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Electronic Supplementary Information

Simultaneous improvement of three parameters using binary processing solvent approach in as-cast non-fullerene solar cells

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Fabrication and Characterization of Solar Cells.

The device structure was ITO/ZnO/Polymer:(acceptor or acceptor pair) (=1:1 by weight) /MoO_3/Al. Patterned indium tin oxide (ITO)-coated glass with a sheet resistance of 15–20 ohm/square was cleaned by a surfactant scrub and then underwent a wet-cleaning process inside an ultrasonic bath that began with deionized water, followed by acetone and 2-propanol. A solution-processed zinic oxide (ZnO) interlayer of 30 nm was prepared according to a recent report. The CF and CF:MES solution were dissolved in ambient temperature. The active layer based on PTQ10:IT-4Cl blend solution was then deposited on top of the ZnO interlayer by casting. The thickness of the active layer was verified by a surface profilometer (Tencor Alpha-500, USA). A 5 nm molybdenum oxide

(MoO_x) layer and a 100 nm Al layer were subsequently evaporated through a shadow mask to define the active area of the devices (~5.8 mm^2) and form a top anode. All of the fabrication processes were performed inside a controlled atmosphere of nitrogen dry box (Vacuum Atmosphere, USA) that contained less than 10 ppm oxygen and moisture. The power conversion efficiencies of the resulting polymer solar cells were measured under 1 sun, AM 1.5G (air mass 1.5 global) spectrum from a solar simulator (Oriel model 91192, USA) set to 100 mW/cm^2. The *J-V* characteristics were recorded with a Keithley 2400 source unit (USA). The external quantum efficiencies of the conventional solar cells were measured with a commercial photo modulation spectroscopic setup that included a xenon lamp, an optical chopper, a monochromator, and a lock-in amplifier operated by a PC computer. A calibrated Si photodiode was used as a standard. Note: Non-fullerene acceptor IT-4Cl was purchased from Derthon Optoelectronic Materials Science Technology Co LTD.

Space-Charge Limited Current (SCLC) Measurement.

Hole-only and electron-only devices were fabricated to measure the hole and electron mobilities of active layers using the space charge limited current (SCLC) method with hole-only device of ITO/PEDOT:PSS/Active layer/Au and electron-only device of ITO/ZnO/Active layer/PFN (poly[(9,9-dioctyl-2,7-fluorene)-alt-(9,9-bis(30-(N,N-dimethylamino)propyl)-2,7-fluorene)])/Al. The mobilities (μ) were determined by fitting the dark current to the model of a single carrier SCLC, described by the equation:

$$J = \frac{9}{8}\varepsilon_0\varepsilon_r\mu\frac{V^2}{d^3}$$

where J is the current, E is the effective electric field, ε_0 is the permittivity of free space, ε_r is the material relative permittivity, d is the thickness of the active layer, and V is the effective voltage. The effective voltage can be obtained by subtracting the built-in voltage (V_{bi}) and the voltage drop (V_s) from the substrate's series resistance from the applied voltage (V_{appl}), $V = V_{appl} - V_{bi} - V_s$. The mobility can be calculated from the slope of the $J^{1/2} \sim V$ curves.

Atomic Force Microscopy and Transmission Electron Microscopy

Tapping-mode atomic force microscopy (AFM) images were obtained using a Nano Scope NS3A system (Digital Instrument) to observe the surface morphology of as-cast active layers. Transmission electron microscopy (TEM) images were obtained using JEM-2100F with an accelerating voltage of 30 kV.

X-ray Photoelectron Spectroscopy/ESCA and Grazing Incidence Wide-Angle X-ray Scattering (GIWAXS)

The XPS measurements were performed on the Axis Ultra DLD system. GIWAXS measurements were conducted by Xeuss 2.0 of Xenocs. The scattered x-rays were detected using a Pilatus3R 1M Dectris area detector. Samples were prepared on Si substrates using identical blend solutions as those used in the as-cast devices.

