

Supporting Information

Efficiency Enhancements in Non-Fullerene Acceptor-Based Organic Solar Cells by Post-Additive Soaking

Hoyeon Choi^{a†}, Jihoon Lee^{b†}, Chang-mok Oh^a, Soyeong Jang^b, Hyojung Kim^c, Munseok Jeong^c,
Sung Heum Park^{b*}, and In-Wook Hwang^{a*}

^a Advanced Photonics Research Institute, Gwangju Institute of Science and Technology,
Gwangju 61005, Republic of Korea

^b Department of Physics, Pukyong National University, Busan 48513, Republic of Korea

^c Department of Energy Science, Sungkyunkwan University, Suwon 16419, Republic of Korea

[†]These authors contributed equally to this work

AUTHOR INFORMATION

Corresponding Authors

*Emails: hwangiw@gist.ac.kr (I.-W. Hwang); spark@pknu.ac.kr (S.H. Park)

EXPERIMENTAL METHODS

Device fabrication. ITIC-m, PBDB-T, and PDINO were purchased from 1-Materials, while PEDOT:PSS was obtained from Heraeus. OSCs with a p-i-n structure of ITO/PEDOT:PSS/PBDB-T:ITIC-m/PDINO/Al were fabricated as follows. The ITO substrates were sequentially sanitized with water, acetone, and isopropyl alcohol for 30 min, before being placed in hot oven (80 °C) for 12 h to completely remove residual alcohol. PEDOT:PSS (PH) was then spin-coated on top of the ITO substrates at 4500 rpm for 45 s. The PBDB-T:ITIC-m (1:1) composites were dissolved in CB (20 mg/ml) and spin-coated on top of the PEDOT:PSS layer at 2500 rpm for 60 s. The thickness of the PBDB-T:ITIC-m BHJ films was ~100 nm. The PBDB-T:ITIC-m BHJ films were dried under a nitrogen atmosphere for 30 min to evaporate CB. We performed PAS treatments to these pristine BHJ films by spin-coating cosolvents (~1 ml) comprised of DIO (1, 0.5, 0.25, and 0.125 vol.%) and *n*-hexane (bal.) at 4000 rpm for 60 s. To remove residual DIO and defective traps on surface, we rinsed the PAS-treated BHJ films by spinning them with methanol (99.8%, anhydrous, Sigma-Aldrich) solvent at 2000 rpm for 30 s, following the literature.^[1,2] On top of the BHJ films, PDINO (dissolved in methanol (0.1 mg/ml)) was spin-coated at 4000 rpm for 30 s.^[3] Finally, an aluminum electrode with a thickness of ~100 nm was deposited at $< 1 \times 10^{-6}$ Torr by using a thermal evaporator.

Measurements. Steady-state absorption and PL spectra were recorded using a Hitachi UH5300 UV-vis absorption spectrometer and Hitachi F-7000 fluorescence spectrometer, respectively. BHJ films coated on quartz substrates were used for the PL and absorption measurements. The thickness of the films was measured using an alpha step thickness monitor system.

Transmission electron microscopy (TEM) images were recorded using a Hitachi H-7500 image analyzer. For TEM measurement, we transferred BHJ films onto a TedPella copper grid by immersing BHJ films coated on glass/PEDOT:PSS into deionized water. The J - V characteristics were recorded using an AAA solar simulator coupled with a Keithley 2400 source measure unit under AM 1.5 G illumination at 100 mW/cm^2 . Incident photon-to-electron conversion efficiency (IPCE) spectra were recorded using a xenon lamp with a monochromator and a multimeter.

Femtosecond transient absorption spectra and decays were recorded using a commercial transient absorption spectrometer (HELIOS, Ultrafast Systems) combined with a 1 kHz repetition rate femtosecond Ti:sapphire regenerative amplifier system (Legend, Coherent). A pump pulse at 750 nm with a power density of $6.4 \mu\text{J/cm}^2$ was produced using an optical parametric amplifier (TOPAS-OPA, Coherent) and a neutral density filter, while a broadband white-light continuum probe pulse was generated by focusing a small portion of the 800 nm amplifier output into a sapphire window. Transient absorption signals in respective optical delays of pump and probe pulses were collected using an optical fiber coupled to a multichannel spectrometer.

