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Supporting Information

Asymmetric Side-Chain Engineering of Conjugated Polymers with Improved Performance and Stability in Organic Electrochemical Transistors

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Table S1. Optical and electrochemical parameters of the polymer films.

Polymer	P(C,C)	P(C,O)	P(O,O)
λ_{max} (nm)	703	710	718
(0-0 / 0-1)	0.767	0.806	0.847
Eg (eV)	1.64	1.61	1.58
HOMO (eV)	−5.52	−5.31	−5.08
LUMO (eV)	−3.88	−3.70	−3.50
ΔE_{onset} (eV)	0.202	0.606	0.695

Table S2. GIXD crystallographic parameters of the as-cast and electrolyte-swelled polymer films derived from the OOP line-cutting profiles.

OOP	As-cast			Swelled		
	P(C,C)	P(C,O)	P(O,O)	P(C,C)	P(C,O)	P(O,O)
d_{200} (Å)	12.04	12.91	11.61	12.09	12.91	12.09
FWHM (Å)	0.089	0.063	0.054	0.092	0.067	0.040
$L_{c,200}$ (Å)	63.25	90.05	104.53	61.80	83.89	139.97
g_{200}	0.183	0.159	0.140	0.186	0.165	0.123

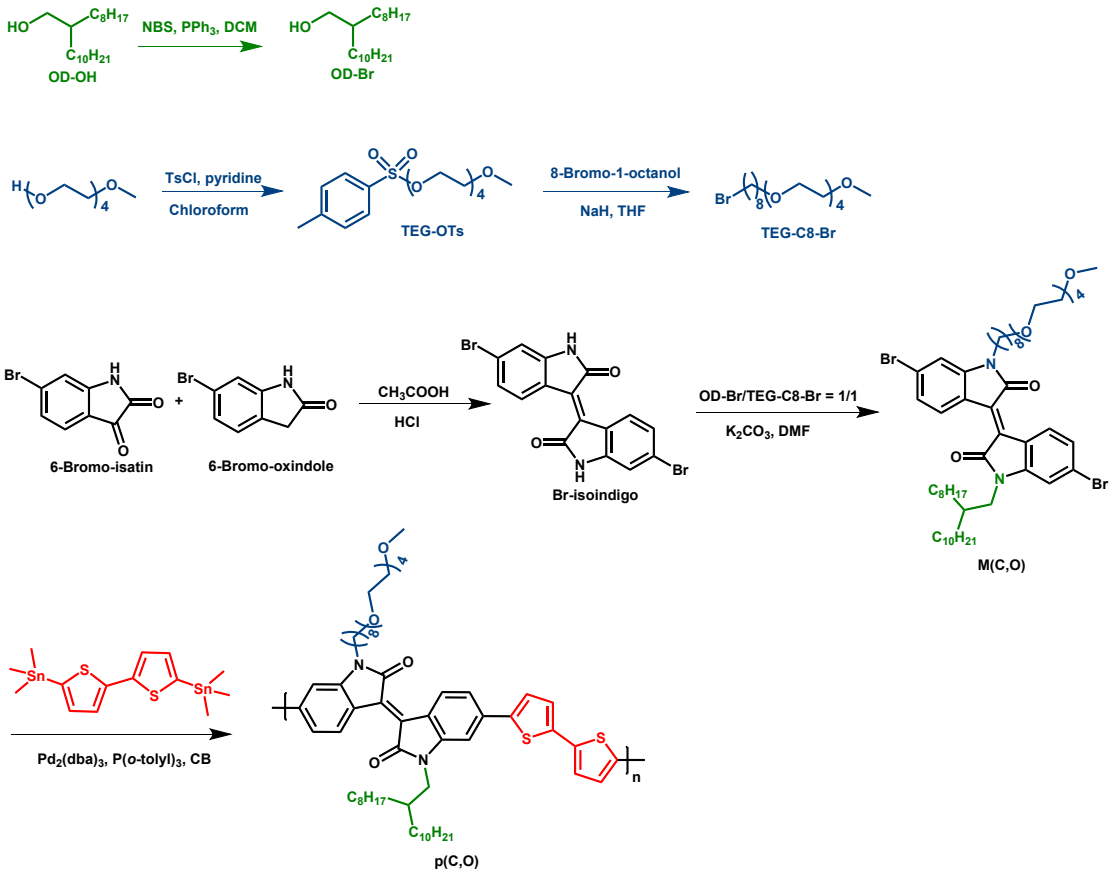
Table S3. GIXD crystallographic parameters of the as-cast and electrolyte-swelled polymer films derived from the IP line-cutting profiles.

