Electronic Supplementary Material (ESI) for Materials Horizons.

Supporting Information

Backbone Coplanarity Manipulation via Hydrogen Bonding to Boost the n-Type Performance of Polymeric Mixed Conductors Operating in Aqueous Electrolyte

Junxin Chen, Shengyu Cong, Lewen Wang, Yazhou Wang, Liuyuan Lan, Chaoyue Chen, Yecheng Zhou, Zhengke Li, Iain McCulloch, Wan Yue **

Table of Contents

1. General Procedures

Reagents and solvents were purchased from Sigma-Aldrich and used without any further purification. The trans-1,2-bis(tributylstannyl)ethene and 2,5-bis(trimethylstannyl)thiophene were purchased from Nanjing Zhiyan Technology Co., Ltd. and used without further purification. The intermediates and monomers were purified by column chromatography using silica gel (General-Regent, 300-400 mesh) and the copolymers were purified by Soxhlet extraction. ^{1}H NMR was conducted in chloroform-d or 1, 1, 2, 2-tetrachloroethane-d_2 (TCE-D_2) with a 400 HMZ or 500 HMZ Bruker Avance III spectrometer.

Two NDI-based copolymers' molecular weight was calculated by Matrix-assisted Laser Desorption/ionization time-of-flight (MALDI-TOF) mass spectra (matrix is trans-2-[3-(4-t-butyl-phenyl)-2-methyl-2-propenylidene]malononitrile (DCTB)), using a Bruker Daltonik (solariX) mass spectrometer. Besides, the number-average molecular weights (Mn) of copolymers were determined by a PL-GPC 220 in chlorobenzene (CB) at 45 °C using a calibration curve with standard linear polystyrene as a reference.

UV-Vis-NIR absorption spectra was measured by an Agilent Technologies Cary 5000/6000i spectrophotometer. Thin films were prepared by spin-coating on ITO glass substrates from a chloroform/chlorobenzene (9/1, v/v) solution (10 mg mL^{-1}) under ambient conditions, following by annealing treatment at 100 °C for 1 h.

Cyclic voltammetry was performed employing a CHI760E electrochemical workstation (Shanghai Chenhua Instrument co., Ltd.) with a standard three-electrode configuration, including copolymer coated ITO as the working electrode, a platinum wire as the counter electrode and an Ag/AgCl electrode as the reference electrode. The copolymer coated electrodes were prepared as UV-Vis-NIR thin film samples. The supporting electrolyte was either an acetonitrile solution containing 0.1 M $n\text{-}Bu_4NPF_6$ or 0.1 M KCl aqueous solution.

Spectroelectrochemical measurements were conducted by fitting the CV setup inside a quartz cuvette in the UV–vis–NIR absorption spectrophotometer. The copolymers were spin-casted ITO-coated glass from a solution (10 mg mL^{-1}) of chloroform/chlorobenzene (9/1, v/v). Then potentials were applied for 30 s before recording the UV–vis–NIR absorption spectrum at each voltage to ensure stabilization of the optical trace. UV-vis-NIR spectra was continuously recorded within a potential range from 0.1 V to -0.7 V for g**NDI-V** and g**NDI-T** with potential interval of 0.1 V. All spectroelectrochemical measurements were performanced using a 0.1 M KCl solution as the supporting electrolyte.

Thermogravimetric analysis (TGA) was conducted on a Netzsch STA 449F3 apparatus. Samples placed into small ceramic crucibles were heated under nitrogen atmosphere from 25 °C to 700 °C at a heating rate of 10 °C min^{-1}.

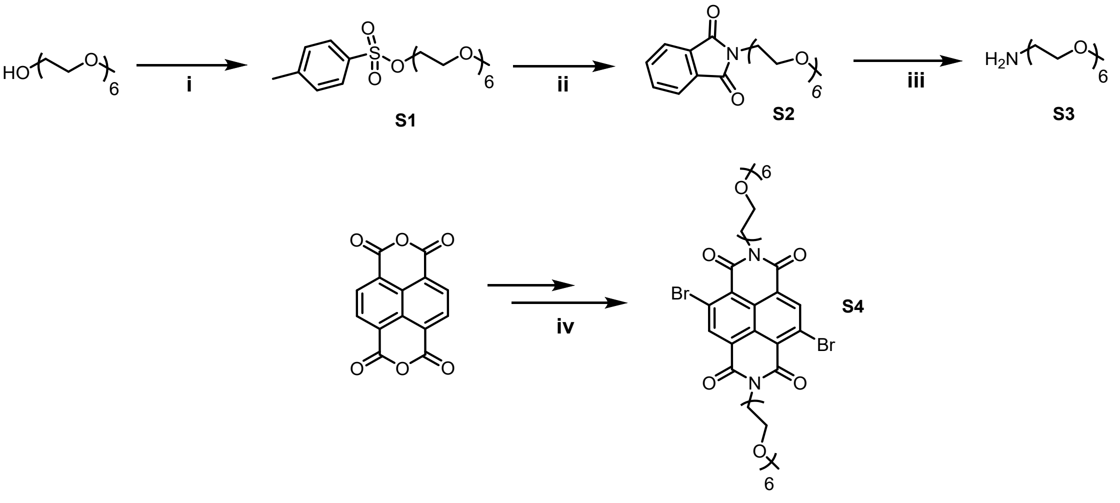
Electrochemical impedance spectroscopy (EIS) was performed using a three-terminal set-up same as CV measurement, employing copolymer coated gold electrodes (the area of 0.2 cm^2) as the working electrode, a Pt foil as the counter electrode and an Ag/AgCl electrode as the reference electrode. A sinusoidal AC amplitude of 10 mV and a frequency range between 10 kHz to 0.1 Hz was chosen. Fitting the EIS spectrums by using the Randles circuit $R_s(R_p//C)$ in ZSimDemo software were to extract capacitance.

Grazing incidence wide-angle X-ray scattering (GIWAXS) was conducted in the Beijing Synchrotron Radiation Facility. A photon beam energy of 8 KeV and an incidence angle of ~ 0.18° were employed to record the scattering patterns.

Density functional theory (DFT) calculations were performed using Gaussian 09 with the level of B3LYP/6-31G(d). The long EG side chain on the NDI core was replaced with methyl group and three repeating units of the NDI-based copolymers were used to simplify the calculation.

Electrode contacts were prepared by subsequently evaporating titanium (as an adhesion layer) and gold on glass using a custom-made mask. The channel length L and width W was 20 μm and 39000 μm, respectively. After cleaning the substrate using piranha solution, nitrogen drying followed. Solutions of the NDI-based copolymers (concentration = 10 mg mL^{-1}) in chloroform/chlorobenzene mixed solvent (9/1, v/v) were spin-coated on the cleaned substrates (500 rpm /10 s + 1000 rpm /30 s). The pristine thin films were heated at 100 °C for 1 hour under nitrogen atmosphere. Film thicknesses were measured by atomic force microscopy. OECTs were gated with 0.1 M KCl aqueous solution using an Ag/AgCl pellet as the gate electrode. Electrical characterization (output, transfer, and pulsed stability) of the OECTs was performed at room temperature using a semiconductor parameter analyzer (Keysight B1500A) and an electrical probe station.

2. Monomers and Copolymers Synthesis



Scheme S1. Monomers synthesis

Reaction conditions: (i) TEA, DCM, 4-toluenesulfonyl chloride, 0 °C – rt, 24h; (ii) anhydrous DMF, 90 °C, 18h; (iii) N_2H_4, Ethanol; (iv) (a) 1, 3-dibromo-5, 5-dimethylhydantoin, H_2SO_4, 60 °C, 18h; (b) CH_3COOH, 130 °C, 4h. **S1-S4** were synthesized according to the literature.[S1]

Monomer S1

2, 5, 8, 11, 14, 17-hexaoxanonadecan-19-ol (15.0 g, 50.6 mol) and triethylamine (8.0 mL, 56.4 mmol) were added to a 500 ml round-bottom flask under nitrogen, then 100 mL of anhydrous dichloromethane (DCM) was added. Tosyl chloride (10.2 g, 53.6 mol) dissolved in anhydrous DCM (50 mL) was then added slowly to the reaction flask. The solution was stirred at room temperature overnight. The reaction solution was cooled to 0 °C to precipitate out the triethylamine hydrochloride salt, which was then removed by filtration. The target product was purified using silica gel flash chromatography to yield colorless oil. (20.9 g, yield 92 %) ^1H NMR (400 MHz, chloroform-d, 300 K), δ (ppm): 7.79 (d, J = 8.16 Hz, 2H), 7.34 (d, J = 8.04 Hz, 2H), 4.15 (t, J = 4.72Hz, 2H), 3.68-3.53 (m, 22H), 3.36 (s, 3H), 2.44 (s, 3H).

