Electronic Supplementary Information (ESI)

Boosting Photoprogramming Performance of Molecular-switch-embedded Organic Transistors via Structural Optimization of Polymer Semiconductors

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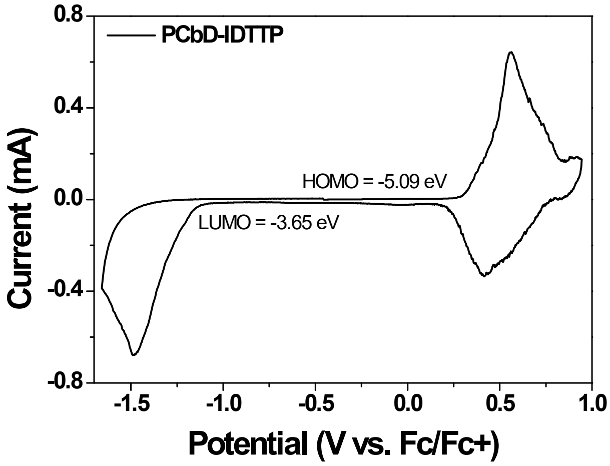

Fig. S1. CV profile of PCbD-IDTTP.

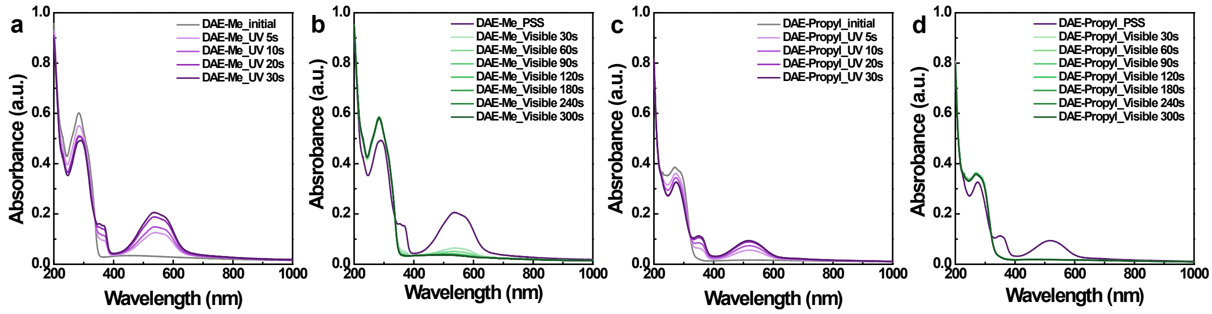

Fig. S2. UV-vis absorption spectra of a) DAE-Me film irradiated upon continuous UV light exposure time, b) DAE-Me film irradiated upon continuous visible light exposure time, c) DAE-Propyl film irradiated upon continuous UV light exposure time and d) DAE-Propyl film irradiated upon continuous visible light exposure time.

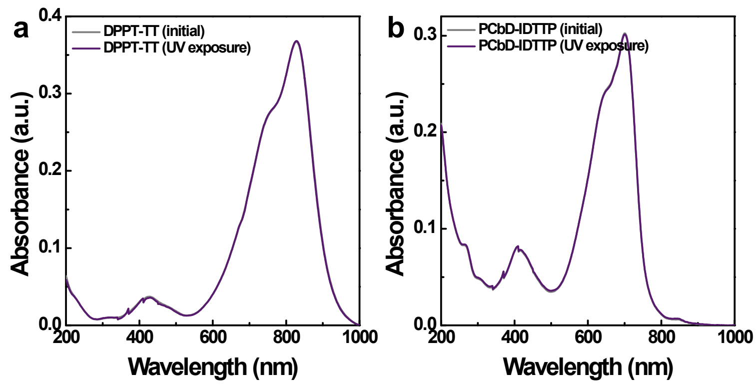

Fig. S3. UV-vis absorption spectra of a) DPPT-TT film and b) PCbD-IDTTP film.