IP	As-cast			Swelled		
	P(C,C)	P(C,O)	P(O,O)	P(C,C)	P(C,O)	P(O,O)
d_{010} (Å)	-	3.55	3.65	-	3.55	3.67
FWHM (Å)	-	0.089	0.104	-	0.091	0.098
$L_{c,010}$ (Å)	-	63.54	54.37	-	62.14	57.70
g_{010}	-	0.099	0.109	-	0.100	0.106

Table S4. GIXD crystallographic parameters of the as-cast and electrolyte-swelled polymer films derived from the geometrically corrected azimuthal integral profiles.

	As-cast			Swelled		
	P(C,C)	P(C,O)	P(O,O)	P(C,C)	P(C,O)	P(O,O)
OOP area	2.292	4.012	12.837	4.999	6.858	15.2
IP area	0.322	0.378	0.256	0.803	0.152	0.178
OOP / (IP+OOP)	0.877	0.914	0.980	0.862	0.978	0.988
rDOC (%)	15.1	26.4	84.5	32.9	45.1	100.0

Scheme S1. Synthetic routes of the side chains, isoindigo monomers, and P(C,O) with asymmetric side chains.



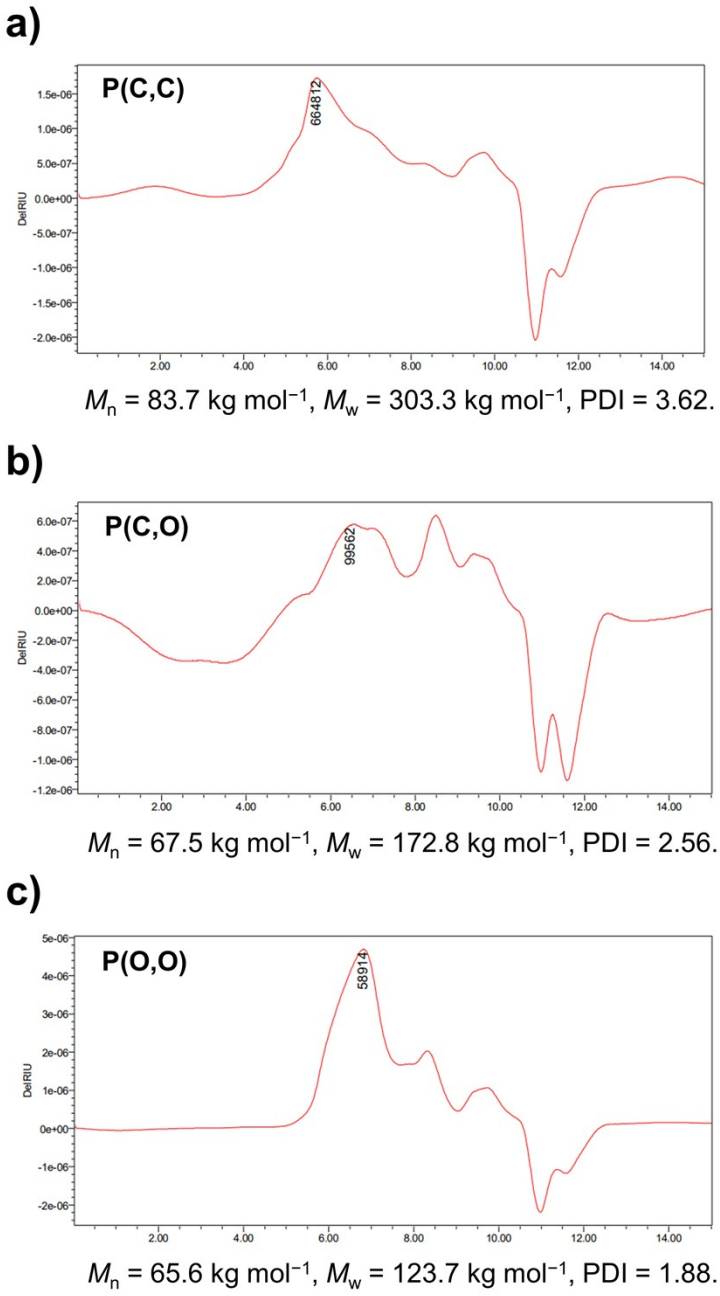


Fig. S1.GPC curves of (a) P(C,C), (b) P(C,O), and (c) P(O,O).

