Supporting information

Flexible large-area organic tandem solar cells with high defect tolerance and device yield

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A separate video has also been uploaded as a part of the supporting information.

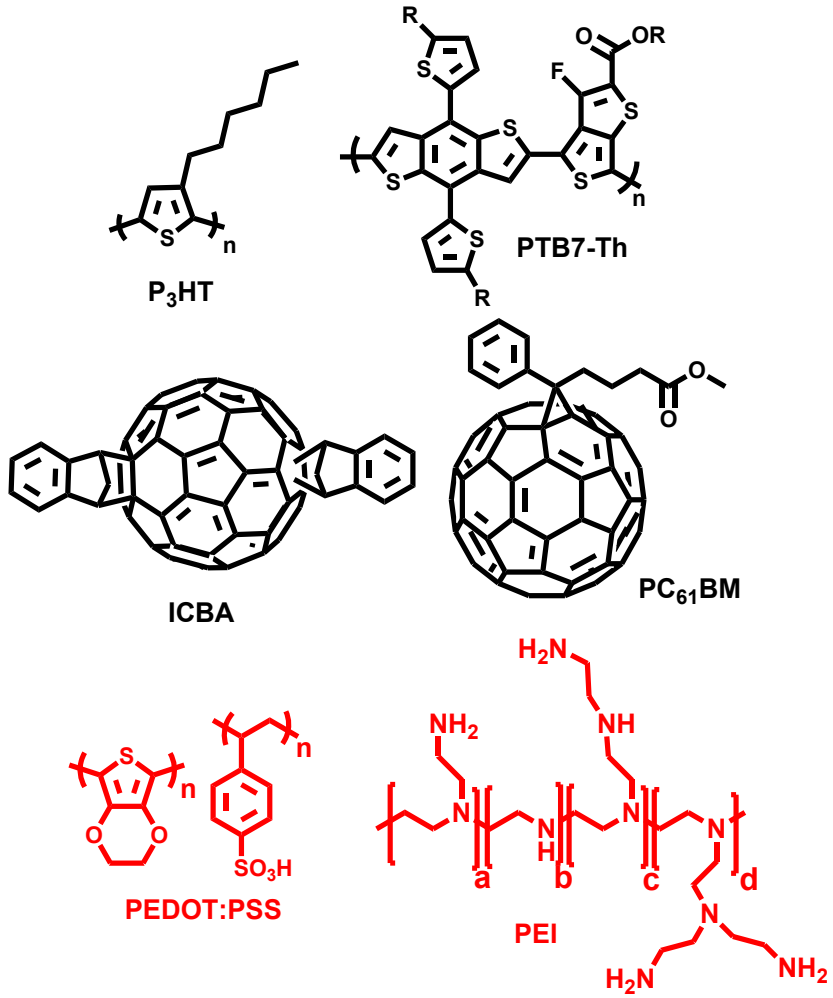

Figure S1. Chemical structures of active layer and interfacial layer materials: P3HT and PTB7-Th were applied as electron donors, ICBA and $PC_{61}BM$ as electron acceptors, PEI as electron transporting layer, and PEDOT:PSS as interconnecting layer/transparent electrode.

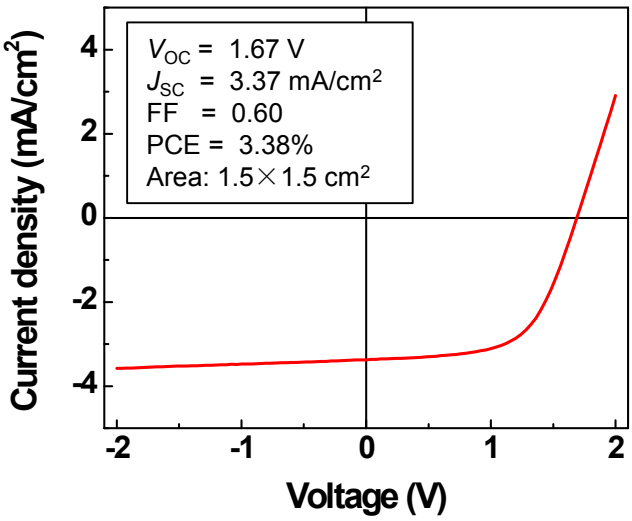

Figure S2. *J-V* characteristics of a tandem cell without metal electrodes (device area of 1.5×1.5 cm^2): glass/ITO/PEI/P3HT:ICBA/m-PEDOT:PSS/PEI/P3HT:ICBA/*hc*-PEDOT:PSS

Experimental section

Materials.