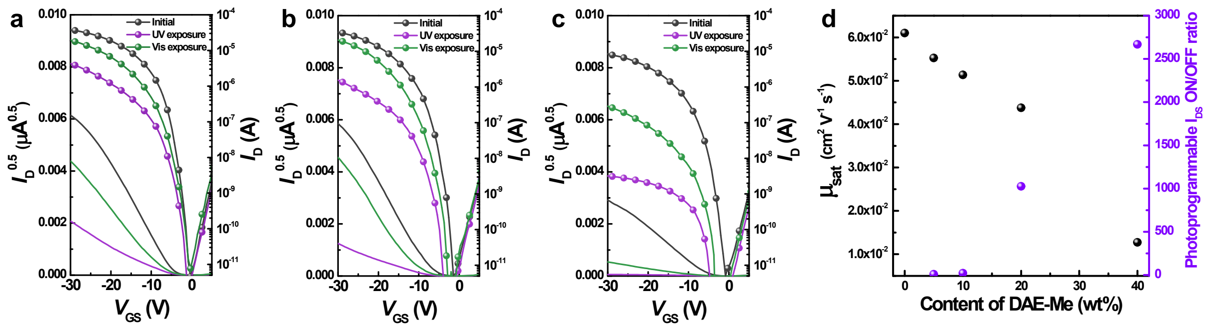

Fig. S4. Photoprogrammable switching behaviour of FETs based PCbD-IDTTP with a) 5 wt%, b) 10 wt% and c) 40 wt% of DAE-Me. d) Changes in saturation mobility and photoprogrammable I_{DS} ON/OFF ratio according to DAE-Me content. All values were extracted from transfer curves of Fig. S4 a-c and Fig. 5d at V_{GS}=-30 V and V_{DS}=-30 V.

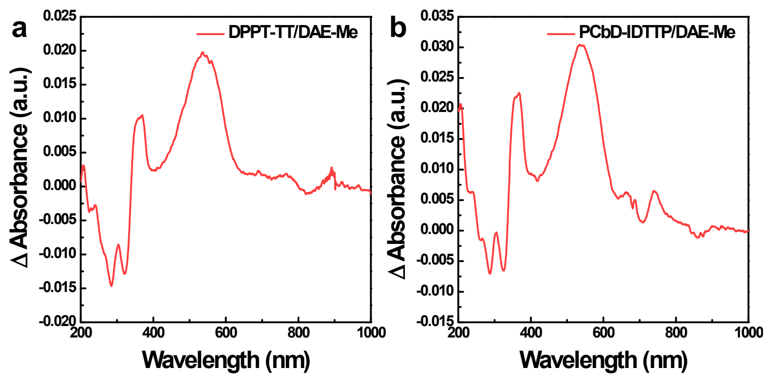

Fig. S5. The differential changes in absorbance after UV exposure (312 nm) for a) DPPT-TT/DAE-Me and b) PCbD-IDTTP/DAE-Me film.

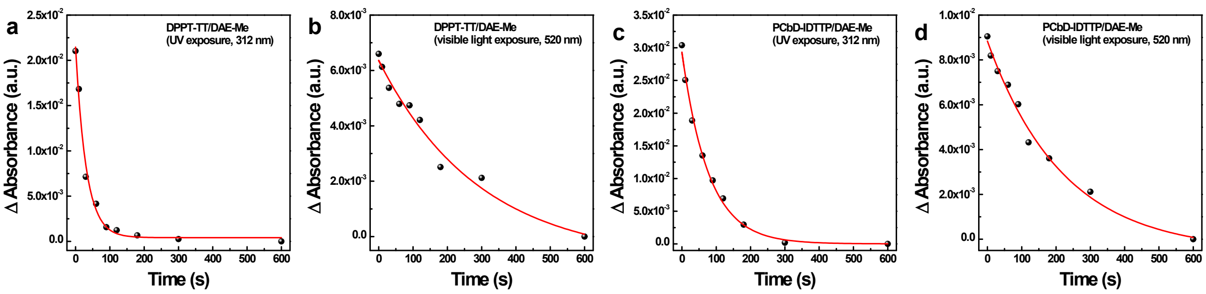

Fig. S6. Changes in absorbance at 537 nm as a function of irradiation time (black symbol) and fitted curves (red line) in a and b) DPPT-TT/DAE-Me and c and d) PCbD-IDTTP/DAE-Me films.

