

## Supporting Information

### **NIR-Sensing Ambipolar Organic Phototransistors with Conjugated Terpolymer Layers Based on Diketopyrrolopyrrole-Benzothiadiazole-Naphthalenediimide Comonomer Units**

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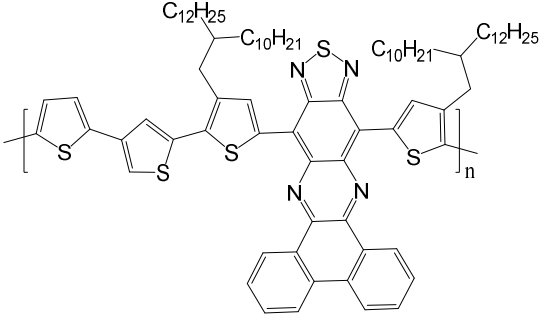
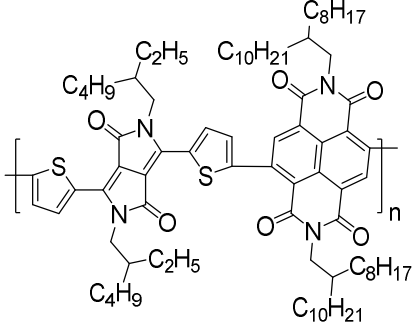
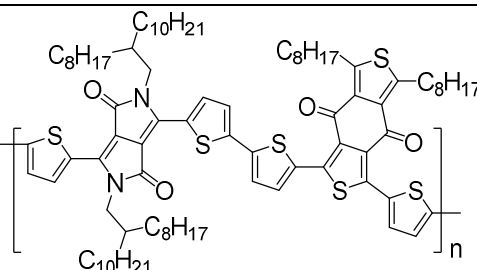
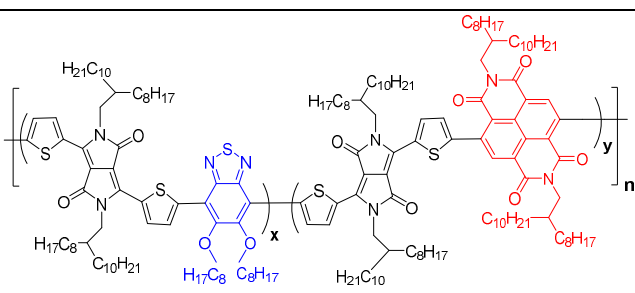
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**Table S1.** Summary of ambipolar-type NIR-absorbing conjugated polymers reported to date.

Authors	Polymer Structure	Device Performances	Refs. (Year)
M. Li, et al.	 <p>PPhTQ (Copolymer)</p>	<p>[Wavelength] 500, 1200 nm</p> <p>[Voltage Window] ±80V</p> <p>[OPTR Application] YES</p>	SR-1 (2015)
P. Wang, et al.	 <p>PNDI-DPP (Copolymer)</p>	<p>[Wavelength] 900 nm</p> <p>[Voltage Window] ±80V</p> <p>[OPTR Application] NO</p>	SR-2 (2015)
G. Zhang, et al.	 <p>P3 (Copolymer)</p>	<p>[Wavelength] 800 nm</p> <p>[Voltage Window] ±80V</p> <p>[OPTR Application] YES</p>	SR-3 (2016)
Y. Cho, et al.	 <p>PDPP-8OBT-NDI (Terpolymer)</p>	<p>[Wavelength] 810 nm, 1100 nm (edge)</p> <p>[Voltage Window] ±10V</p> <p>[OPTR Application] YES</p>	This work (2022)