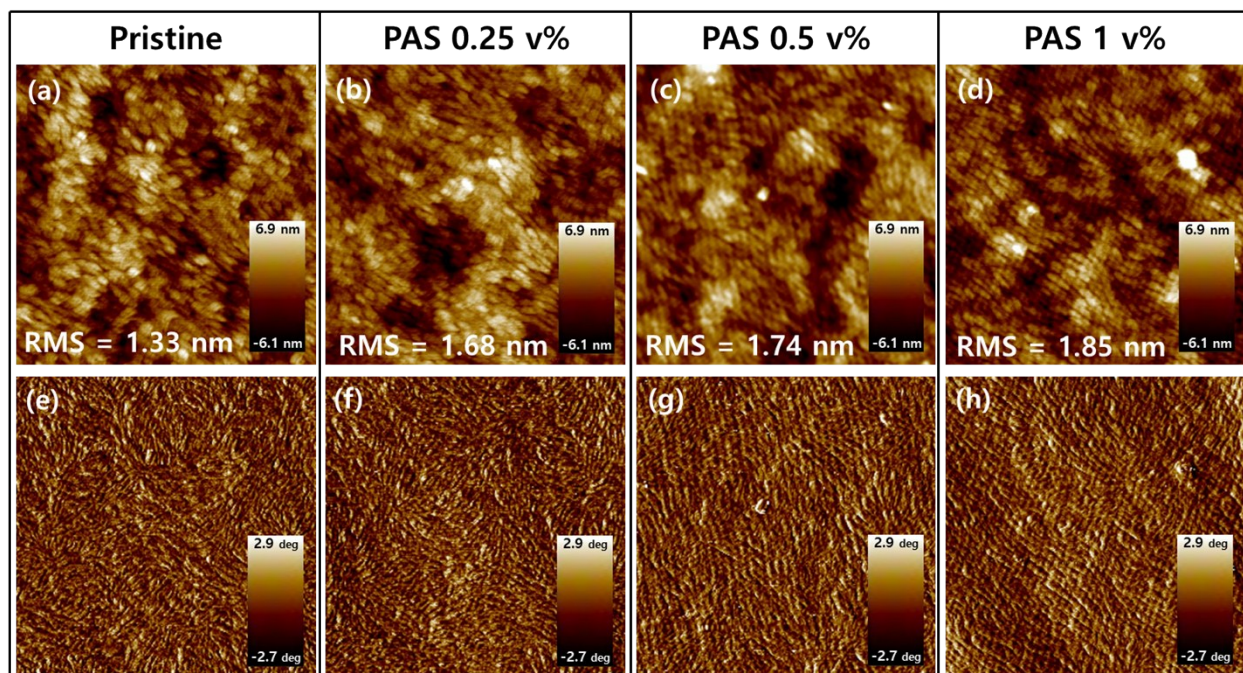


Figure S1. (a-d) Tapping mode AFM topography images ($2.5 \mu\text{m} \times 2.5 \mu\text{m}$) of the PBDB-T:ITIC-m (1:1) films before (pristine) and after PAS treatment using 1, 0.5, and 0.25 vol.% DIOL in n-hexane. (e-h) Corresponding AFM phase images.