Monomer S2

Phthalimide (7.8 g, 53.0 mmol) and K_2CO_3 (7.4 g, 53.6 mmol) were added to a 500 ml round-bottom flask under nitrogen. Then **S1** (20.0 g, 44.4 mmol) dissolved in anhydrous DMF (150 mL) was added to the reaction flask. The reaction mixture was heated at 90 °C and stirred overnight before cooling to room temperature. The solvent was removed in vacuo, water (300 mL) was added, and the mixture was extracted with DCM (3 × 200 mL). The combined organic layers were dried over $MgSO_4$, filtered, and concentrated in vacuo. The crude product was purified by column chromatography to afford **S2** as colorless oil (17.8 g, 94 % yield). ^1H NMR

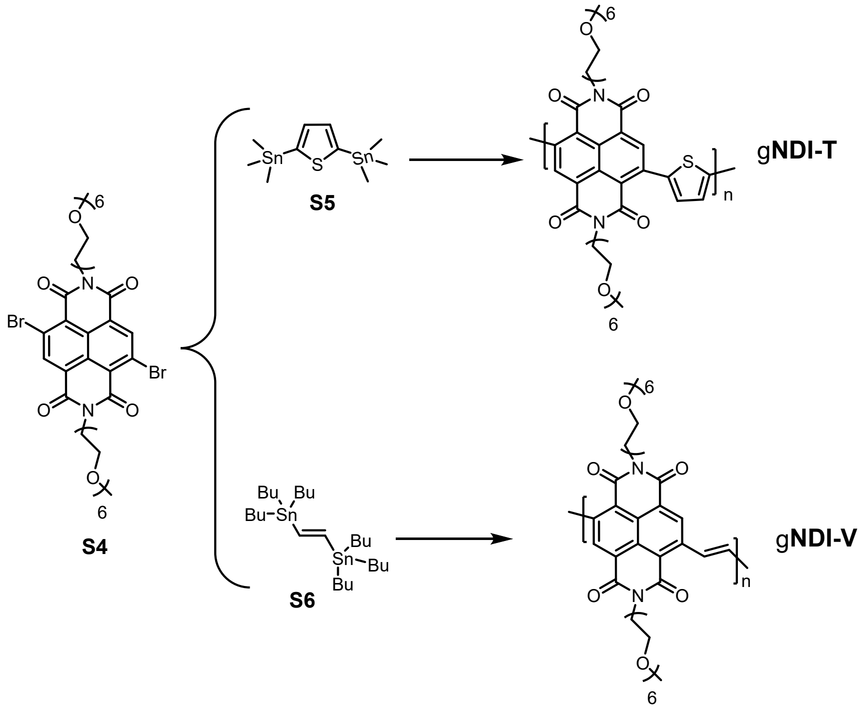
(400 MHz, chloroform-d, 300 K), δ (ppm): 7.80 (m, 2H), 7.68 (m, 2H), 3.85 (t, *J* = 5.12 Hz, 2H), 3.70 (t, *J* = 5.28 Hz, 2H), 3.57 (m, 20H), 3.33 (s, 3H).

Monomer S3

S2 (13.0 g, 30.6 mmol), hydrazine hydrate (2.8 ml, 92.4 mmol) and 150 mL ethanol were stirred at 90 ºC overnight. After disappearance of the starting imide, the methanol was evaporated under reduced pressure, the residue diluted with DCM and washed with 10% KOH. Aqueous layers were combined and extracted with DCM. The combined organic layers were washed with brine and dried over $MgSO_4$. The removal of DCM afforded colorless oil as product which was used without further purification. Yield: 83 % (7.4 g). ^{1}H NMR (400 MHz, chloroform-d, 300 K), δ (ppm): 3.67-3.59 (m, 18H), 3.55-3.45 (m, 4H), 3.35 (s, 3H), 2.84 (t, *J* = 5.16 Hz, 2H), 1.63 (br, 2H).

Monomer S4

Under nitrogen, 1,3-dibromo-5,5-dimethylhydantoin (35.3 g, 123.2 mmol) was added portion wise to 1, 4, 5, 8-naphthalenetetracarboxylic dianhydride (15 g, 56.0 mmol) in sulphuric acid (150 mL) and the reaction was heated to 60 ºC for 12 h. The resulting reaction solution was poured carefully into ice and H_2O (500 mL) mixture. The mixture was stirred in ice water for half an hour. Next, the yellow solids were collected by filtration, sonicated in methanol and filtered again to yield brominated products for next step. 3.0 g brominated products and **S3** (4.7 g, 15.8 mmol) were added in acetic acid (100 mL). The reaction mixture was refluxed under nitrogen for 3 h. Then the reaction mixture was poured into water and extracted with DCM. The crude product was purified via column chromatography over silica gel. Monomer **S4** was obtained as an orange solid. Yield: 25 % (1.8 g). ^{1}H NMR (400 MHz, chloroform-d, 300 K), δ (ppm): 8.95 (s, 2H), 4.44 (t, *J* = 5.6 Hz, 4H), 3.82 (t, *J* = 6.0 Hz, 4H), 3.69-3.64 (m, 4H), 3.64-3.54 (m, 32H), 3.53-3.46 (m, 4H), 3.34 (s, 6H).



Scheme S2. NDI-based copolymers synthesis

g**NDI-T**

S4 (50.22 mg, 0.051 mmol), **S5** (20.98 mg, 0.051 mmol), $Pd_2(dba)_3$ (0.93 mg, 0.001 mmol) and tri(o-tolyl)phosphine (1.87 mg, 0.006 mmol) were added to a microwave vial. The tube was sealed and flushed with nitrogen, and then the degassed toluene (1 mL) was added. The tube was stirred at 110 °C for 24 h. After cooling to room temperature, the solution was precipitated into methanol, and filtered through a Soxhlet thimble. The polymer was extracted using Soxhlet apparatus with methanol, acetone, hexane, and chloroform. The chloroform solution was concentrated and precipitated into methanol again. The precipitate was filtered and dried under vacuum to afford g**NDI-T** (37.8 mg, 82 %). ^{1}H NMR (500 MHz, 1, 1, 2, 2-tetrachloroethane-d_2, 373K), δ (ppm)： 8.50-8.48 (s, 2H), 7.10-7.00 (s, 2H), 4.06-3.90 (m, 4H), 3.44-3.36 (m, 4H), 3.25-3.21 (m, 4H), 3.19-3.07 (m, 32H), 3.06-3.01 (m, 4H), 2.89-2.84 (s, 6H).

g**NDI-V**

S4 (49.22 mg, 0.050 mmol), **S6** (30.42 mg, 0.050 mmol), $Pd_2(dba)_3$ (0.92 mg, 0.001 mmol) and tri(o-tolyl)phosphine (1.84 mg, 0.006 mmol) were added to a microwave vial. The tube was sealed and flushed with nitrogen, and then the degassed toluene (1 mL) was added. The tube was stirred at 110 °C for 24 h. After cooling to room temperature, the solution was precipitated into methanol, and filtered through a Soxhlet thimble. The polymer was extracted using Soxhlet apparatus with methanol, acetone, hexane, and chloroform. The chloroform solution was concentrated and precipitated into methanol again. The precipitate was filtered and dried under vacuum to afford g**NDI-V** (31.0 mg, 73 %). ^{1}H NMR (500 MHz, 1, 1, 2, 2-tetrachloroethane-d$_2$, 373K), δ (ppm)：8.89-8.69 (s. 2H), 8.69-8.45 (s, 2H), 4.26-3.92 (m, 4H), 3.52-3.42 (m, 4H), 3.32-3.25 (m, 4H), 3.17-3.04 (m, 32H), 3.04-2.98 (m, 4H), 2.89-2.82 (s, 6H).

