Supporting information

High Efficiency Non-Fullerene Organic Tandem Photovoltaics Based on Ternary Blend Sub-Cells

*Wenchao Huang[12], Sheng-Yung Chang[12], Pei Cheng[12], Dong Meng[123], Bowen Zhu[12], Selbi Nuryyeva[12], Chenhui Zhu[4], Lijun Huo[5], Zhaohui Wang[3], Mingkui Wang[6], Yang Yang[12]**

[1] Department of Materials Science and Engineering, University of California Los Angeles, California, 90095, USA

[2] California NanoSystems Institute, University of California Los Angeles, California, 90095, USA

[3] Beijing National Laboratory for Molecular Sciences, Key Laboratory of Organic Solids, Institute of Chemistry, Chinese Academy of Sciences (ICCAS), Beijing, 100190, P. R. China

[4] Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California, 94720, USA

[5] Key Laboratory of Bio-Inspired Smart Interfacial Science and Technology of Ministry of Education, Heeger Beijing Research and Development Center, School of Chemistry, Beihang University, Beijing 100191, P.R. China

[6] Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, Wuhan, 430070, P.R. China

* Corresponding author. Email: yangy@ucla.edu

KEYWORDS: Organic tandem photovoltaics, non-fullerene, ternary sub-cell, current matching, morphology

Experimental Section

Materials

PDBT-T1 and TPH-Se were synthesized according to the literature.[1, 2] PBDTTT-EFT, PBDB-T, ITIC and IEICO-4F were purchased from 1-Materials. 1,8-diiodooctance (DIO), 1-chloronaphthalene (CN) and chlorobenzene (CB) were purchased from Sigma-Aldrich.

Device fabrication

The single-junction organic solar cells were fabricated in an inverted structure of ITO/ZnO/active layer/MoO_3/Ag. The ITO glasses were washed in an ultrasonic bath of acetone and isopropanol and treated with ultraviolet-ozone for 15 min after. The ZnO layer was prepared via sol-gel process with a thickness of 30 nm. The PDBT-T1:TPH-Se:ITIC ternary blends with different ratios were spun from a mixed solvent of CB and DIO (99.25:0.75 vol.%) at 1000 rpm for 45 s, followed by thermal annealing at 100 °C for 5 min. The concentration of PDBT-T1 was fixed at 7 mg/ml. The PBDB-T:ITIC (11 mg/ml: 11 mg/ml) blends were spin-coated from a mixed solvent of CB and DIO (99.5:0.5 vol.%) at 2000 rpm for 45 s, followed by thermal annealing at 100 °C for 1 min. The PBDTTT-EFT:IEICO-4F blends (10 mg/ml: 15mg/ml) were spin-coated from a mixed solvent of CB and CN (97:3 vol.%) at 1500 rpm for 45 s. MoO_3 (10 nm) and Ag (100 nm) were evaporated onto the active layer through a shadow mask. The active area of the device was 0.1 cm^2. For the fabrication of interconnecting layer (ICL) in tandem devices, MoO_3 was thermally evaporated with a thickness of 6 nm. A thin layer of modified PEDOT:PSS was then spin-coated, followed by thermal annealing at 120 °C for 90 s and another layer of np-ZnO was deposited from ZnO nanocrystal solution, followed by thermal annealing at 120 °C for 90 s.[3]

Device characterization

The PCEs of solar cells were measured under 1 sun AM 1.5G spectrum with an Oriel 9600 solar simulator. The intensity of solar simulator was calibrated by using a calibrated silicon reference cell. EQE spectra were measured using an integrated system (Enlitech). The short circuit current was recorded with a lock-in amplifier. The device characterization of tandem solar cells was measured following a previously reported methodology.[3] In order to avoid the parasitic current during device measurements, each device was absolutely isolated from surround devices. For EQE spectrum measurement of each sub-cell, a bias light with the selected wavelength was applied.

Optical characterization

The samples for UV-Vis measurements were prepared on the glass slide. The absorption spectra were collected using a 4100 Hitachi spectrofluorophotometer.

Microstructural characterization

GIWAXS experiments were conducted at the beamline 7.3.3 of the Advanced Light Source at Lawrence Berkeley National Laboratory. All samples were deposited on silicon wafers following the same procedure as described in device fabrication. All samples were irradiated with 10 KeV X-ray at a fixed angle of 0.14°. 2D scattering signals were probed using a Dectris Pilatus 1M detector. The results were analyzed by an altered version of the NIKA 2D based in IgorPro.