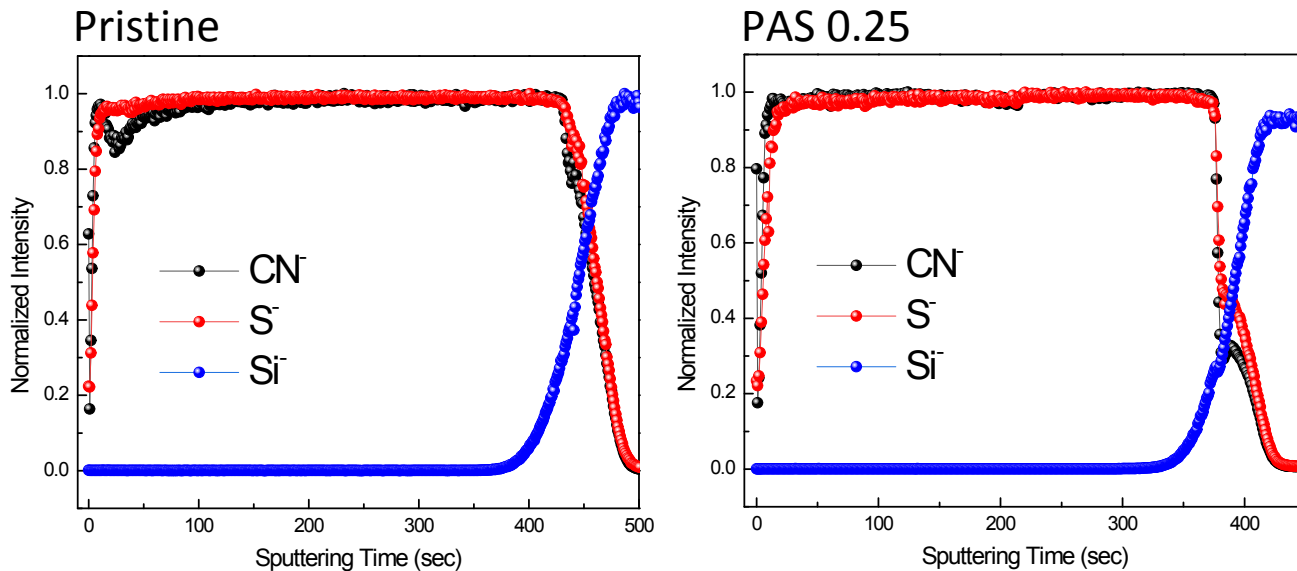


Figure S2. In-depth compositional distributions of the PBDB-T:ITIC-m (1:1) BHJ film before (left) and after (right) PAS, which were obtained using a ToF-SIMS (time of flight secondary ion mass spectroscopy) depth profiler (TOF-SIMS5, ION-TOF GmbH). The BHJ samples were prepared on glasses. The plots for CN^- in 0~100 sec are indicative of disappearance of local surface segregation of ITIC-m moieties after the PAS.

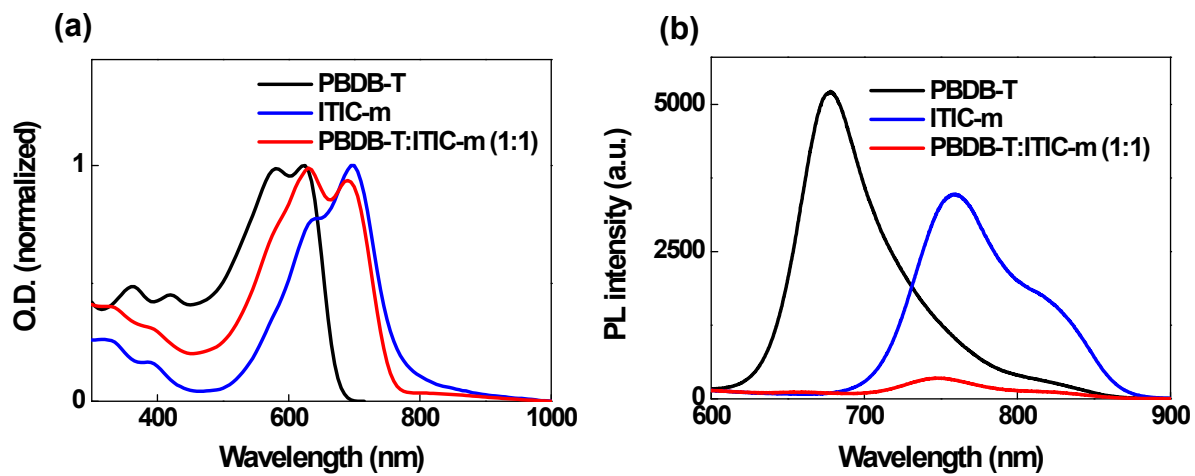


Figure S3. (a) Absorption and (b) PL spectra of films of PBDB-T, ITIC-m, and PBDB-T:ITIC-m (1:1).