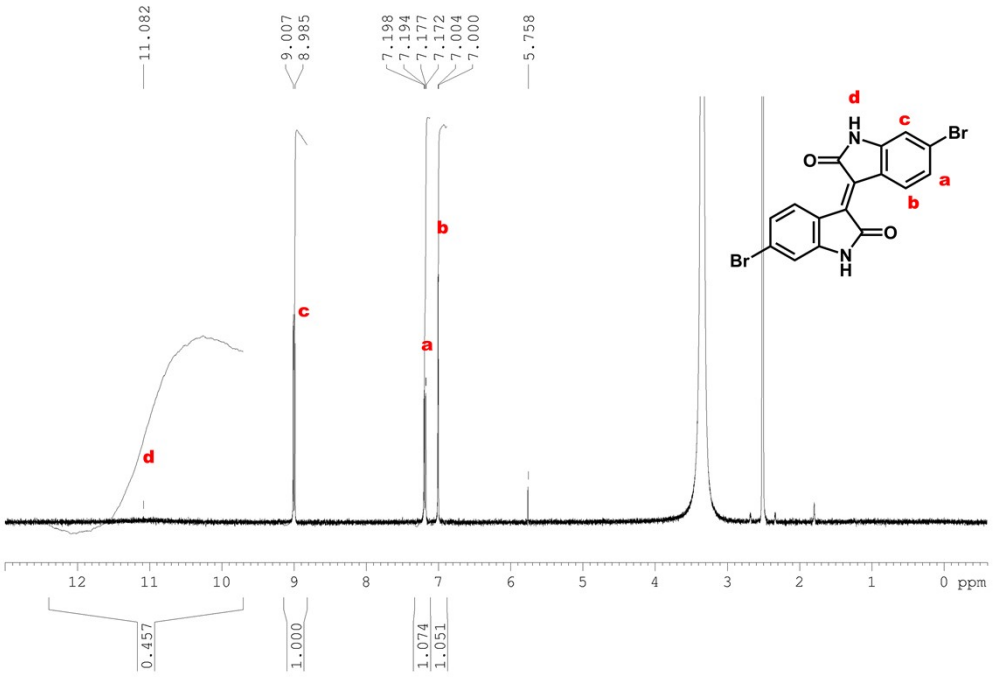


Fig. S2. ^{1}H NMR spectrum of **Br-isoindigo** in d_6-DMSO.

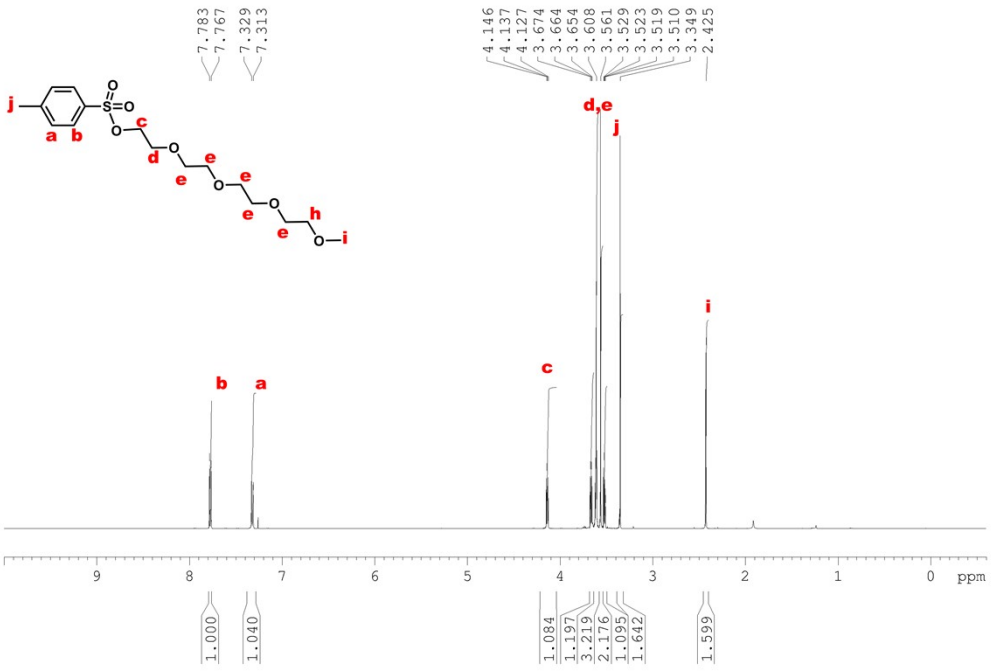


Fig. S3. ^{1}H NMR spectrum of **TEG-OTs** in CdCl$_3$.

