Supporting Information

One key issue in characterization of organic solar cells with solution

processed interfacial layer

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Device Fabrication: OSCs were fabricated with a structure ITO/PEDOT:PSS/active layers/PDIN/Al. The patterned indium tin oxide (ITO) glass coated substrates (sheet resistance 15 Ω/\Box) were pre-cleaned by sequential ultrasonic treatment in detergent, deionized water and ethanol, respectively. The cleaned ITO substrates were blowdried by high pure nitrogen gas and then treated by oxygen plasma for 1 min to function improve its work and clearance. Subsequently, poly(3.4ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, clevios PVP Al 4083, purchased from H.C. Starck Co., Ltd.) solution was spin-coated on the ITO substrates at 5000 round per minute (RPM) for 30 s and then annealed at 150° C for 15 min in air. After annealing treatment, the ITO substrates coated PEDOT:PSS films were transferred to a high-purity nitrogen-filled glove box to fabricate active layers. The polymer donor J71 (purchased from Solarmer Materials Inc), nonfullerene acceptors IT-2Cl (synthesized by Yang's group of Wuhan University) with 1:1 weight ratio were dissolved in chloroform to prepare 17 mg/ml blend solutions. After heated and stirred at 40°C about 3 h, the blend solutions were spin-coated on PEDOT:PSS films at 2500 RPM for 30 s and then the active layer on the substrate were placed in a small petri dish to perform solvent vapor annealed (SVA) treatment by carbon disulphide for 30 s and then annealed at 80°C for 5 min. After that, PDIN solution (2 mg/ml in methanol with 0.25 vol% acetic acid) was spin-coated on the top of active layers at 5000 RPM for 30 s to prepare cathode interlayer. Finally, the cathode of Al was

deposited by thermal evaporation with a shadow mask under 10⁻⁴ Pa and the thickness of 100 nm was monitored by a quartz crystal microbalance. The active area is approximately 3.65 mm², which is defined by the overlapping area of ITO anode and Al cathode.

Device Measurement: The shadow mask with well-defined aperture (2.89 mm²) is utilized to define the active area in OSCs measured with mask. The current density-voltage (J-V) curves of all the organic solar cells were measured by a Keithley 2400 unit in high-purity nitrogen-filled glove box. The AM 1.5G irradiation was provided by an XES-40S2 (SAN-EI ELECTRIC Co., Ltd) solar simulator (AAA grade, 70×70 mm² photobeam size) with light intensity of 100 mW/cm², which was calibrated by standard silicon solar cells (purchased from Zolix INSTRUMENTS CO. LTD). The external quantum efficiency (EQE) spectra of organic solar cells were measured in air conditions by a Zolix Solar Cell Scan 100. The ultraviolet-visible (UV-Vis) absorption spectra of neat and blend films were obtained using a Shimadzu UV-3101 PC spectrometer. The morphology of the PEDOT:PSS films was investigated by atomic force microscopy (AFM, Dimension Icon). Transmission electron microscopy (TEM) images of active layers were obtained by a JEOL JEM-1400 transmission electron microscope operated at 80 kV.



Fig. S1 absorption spectra of neat J71, IT-2Cl and J71:IT-2Cl blend films.



Fig. S2 The *J*-*V* curves of OSCs under different measurement condition, a) without mask, b) with mask.

| SVA | Spin-coating PDIN | Without mask | | | | With mask | | | |
|-----|----------------------|------------------------|------|-------|--------------------|------------------------|------|-------|--------------------|
| | | J_{SC} | Voc | FF | РСЕ | J _{SC} | Voc | FF | РСЕ |
| | | (mA cm ⁻²) | (V) | (%) | (%) | (mA cm ⁻²) | (V) | (%) | (%) |
| DCM | S-PDIN | 27.48 | 0.84 | 56.30 | 13.00 (12.91±0.12) | 18.74 | 0.82 | 72.74 | 11.17 (11.05±0.14) |
| | D-PDIN | 19.09 | 0.84 | 70.82 | 11.36 (11.24±0.13) | 18.79 | 0.82 | 74.39 | 11.46 (11.32±0.15) |
| CF | S-PDIN | 26.33 | 0.85 | 56.36 | 12.61 (12.53±0.11) | 18.45 | 0.83 | 72.38 | 11.08 (10.94±0.15) |
| | D-PDIN | 18.76 | 0.85 | 68.14 | 10.86 (10.70±0.17) | 18.37 | 0.83 | 72.90 | 11.11 (11.03±0.13) |
| | S-PDIN | 25.23 | 0.86 | 53.50 | 11.61(11.49±0.14) | 18.12 | 0.84 | 67.42 | 10.26 (10.16±0.14) |
| | D-PDIN | 18.68 | 0.86 | 64.95 | 10.43 (10.31±0.15) | 18.35 | 0.84 | 67.25 | 10.36 (10.22±0.16) |

Table S1 Photovoltaic parameters of the OSCs measured with or without mask.

Remarks: The average PCEs and standard deviations were calculated with 10 cells prepared from different batches.



Fig. S3 TEM images of a) pristine blend films; SSC method.

b) blend films post-treated with methanol by