An excitation wavelength of 400 nm was used in the PL measurements.

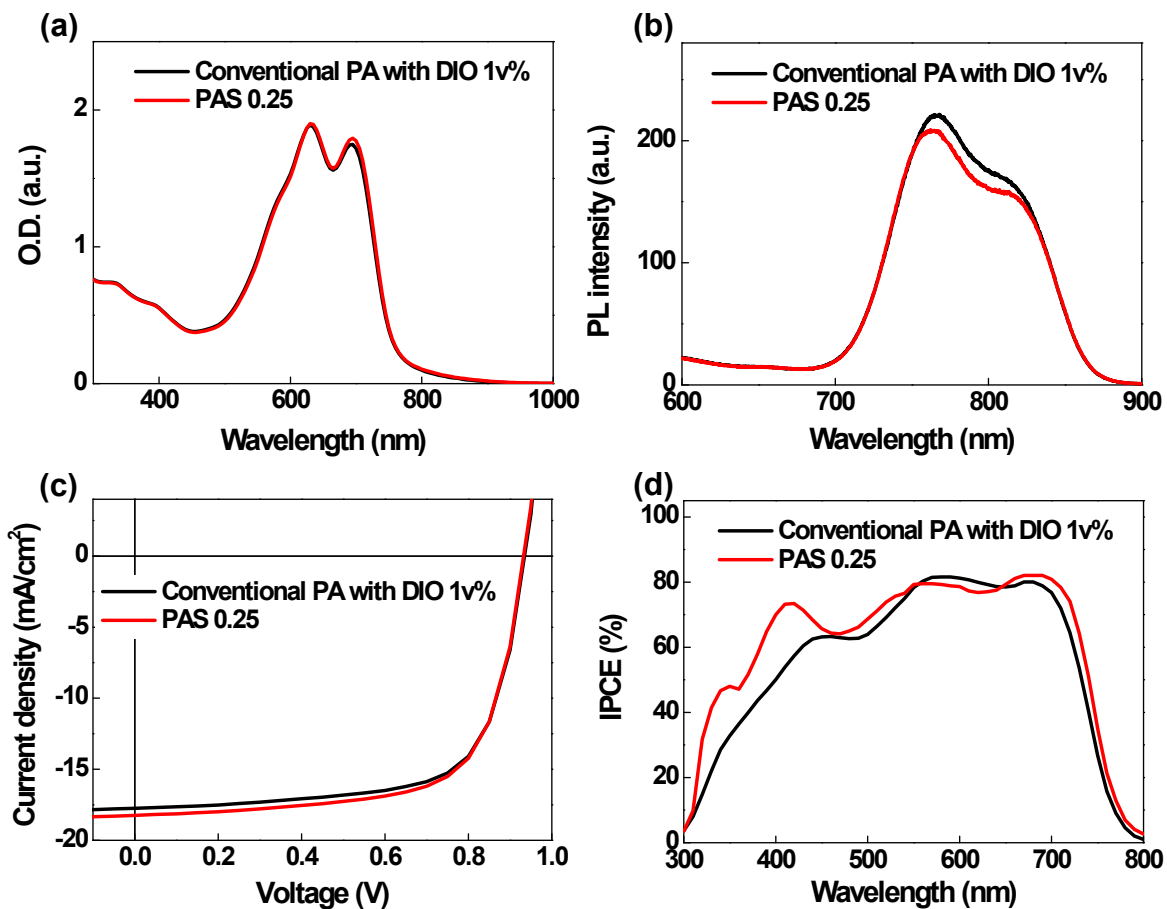


Figure S4. (a) Absorption and (b) PL spectra of films fabricated using conventional processing additive adding (PA) into PBDB-T:ITIC-m (1:1) solution i.e., 1 vol.% DIO adding into PBDB-T:ITIC-m (1:1) chlorobenzene solution and PAS 0.25. An excitation wavelength of 400 nm was used in the PL measurements. (c) J-V and (d) EQE plots for the OSCs fabricated using films of PA and PAS 0.25.