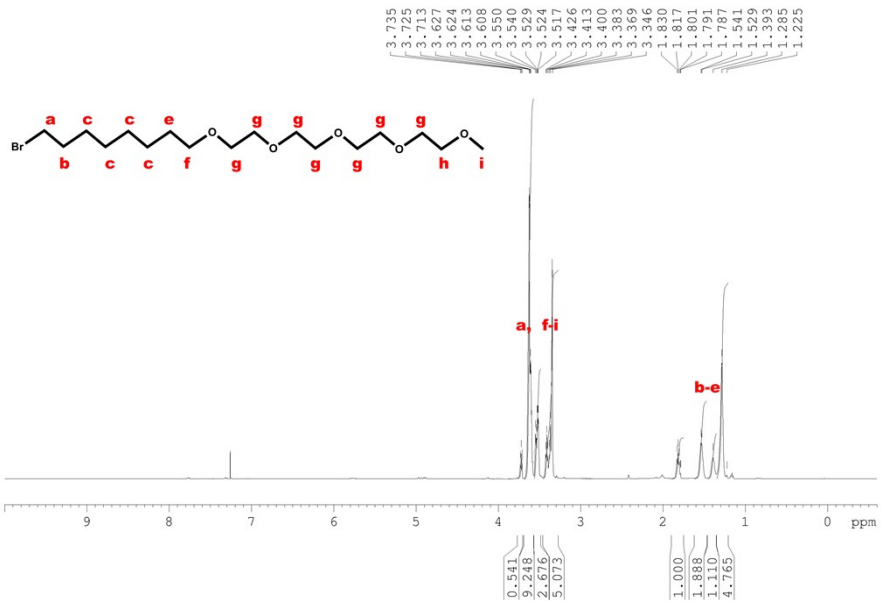


Fig. S4. ^{1}H NMR spectrum of **TEG-C8-Br** in C*d*Cl$_3$.

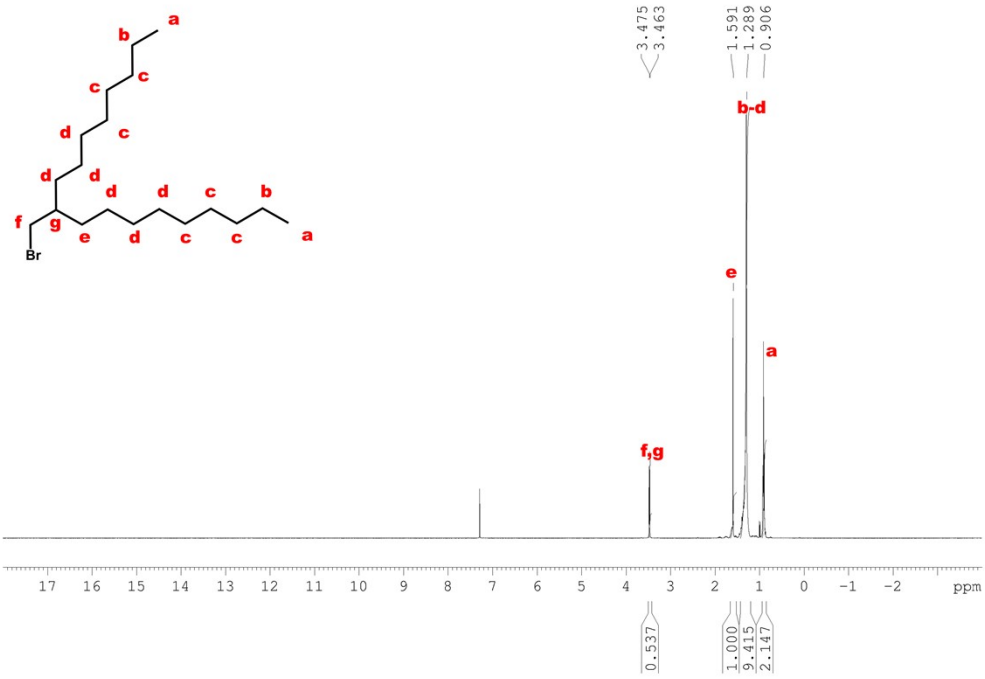


Fig. S5. ^{1}H NMR spectrum of **OD-Br** in C*d*Cl$_3$.

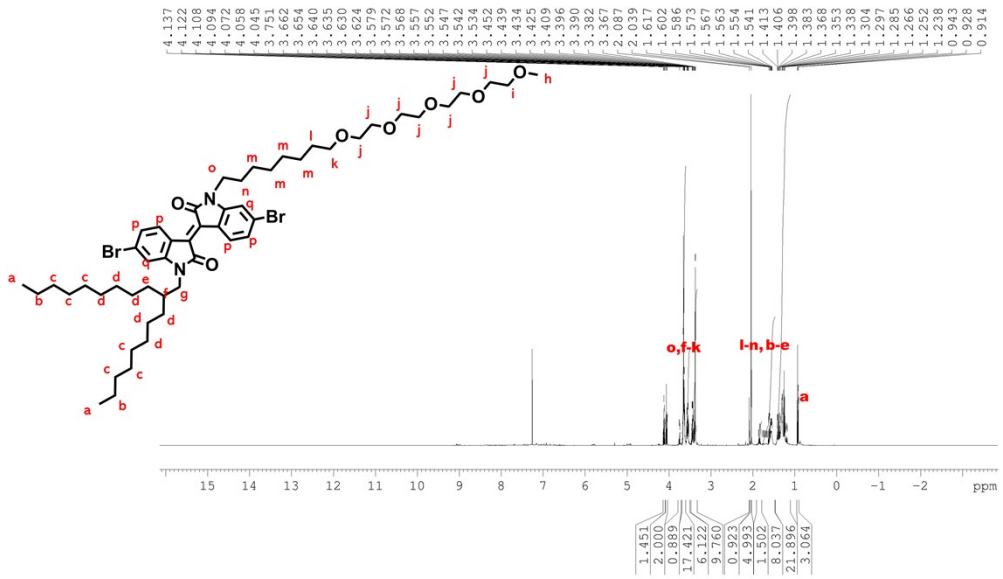


Fig. S6. ^{1}H NMR spectrum of **M(C,O)** in C*d*Cl$_3$.

