Highly Efficient and Reproducible Non-Fullerene Solar Cells from Hydrocarbon Solvents

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SUPPORTING INFORMATION

Experimental methods

Materials. PffBT4T-2DT and EH-IDTBR were synthesised according to literature procedures,\textsuperscript{1-3} and reactions were carried out under an inert argon atmosphere using conventional Schlenk techniques. All other chemicals and solvents were purchased from Sigma Aldrich and Acros Organics and used as received.

OPV device fabrication and characterisation. Bulk heterojunction solar cells were fabricated in an inverted architecture (glass/ITO/ZnO/PffBT4T-2DT:EH-IDTBR/MoO\textsubscript{3}/Ag). Glass substrates, pre-patterned with ITO (15 Ω sheet resistance per square), were cleaned by sonication in acetone, detergent, deionized water and isopropanol before ozone plasma treatment for 10 min. A layer of ZnO, approximately 30 nm, (from Zn(OAc)\textsubscript{2} in 60 μL monoethanolamine and 2 mL of 2-methoxyethanol) was deposited by spin-coating onto the ITO substrate at 4,000 r.p.m. for 40 s, followed by annealing at 150 °C for 20 min. Active layer solutions (PffBT4T-2DT:EH-IDTBR, weight ratio 1:1) were prepared from CB, TMB, mesitylene and o-xylene, all 15 mg mL\textsuperscript{-1} concentration. The CB solution was heated to 80 °C overnight, whereas the TMB, mesitylene and o-xylene solutions were heated to 60 °C. For devices processed from CB, the solutions and substrates were preheated to 100 °C and the active layer was deposited by spin-coating at 2,500 r.p.m. for 1 min. For devices processed from TMB and o-xylene, the active layer was deposited from r.t. solutions and substrates by spin-coating at 2,000 r.p.m. for 1 min. For devices processed from mesitylene, the active layer was deposited from r.t. solutions and substrates by spin-coating at 1,750 r.p.m. for 1 min. The active layers were then dried at 80 °C for 5 min, under an inert atmosphere. A MoO\textsubscript{3} anode interlayer (10 nm) and Ag anode (100 nm) were then deposited by thermal evaporation through a shadow mask, giving an active area of
0.045 cm$^2$ per device. The J-V characteristics were measured under AM1.5G (100 mW cm$^{-2}$) irradiation using an Oriel Instruments Xenon lamp calibrated to a Si reference cell to correct for spectral mismatch, and a Keithley 2400 source meter. EQE was measured by comparing the photocurrent from a calibrated photodiode to the photocurrent of the sample cell at the same monochromatic light intensity within the same spectral range. The monochromatic light was achieved using a CVI DIGIKROM 240 grating spectrometer in combination with a xenon arc light source.

**Morphological analysis.** Grazing incidence wide angle x-ray scattering (GIWAXS) measurements were performed at beam-line D1 at the Cornell High Energy Synchrotron Source, Wilson Lab, NY, USA. Active layer blends were prepared using the optimized device processing conditions for each solvent onto cleaned Si substrates. A fast 2D detector (PILATUS 200 k from Dectris) was used to record the scattering pattern with an exposure time of 0.2 s. An X-ray beam with wavelength of 1.17 Å was used to perform the GIWAXS experiments. The experiments were performed at sample-to-detector-distance of 167.15 mm and an incident angle of 0.15° with respect to the sample plane. Silver behenate was used to calibrate the lengths in the reciprocal space. Dark-field transmission electron microscopy (DF-TEM) measurements were carried out using a FEI Tecnai Bio-twin Transmission Electron Microscope (TEM) operated at 120 kV and a Titan 80–300 kV (ST) equipped with GIF TridiemER energy filtered detector and 2 CDD cameras, operating at 300 kV in Dark field mode. Samples for TEM were prepared by spin-coating the active layer blends, using the optimized device processing conditions, on top of a water-soluble sacrificial layer and then emerged in water. Floating layers were placed on copper grids (300 Mesh) with lacey carbon film and were dried overnight under vacuum to remove any water residue.
**Stability studies.** Storage lifetime was probed by leaving devices under an inert atmosphere in the dark and measuring the J-V characteristics of the cells every 10 days. For the photo-stability study, devices were aged in a nitrogen filled chamber with inert conditions maintained by an over pressure of bottled nitrogen (oxygen-free) gas. During ageing of the device, a continuous white LED array was used to measure the performance with the light intensity such that the short circuit current matched characterisation of the device from an AM1.5G solar simulator. Solar cell current-voltage characteristics were taken every 20 min with a Keithley 2401 Sourcemeter and custom written Igor procedure. White light and UV photo-bleaching studies were carried out in thin films, spin-coated from 5 mg mL\(^{-1}\) solutions of PfluBT4T-2DT:EH-IDTBR (1:1) blends at 2000 r.p.m. For white light photo-bleaching experiments, the films were aged using a continuous white LED array in an inert atmosphere. For UV photo-bleaching experiments the films were aged using 100 W, 365 nm UV lamp in an inert atmosphere. UV-vis absorption spectra were measured using a UV-1601 Shimadzu UV-vis spectrometer.