Table S1. Device parameters for OSCs fabricated using PA (1 vol.% DIO adding into PBDB-T:ITIC-m (1:1) chlorobenzene solution) and PAS 0.25 films.

	<i>J_{sc}</i> (/)	<i>V_{oc}</i> (V)	<i>FF</i>	<i>PCE (%)</i>
PA	17.74	0.94	0.69	11.45
PAS 0.25	18.47	0.94	0.71	12.27

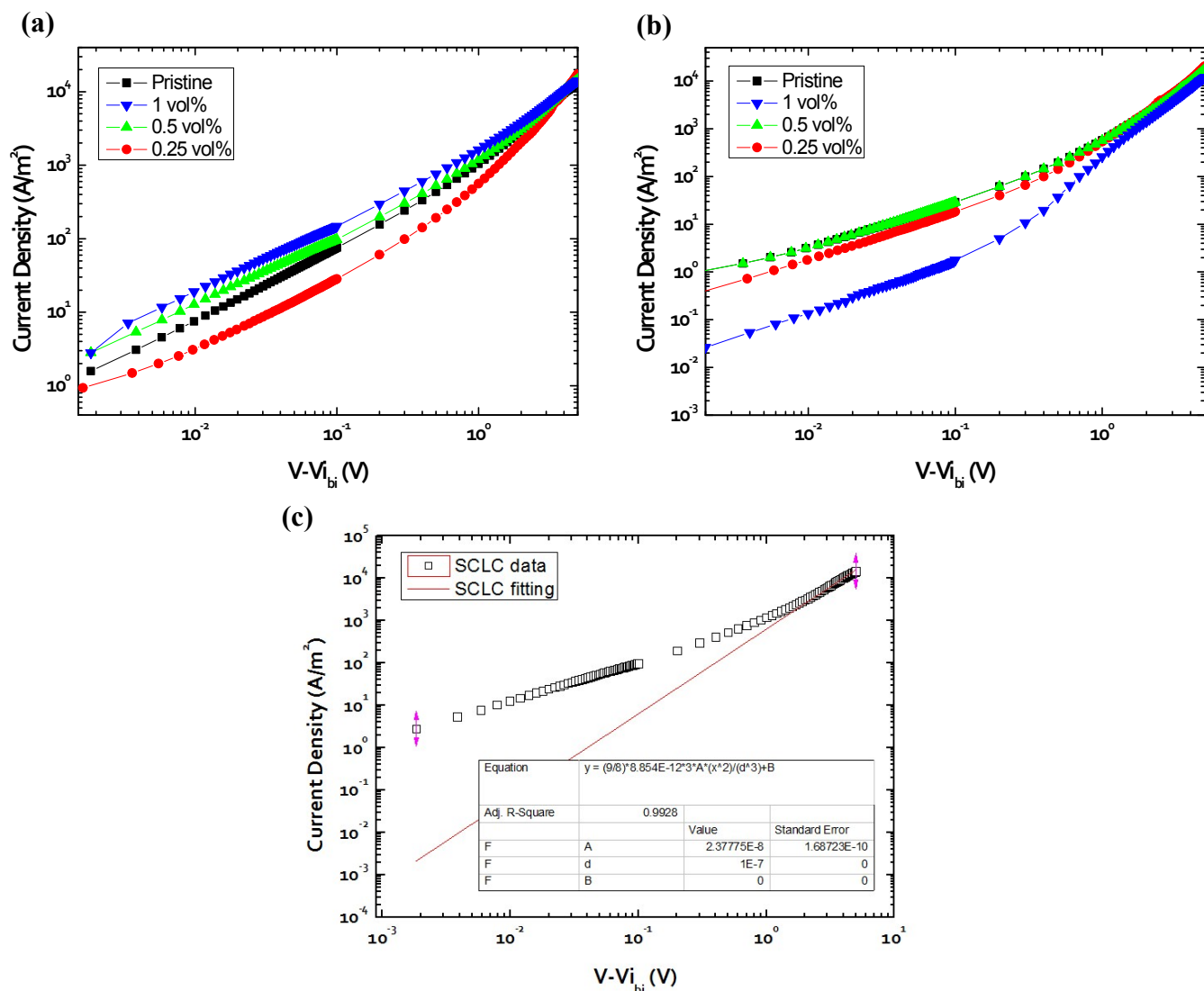


Figure S5. J - V characteristics of (a) hole-only and (b) electron-only devices fabricated using pristine and PAS-treated PBDB-T:ITIC-m films. Device structures of ITO/PEDOT:PSS/PBDB-T:ITIC-m/MoO_x/Ag and ITO/ZnO/PBDB-T:ITIC-m/PDINO/Al were used for the hole- and electron-only devices, respectively. (c) To obtain carrier mobility, a space-charge limited current (SCLC) model with equation $J = (9/8) \epsilon_0 \epsilon_r \mu (V - V_{bi})^2 / d^3$ was used, where J is current density, V is applied voltage, V_{bi} is built-in voltage, d is active layer thickness ($d = 85$ nm for PAS-treated films; $d = 100$ nm for pristine film), ϵ_r