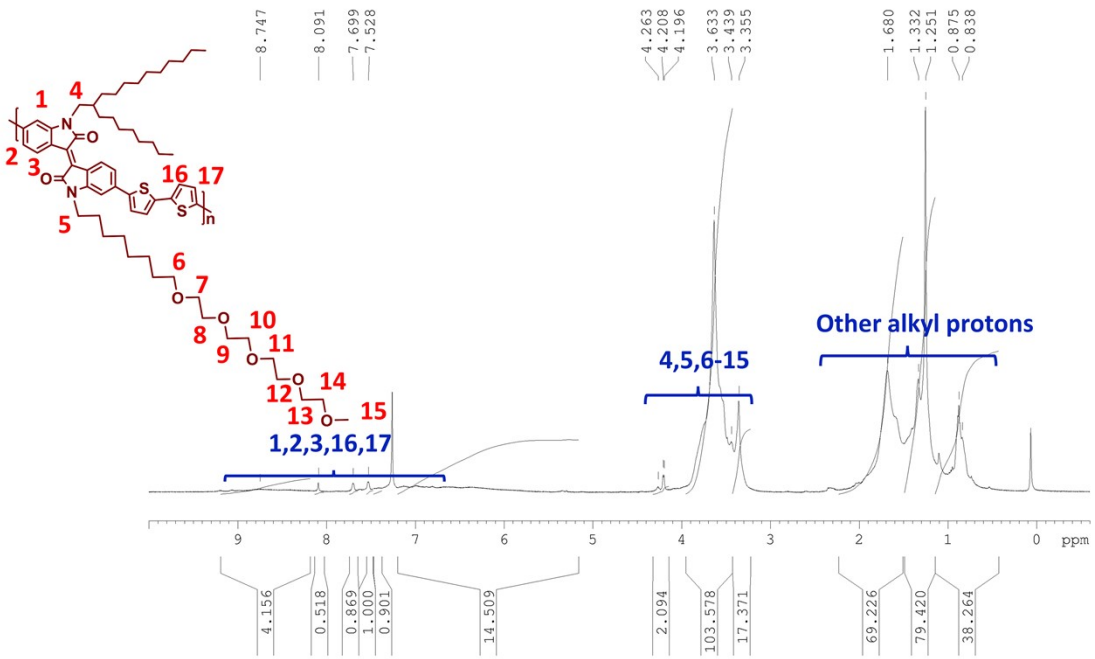


Fig. S7. ^{1}H NMR spectrum of **P(C,O)** in C*d*Cl$_3$.

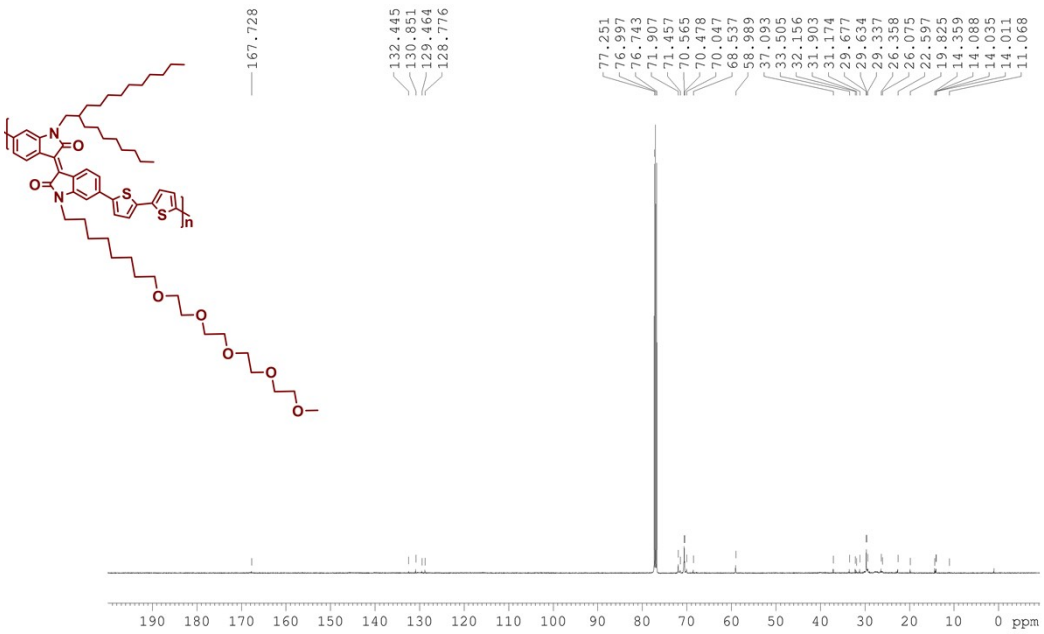


Fig. S8. ^{13}C NMR spectrum of **P(C,O)** in C*d*Cl$_3$.

