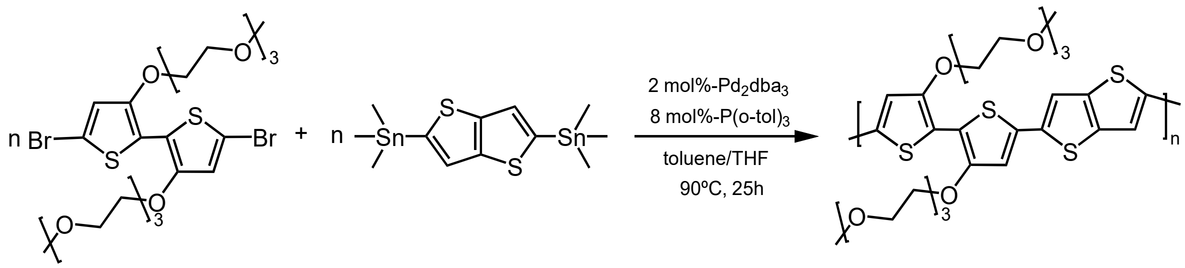

Scheme S1 The synthesis of the polymer p(g2T-TT).[S1]

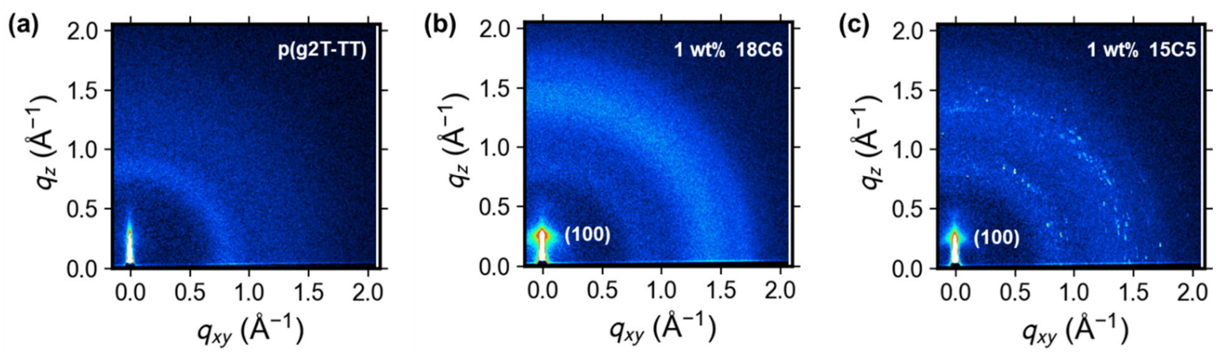

Fig. S1 Comparison of grazing incidence wide-angle X-ray scattering (GIWAXS) 2D patterns of three polymer thin films without annealing: (a) p(g2T-TT), (b) 1wt% 18C6-mixed p(g2T-TT), and (c) 1wt% 15C5-mixed p(g2T-TT).