## **References for Table S1**

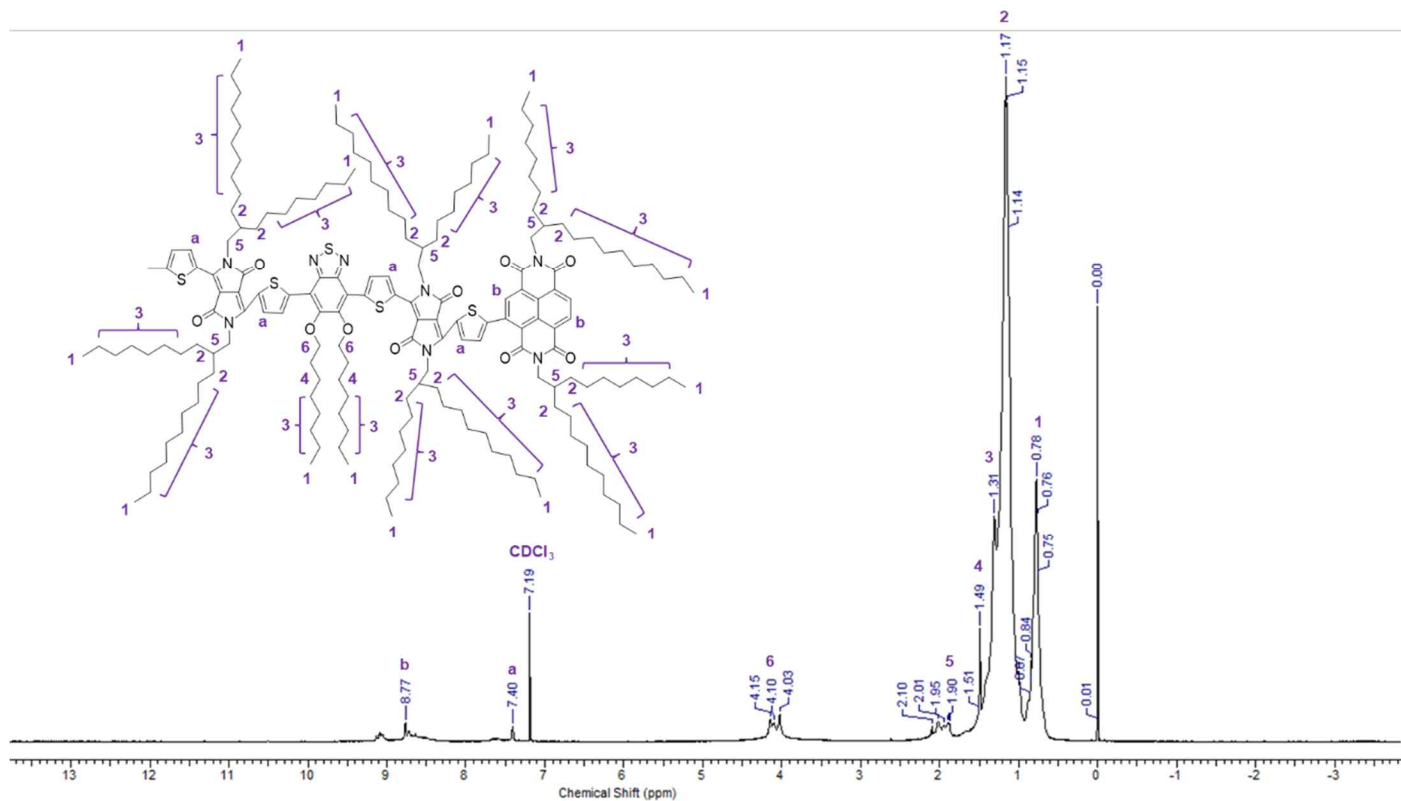
- SR-1. M. Li, C. An, T. Marszalek, X. Guo, Y. Long, H. Yin, C. Gu, M. Baumgarten, W. Pisula, K. Mullen, *Phenanthrene condensed thiadiazoloquinoxaline donor–acceptor polymer for phototransistor applications*, *Electrochem. Commun.* 74 (2017) 33-37.
- SR-2. P. Wang, H. Li, C. Gu, H. Dong, Z. Xu, H. Fu, *Air-stable ambipolar organic field-effect transistors based on naphthalenediimide–diketopyrrolopyrrole copolymers*, *RSC Adv.* 5 (2015) 19520.
- SR-3. G. Zhang, J. Guo, J. Zhang, W. Li, X. Wang, H. Lu, L. Qiu, *Benzodithiophenedione and diketopyrrolopyrrole based conjugated copolymers for organic thin-film transistors by structure modulation*, *Dyes Pigm.* 126 (2016) 20-28.

**Table S2.** Summary of device parameters under illumination with NIR light ( $\lambda = 810$  nm) for the OPTRs with the PDPP-8OBT-NDI sensing channel layers. Note that data were taken from the transfer curves in Figure 4a ( $V_G = V_D = \pm 10$  V), respectively.