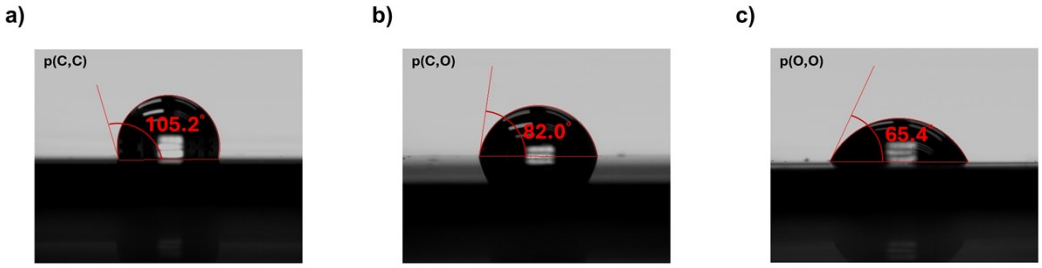


Fig. S9. Contact angle measurements of water droplets on the polymer films: (a) P(C,C), (b) P(C,O), and (c) P(O,O).

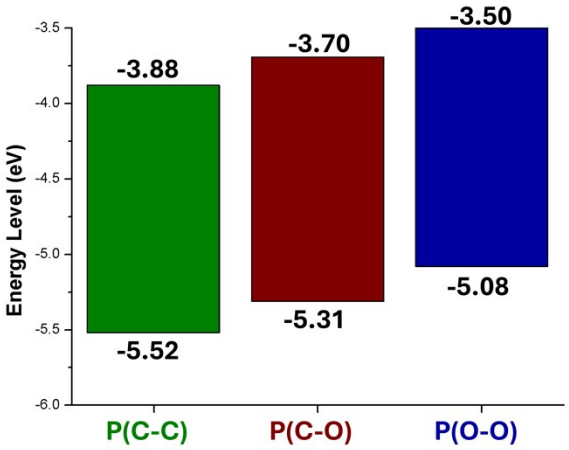


Fig. S10. Frontier energy level diagram of the polymers studied.

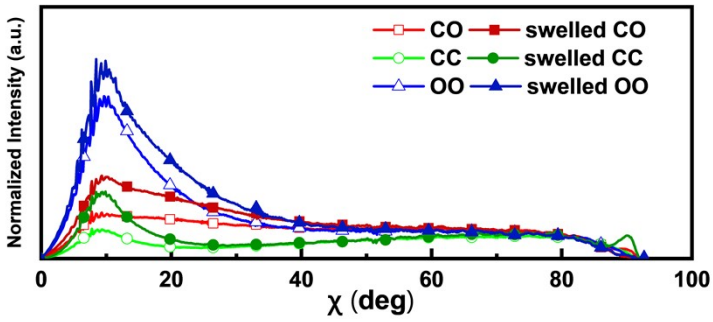


Fig. S11. Geometrically corrected pole figures of the polymer films derived from the azimuthal integral of their (100) diffractions in 2D GIXD patterns.

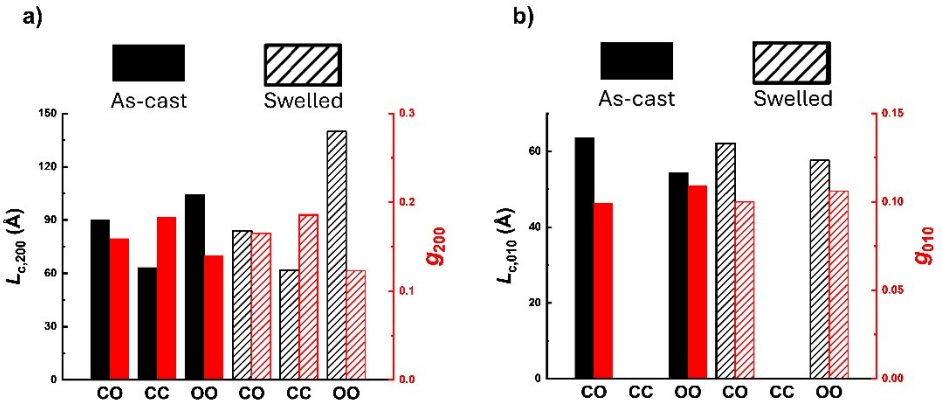


Fig. S12. Summary of the crystallographic parameters: L_c and g of (a) (200) diffractions and (b) (010) diffractions of the as-cast and electrolyte-swelled polymer films.