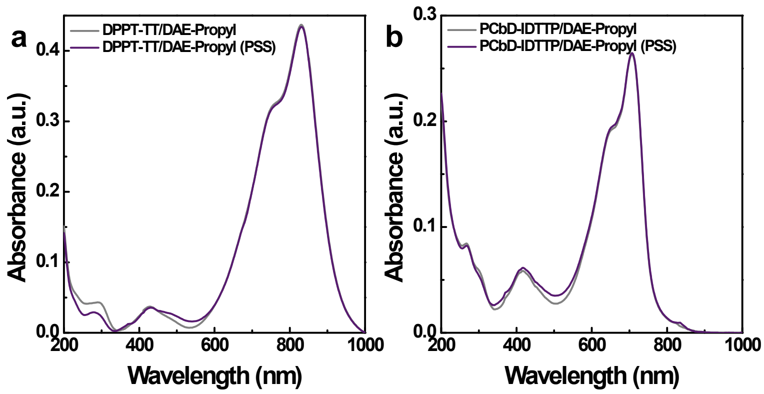

Fig. S7. UV-vis absorption spectra of a) DPPT-TT/DAE-Propyl film and b) PCbD-IDTTP/DAE-Propyl film.

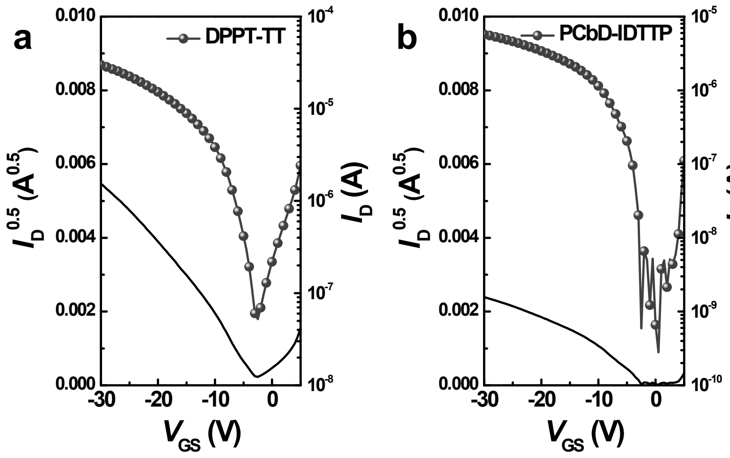

Fig. S8. Transfer curves of the FET with bottom-gate top-contact structure based on a) DPPT-TT and b) PCbD-IDTTP.

