

## Supporting Information

### **Enhancing Charge Transport in Isoindigo-based Donor-Acceptor Copolymers by Combining Ionic Doping with Polar Alkoxy Side Chains**

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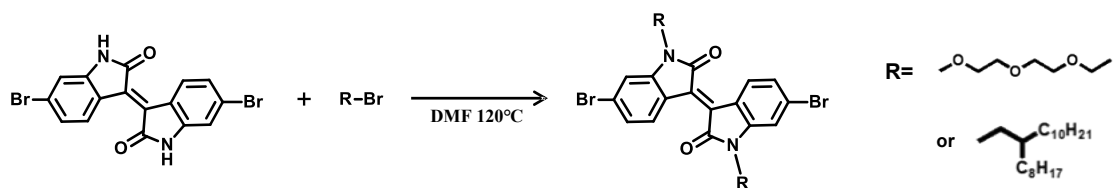
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**Scheme S1.** Synthesis of N-alkylation of 6,6-Dibromoisoindigo with alkoxy chains and branch alkyl side chains.

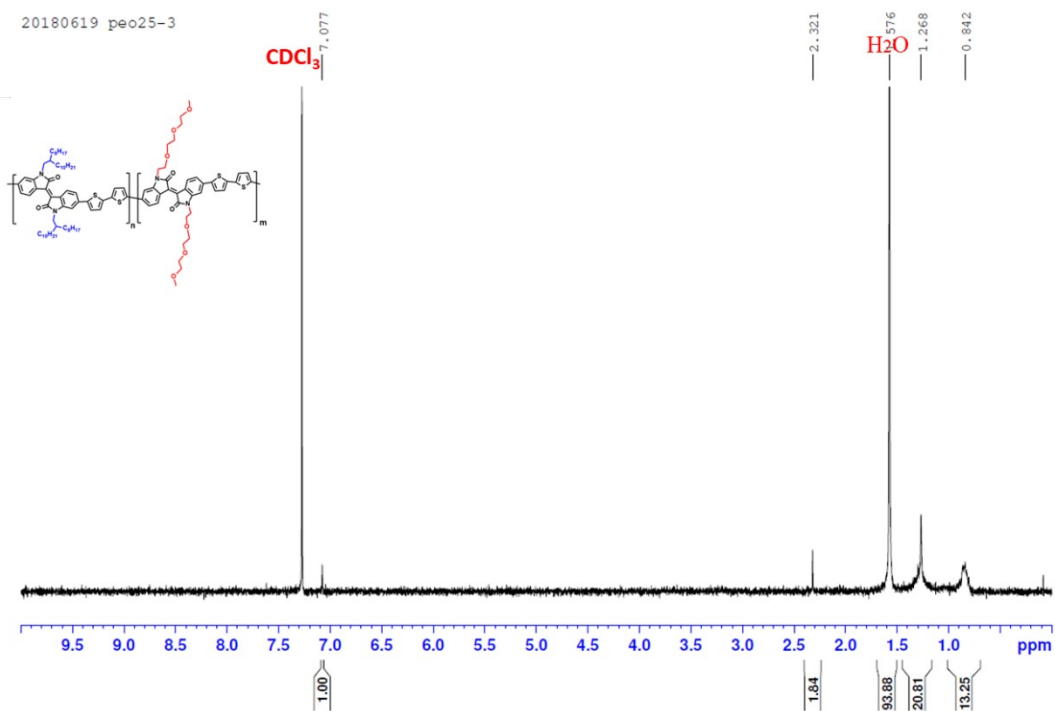
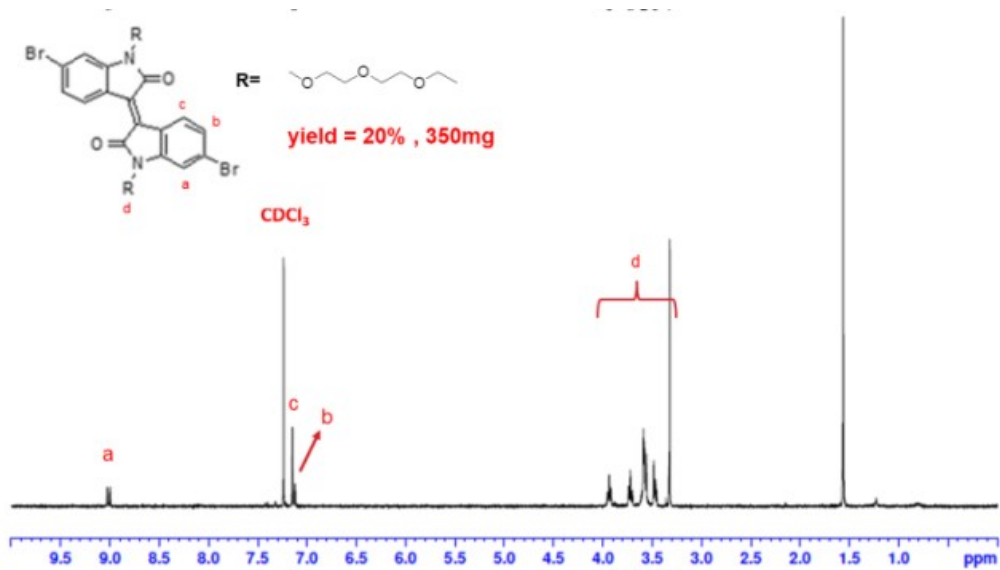
### Characterizations of polymers