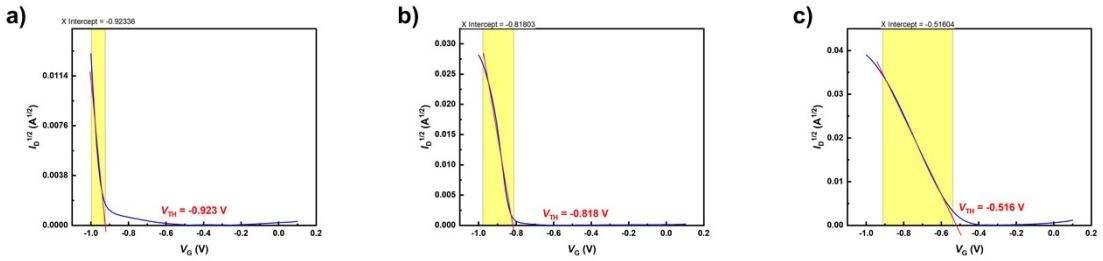


Fig. S13. The relationship of $I_d^{1/2}$ vs. V_g of (a) P(C,C), (b) P(C,O), and (c) P(O,O) based OECT devices.

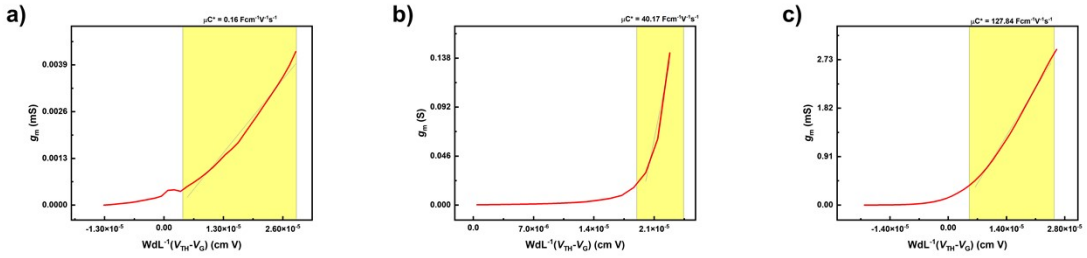


Fig. S14. The relationship of g_m vs. $WdL^{-1}(V_{th} - V_g)$ of (a) p(C,C), (b) p(C,O), and (c) P(O,O) based OECT devices.

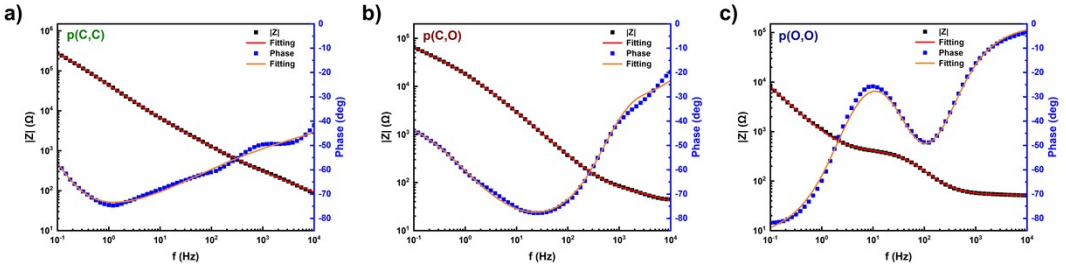


Fig. S15. Bode plots derived from the EIS characteristics of the polymer films: (a) P(C,C), (b) P(C,O), and (c) P(O,O).

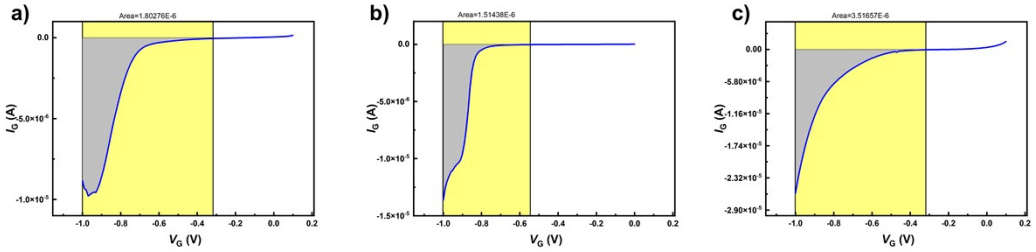


Fig. S16. The relationship of I_g vs. V_g of (a) P(C,C), (b) P(C,O), and (c) P(O,O) based OECT devices.