is relative dielectric constant (assumed to be 3), ϵ_0 is permittivity of free space, and μ is carrier mobility. Note that the fitting was conducted in the SCLC regime. The R^2 values for the fitting were 0.99-1.00.

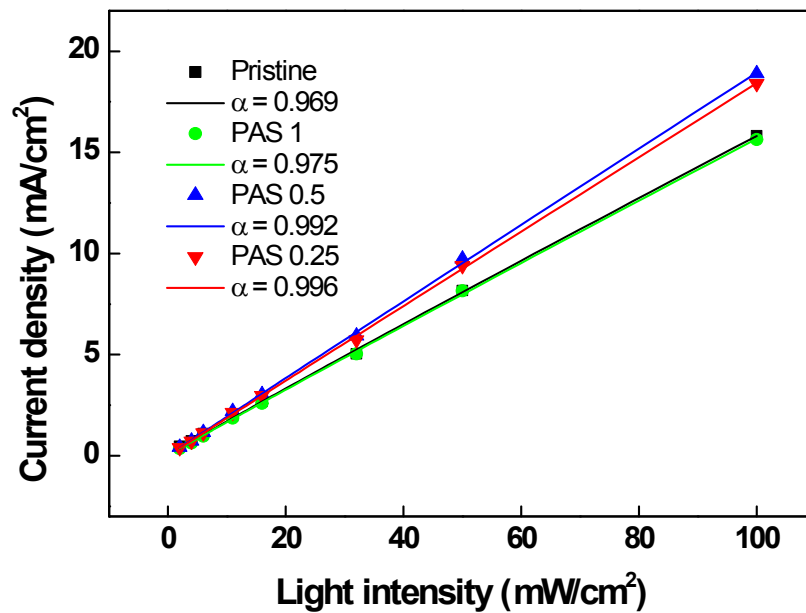


Figure S6. Photocurrents of OSCs plotted as a function of irradiation light intensity. For fitting the data, a power law function, $J_{ph} \propto P^\alpha$, was used. The fit lines and fitted α values for OSCs fabricated using pristine and PAS- treated PBDB-T:ITIC-m films are indicated.

References

- [1] L. Ye, Y. Jiang, X. Guo, H. Sun, S. Zhang, M. Zhang, L. Huo, and J. Hou, *J. Phys. Chem. C*, 2013, **117**, 14920-14928.
- [2] H. Zhou, Y. Zhang, J. Seifert, S. D. Collins, C. Luo, G. C. Bazan, T.-Q. Nguyen, and A. J. Heeger, *Adv. Mater.*, 2013, **25**, 1646-1652.
- [3] Z.-G. Zhang, B. Qi, Z. Jin, D. Chi, Z. Qi, Y. Li, and J. Wang, *Energy Environ. Sci.*, 2014, **7**, 1966-1973.