## Electronic Supplementary Information

## **Biaxially Extended Thiophene-Isoindigo Donor-Acceptor Conjugated Polymers for High-Performance Flexible Field-Effect Transistors**

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Polymer	Lamellar spacing (Å)	In-plane $\pi$ - $\pi$ stacking distance (Å)
PIITT4T	23.4	_
PIITT4TSi	26.5	3.59
PII2T4T	21.8	4.13
PII2T4TSi	23.9	3.58
PII2T8T	-	-
PII2T8TSi	29.4	-

Table S1. Relevant Crystallographic Properties for the Studied Polymer Thin Films



Fig. S1. <sup>1</sup>H-NMR spectrum of (a) TT4T, (b) 2T4T, and (c) 2T8T in  $CD_2Cl_2$  (x:  $CD_2Cl_2$ , y:  $H_2O$ ).



**Fig. S2.** <sup>1</sup>H-NMR spectrum of (a) TT4T-ditin, (b) PIITT4T, and (c) PIITT4TSi in  $CD_2Cl_2$  (x:  $CD_2Cl_2$ ; y:  $H_2O$ ).



**Fig. S3.** <sup>1</sup>H-NMR spectrum of (a) 2T8T-ditin in  $CD_2Cl_2$  (x:  $CD_2Cl_2$ ; y:  $H_2O$ ), (b) PII2T8T in  $CDCl_3$  (x:  $CDCl_3$ ; y:  $H_2O$ ), and (c) PII2T8TSi in  $CD_2Cl_2$  (x:  $CD_2Cl_2$ ; y:  $H_2O$ ).



Fig. S4. DSC curves of the studied polymers under a nitrogen atmosphere.



**Fig. S5.** UV-Vis absorption spectra of the studied polymers in dilute chloroform solution.



**Fig. S6.** FET output curves of (a) PIITT4T-, (b) PIITT4TSi-, (c) PII2T4T-, (d) PII2T8T-, and (e) PII2T8TSi-based device, respectively.