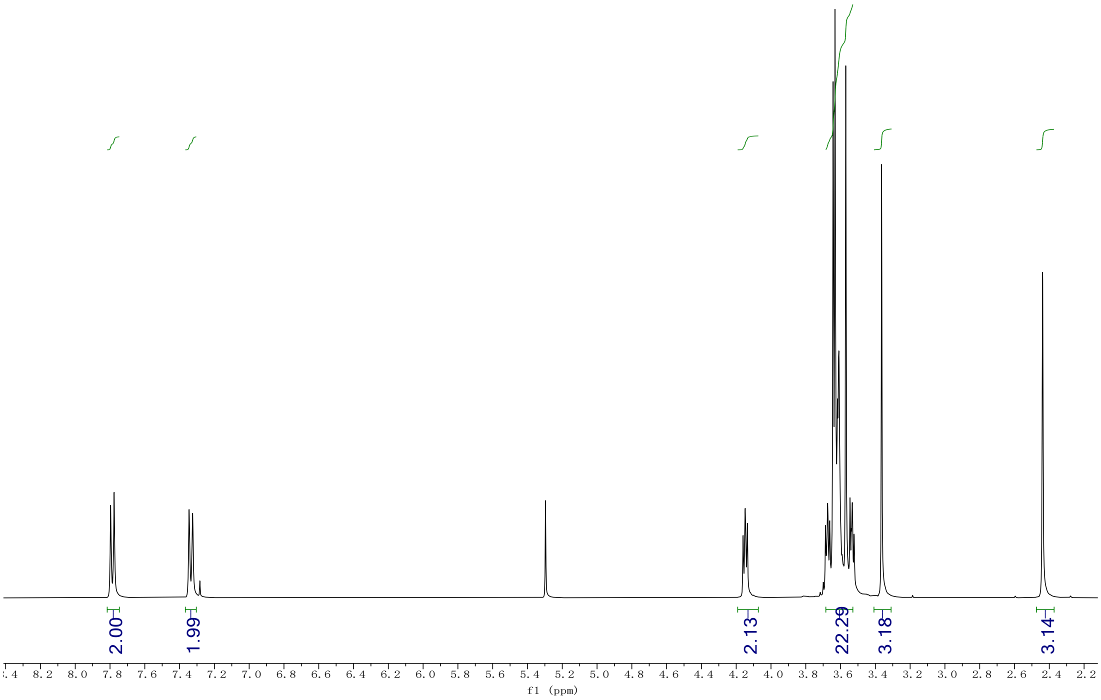


Figure S1. ^{1}H NMR of monomer **S1**

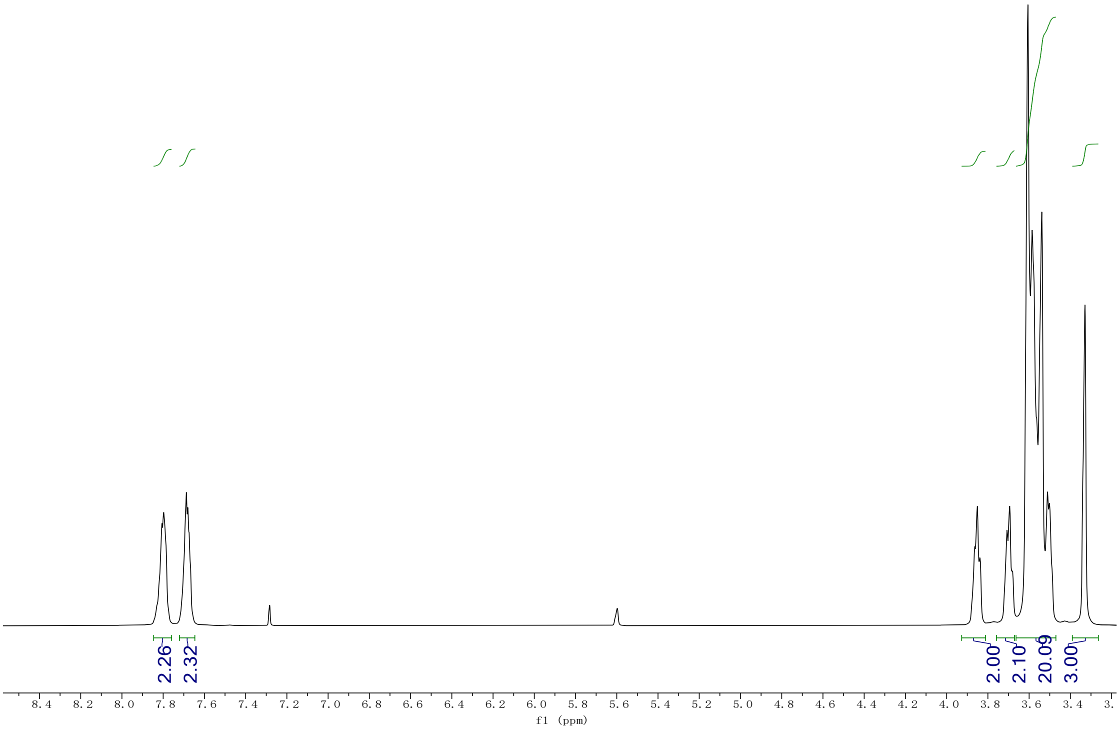


Figure S2. ^{1}H NMR of monomer **S2**

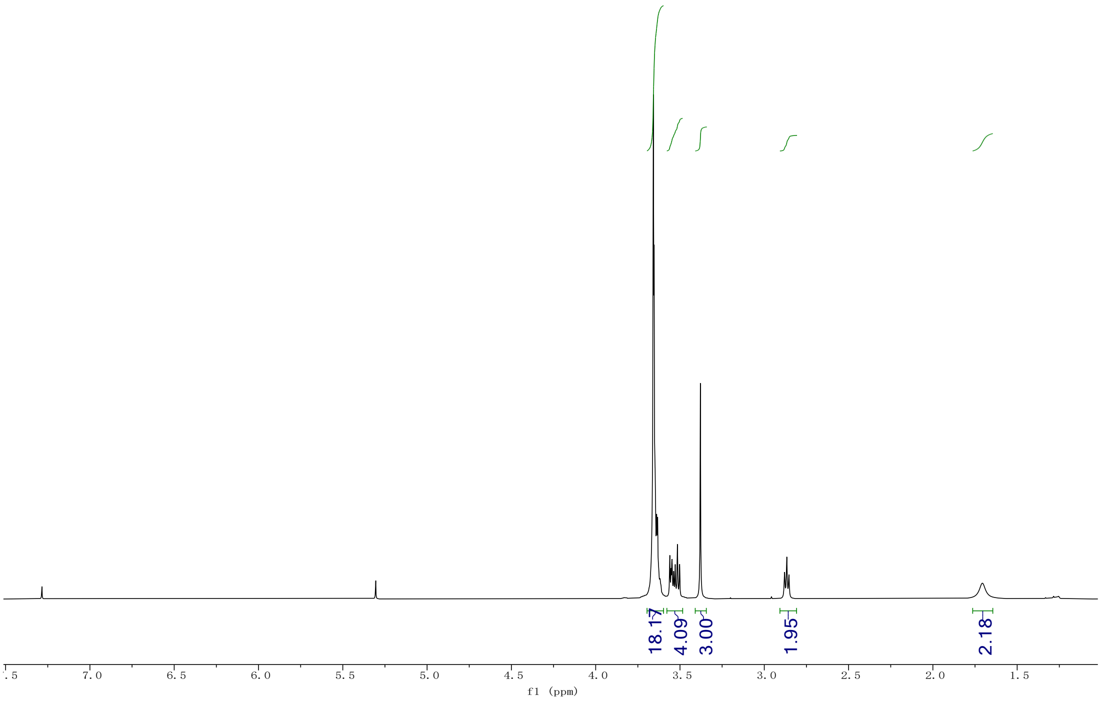


Figure S3. ^{1}H NMR of monomer **S3**

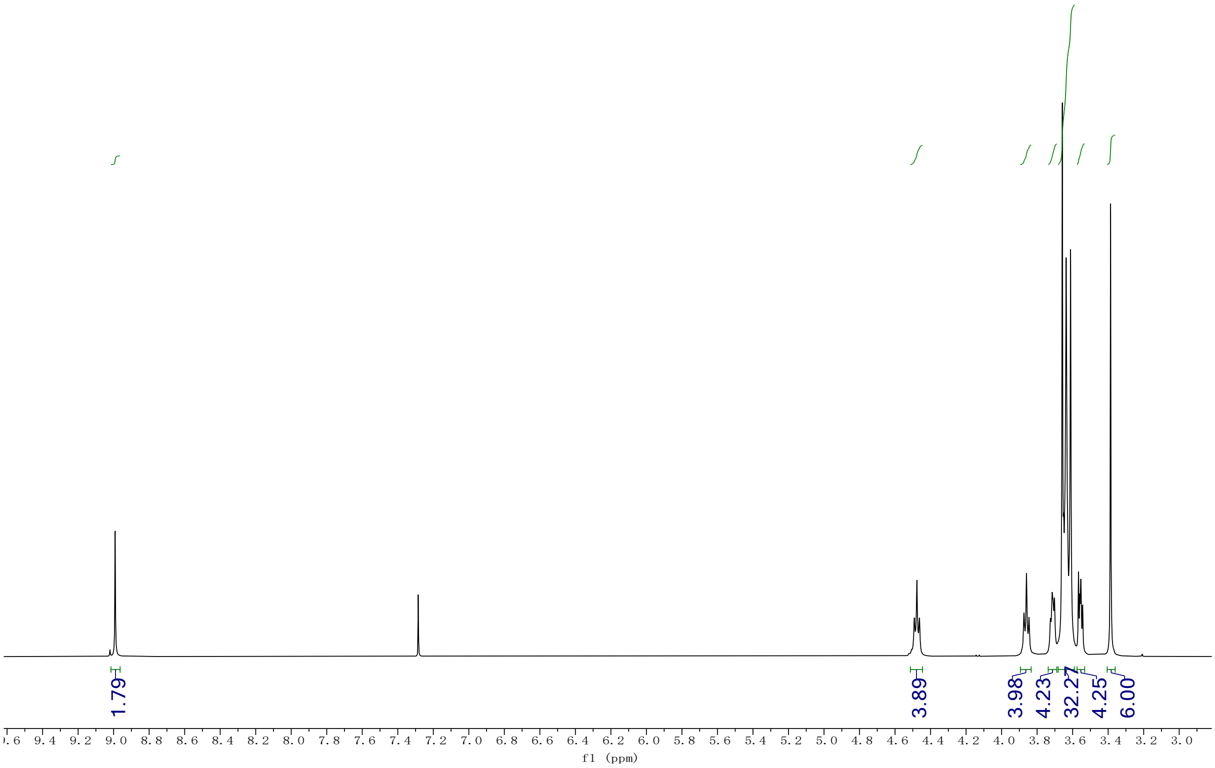


Figure S4. ^{1}H NMR of monomer **S4**

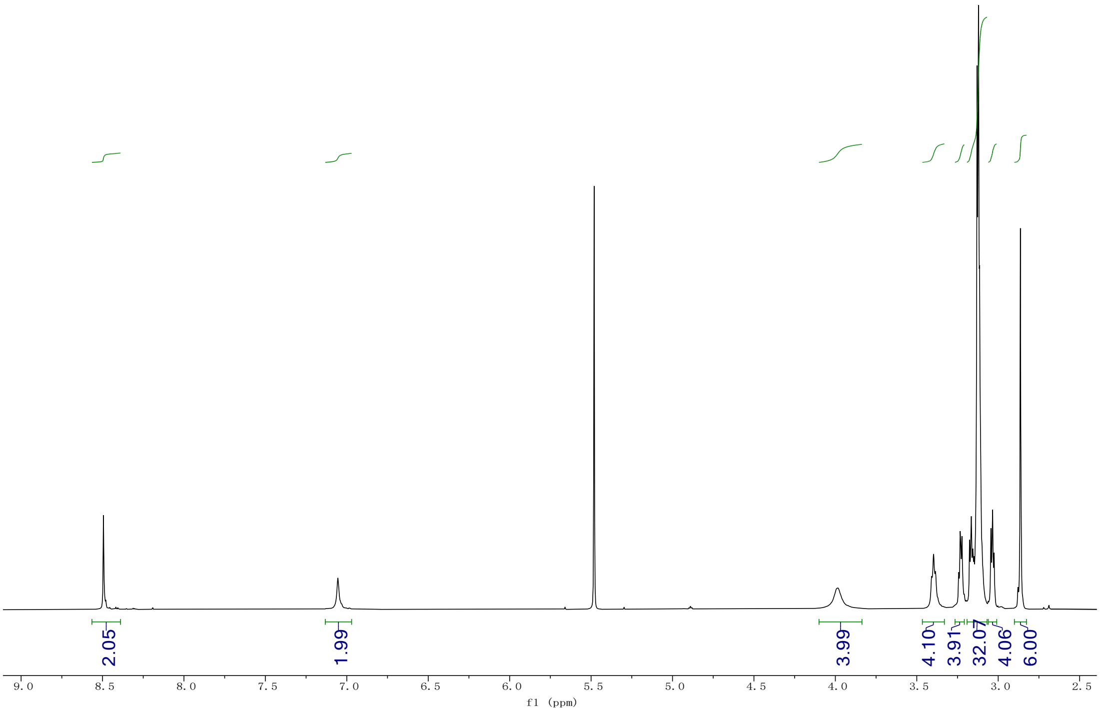


Figure S5. ^{1}H NMR of g**NDI-T** (1, 1, 2, 2-tetrachloroethane-d_2, 500 MHz, 373 K)