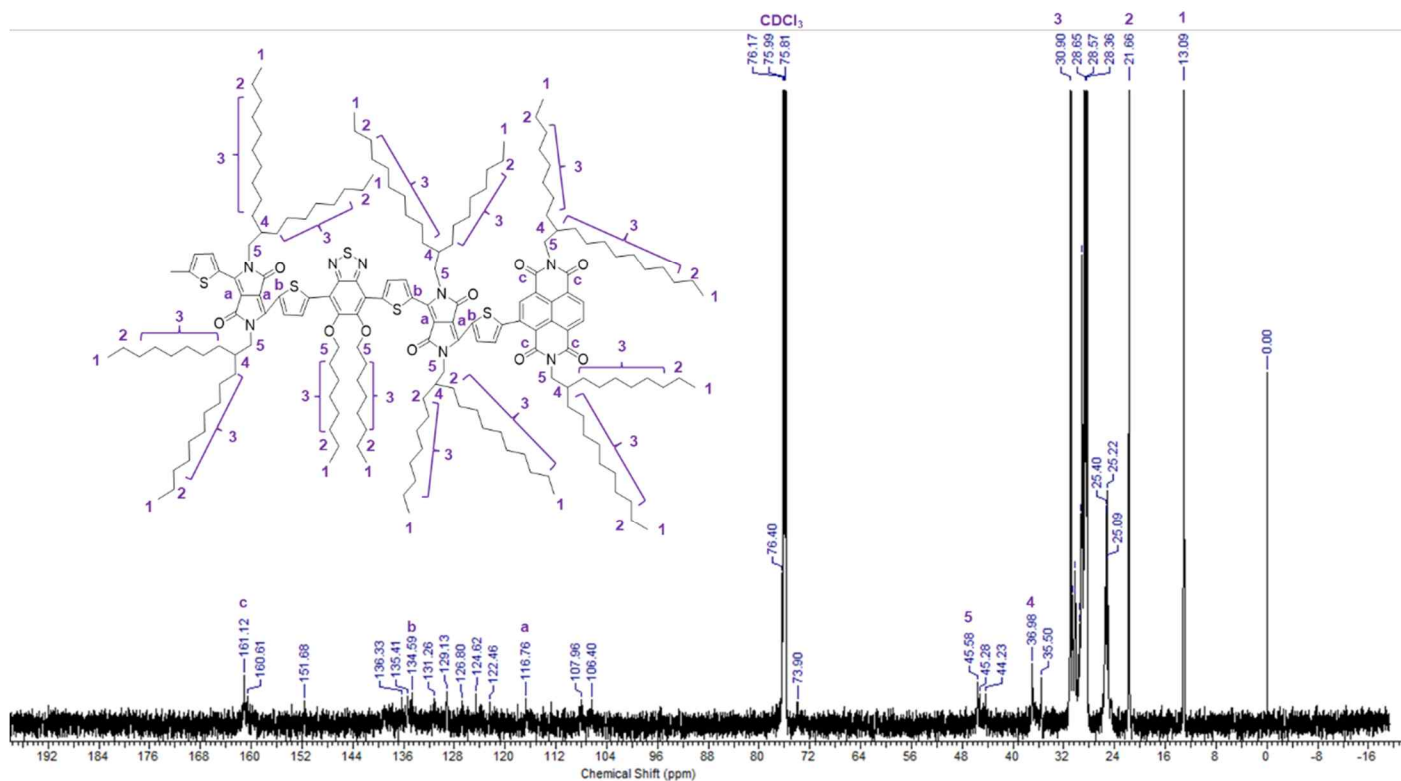
$P_{IN}$ ( $\mu W/cm^2$ )	$t$ (nm)	Channel	$\Delta V_{TH}$ (V)	$S_p$ (%)	$R_C$ (mA $W^{-1}$ )	DCR
56	40	p	-3.24	78.04	0.33	1.10
		n	3.38	36.02	0.16	
	80	p	-2.25	169.60	1.14	1.21
		n	2.81	165.57	0.98	
169	40	p	-2.87	114.19	0.16	1.11
		n	2.90	62.99	0.10	
	80	p	-2.03	249.01	0.48	1.22
		n	2.43	206.55	0.46	
394	40	p	-2.33	195.85	0.11	1.12
		n	2.37	124.26	0.08	
	80	p	-1.98	335.29	0.29	1.23
		n	2.13	294.26	0.27	
563	40	p	-2.17	245.38	0.10	1.14
		n	2.34	163.48	0.75	
	80	p	-1.25	413.72	0.25	1.27
		n	1.78	373.36	0.24	