**PII2TC8C10.** 196.74 mg (0.4 mmol) of 5,5'-bis(trimethylstannyl)-2,2'-bithiophene, 392.6 mg (0.4 mmol) of IID-C8C10, and 5 ml of chlorobenzene were used to afford a deep red solid (yield: 220 mg, 55.8%). The number average molecular weight ( $M_n$ ) and polydispersity index (PDI) estimated from GPC are 30.8 kg mol<sup>-1</sup> and 2.39, respectively. Found (%): C, 76.88±0.4; H, 9.02±0.4; N, 2.84±0.4; S, 6.62±0.4; O, 3.24±0.4.

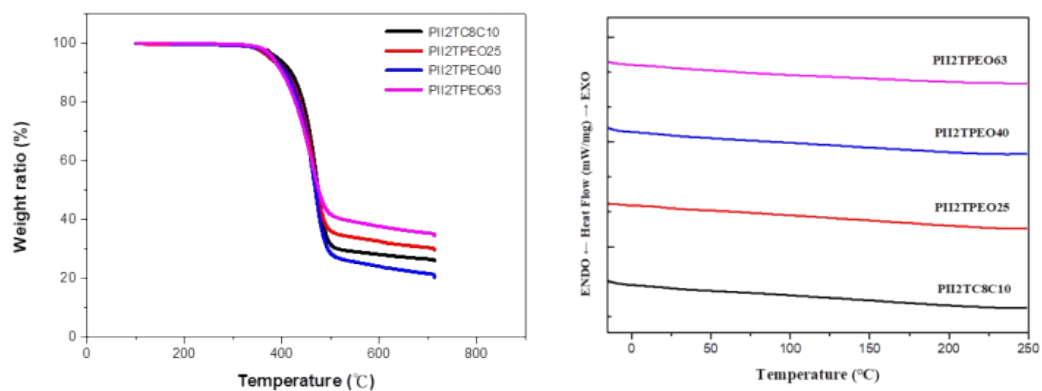
**PII2TPEO25.** 196.74 mg (0.4 mmol) of 5,5'-bis(trimethylstannyl)-2,2'-bithiophene, 342.44 mg (0.36 mmol) of IID-C8C10, 28.49 mg (0.04 mmol) of IID-PEO, and 5 ml of chlorobenzene were used to afford a deep red solid (yield: 300 mg, 78.2%). The  $M_n$  and PDI estimated from GPC are 21.0 kg mol<sup>-1</sup> and 2.00, respectively. Found (%): C, 75.71±0.4; H, 9.42±0.4; N, 2.94±0.4; S, 6.79±0.4; O, 4.96±0.4.