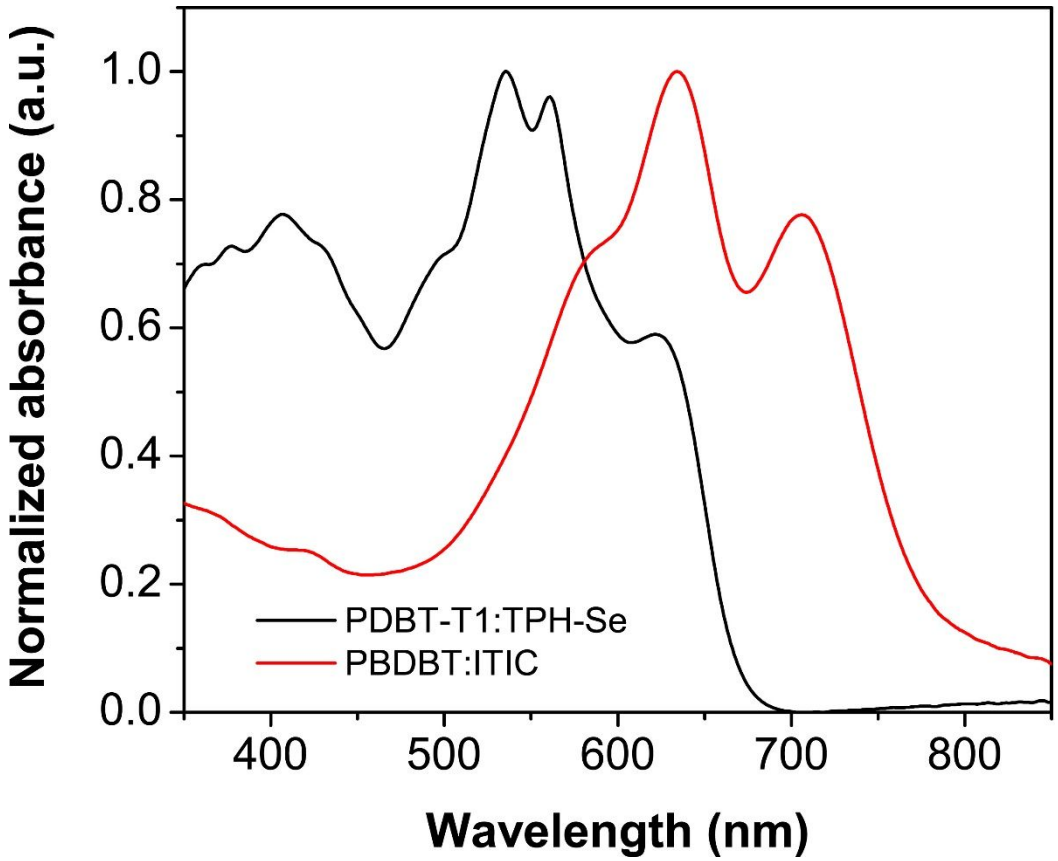

Figure S1. UV-vis spectra of PDBT-T1:TPH-Se and PBDB-T:ITIC blends.

Table S1. Photovoltaic parameters of organic solar cells based on the PDBT-T1 wide-bandgap polymer.

Active layer	Efficiency (%)	V_{OC} (V)	J_{SC} (mA/cm^2)	Fill factor
PDBT-T1: $PC_{71}BM$	7.2	0.85	12.4	0.68
PDBT-T1:TPH-Se	8.0	1.00	11.9	0.67