**Table S3.** Summary of device parameters under illumination with NIR light ( $\lambda = 905$  nm) for the OPTRs with the PDPP-8OBT-NDI sensing channel layers. Note that data were taken from the transfer curves in Figure 4a ( $V_G = V_D = \pm 10$  V), respectively.

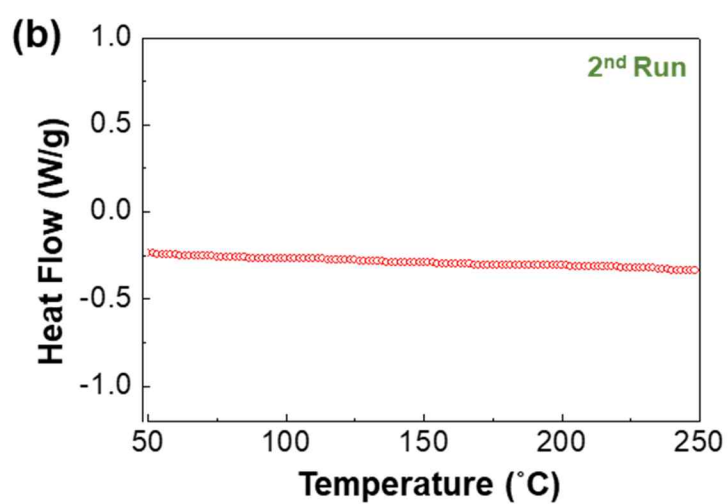
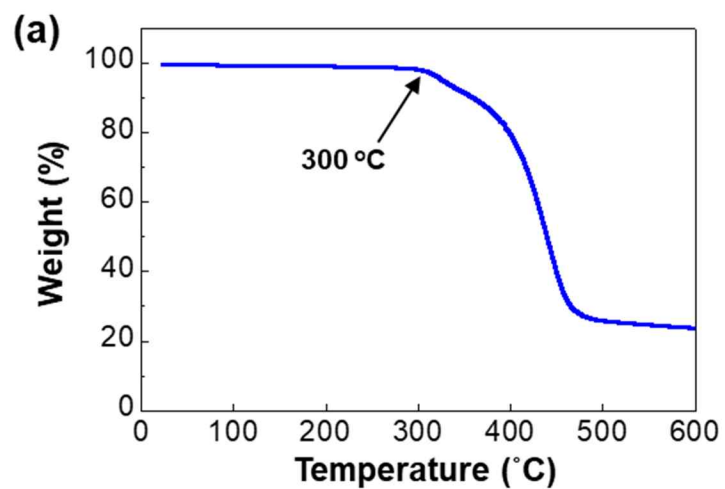
$P_{IN}$ ( $\mu W/cm^2$ )	$t$ (nm)	Channel	$\Delta V_{TH}$ (V)	$S_p$ (%)	$R_C$ (mA $W^{-1}$ )	DCR
40	40	p	-3.26	261.78	1.37	1.10
		n	3.81	180.70	1.03	
	80	p	-2.54	203.92	1.63	1.21
		n	2.59	190.98	1.45	
136	40	p	-2.84	303.17	0.52	1.14
		n	3.37	222.30	0.42	
	80	p	-2.51	360.78	0.92	1.24
		n	2.55	325.40	0.85	
319	40	p	-2.31	419.41	0.31	1.16
		n	2.37	311.76	0.25	
	80	p	-2.49	495.09	0.58	1.25
		n	2.54	481.96	0.50	
456	40	p	-2.28	522.48	0.27	1.18
		n	2.32	415.93	0.24	
	80	p	-2.38	665.57	0.56	1.27
		n	2.47	622.54	0.44	