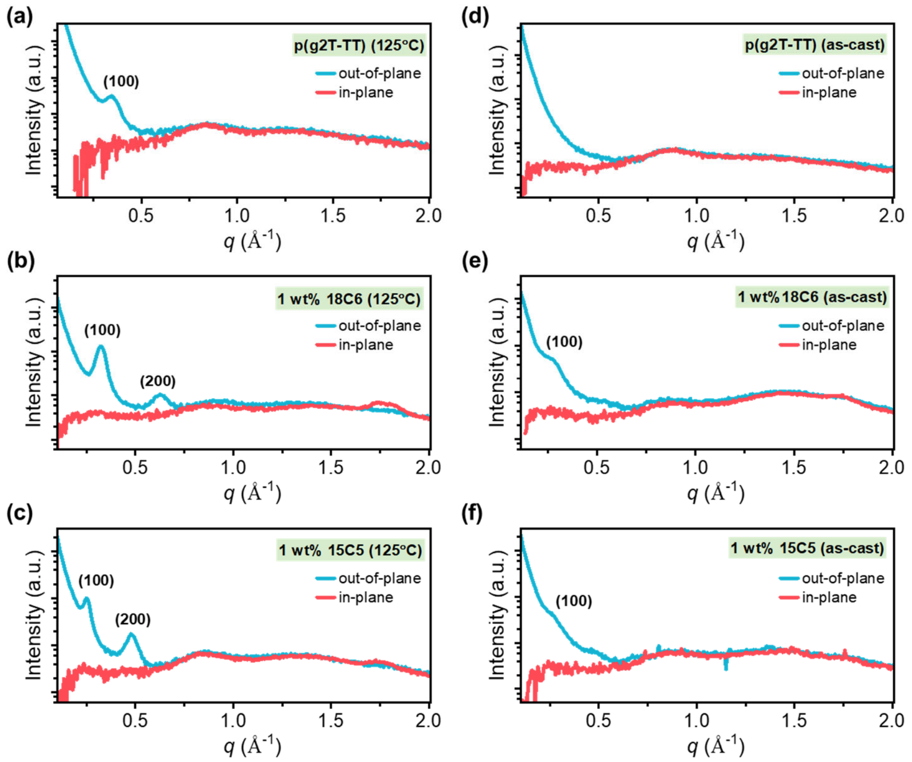

Fig. S2 The corresponding 1D profiles from 2D GIWAXS patterns (–90°±10° for out-of-plane; –5°±5° and –175°±5° for in-plane) of (a, d) p(g2T-TT), (b, e) 1wt% 18C6-mixed p(g2T-TT) and (c, f) 1wt% 15C5-mixed p(g2T-TT) films (a-c) before annealing or (d-f) after annealing at 125 °C for 10 min.

Table S1. Extracted peak values and calculated parameters from 1D GIWAXS profiles

$T_{annealing}$ (°C)	Materials	lamellar spacing (100)				lamellar spacing (200)			
		q (Å^{-1})	d-spacing[a] (Å)	FWHM[b] (Å^{-1})	L_c (Å)	q (Å^{-1})	d-spacing[a] (Å)	FWHM[b] (Å^{-1})	L_c (Å)
125°C	p(g2T-TT)	0.338	18.59	0.10	56.5	--	--	--	--
	1wt% 18C6	0.323	19.45	0.05	113.1	0.627	10.02	0.07	80.8
	1wt% 15C5	0.252	24.93	0.04	141.4	0.479	13.12	0.07	80.8
As-cast	p(g2T-TT)	--	--	--	--	--	--	--	--
	1wt% 18C6	0.246	25.54	0.13	43.5	--	--	--	--
	1wt% 15C5	0.258	24.35	0.10	56.5	--	--	--	--

[a] d-spacing = 2π/q; [b] L_c = 2πK/FWHM (K = 0.9).