**Supplementary figures and tables**

![Figure S1](image)

Figure S1. (a) Active layer blends processed from CB (left) and mesitylene (right) with spin coating from r.t. substrates. (b) High viscosity active layer solution in CB at r.t. (total concentration = 15 mg mL\(^{-1}\)). (c) Low viscosity active layer solution in mesitylene at r.t. (total concentration = 15 mg mL\(^{-1}\)).
Table S1. J-V Characteristics of the inverted structure PffBT4T-2DT:EH-IDTBR devices.

<table>
<thead>
<tr>
<th>Processing Conditions</th>
<th>V_{oc} (V)</th>
<th>J_{sc} (mA cm^{-2})</th>
<th>FF</th>
<th>PCE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TMB</td>
<td>1.01</td>
<td>16.9</td>
<td>0.61</td>
<td>10.4</td>
</tr>
<tr>
<td>TMB/PN (97.5:2.5)</td>
<td>0.90</td>
<td>11.6</td>
<td>0.49</td>
<td>5.1</td>
</tr>
</tbody>
</table>

Devices were measured under AM1.5G illumination at 100 mW cm^{-2}, FF is the fill factor, PCE is the power conversion efficiency.

Figure S2. Thin film UV-Vis absorption coefficient spectra (corrected for thickness) of the PffBT4T-2DT:EH-IDTBR blend in CB and mesitylene spin-coated from 5 mg mL^{-1} solutions in their respective solvents at 3000 r.p.m.
Figure S3. (a) Thin film UV-Vis absorption spectra of the neat PffBT4T-2DT, EH-IDTBR and the active layer blend spin-coated from 5 mg mL$^{-1}$ solutions in their respective solvents at 1500 r.p.m. (b) Thin film UV-Vis absorption of the active layer blends spin-coated from 5 mg mL$^{-1}$ solutions in their respective solvents at 1500 r.p.m.

Figure S4. Temperature-dependent solution UV-Vis absorption spectra for a 0.1 mg mL$^{-1}$ solution of PffBT4T-2DT in mesitylene.
Figure S5. Pole figures for the GIWAXS measurements of PffBT4T-2DT:EH-IDTBR blend films from CB and mesitylene for, (a) Scattering from the \( \pi \)-stacking of the polymer. (b) Scattering from the lamellar stacking of the polymer.

Figure S6. Line-cuts of integrated scattering intensity of GIWAXS measurements of PffBT4T-2DT:EH-IDTBR blend films from CB and mesitylene; integration was performed for (a) All crystallite orientations (angles from 0° to 90). (b) In the in-plane direction and (c) Out-of-plane direction.
Figure S7. Dark field TEM images of PffBT4T-2DT:EH-IDTBR active layers from CB, (a) Before and (b) After aging for 150 days, in a glovebox under N₂.

Figure S8. Dark field TEM images of PffBT4T-2DT:EH-IDTBR active layers from mesitylene, (a) Before and (b) After aging for 150 days, in a glovebox under N₂.
Figure S9. Photo-stability of the $J_{SC}$ for PffBT4T-2DT:EH-IDTBR solar cells kept under constant white light illumination for 10 days.

Figure S10. Photo-stability of the $V_{OC}$ for PffBT4T-2DT:EH-IDTBR solar cells kept under constant white light illumination for 10 days.
Figure S11. Photo-stability of the FF for PffBT4T-2DT:EH-IDTBR solar cells kept under constant white light illumination for 10 days.

Figure S12. (a) UV-Vis absorption spectrum of the PffBT4T-2DT:EH-IDTBR blend processed from CB upon exposure to 365 nm UV radiation for 10 days in an inert atmosphere. (b) UV-Vis absorption spectrum of the PffBT4T-2DT:EH-IDTBR blend processed from CB upon exposure to white light from an LED array for 10 days.
Figure S13. (a) UV-Vis absorption spectrum of the PffBT4T-2DT:EH-IDTBR blend processed from mesitylene upon exposure to 365 nm UV radiation for 10 days in an inert atmosphere. (b) UV-Vis absorption spectrum of the PffBT4T-2DT:EH-IDTBR blend processed from mesitylene upon exposure to white light from an LED array for 10 days.

REFERENCES