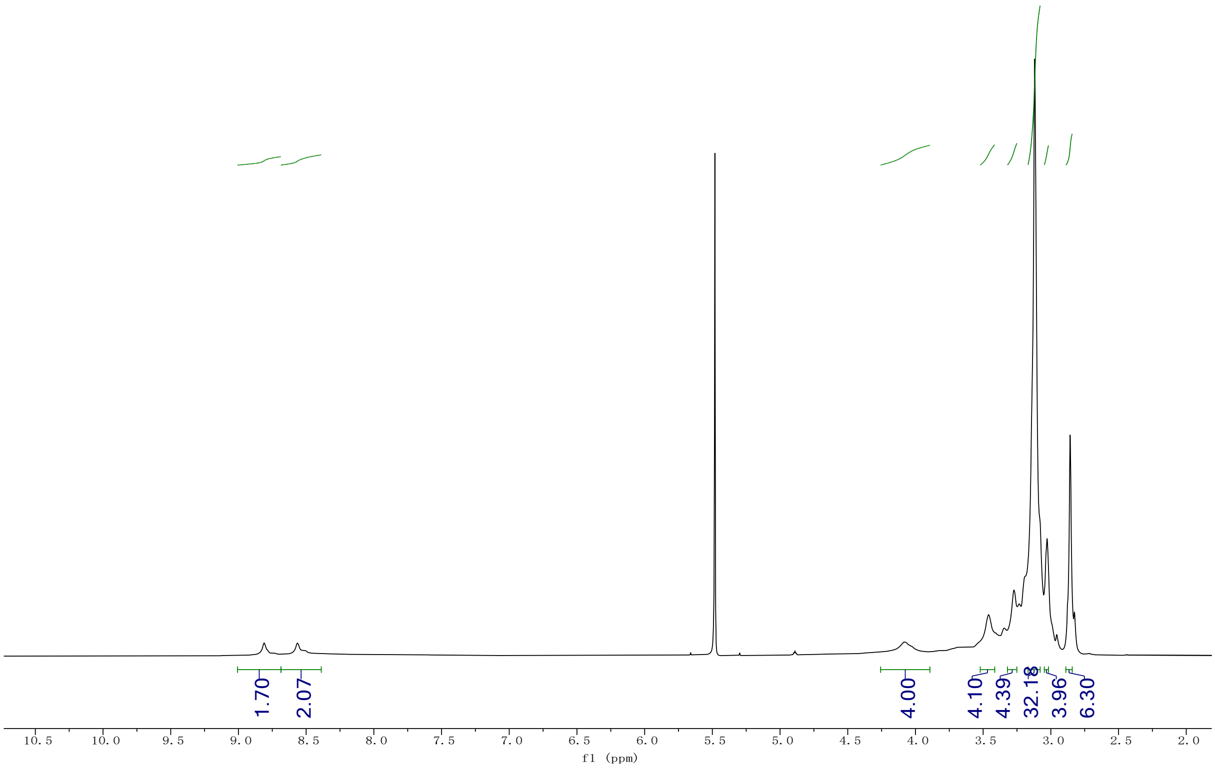


Figure S6. ^{1}H NMR of g**NDI-V** (1, 1, 2, 2-tetrachloroethane-d_2, 500 MHz, 373 K)

3. Molecular Weight

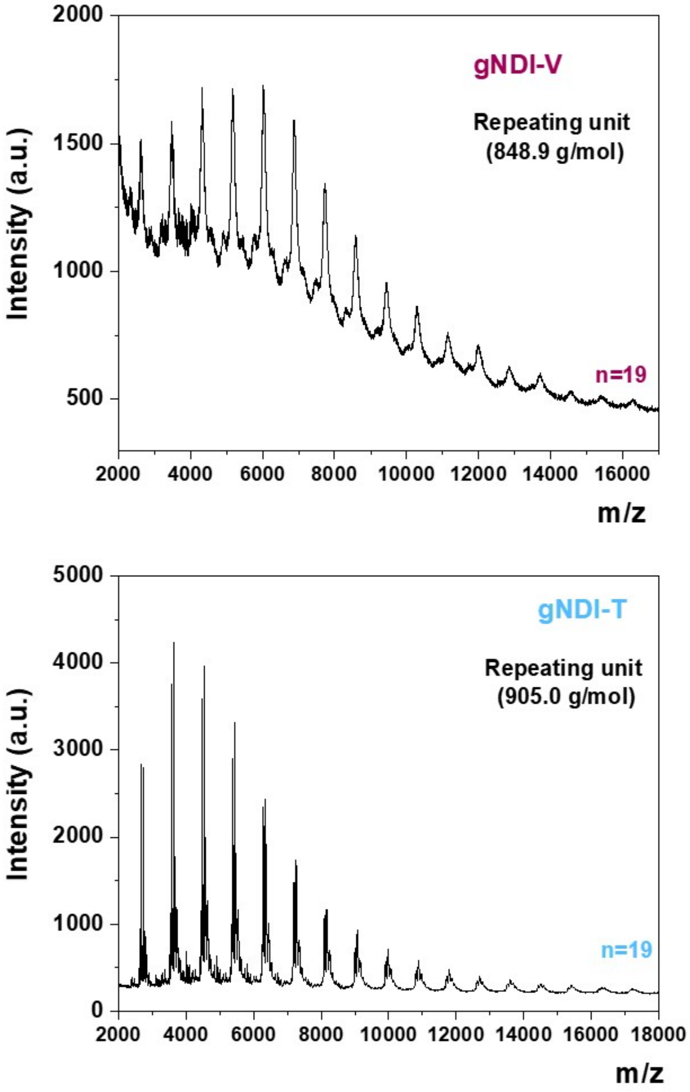
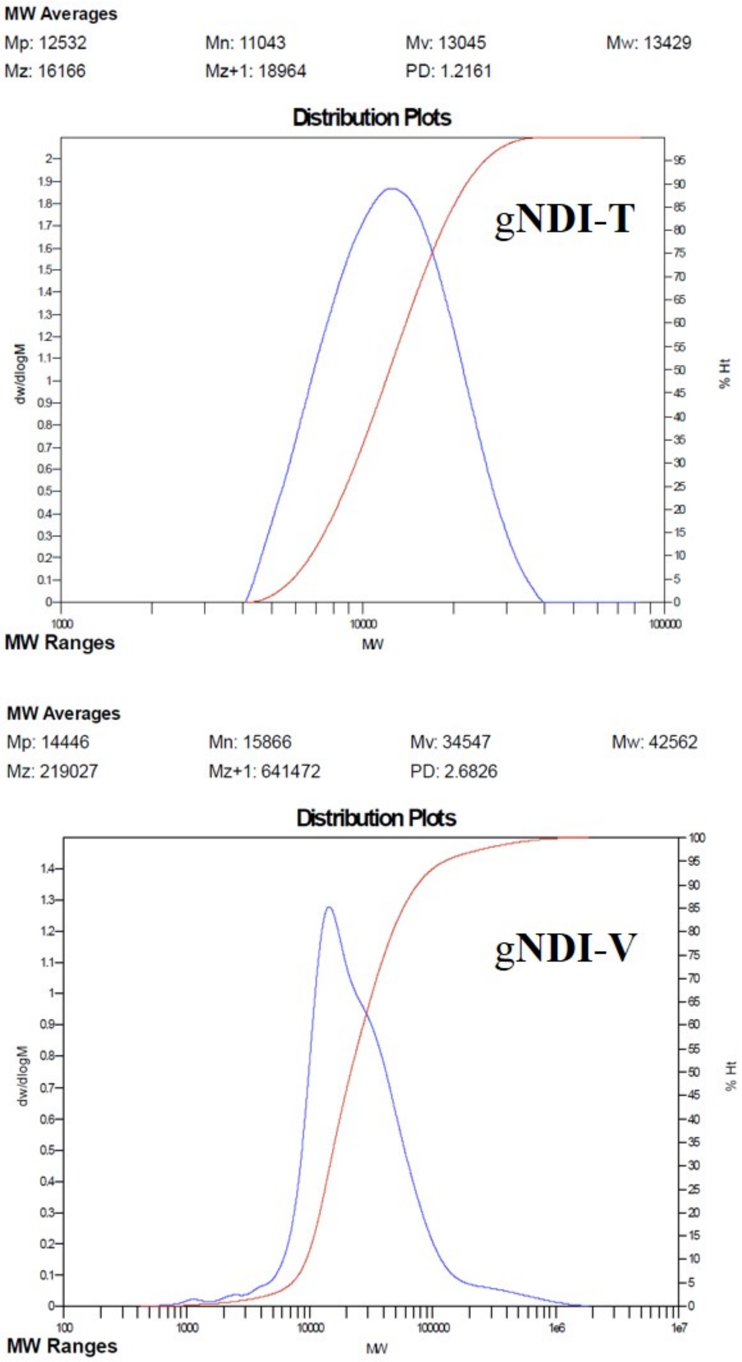


Figure S7. MALDI-TOF data of g**NDI-V** and g**NDI-T**



MW Averages

Mp: 12532
Mn: 11043
Mv: 13045
Mw: 13429
Mz: 16166
Mz+1: 18964
PD: 1.2161

MW Averages

Mp: 14446
Mn: 15866
Mv: 34547
Mw: 42562
Mz: 219027
Mz+1: 641472
PD: 2.6826

Figure S8. GPC data of g**NDI-T** and g**NDI-V**.