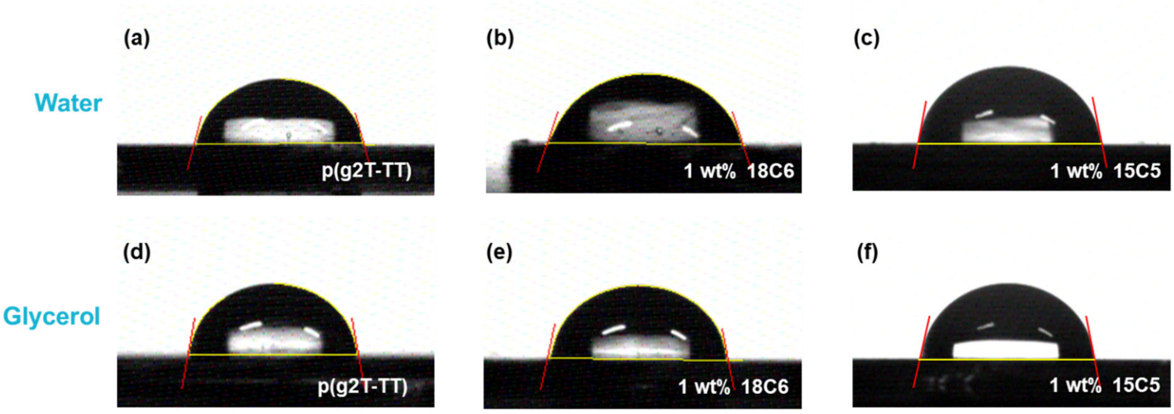

Fig. S3 Contact angles of polymer thin films on Si^{++}/SiO_2 substrate in air: (a, d) p(g2T-TT), (b, e) 1wt% 18C6-mixed p(g2T-TT), and (c, f) 1wt% 15C5-mixed p(g2T-TT).

Table S2. Contact angles and surface energy of the polymer thin films with water and glycerol drop

Materials	θ_{water} (°)	$\theta_{glycerol}$ (°)	γ (mN m^{-1})
p(g2T-TT)	74.1	77.5	31.2
18C6-mixed p(g2T-TT)	68.8	77.7	40.4
15C5-mixed p(g2T-TT)	86.5	79.1	22.4

Table S3. Summary of average threshold voltage (V_{th}), transconductance (g_m), and μC^* value in OECT.

Materials	p(2gT-TT)	18C6-mixed p(2gT-TT)	15C5-mixed p(2gT-TT)	p(2gT-TT)	18C6-mixed p(2gT-TT)	15C5-mixed p(2gT-TT)
	Na^+			K^+		
V_{th} (V)	−0.53 ± 0.067	−0.24 ± 0.048	−0.11 ± 0.054	−0.51 ± 0.065	−0.22 ± 0.068	−0.069 ± 0.079
g_m (mS)	0.62 ± 0.014	0.33 ± 0.022	0.37 ± 0.024	0.63 ± 0.028	0.37 ± 0.024	0.40 ± 0.019
μC^* (F cm^{-1} V^{-1} s^{-1})	24.23 ± 2.21	10.51 ± 0.92	13.99 ± 1.69	23.69 ± 2.36	15.21± 0.55	15.05 ± 1.17

Table S4. Summary of the ion sensitivity, linear range of detection, and limit of detection (LOD) in OECTs

Active Materials	Cations	I_D Sensitivity (μA dec^{-1})	Linear range (M)	LOD (M)
p(g2T-TT)	Na^+	10.63	$10^{-4} \sim 1$	1.65×10^{-5}
	K^+	10.45	$10^{-4} \sim 1$	3.34×10^{-5}
18C6-mixed p(2gT-TT)	Na^+	7.84	$10^{-3} \sim 1$	4.06×10^{-4}
	K^+	12.46	$10^{-4} \sim 1$	6.22×10^{-5}
15C5-mixed p(2gT-TT)	Na^+	10.3	$10^{-4} \sim 1$	1.81×10^{-4}
	K^+	13.9	$10^{-4} \sim 1$	9.93×10^{-5}