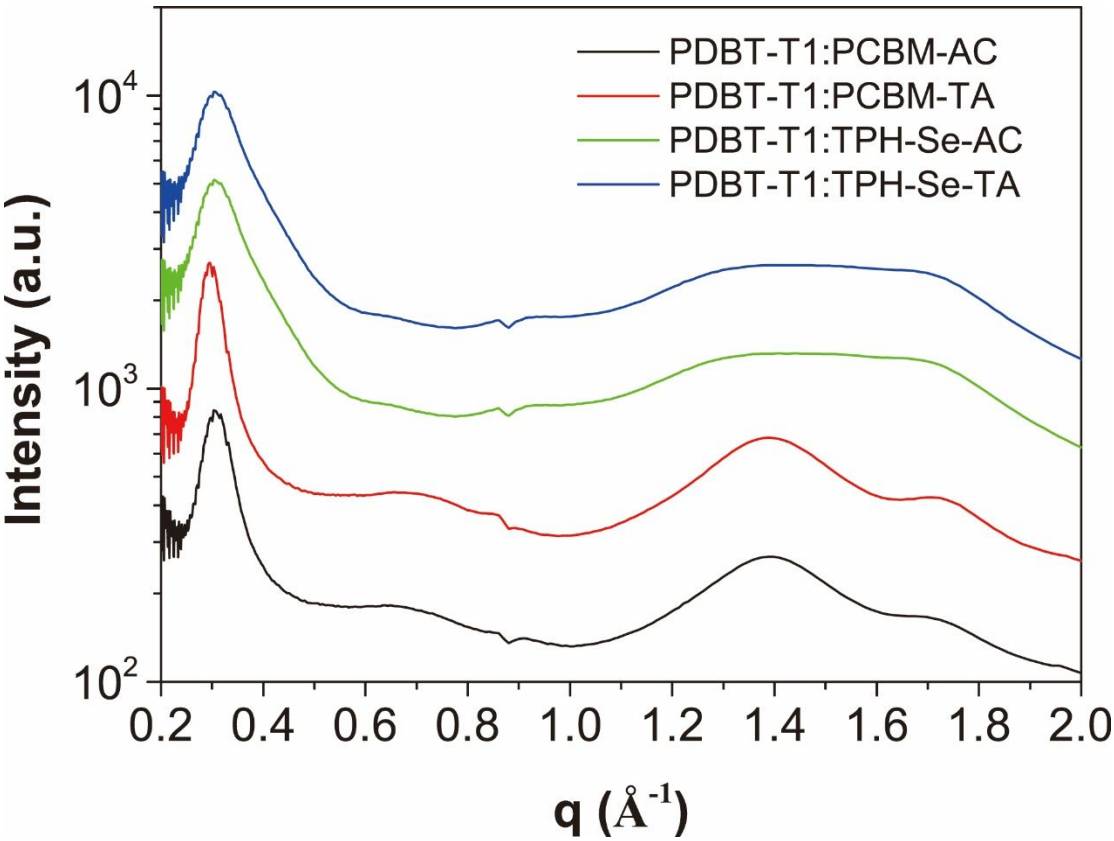

Figure S2. 1D line profiles cut from 2D GIWAXS patterns of PDBT-T1:PCBM and PDBT-T1:TPH-Se blends before and after heat treatment.