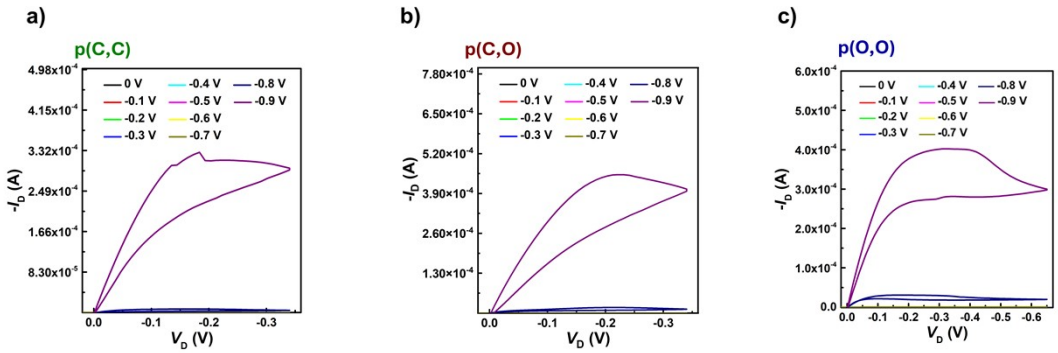


Fig. S17. Output characteristics of (a) P(C,C), (b) P(C,O), and (c) P(O,O) based OECT devices with a forward V_D sweeping from 0 to −0.3 ~ −0.6 V at V_G = 0 ~ −0.9 V.

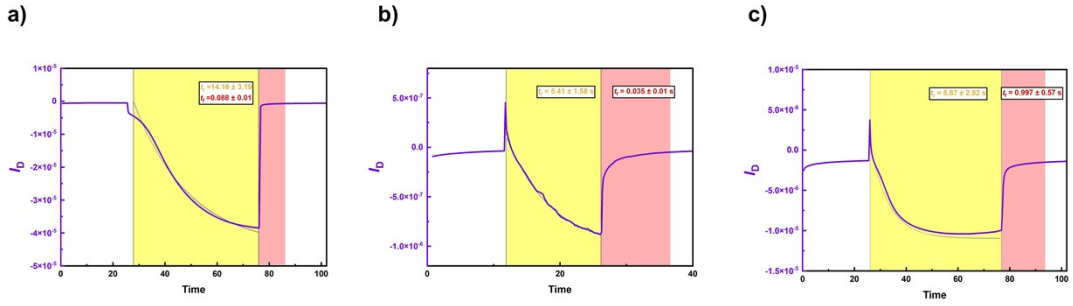


Fig. S18. Transient curves of (a) P(C,C), (b) P(C,O), and (c) P(O,O) based OECT devices. The x (time) and y (I_D) axes are in units of [s] and [A]. The exponential fittings indicate their characteristic response time.

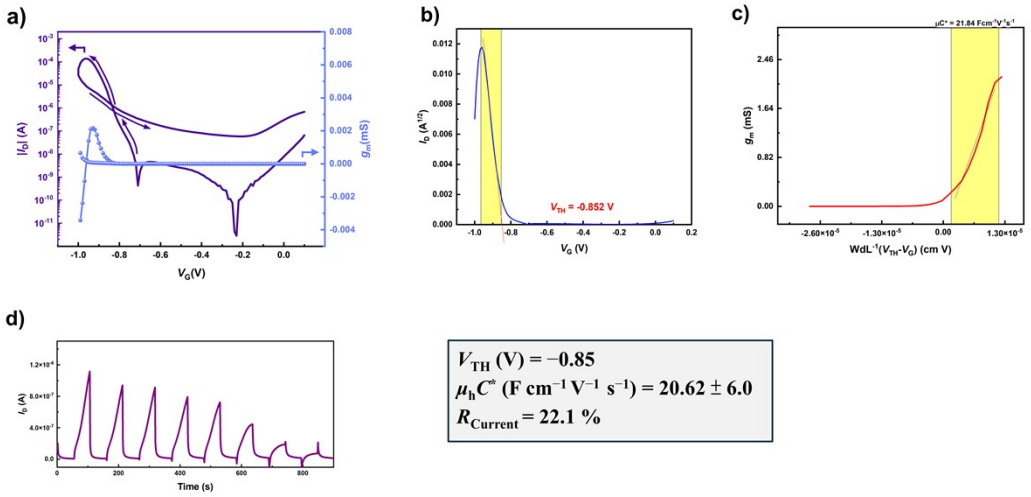


Fig. S18. OECT transfer characteristics of the reference device comprising the polymer blend of P(C,C) and P(O,O): (a) Transfer curve with a forward V_G sweeping from 0.1 to −1.0 V at V_D = −0.3 V. (b) The relationship of $I_d^{1/2}$ vs. V_g. (c) The relationship of g_m vs. $WdL^{-1}(V_{th} - V_g)$. (d) Transient characteristics of the polymer films with V_G pulse of −0.85 V at V_D = −0.3 V for 8 cycles.