4. Thermogravimetric Analysis

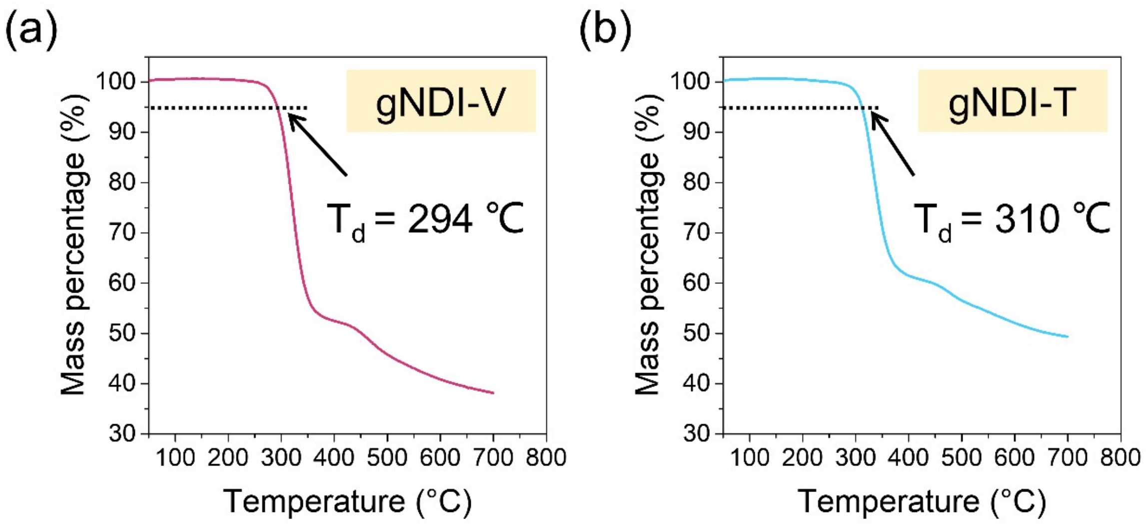


Figure S9. TGA analysis of (a) g**NDI-V** and (b) g**NDI-T** under nitrogen atmosphere at a scan rate of 10 °C min^{-1}.

5. GIWAXS

Table S1. Pristine state packing parameters for the NDI-based copolymers

Sample	**orientation**	**peak**	**q a) [Å^{-1}]**	**π–π distance b) [Å]**	**FWHM [Å^{-1}]**	**L_c c) [Å]**	***g* d)**
g**NDI-V**	Out-of-plane	(010)	1.75	3.59	0.38	14.87	0.19
	In-plane	–	–	–	–	–	–
g**NDI-T**	Out-of-plane	(100)	0.25	25.12	0.07	80.74	–
		(200)	0.47	13.36	0.11	51.38	–
		(010)	1.58	3.97	0.42	13.46	0.21
	In-plane	(001)	0.61	10.30	0.07	80.74	–
		(010)	1.58	3.97	0.39	14.49	0.20

a) Peak location; b) π–π stacking distance $d = 2\pi/q$; c) Correlation length $L_c = 2\pi K/\mathrm{FWHM}$, $K=0.9$; d) Paracrystalline disorder $g = (\mathrm{FWHM}/2\pi q)^{1/2}$.

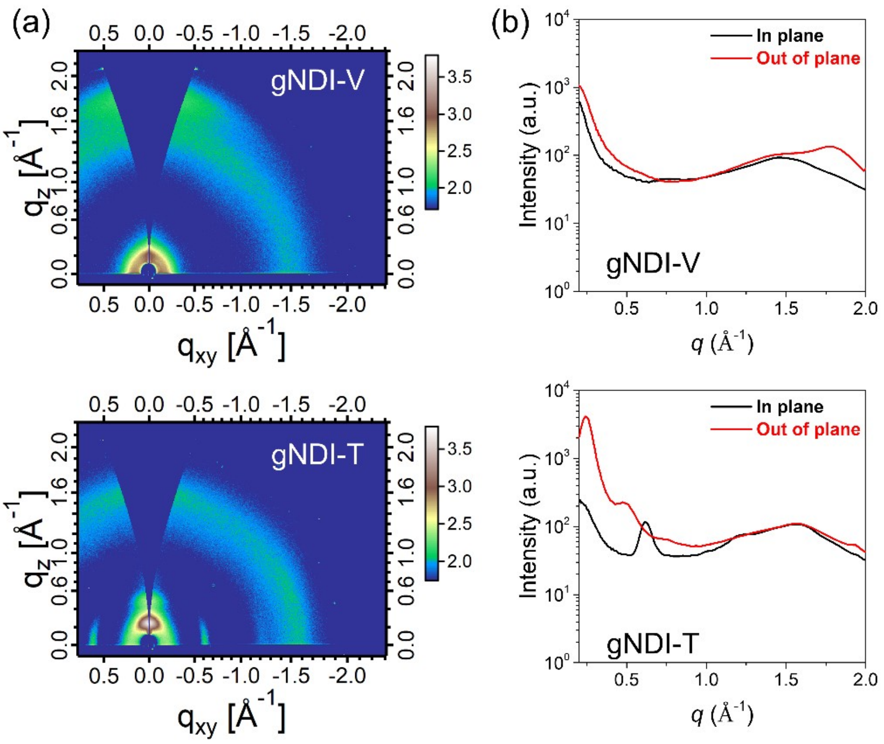


Figure S10. 2D GIWAXS patterns of doped/dedoped films of g**NDI-V** and g**NDI-T**, corresponding line-cut profiles along the in-plane and out-of-plane directions;

Table S2. Reduced state packing parameters for the NDI-based copolymers

Sample	**orientation**	**peak**	**q a)** **[Å^{-1}]**	**π–π distance b)** **[Å]**	**FWHM** **[Å^{-1}]**	**L_c c)** **[Å]**	***g* d)**
g**NDI-V**	Out-of-plane	(010)	1.76	3.57	0.44	10.87	0.20
	In-plane	–	–	–	–	–	–
g**NDI-T**	Out-of-plane	(100)	0.24	26.17	0.08	70.65	–
		(200)	0.47	13.36	0.18	31.40	–
		(010)	1.58	3.97	0.47	12.03	0.22
	In-plane	(001)	0.62	10.13	0.07	80.74	–
		(010)	1.58	3.97	0.41	13.79	0.20

a) Peak location; b) π–π stacking distance $d = 2\pi/q$; c) Correlation length $L_c = 2\pi K/\text{FWHM}$, $K=0.9$; d) Paracrystalline disorder $g = (\text{FWHM}/2\pi q)^{1/2}$.

6. Cyclic Voltammetry

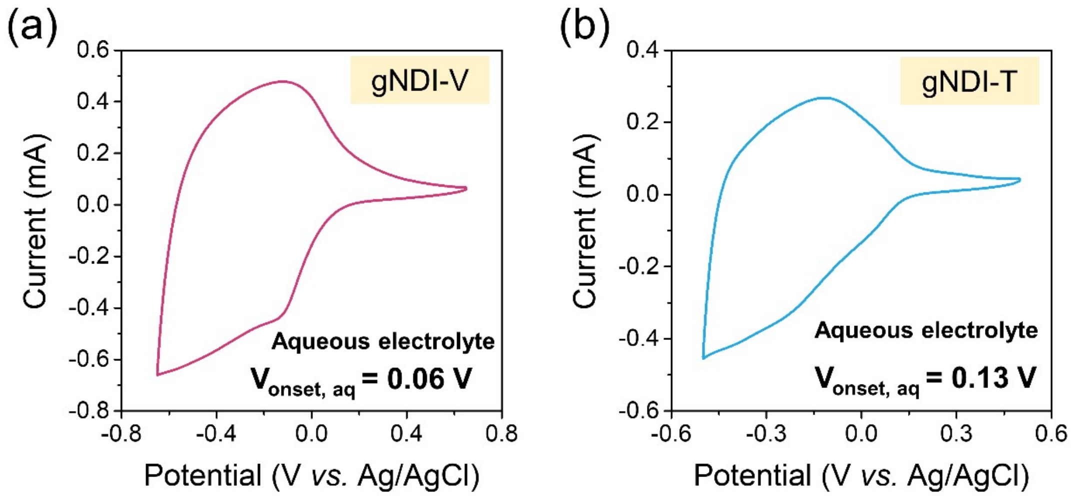


Figure S11. Cyclic voltammetry of (a) g**NDI-V** and (b) g**NDI-T** thin films in 0.1 M KCl solution.

7. Charging Stability

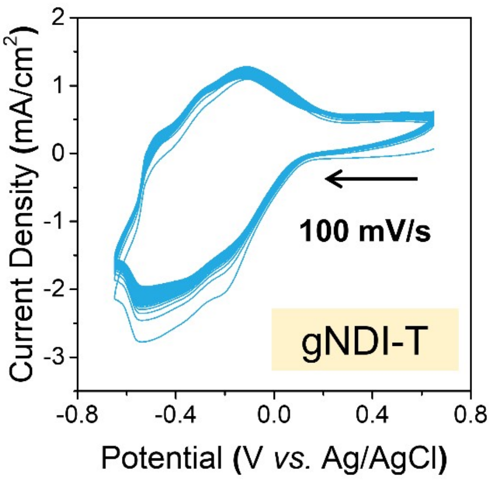


Figure S12. Reversible charging of g**NDI-T** (-0.65 V to 0.65 V) with 0.1 M $KCl_{(aq)}$ electrolyte over 150 cycles at a scan rate of 100 mV/s.