**PII2TPEO40.** 196.74 mg (0.4 mmol) of 5,5'-bis(trimethylstannyl)-2,2'-bithiophene, 313.95 mg (0.32 mmol) of IID-C8C10, 56.98 mg (0.08 mmol) of IID-PEO, and 5 ml of chlorobenzene were used to afford a deep red solid (yield: 322 mg, 86.4%). The  $M_n$  and PDI estimated from GPC are 17.6 kg mol<sup>-1</sup> and 2.10, respectively. Found (%): C, 74.65±0.4; H, 8.97±0.4; N, 2.99±0.4; S, 7.18±0.4; O, 6.67±0.4.

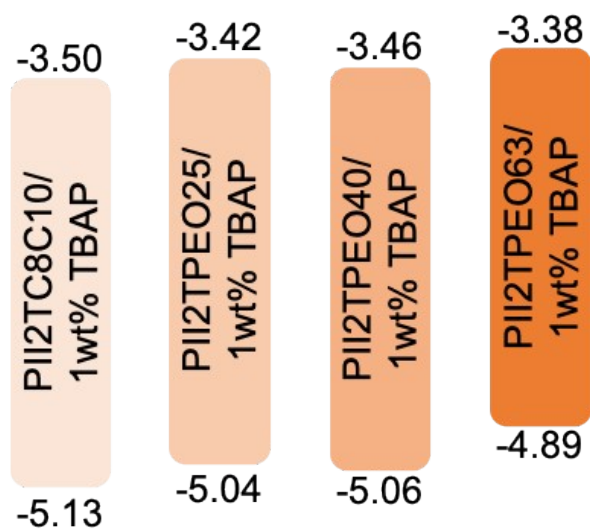
**PII2TPEO63.** 196.74 mg (0.4 mmol) of 5,5'-bis(trimethylstannyl)-2,2'-bithiophene, 196.24 mg (0.2 mmol) of IID-C8C10, 142.48 mg (0.2 mmol) of IID-PEO, and 5 ml of chlorobenzene were used to afford a deep red solid (yield: 201 mg, 59%). The  $M_n$  and PDI estimated from GPC are unavailable because the solubility of the polymer is not as well as expected. Found (%): C, 75.71±0.4; H, 9.42±0.4; N, 2.94±0.4; S, 6.79±0.4; O, 4.96±0.4.



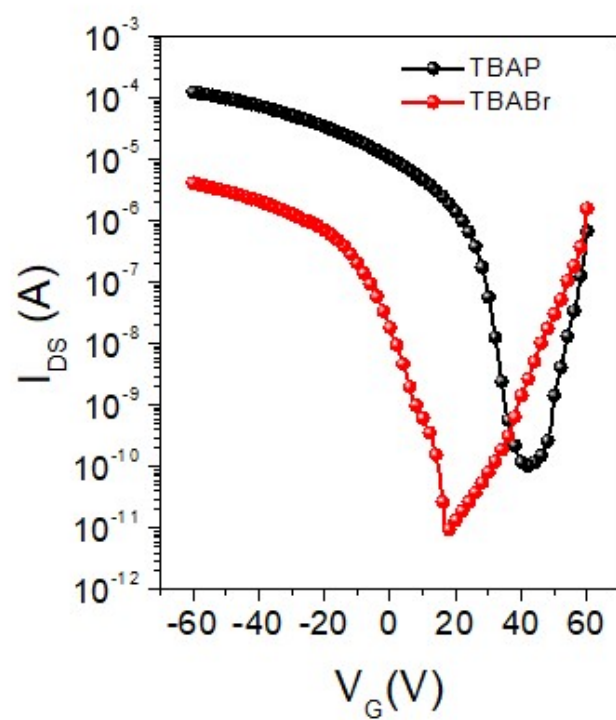
**Fig. S1.** <sup>1</sup>H NMR spectra of (top) the alkoxy PII2T monomer and (bottom) PII2TPEO25 in CDCl<sub>3</sub>.