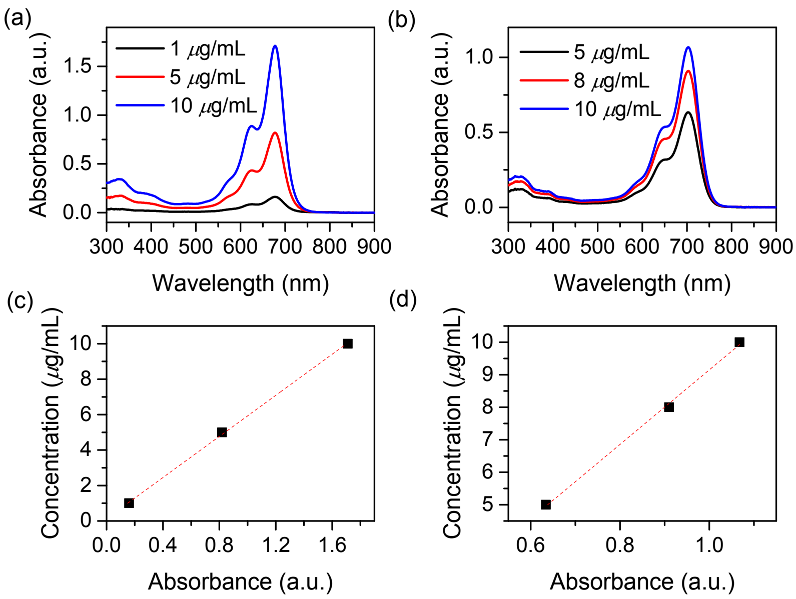

Fig. S1 Calibration spectra used to calculate solubility. (a) and (b) Absorption spectra of standard solutions of IT-4Cl in MES and CF, respectively. (c) Plot of IT-4Cl absorbance at 677 nm *vs*. concentration. (d) Plot of IT-4Cl absorbance at 703 nm *vs*. concentration.

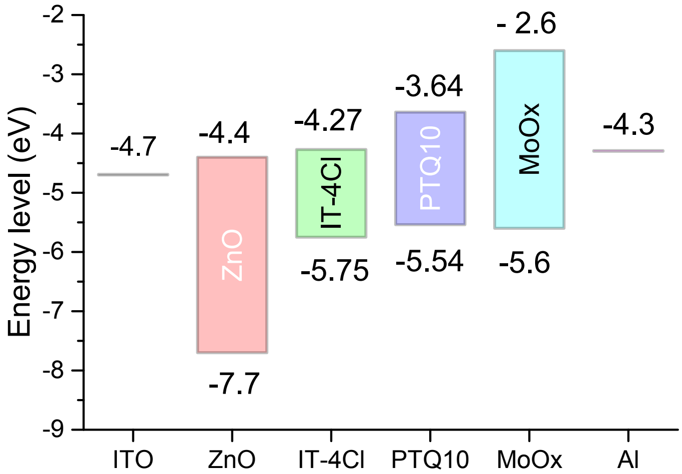

Chart S1. The energy level diagrams for the materials used in the inverted devices.

Table S1. Photovoltaic parameters of PTQ10:IT-4Cl (1:1, w/w) based devices fabricated by the CF:MES binary solvents with different MES volume contents.

MES content	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
10%	0.91	19.43	68.67	12.14
20%	0.91	19.47	69.66	12.34
30%	0.91	19.19	69.87	12.20
50%	0.91	19.61	73.01	13.04
100%	0.89	16.72	61.68	9.18