P3HT and ICBA were received from Luminescence Technology Corp. PTB7-Th and $PC_{61}BM$ were purchased from 1-Material and American Dye Source Inc., respectively. PEDOT:PSS Clevios P VP AI 4083 and PH1000 were supplied by Heraeus. PEI (Mw =25,000) was purchased from Sigma-Aldrich. Silver slug (99.999%) for evaporation was received from Alfa Aesar. All the materials were used without further purification.

Fabrication of single and tandem OSCs.

For single junction solar cells, PEI in isopropanol (0.1 wt.%) was spin-coated onto pre-cleaned ITO glass at 5000 rpm for 1 min and annealed at 100 ºC for 10 min in air. The P3HT and ICBA blend film was prepared by dissolving in 1,2-dichlorobenzene a mixture of P3HT:ICBA (20 mg:20 mg per 1 ml), then spin-coated at 700 rpm for 1 min and thermal annealing at 150 ºC for 10 min in N_2-filled glove box. When fabricating the 10.5-cm^2 cells, the solutions were first dropped on the substrates and paved uniformly covering the surface using pipettes. Then, the substrates were spin coated to get uniform films. For tandem solar cells, a layer of *m*-PEDOT:PSS was spin-coated on BHJ layer and annealed at 100 ºC for 5 min. The *m*-PEDOT:PSS was prepared by mixing PEDOT:PSS formulation of AI 4083 and PH1000 at a mixing ratio of 3:1 as we reported earlier.[1] Then we repeated the previous procedure to deposit PEI/P3HT:ICBA and form the second junction. To finish the devices, film-transfer laminated

PEDOT:PSS PH1000 with PDMS as the transfer medium was used as the top electrodes as described in previous literature.[2] Before any deposition of PEDOT:PSS aqueous, the hydrophobic surface of BHJ layer need a brief flash (5 s) of plasma treatment (PDC-002, Harrick).

Flexible tandem OSCs are fabricate on PES substrates. A layer of silver film with a thickness of 80 nm was thermally deposited with a shadow mask. The subsequent organic layers (PEI/P3HT:ICBA/m-PEDOT:PSS/PEI) were prepared in the same way as described in the previous paragraph. The BHJ of PTB7-Th: $PC_{61}BM$ (1:1.5, wt.%) was deposited by spin-coating from a chlorobenzene/1,8-diiodoctane (97:3 by volume) solution (a total concentration of 25 mg ml^{-1}) in the case of hetero-tandem OSCs. Eventually, after the transfer of the top PEDOT:PSS PH1000 electrode, silver grids with thickness of 80 nm was evaporated through a shadow mask to finish the device. The grids is spaced 2 mm apart and each sub-line is 100 μm wide.

Defect in the BHJ layer was created via the process of transferring P3HT:ICBA film (bottom BHJ). The film was first spin-coated on clean silicon wafer and transferred onto PDMS by immersing into distilled water; After removing a small portion of P3HT:ICBA (approximately 1 mm^2) with tweezers, the PDMS/P3HT:ICBA was put onto PEI modified ITO and then the PDMS was peeled off. Then the bottom P3HT:ICBA film with a small portion missing was fabricated. The small missing portion is the designed defects. More details of the procedure of transfer printing P3HT:ICBA can be found elsewhere.[1,2] To create a defect within the CRL, a PDMS square with size of approximately 1 mm^2 was placed on BHJ layer prior to plasma

treatment to create a hydrophobic spot (while other area is hydrophilic) where the subsequent deposited aqueous PEDOT:PSS formulation would be repelled, therefore creating a defect in the CRL.

Characterization.

Current density–voltage (*J–V*) characteristics were measured inside a N_2-filled glove box using a source meter (2400, Keithley Instruments) controlled by a LabVIEW program under AM1.5G (100 mW cm-2) and a Newport solar simulator. Optical microscopic images were captured using an optical microscope (DM4000 M, Leica).

References：

1. J. Tong, S. Xiong, Y. Zhou, L. Mao, X. Min, Z. Li, F. Jiang, W. Meng, F. Qin, T. Liu, R. Ge, C. Fuentes-Hernandez, B. Kippelen and Y. Zhou, *Mater. Horiz.*, 2016, **3**, 452-459.

2. Y. Zhou, T. M. Khan, J. W. Shim, A. Dindar, C. Fuentes-Hernandez and B. Kippelen, *J. Mater. Chem. A*, 2014, **2**, 3492-3497.