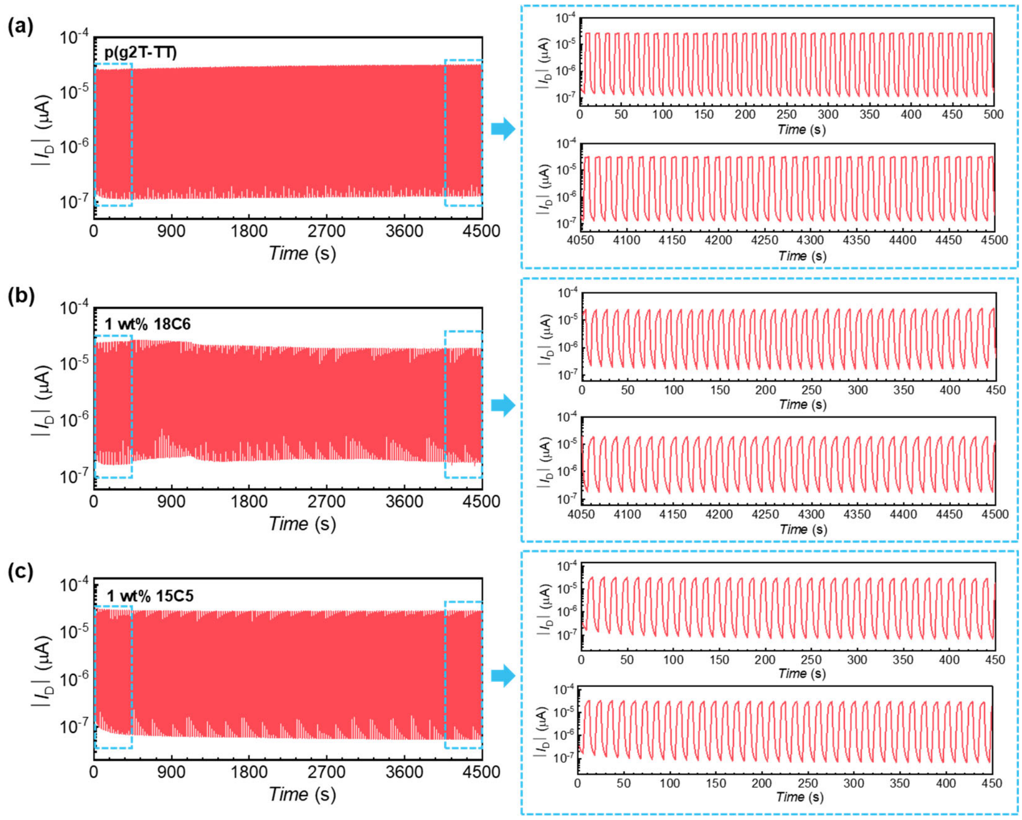

Fig. S4 Operation stability of drain current for IS-OECTs. Transient response of drain current for V_D is –0.4 V and square pulse V_G switching between 0 and −0.4 V with the pulse width of 5 s for 450 cycles in 0.1 M KCl aqueous solution. The right figures show the first 45 cycles (top) and the last 45 cycles (bottom): (a) pristine p(g2T-TT); (b) 18C6-mixed p(g2T-TT), and (c) 15C5-mixed p(g2T-TT).

Table S5. Average drain current of "ON" and "OFF" states at first 45 cycles and last 45 cycles in transient response with 0.1 M NaCl or KCl aqueous solutions as the electrolyte under switching gate voltage as 0 V/−0.4 V in OECTs

Testing Cycles \ Materials		p(2gT-TT)	18C6-mixed p(2gT-TT)	15C5-mixed p(2gT-TT)	p(2gT-TT)	18C6-mixed p(2gT-TT)	15C5-mixed p(2gT-TT)
		Na^{+}			K^{+}		
First 45 Cycles	I_{on} (μA)	-24.3	-70.2	-30.7	-26.1	-23.3	-28.5
	I_{off} (μA)	-0.114	-0.731	-0.0692	-0.134	-0.213	-0.970
Last 45 Cycles	I_{on} (μA)	-24.5	-54.4	-30.9	-31.6	-17.5	-27.0
	I_{off} (μA)	-0.141	-1.42	-0.0136	-0.145	-0.222	-0.0629
Retention		**100%**	**76.3%**	**100%**	**100%**	**74.8%**	**97.8%**

Reference

S1 A. Giovannitti, D. T. Sbircea, S. Inal, C. B. Nielsen, E. Bandiello, D. A. Hanifi, M. Sessolo, G. G. Malliaras, I. McCulloch and J. Rivnay, *Proc. Natl. Acad. Sci. U.S.A.*, 2016, **113**, 12017-12022.