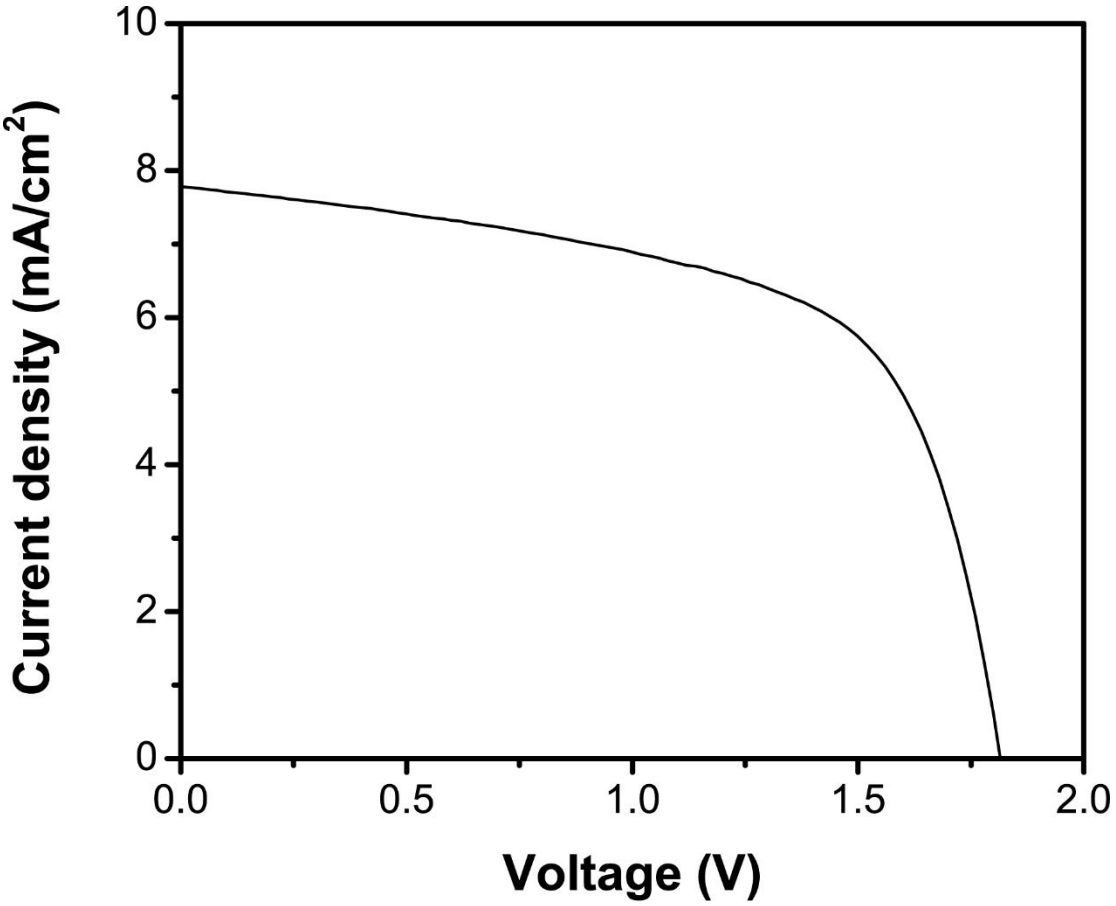

Figure S3. Photovoltaic characteristics of tandem solar cells based on PDBT-T1:ITIC front sub-cell and PBDBT:ITIC rear sub-cell.

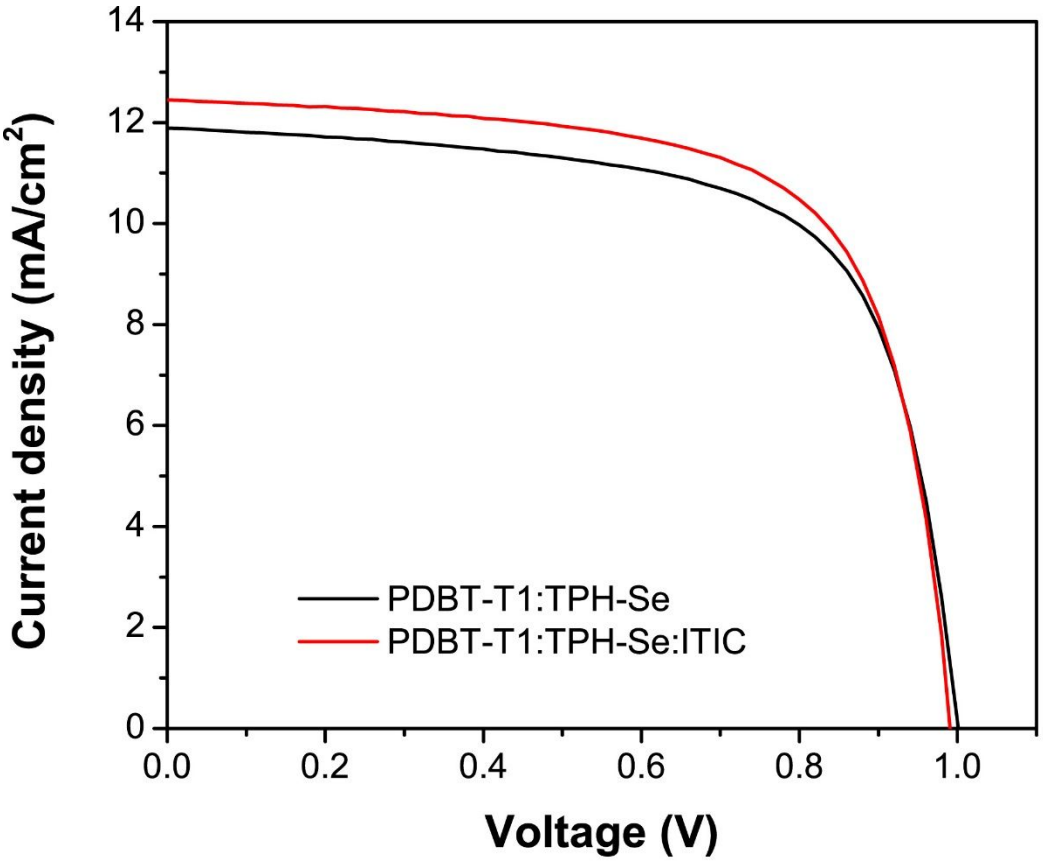

Figure S4. Photovoltaic characteristics of PDBT-T1:TPH-Se binary solar cells and PDBT-T1:TPH-Se:ITIC (1:0.9:0.1) ternary solar cells.