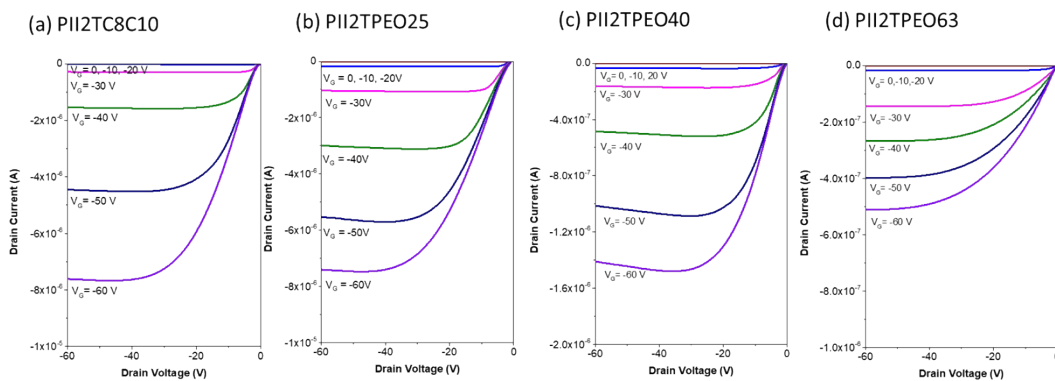
**Fig. S2.** (a) TGA curves and (b) DSC traces of the studied polymers with a heating rate of  $10\text{ }^{\circ}\text{C min}^{-1}$  under nitrogen atmosphere.



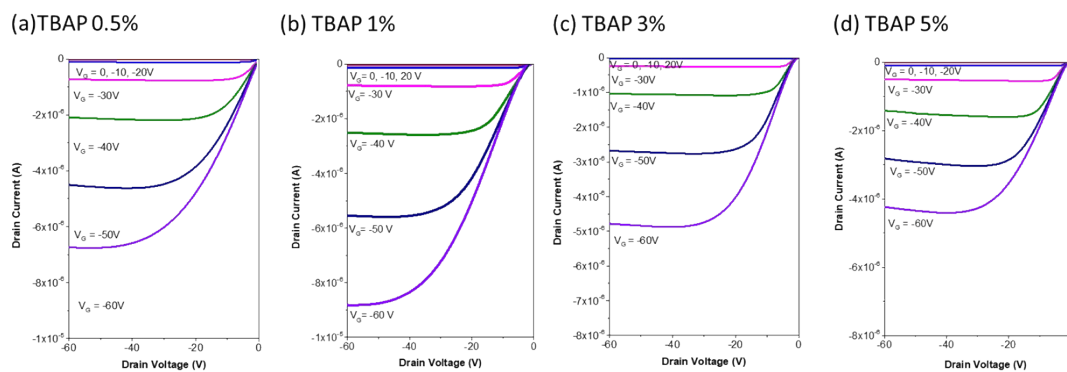
**Fig. S3.** Energy levels of TBAP-doped polymer films.



**Fig. S4.** Transfer curves of poly(diketopyrrolopyrrole-thienothiophene) (PDPPTT)-based FETs doped with TBAP and TBABr, respectively.



**Fig. S5.** Output characteristics of isoindigo-based copolymers with different side chains: (a) PII2TC8C10; (b) PII2TPEO25; (c) PII2TPEO40; (d) PII2TPEO64.



**Fig. S6.** Output characteristics of PII2TPEO25 doped with different amounts of TBAP: (a) 0.5 wt%, (b) 1 wt%, (c) 3wt%, and (d) 5wt%.