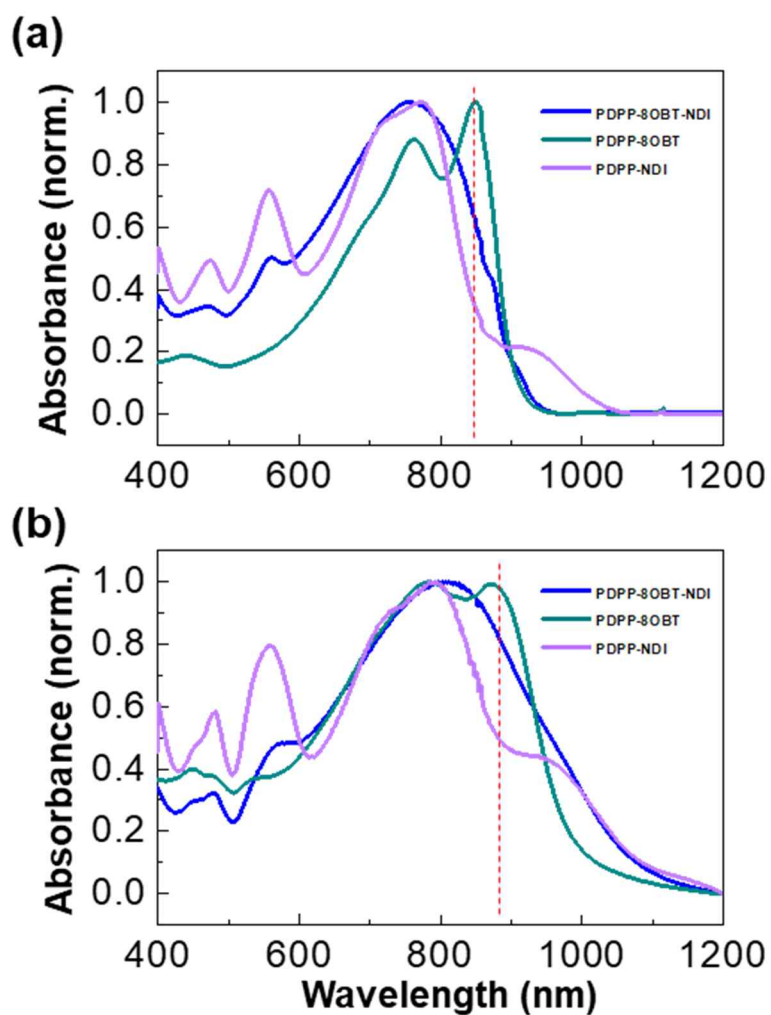
**Figure S1.**  $^1\text{H-NMR}$  (700 MHz) spectra for the PDPP-8OBT-NDI polymer (solvent:  $\text{CDCl}_3$ ) synthesized in this work. A detailed interpretation of the major peaks is given in the experimental section.



**Figure S2.**  $^{13}\text{C-NMR}$  (700 MHz) spectra for the PDPP-8OBT-NDI polymer (solvent:  $\text{CDCl}_3$ ) synthesized in this work. A detailed interpretation of the major peaks is given in the experimental section.

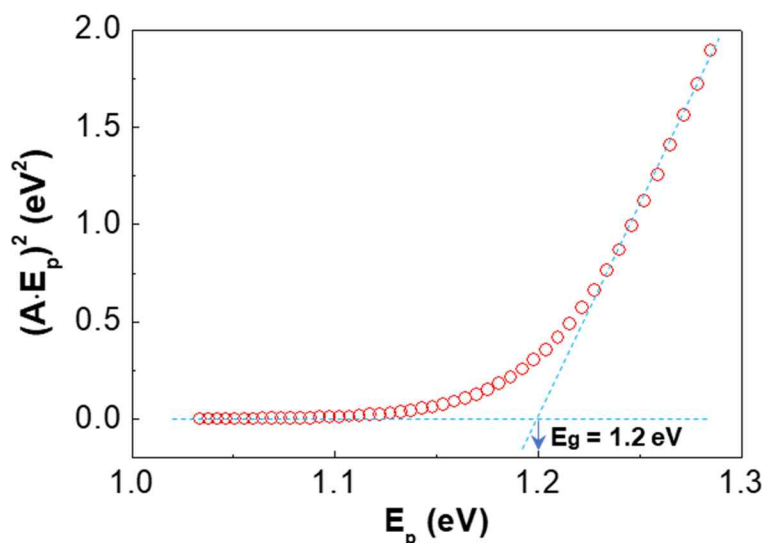


**Figure S3.** TGA (a) and DSC (b) thermograms (nitrogen environment) for the PDPP-8OBT-NDI polymer. The TGA measurement was carried out by heating at a ramp rate of 10 °C/min, while the DSC measurement was performed by scanning at a heating rate of 5 °C/min (second run).