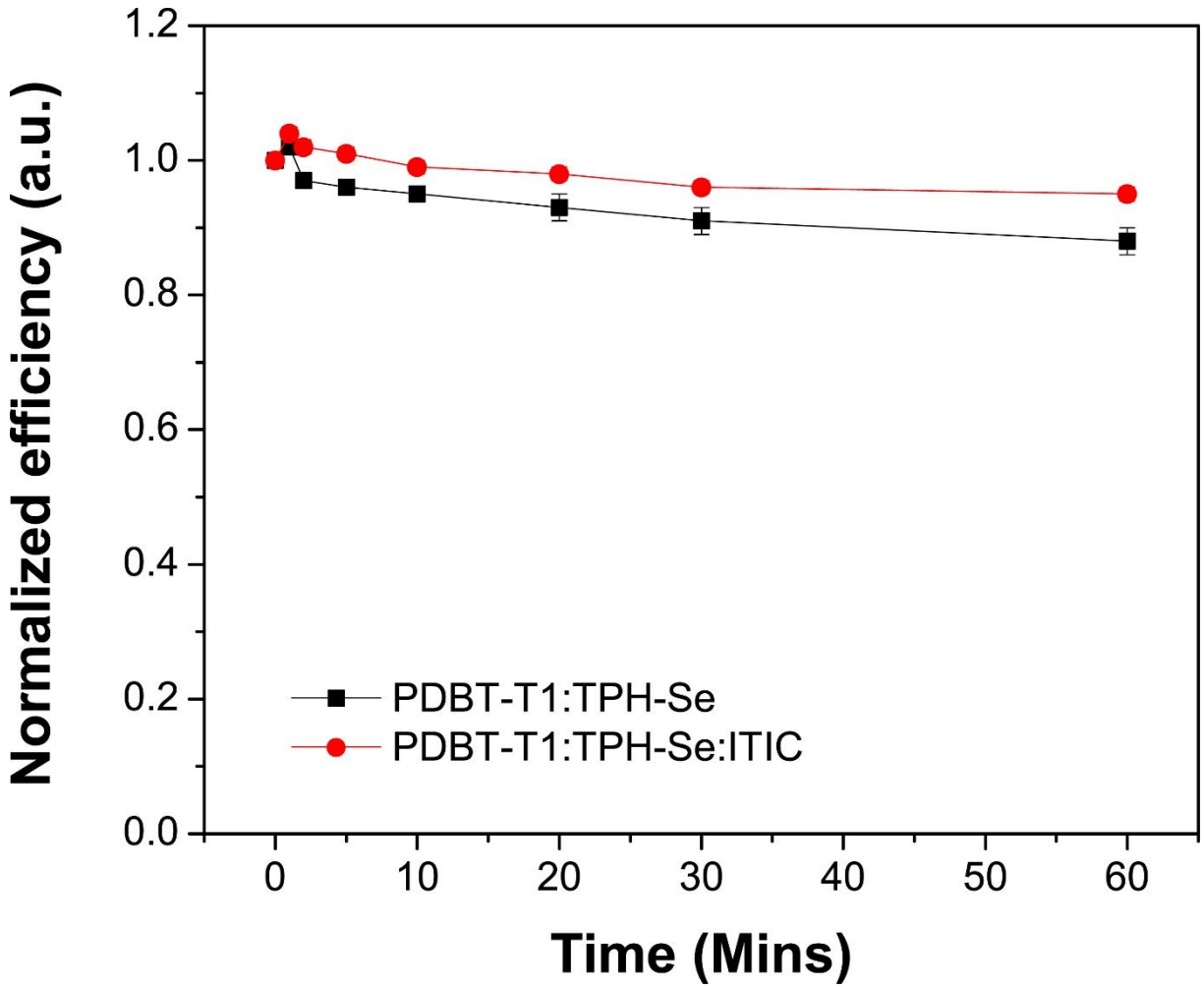

Figure S5. Thermal stability of PDBT-T1:TPH-Se binary solar cells and PDBT-T1:TPH-Se:ITIC (1:0.9:0.1) ternary solar cells. The devices were thermally aged at 120 °C.