8. Cyclic Voltammetry Measurement at Different Scan Rates

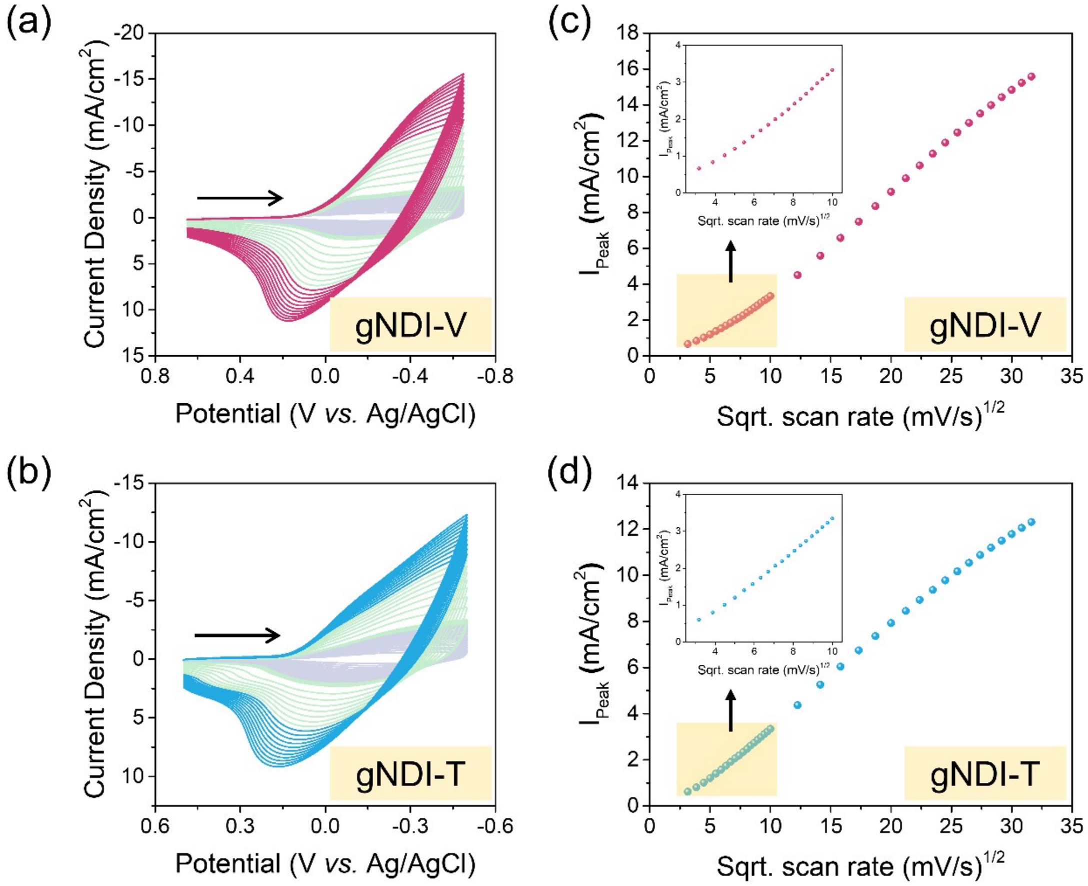


Figure S13. 10-1000 mV/s scan rate dependence CV of (a) g**NDI-V** and (b) g**NDI-T**; The peak current (I_{peak}) recorded for thin film (c) g**NDI-V** and (d) g**NDI-T** as a function of the square roots of scan rates. All measurements were conducted in 0.1 M KCl aqueous electrolyte.

9. Absorption Spectra Difference

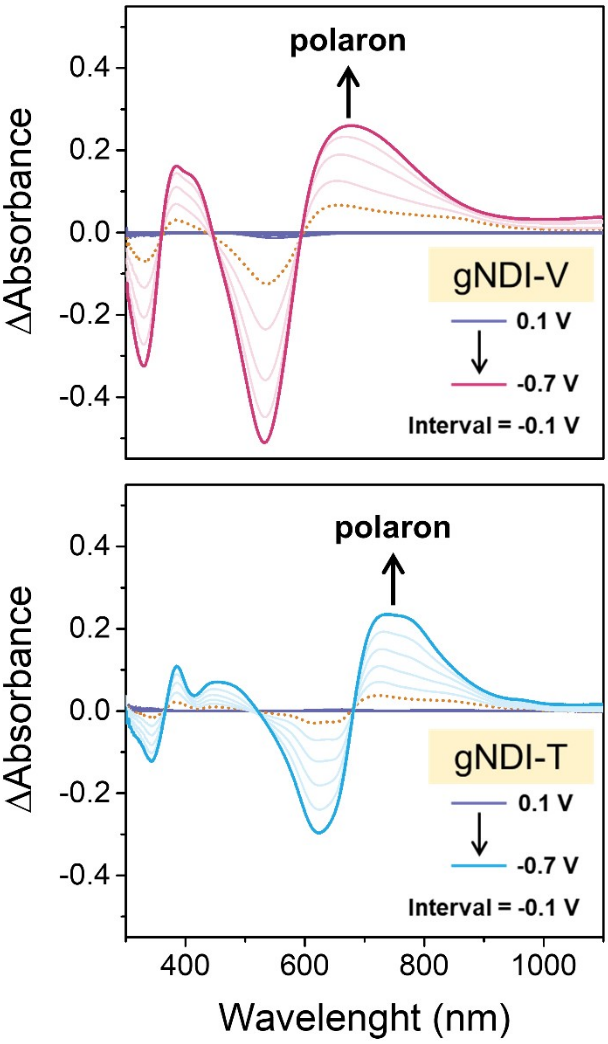


Figure S14. Absorption spectra difference of **gNDI-V** and **gNDI-T** thin film (0.1 V to -0.7 V).

10. Coloration Efficiency

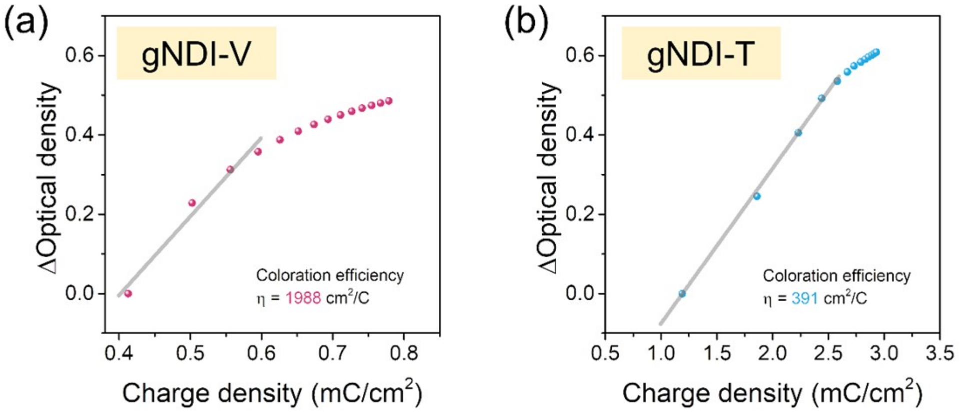


Figure S15. Correlation between the change of optical density (ΔOD) versus injected charge density for (a) g**NDI-V** and (b) g**NDI-T**-based ECDs.

Table S3. Comparison of electrochromic characteristics for copolymer-based ECDs

Copolymer	**ΔT_{max} a) [%]**	**T_c b) [s]**	**T_b c) [s]**	**η d) [cm^2/C]**	**Reference**
gNDI-V	**~ 30**	**0.4**	**0.4**	**~ 1988**	**This work**
gNDI-T	**~ 19**	**1.5**	**0.4**	**~ 391**	
gNDI-EDBT	~ 20	1.4	6.3	-	*Adv. Funct. Mater.* **2022**, 32, 2201821
gNDI-BT	~ 44	1.6	6.3	-	
gNDI-FBT	~ 21	1.7	6.3	-	
PDTBF-Th	~ 32	1.8	1.0	~ 165	*Dyes and Pigments.* **2022**, 204, 110432
PDTBF-ThSe	~ 33	1.6	1.3	~ 256	
PDTBF-DTh	~ 48	3.4	3.1	~ 149	
PIC	~ 33	55.1	4.7	~ 694	*Chemical Engineering Journal.* **2022**, 433, 133808
PID	~ 44	30.8	0.4	~ 733	
PIDF	~ 46	5.6	0.6	~ 1563	
PTCD-1	~ 48	1.3	0.4	~ 541	*Organic Electronics.* **2022**, 105, 106514
PTCD-2	~ 53	0.8	0.4	~ 657	
PBTz-E	~ 31	1.4	0.6	~ 774	*New J. Chem.* **2022**, 46, 16684–16692
P2F-BTz-E	~ 42	2.2	0.5	~ 150	

a) The maximum transmittance change; b) Coloration time; c) Bleaching time; d) Composite colouration efficiency.

Table S4. The detailed OECT parameters for polymers.

Copolymer	**d a) [nm]**	**$I_{on/off}$**	**V_{Th} b) [V]**	**$\tau_{on,\ norm}$ c) [ms/μm^2]**	**$\tau_{off,\ norm}$ c) [ms/μm^2]**	**$T_{on,\ norm,\ 90\%}$ d) [ms/μm^2]**	**$T_{off,\ norm,\ 90\%}$ d) [ms/μm^2]**
g**NDI-V**	119 ± 7	~ 10^5	0.3	2.90	0.32	7.61	0.65
g**NDI-T**	145 ± 5	~ 10^4	0.2	0.87	0.18	2.33	0.42

a) Thin film thickness; b) Determined by extrapolating the corresponding $I_{DS}^{1/2}$~V_{GS} plots. c) Normalized on/off-time constant $\tau_{norm} = \tau/(d(WL)^{1/2})$; d) Normalized rise/fall time $T_{norm,\ 90\%} = T/(d(WL)^{1/2})$.

11. Threshold Voltage

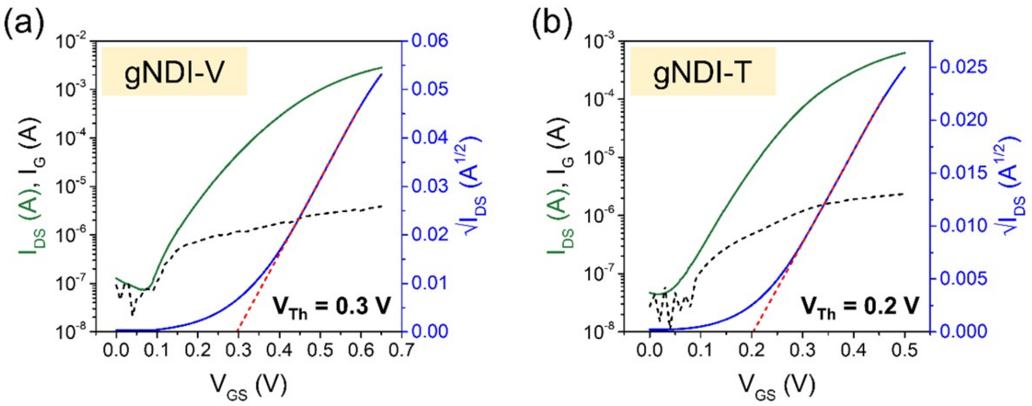


Figure S16. The square roots of I_{DS} (right axis) as a function of gate bias for (a) g**NDI-V** and (b) g**NDI-T** to obtain the on-off ratio as well as threshold voltage. Note that, the black dot lines represent the leakage currents.

