## **Electronic Supplementary Information**

Improved Electron Transport Property of *n*-Type Naphthalenediimide Polymers through Refined Molecular Ordering and Orientation Induced by Processing Solvents

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**Figure S1.** Cyclic voltammograms of  $P(NDI2SiC_5-T2)$  (a) and  $P(NDI2SiC_5-TTh)$  (b) thin films prepared by drop-casting on a platinum working electrode using CF solution and measured in *n*-Bu<sub>4</sub>NPF<sub>6</sub>/CH<sub>3</sub>CN solution (scan rate = 100 mV/s).



**Figure S2.** Dihedral angles and isosurfaces of the dimer models for  $P(NDI2SiC_5-T2)$  (a) and  $P(NDI2SiC_5-TTh)$  (b) calculated by DFT, respectively (B3LYP/6-31G\*).



**Figure S3.** AFM height images of the as-cast  $P(NDI2SiC_5-T2)$  (a) and  $P(NDI2SiC_5-TTh)$  (b) films depending on the processing solvents.



**Figure S4.** GIXD images of the as-cast  $P(NDI2SiC_5-T2)$  (a) and  $P(NDI2SiC_5-TTh)$  (b) films depending on the processing solvents.

Preparation condition	Crystallographic parameters		P(NDI2SiC <sub>5</sub> -T2)	P(NDI2SiC <sub>5</sub> -TTh)
CF	(100)	q (Å-1)	0.228	0.229
		d-spacing (Å)	27.5	27.4
	π-π	q (Å <sup>-1</sup> )	1.65	1.60
		d-spacing (Å)	3.80	3.92
CN	(100)	q (Å <sup>-1</sup> )	0.218 <sup>a</sup>	0.243 <sup>a</sup>
		d-spacing (Å)	28.82 <sup>a</sup>	25.85 <sup>a</sup>
	π-π	q (Å <sup>-1</sup> )	$N/A^{b}$	$N/A^{b}$
		d-spacing (Å)	N/A <sup>b</sup>	$N/A^{b}$
DCB	(100)	q (Å <sup>-1</sup> )	0.224 <sup>a</sup>	0.248 <sup>a</sup>
		d-spacing (Å)	28.05 <sup>a</sup>	25.34 <sup>a</sup>
	π-π	q (Å <sup>-1</sup> )	$N/A^{b}$	$N/A^{b}$
		d-spacing (Å)	N/A <sup>b</sup>	$N/A^{b}$
ТСЕ	(100)	q (Å <sup>-1</sup> )	0.235	0.230
		d-spacing (Å)	26.7	27.3
	π-π	q (Å <sup>-1</sup> )	N/A <sup>b</sup>	$N/A^{b}$
		d-spacing (Å)	N/A <sup>b</sup>	$N/A^{b}$

**Table S1.** Crystallographic parameters calculated from GIXD profiles of the as-cast films.

<sup>*a*</sup>The values were predicted from *d*-spacing of (200) peaks. <sup>*b*</sup>The discernable peaks were not detected.