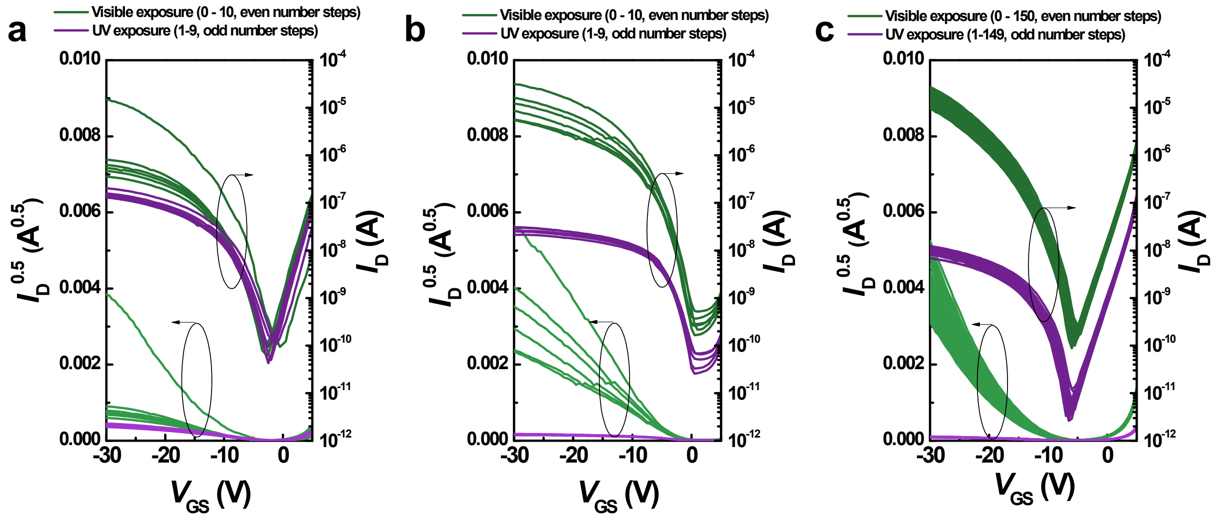

Fig. S9. Photoprogrammable switching behavior of FETs based on a) DPPT-TT/DAE-Me, b) PCbD-IDTTP/DAE-Me and c) PCbD-IDTTP/DAE-Propyl. The corresponding photoprogrammed reversible switching behavior after irradiation with UV (312 nm) and visible light (520 nm) in up to 10 steps for DPPT-TT/DAE-Me and PCbD-IDTTP/DAE-Me and 150 steps for PCbD-IDTTP/DAE-Propyl, respectively. The UV light irradiation time was fixed at 30 s and the visible light irradiation time was fixed at 300 s.

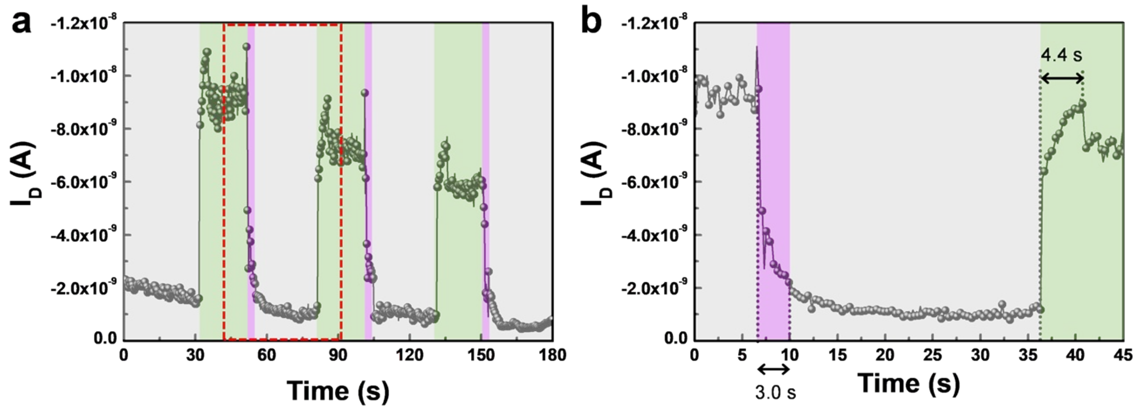

Fig. S10. Dynamic current modulation of PCbD-IDTPP/DAE-Propyl devices for a) three cycles of visible light (light green region), UV (violet region) and dark (light grey region) condition (V_{GS}= -20 V and V_{DS}=−5 V). b) Dynamic current modulation corresponding to the red dotted line area in Fig. S10a.