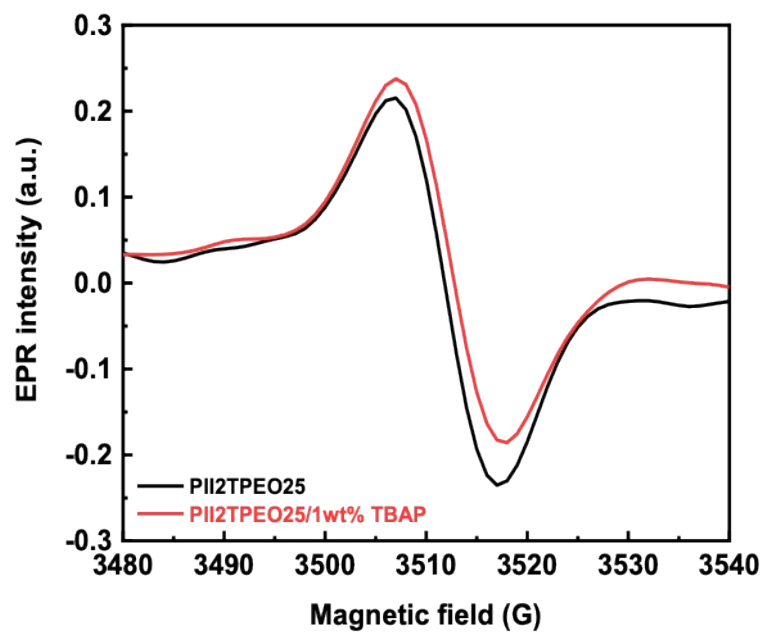
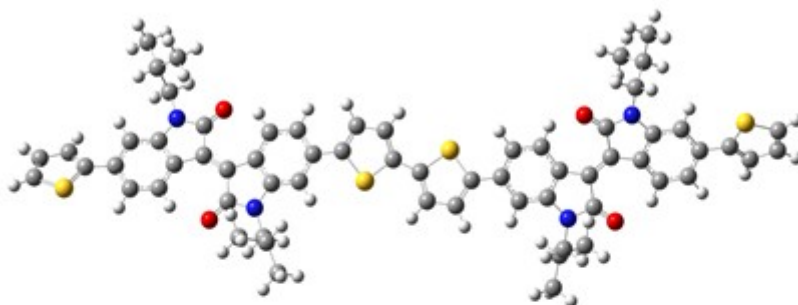
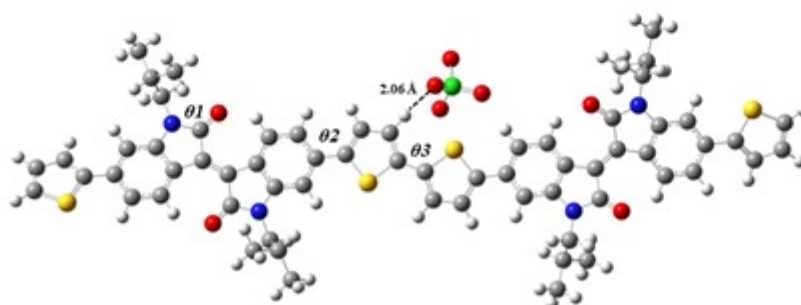


Fig. S7. EPR spectra of undoped and doped PII2TPEO25.