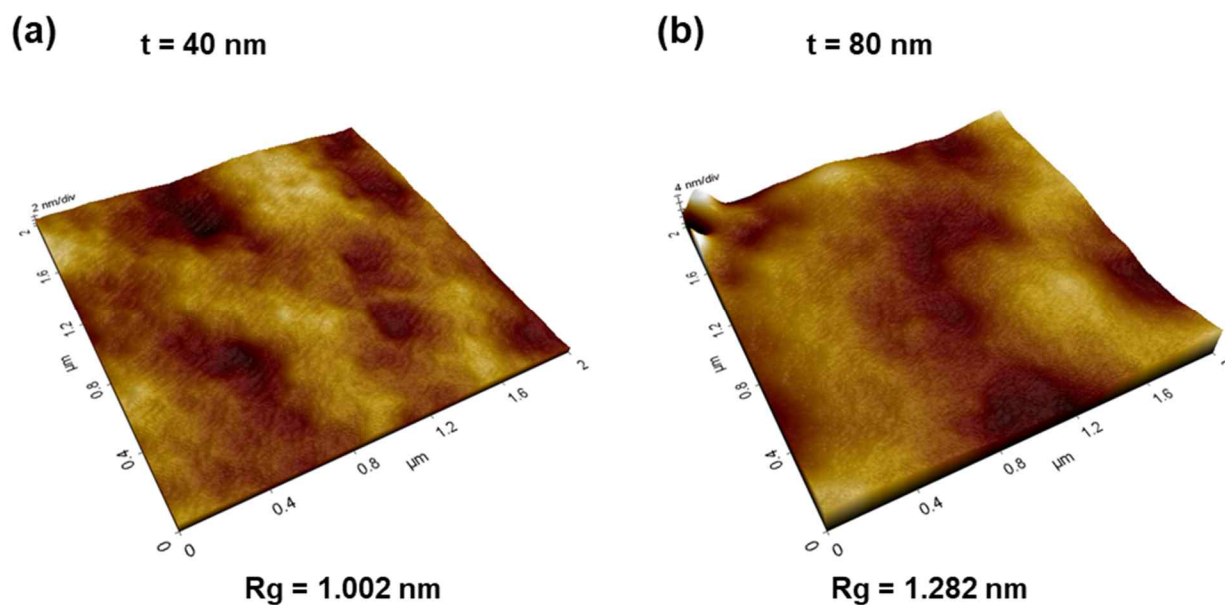


**Figure S4.** Optical absorption spectra of PDPP-8OBT, PDPP-NDI, and PDPP-8OBT-NDI polymers: (a) solutions (solvent: toluene), (b) films. On the basis of optical absorption spectra, the composition ratio of 8OBT to NDI comonomer in the PDPP-8OBT-NDI chains was calculated to 8OBT:NDI = 0.54:0.46 (from solutions) and 0.62:0.38 (from films).





**Figure S5.** Tauc plot of the PDPP-8OBT-NDI film (original data: from the optical absorption spectrum in Figure 1b).  $A$  and  $E_p$  denote absorbance and photon energy, respectively.



**Figure S6.** AFM (height-mode,  $2\ \mu\text{m} \times 2\ \mu\text{m}$ ) images of the PDPP-8OBT-NDI films: (a)  $t = 40\ \text{nm}$ , (b)  $t = 80\ \text{nm}$ . Note that the root-mean-square (rms) roughness ( $R_g$ ) from the AFM images was measured in the range of ca.  $1.0 \sim 1.3\ \text{nm}$ , which corresponds to ca.  $1.6 \sim 2.5\%$  compared to the thickness of films.