12. Transient Response

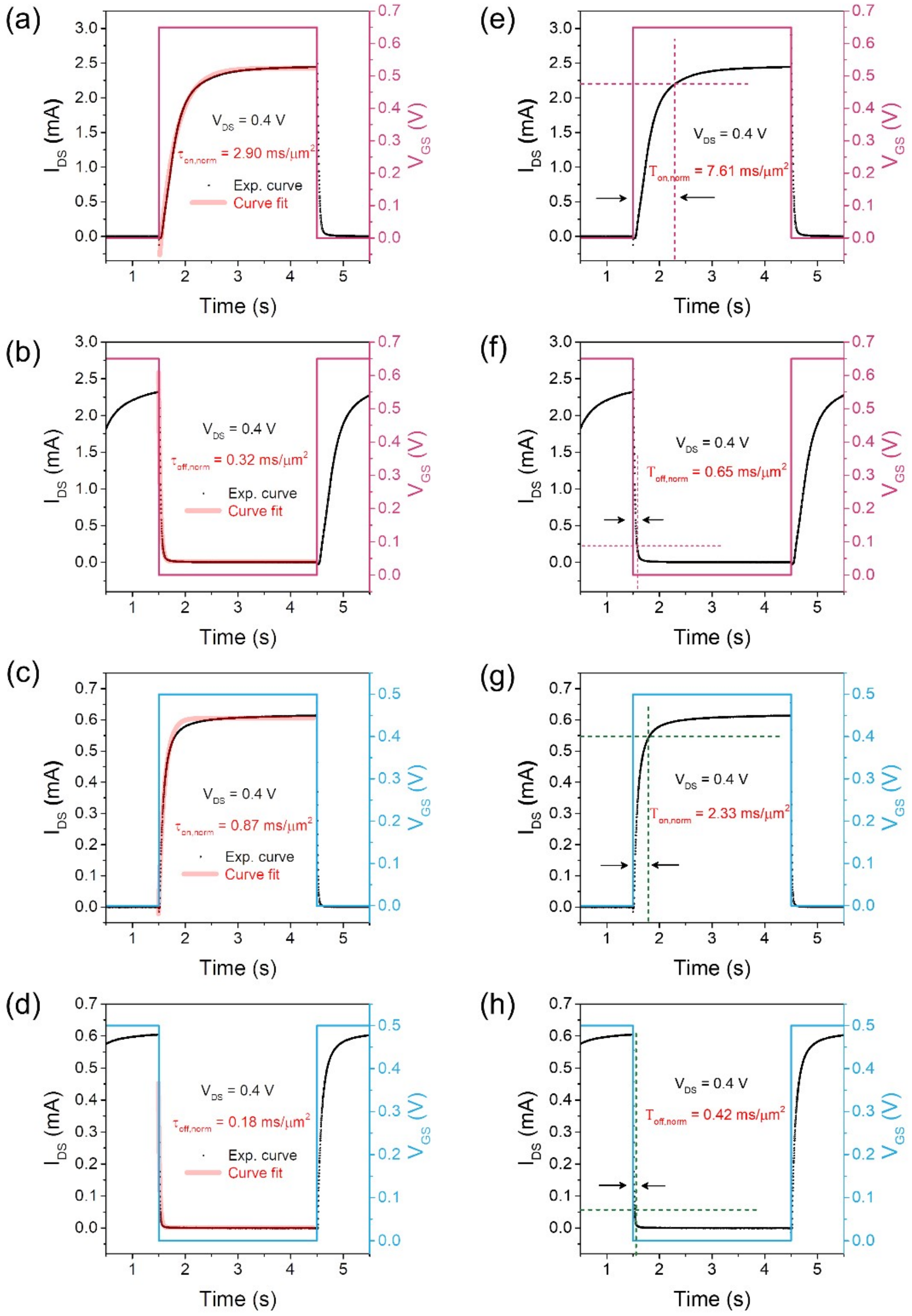


Figure S17. Transient response: Fitted on-time constant $\boldsymbol{\tau_{on}}$ (a), off-time constant $\boldsymbol{\tau_{off}}$ (b), rise time $\mathbf{T_{on,\ 90\%}}$ (e) and fall time $T_{off,\ 90\%}$ (f) for g**NDI-V** (pink line); Fitted on-time constant $\boldsymbol{\tau_{on}}$ (c), off-time constant $\boldsymbol{\tau_{off}}$ (d), rise time $\mathbf{T_{on,\ 90\%}}$ (g) and fall time $T_{off,\ 90\%}$ (h) for g**NDI-V** (blue line); Note that the response time was calculated by two methods: one is fitted by the exponential decay function and the other one is measured the time that it takes for channel current (I_{DS}) to reach 90% of its max ($T_{on,\ 90\%}$) and min ($T_{off,\ 90\%}$) value. Note that the response time was normalized by channel geometry ($\tau_{norm} = \tau/(d\times(W\times L)^{1/2})$ and/or $T_{norm,\ 90\%} = T/(d\times(W\times L)^{1/2})$).

13. Electrochemical Impedance Spectroscopy

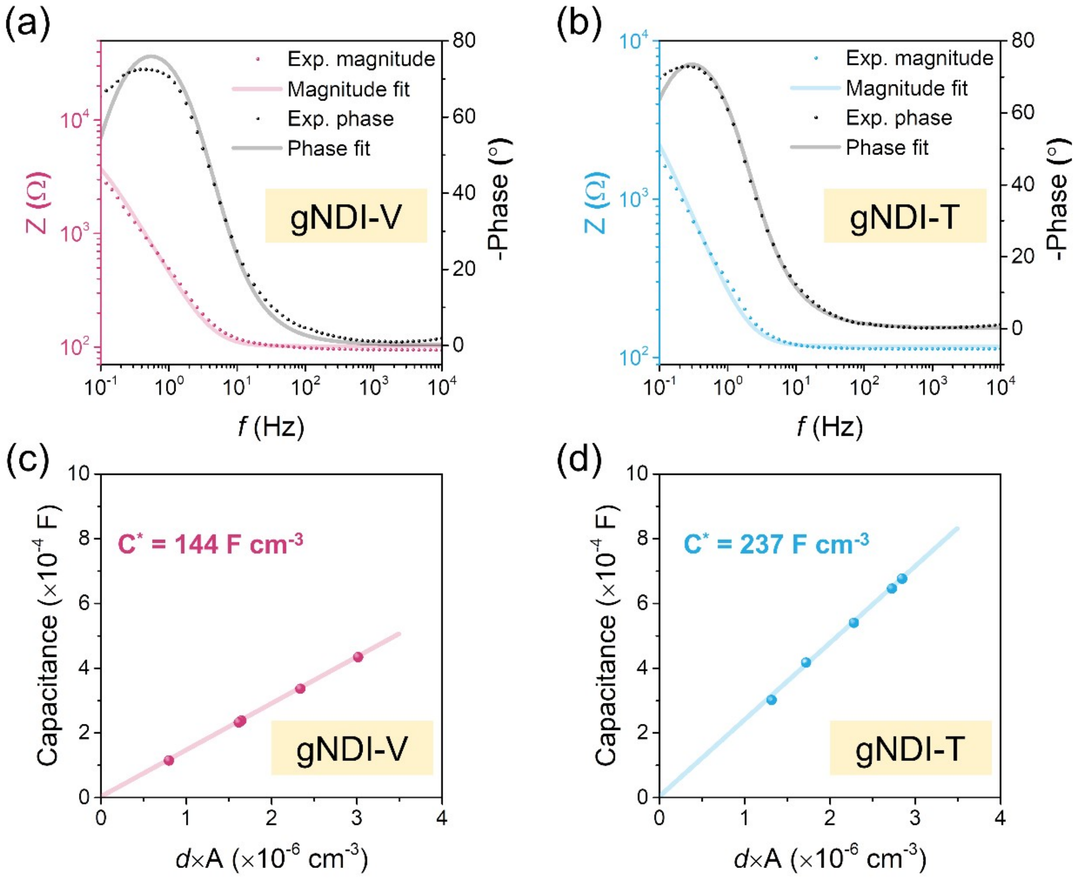


Figure S18. Bode plots acquired using the (a) g**NDI-V** and (b) g**NDI-T** film as a working electrode and the data was fitted with an equivalent circuit R_s (R_p //C). The volumetric capacitance (C^*) of (c) g**NDI-V** and (d) g**NDI-T** was obtained from the slope of the extracted capacitance versus film volume plot.

14. Device Performance Reproducibility

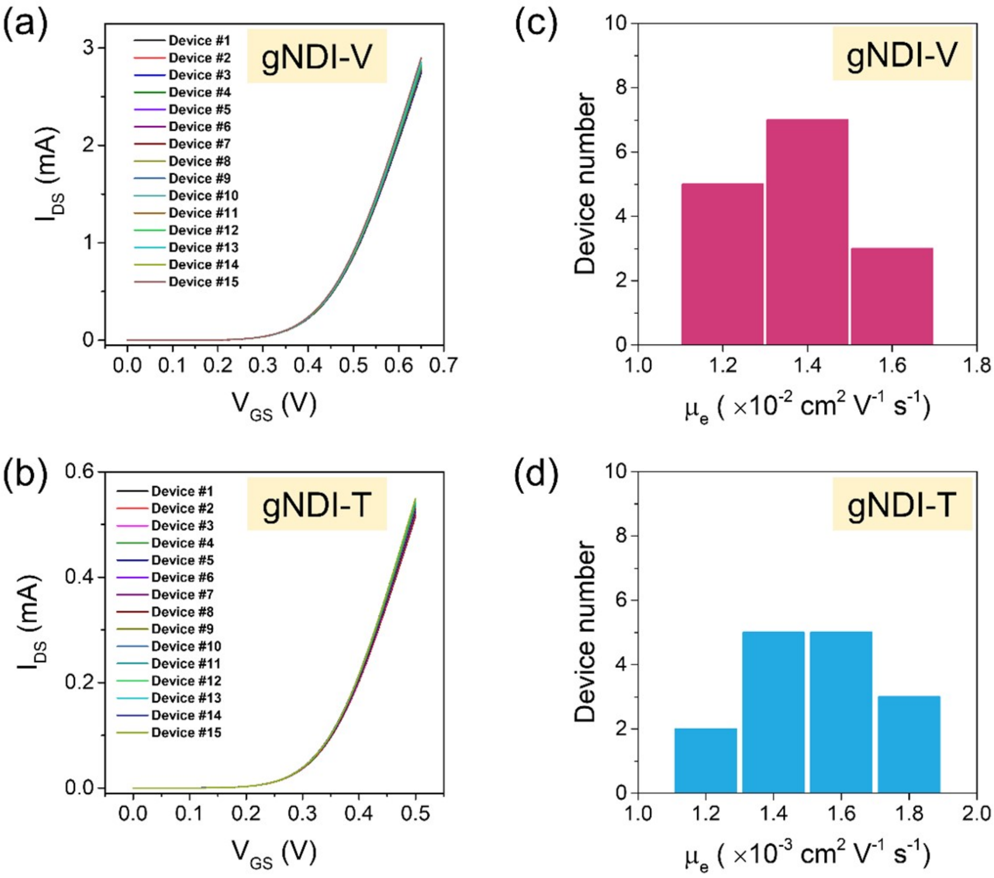


Figure S19. OECT device performance reproducibility based on a) g**NDI-V** and b) g**NDI-T**; Statistics of the electron mobility for 15 individual devices processed with c) g**NDI-V** and d) g**NDI-T**. Note that the electron mobility was calculated by the equation: $g_m = W L^{-1} d \mu_e C^* (V_g - V_{Th})$.