### PII2TC8C10

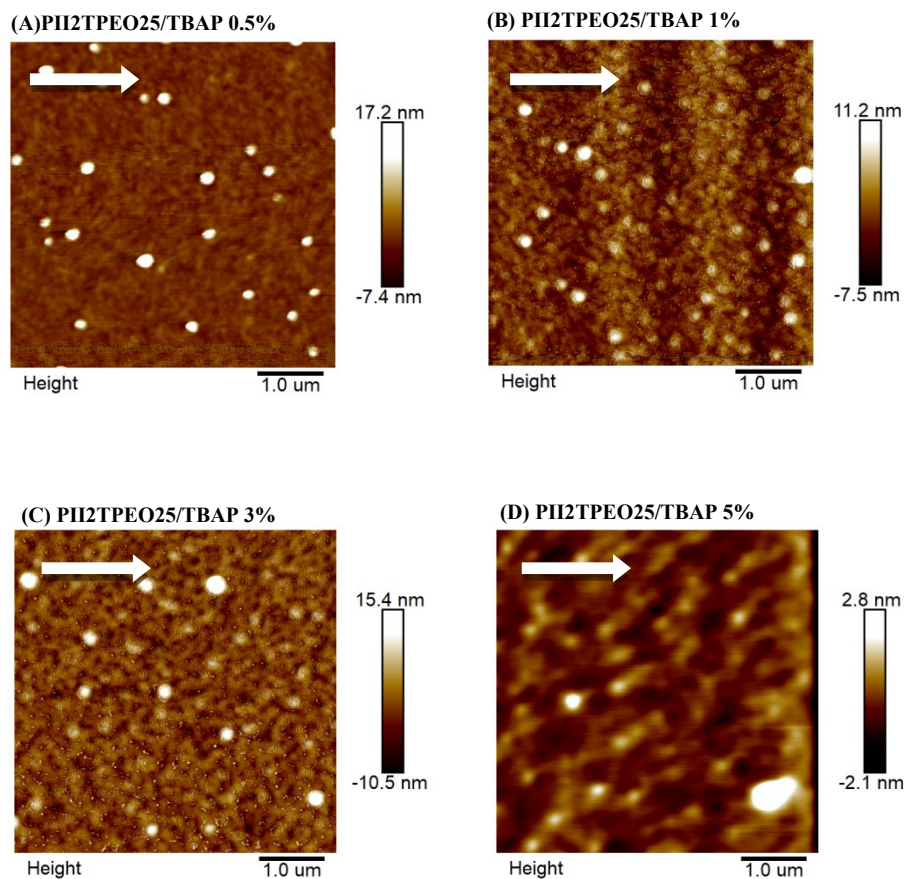


### PII2TC8C10 / TBAP

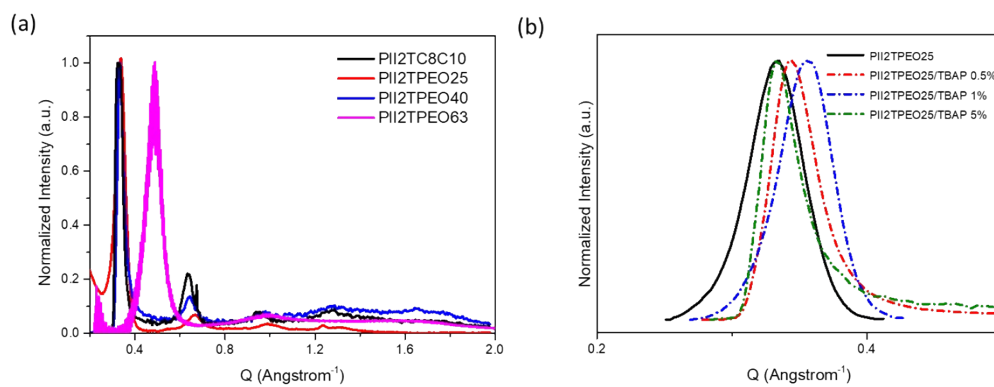


**Fig. S8.** Optimized structural geometries of  $\text{ClO}_4^-$  free and  $\text{ClO}_4^-$  containing branched alkyl side chains.





**Fig. S9.** AFM height images of TBAP-doped PII2TPEO25 films at different ratios of (a) 0.5, (b) 1, (c) 3, and (d) 5 wt%, respectively.



**Fig. S10.** 1D GIXD curves of (a) isoindigo-based copolymers and (b) PII2TPEO25 doped with different amounts of TBAP.