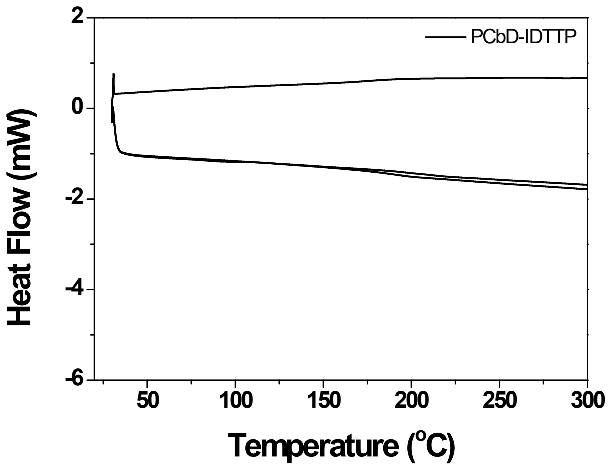

Fig. S11. Differential scanning calorimeter result of PCbD-IDTTP.

Table S1. Photophysical properties of DAEs and polymer/DAE blend films

System	λ_{max} [nm][a)]		PSS [%] [b)]	Φ_{UV} [%][c)]	Φ_{vis} [%][d)]
	open form	closed form			
DAE-Me solution[a)]	279	520	92	43	0.8
DAE-Me film[a)]	286	543	56	42	0.11
DPPT-TT/DAE-Me film	285	537	61	62	0.08
PCbD-IDTTP/DAE-Me film	286	537	42	24	0.28

[a)]Reported value from reference 1; [b)]Amount of the closed from in PSS upon UV light exposure (312 nm); Quantum yields upon [c)]UV and [d)]visible light exposure with the experimental error of 10%.

Table S2. Morphological characteristics for polymer and polymer/DAE blend films

System	R_{RMS} [nm]	Lamellar spacing [Å]	π-π stacking spacing [Å]	
			q_{xy}	q_z
DPPT-TT	0.729	21.71	3.76	3.80
DPPT-TT/DAE-Me	0.787	22.40	3.77	N/A
PCbD-IDTTP	0.293	19.73	4.21	4.19
PCbD-IDTTP/DAE-Me	0.409	19.86	4.20	4.20

Table S3. Summary of photoprogrammable performance of DAE-embedded OFETs.

System	Saturation hole mobility [$cm^2\ V^{-1}s^{-1}$]	Maximum photoprogrammable I_{DS} ON/OFF ratio	Maximum number of repeating steps	Photoprogrammable I_{DS} ON/OFF ratio retention between initial and final step [%]	References
P3HT/DAE-Me	N/A	approximately 10	4	85[a)]	1
P3HT/DAE-^{t}Bu	N/A	approximately 10	4	71[a)]	1
F8T2/DAE-Me	1.5×10^{-4}	approximately 1,000	4	75[a)]	1
F8T2/DAE-^{t}Bu	6×10^{-3}	approximately 1,000	4	90[a)]	1
DPPT-TT/DAE-Me	2×10^{-2}	approximately 100	4	2[a)]	1
DPPT-TT/DAE-^{t}Bu	1×10^{-2}	approximately 100	4	98[a)]	1
DPPT-TT/DAE-Propyl	6.40×10^{-2}	4,405	100	18	2
DPPT-TT/DAE-Me	3.02×10^{-2}	73	10	5	This work
PCbD-IDTTP/DAE-Me	4.37×10^{-2}	1,026	10	26	This work
PCbD-IDTTP/DAE-Propyl	5.38×10^{-2}	2,588	150	46	This work

[a)] Estimated values from output current in reversible modulation step.

References

1 L. Hou, T. Leydecker, X. Zhang, W. Rekab, M. Herder, C. Cendra, S. Hecht, I. McCulloch, A. Salleo, E. Orgiu and P. Samorì, *J. Am. Chem. Soc.*, 2020, **142**, 11050.

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