Table S5. The reported n-type OECTs performance

Category	Polymers	μC^* [F V^{-1} cm^{-1} s^{-1}]	μ_e [cm^2 V^{-1} s^{-1}]	C^* [F cm^{-3}]	$g_{m,norm}$ [g_m/(WdL^{-1})] [S cm^{-1}]	Reference
NDI-based D–A conjugated polymers	gNDI-V	2.31	1.4×10^{-2}	144	0.42	This work
	gNDI-T	0.42	1.5×10^{-3}	237	0.11	
	p(C$_4$-T2)	0.30	1.90×10^{-3}	158	0.063	*ACS Appl. Mater. Interfaces.* **2021**, 13, 3, 4253
	p(C$_4$-T2-OMe)	0.07	3.87×10^{-4}	170	0.010	
	p(C$_4$-T2-C$_0$-EG)	0.22	1.16×10^{-3}	188	0.031	
	p(C$_4$-T2-C$_2$-EG)	0.01	4.99×10^{-5}	200	0.002	
	p(C$_4$-T2-C$_4$-EG)	0.006	5.34×10^{-5}	116	0.001	
	p(C$_2$-T2)	0.20	3.97×10^{-4}	492	0.04	
	p(C$_6$-T2)	1.29	4.74×10^{-3}	272	0.228	
	p(C$_8$-T2)	0.13	3.76×10^{-4}	342	0.015	
	p(gNDI-gT2)	0.06	2.2×10^{-4}	221	0.013	*Adv. Funct. Mater.* **2021**, 31, 2008718.
	p(C3-gNDI-gT2)	0.13	9.2×10^{-4}	72	0.034	
	p(C6-gNDI-gT2)	0.16	6.3×10^{-4}	59	0.037	
	P(NDITEG-T)	0.04	2.4×10^{-4}	165.8	0.008	*Adv. Funct. Mater.* **2022**, 2111950.
	P(NDIDTEG-T)	0.21	9.4×10^{-4}	221.5	0.034	
	P(NDIDEG-T)	0.51	2.12×10^{-3}	239.9	0.09	
	P(NDIMTEG-T)	0.56	2.23×10^{-3}	250.9	0.095	
	PNDIODTEG-2Tz	N/A	N/A	N/A	0.1	*Adv. Funct. Mater.***2022**, 2201593
	PNDI2TEG-2Tz	1.16	3.16×10^{-3}	367	0.49	
	p(NDI-T2-L2)	0.037	3.89×10^{-4}	95	0.0084	*Mater. Horiz.* **2020**, 7, 2348
	P90	0.0343	7.45×10^{-5}	261.5	0.009	*Adv. Funct. Mater.* **2021**, 31, 2010165.
Ladder type BBL-based polymers	BBL	1.99	2.14×10^{-3}	731	0.815	
	BBL15	1.94	3.59×10^{-3}	540	0.617	*Adv.Mater.* **2022**, 34, 2106235.
	BBL60	4.90	9.42×10^{-3}	520	1.92	
	BBL98	10.2	2.05×10^{-2}	499	4.04	
	BBL152	25.9	4.40×10^{-2}	589	11.1	
Lactone-based polymers	p(C-T)	6.7	6.9×10^{-2}	97	0.80	*Adv. Funct. Mater.* **2021**, 2111439.
	p(N-T)	4.3	5.9×10^{-2}	73	0.72	
	p(C-2T)	1.0	1.9×10^{-2}	53	0.14	
Fused electron-deficient lactam backbone	PgNaN	0.662	6.50×10^{-3}	100	0.212	*Angew. Chem. Int. Ed.* **2021**, 60, 9368
	PgNgN	0.037	1.89×10^{-4}	239	0.007	
	p(g$_7$NC$_2$N)	0.36	2.0×10^{-3}	180	0.069	*J. Am. Chem. Soc.* **2022**, 144, 10, 4642
	p(g$_7$NC$_4$N)	0.18	1.46×10^{-3}	126	0.035	
	p(g$_7$NC$_6$N)	0.34	2.29×10^{-3}	150	0.065	
	p(g$_7$NC$_8$N)	1.19	6.01×10^{-3}	199	0.240	
	p(g$_7$NC$_{10}$N)	1.83	1.20×10^{-2}	153	0.370	
	p(g$_7$NC$_{12}$N)	0.66	6.50×10^{-3}	100	0.212	
	p(g$_7$NC$_{16}$N)	0.33	3.80×10^{-3}	86	0.047	
bithiophene Imide dimer-based polymers	f-BTI2TEG-T	2.13	4.1×10^{-2}	47	0.26	*Angew. Chem. Int. Ed.* **2021**, 133, 24400
	f-BTI2TEG-FT	14.71	3.2×10^{-2}	421	4.42	
	f-BTI2g-TVT	1.23	1.1×10^{-2}	110	0.23	*Adv.Mater.***2022**, 2201340

	f-BTI2g-TVTCN	38.4	0.23	170	11.9	
DPP-based D-A conjugated polymers	P(gPzDPP-2T)	0.18	1.6×10^{-3}	134	0.048	*Chem. Mater.* **2022**, 34, 2, 864
	P(gPzDPP-CT2)	1.36	1.9×10^{-2}	91	0.434	
	2DPP-OD-TEG	7	4.79×10^{-2}	146.2	0.73	*Adv. Funct. Mater.* **2021**, 31, 2102903.

15. OECT Device Stability

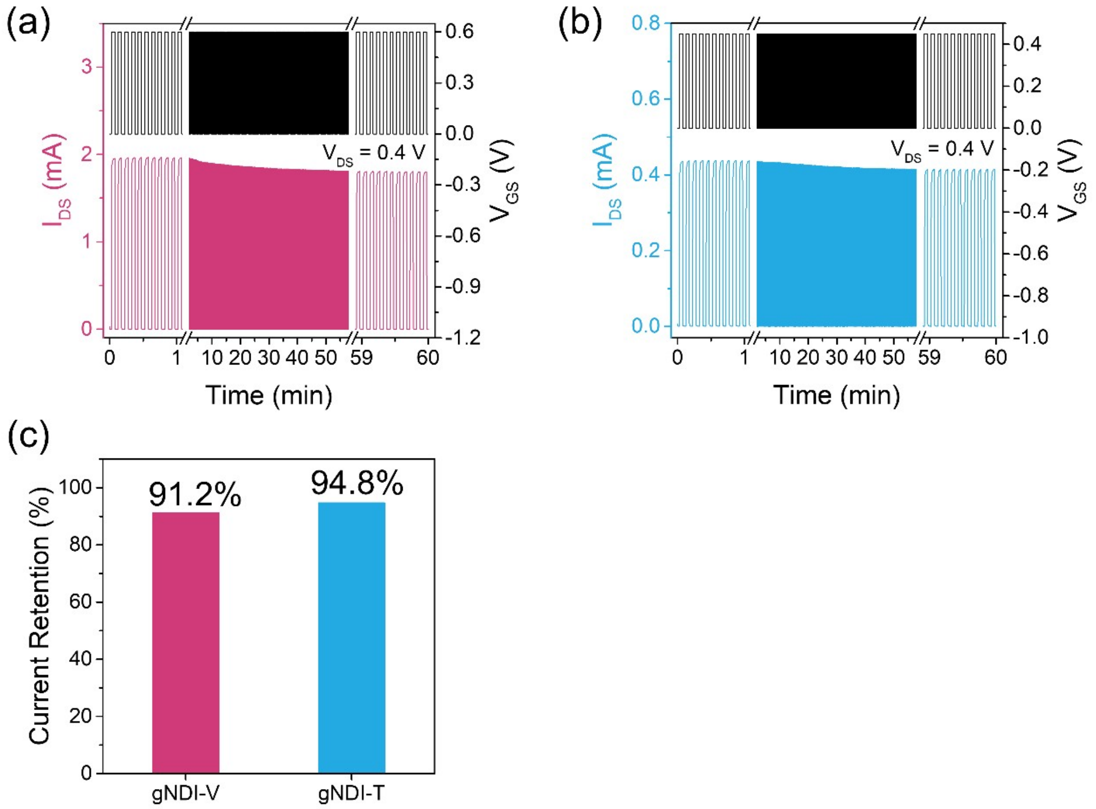


Figure S20. Operation stability of OECTs based on (a) g**NDI-V** (red colour) and (b) g**NDI-T** (blue colour) measured in 0.1 M KCl aqueous solution; (c) The current retention of both copolymers after applying sequential square wave gate voltage for 60 mins.

16. References

[S1] S. Cong, J. Chen, L. Wang, L. Lan, Y. Wang, H. Dai, H. Liao, Y. Zhou, Y. Yu, J. Duan, Z. Li, I. McCulloch, W. Yue, *Adv. Funct. Mater.* **2022**, *32*, 2201821.