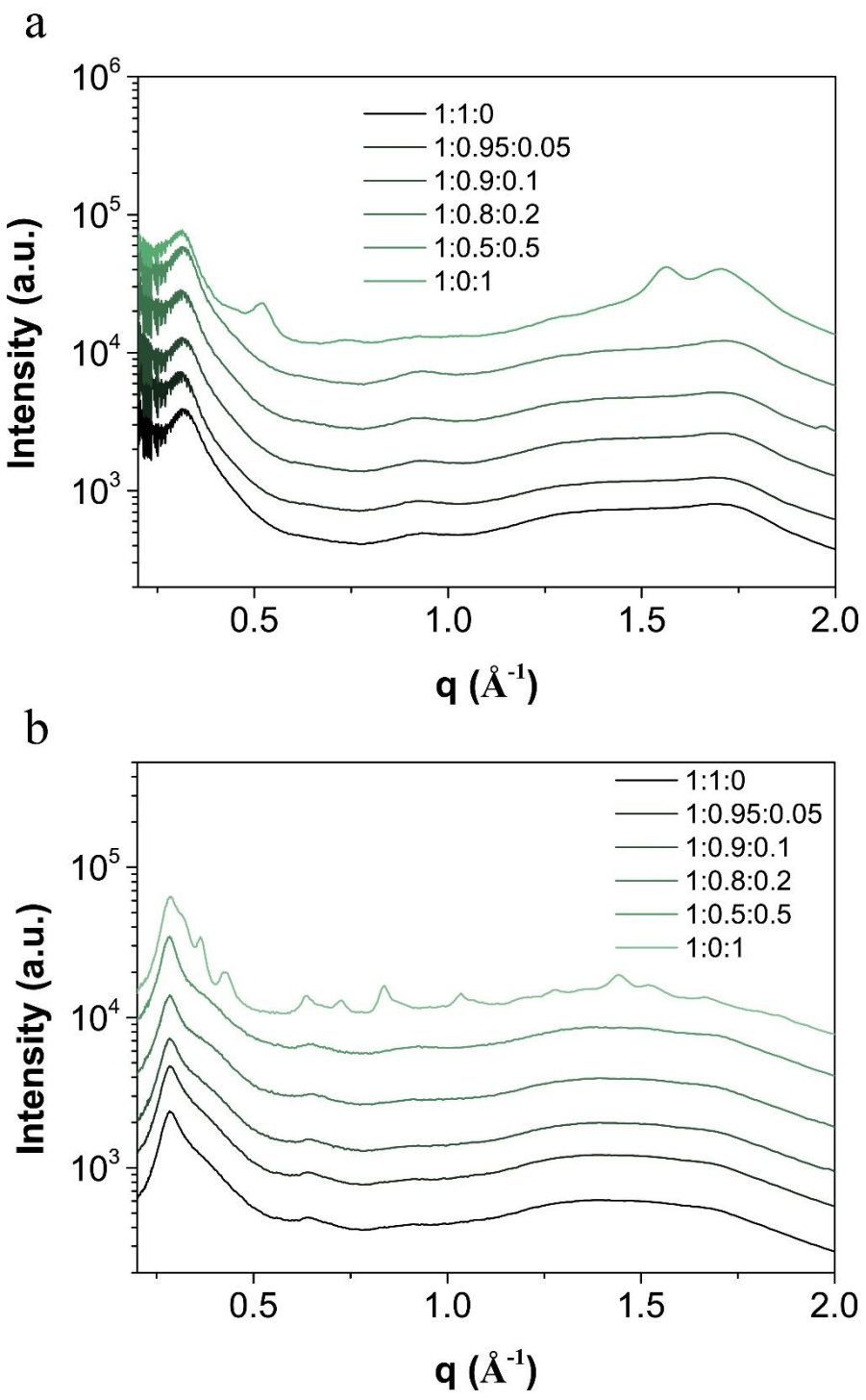

Figure S6. 1D GIWAXS line profile of PDBT-T1:TPH-Se:ITIC ternary blends with different chemical compositions along (a) out-of-plane (OOP) and (b) in-plane (IP).

1. Huo, L.; Liu, T.; Sun, X.; Cai, Y.; Heeger, A. J.; Sun, Y. *Adv. Mater.* 2015, 27, 2938-2944.
2. Meng, D.; Fu, H.; Xiao, C.; Meng, X.; Winands, T.; Ma, W.; Wei, W.; Fan, B.; Huo, L.; Doltsinis, N. L.; Li, Y.; Sun, Y.; Wang, Z. *J. Am. Chem. Soc.* 2016, 138, 10184-10190.
3. You, J.; Dou, L.; Yoshimura, K.; Kato, T.; Ohya, K.; Moriarty, T.; Emery, K.; Chen, C.-C.; Gao, J.; Li, G.; Yang, Y. *Nat. Commun.* 2013, 4, 1446.