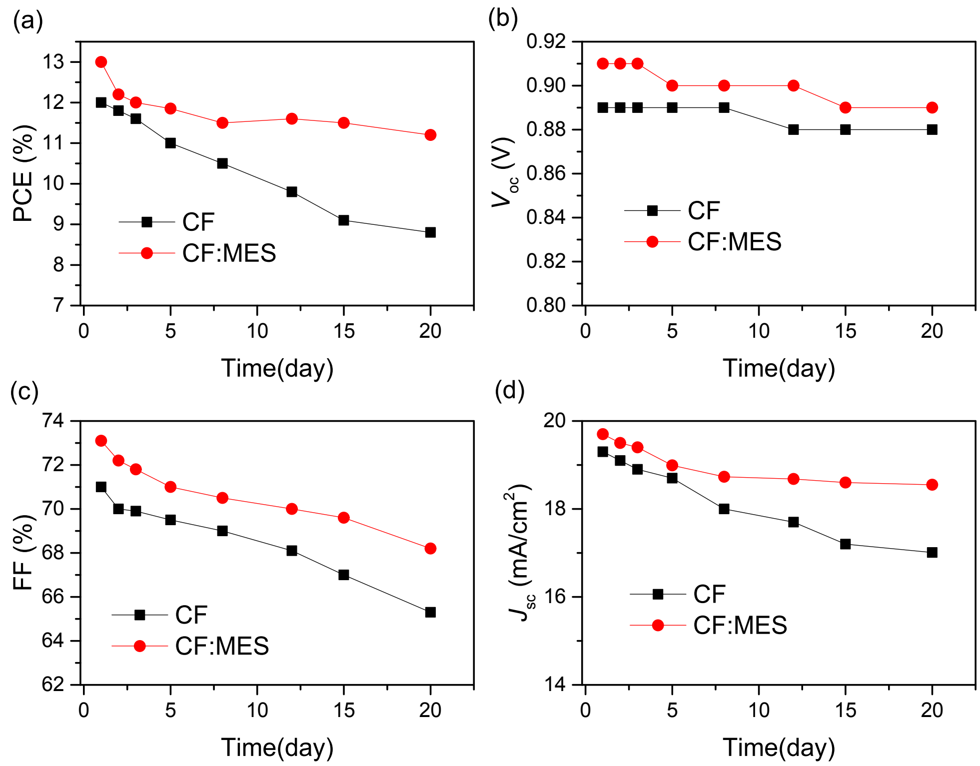

Fig. S2 Shelf stability of the inverted as-cast solar cells kept in the air for 20 days: (a) PCE *vs*. time, (b) V_{oc} *vs*. time, (c) FF *vs*. time and (d) J_{sc} *vs*. time.

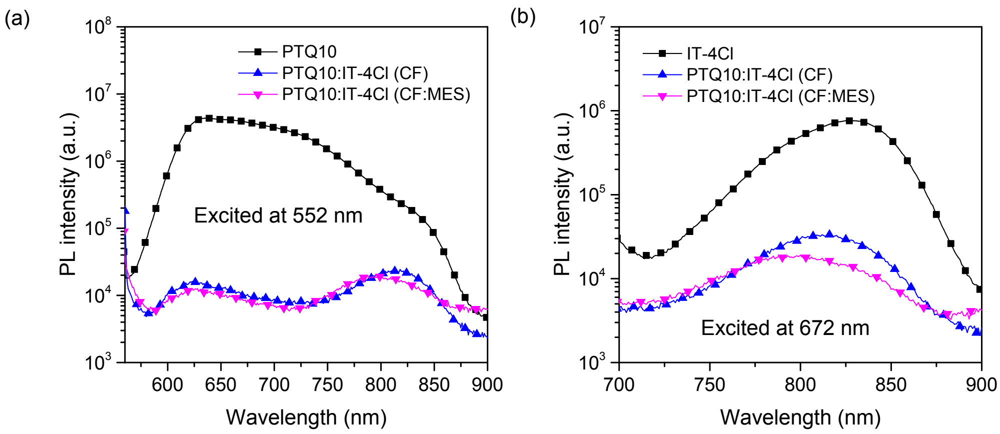

Fig. S3 Photoluminescent spectra of the pristine films of PTQ10, IT-4Cl and their blend films processed by different solvents.

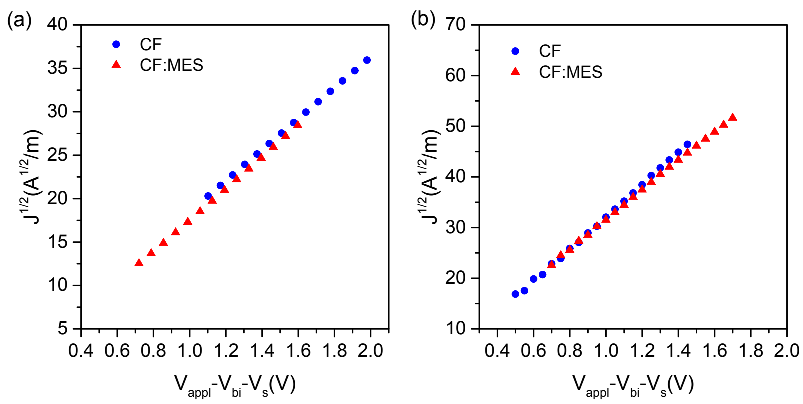

Fig. S4 (a) Electron and (b) hole mobilities of the devices based on different solvents.

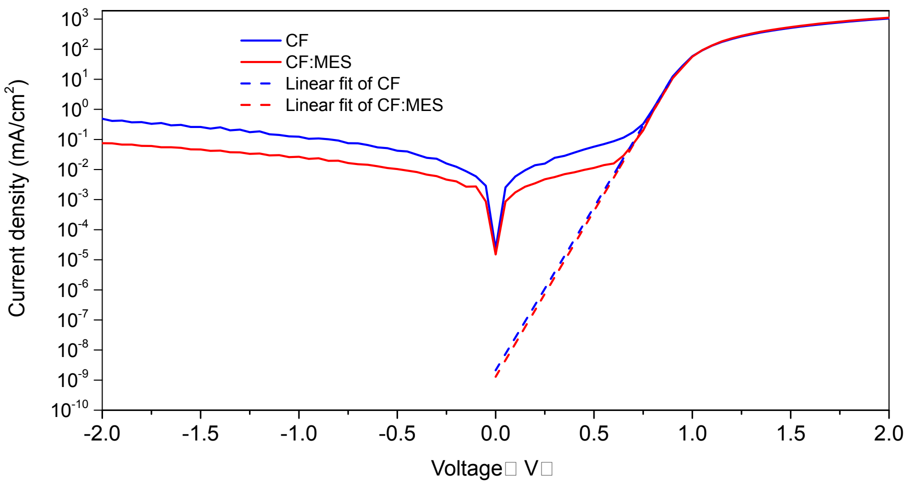

Fig. S5 Solid line shows the dark current *J–V* curve, and the dashed lines show the extrapolated line for determining saturation current density from intercept and diode ideality factor from the slope.

Table S2. Photovoltaic parameters of PTQ10:IT-4Cl (1:1, w/w) based solar cells in a PEDOT:PSS-containing conventional structure with different processing solvents.

Processing solvent	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
CF	0.89	19.01	69.96	11.85 (11.51 ± 0.34)
CF:MES	0.90	18.61	66.71	11.18 (10.86 ± 0.32)

Table S3. Element content of blend films processed by different solvents.

	F (%)	O (%)	N (%)	C (%)	Cl (%)	F/Cl ratio
CF	5.62	1.65	4.26	87.42	1.05	5.35
CF:MES	5.49	2.24	4.35	87.06	0.86	6.38

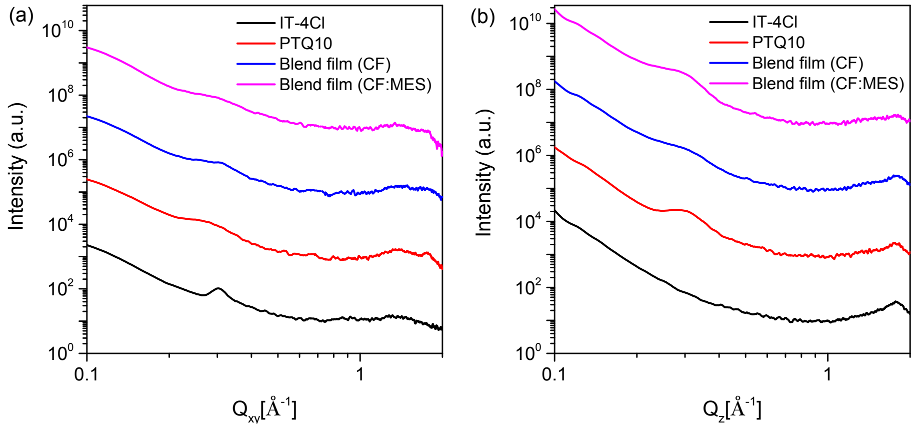

Fig. S6 Corresponding line-cuts profiles in (a) in-plain (IP) and (b) out-of-plain (OOP) directions.

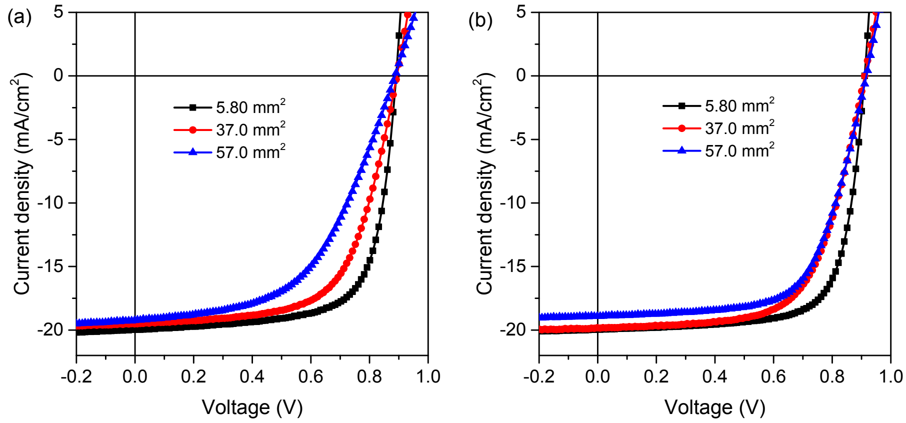

Fig. S7 The *J*–*V* curves of PSCs of different device areas, casted from (a) CF (b) CF:MES (1:1).