### **Electronic Supplementary Information**

For

# In-Situ Modulation of the Vertical Distribution in a Blend of P3HT and PC<sub>60</sub>BM via the Addition of a Composition Gradient for Improving Photovoltaic Performance

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## **Table of Contents**

- 1. Image of F-ADD film (Fig. S1)
- 2. UV-vis spectra of PC<sub>60</sub>BM with F-ADD in chloroform (Fig. S2)
- 3. Contact angle and surface energy (Fig. S3)
- 4. Secondary ion Mass Spectrometry (Fig. S4)
- 5. Device evaluations (Fig. S5, S6, S7, S8 and S9)

#### 1. Image of F-ADD film



**Fig. S1** An image of F-ADD film spun from chloroform (100 mg/mL) on a PEDOT:PSS-coated glass substrate.

#### 2. UV-vis spectra of PC<sub>60</sub>BM with F-ADD in chloroform



**Fig. S2** UV-Vis. absorption spectra of PC<sub>60</sub>BM with F-ADD in 4.0 x  $10^{-4}$  M (a) and 1.3 x  $10^{-5}$  M chloroform solution.

### 2. Contact angle and surface energy

## Contact Angle (Water)

Туре	Droplet	Θ (°)	Туре	Droplet	Θ (°)
PCBM only		87.1	3 wt%		97.9
0.25 wt%		92.1	5 wt%		100.6
0.5 wt%	$\mathbf{O}$	93.8	10 wt%		103.3
1 wt%	0	95.9	P3HT only		101.2

## Contact Angle (Glycerol)

Туре	Droplet	Θ (°)	Туре	Droplet	Θ (°)
PCBM only		53.8	3 wt%		59.4
0.25 wt%		55.6	5 wt%		61.5
0.5 wt%		56.5	10 wt%		66.8
1 wt%		57.1	P3HT only		90.2

Fig. S3 Contact angles of  $PC_{60}BM$  containing various amounts of F-ADD on a PEDOT:PSS-coated substrate.



#### **3. Secondary Ion Mass Spectrometry Result**

**Fig. S4** SIMs of P3HT:PC<sub>60</sub>BM blend films with 0 (a), 0.25 (b), 0.5 (c), 1.0 (d), 3.0 (e), 5.0 (f) and 10 (g) % of F-ADD.





Fig. S5 Deviation of photovoltaic parameters of 8 devices polymer solar cells.



**Fig. S6** Devices data of polymer solar cells with 0 (a), 0.25 (b), 0.5 (c), 1.0 (d), 3.0 (e), 5.0 (f) and 10 (g) % of F-ADD.



**Fig. S7** IPCE curves of the polymer solar cells with 0 (a), 0.25 (b), 0.5 (c), 1.0 (d), 3.0 (e), 5.0 (f) and 10 (g) % of F-ADD



**Fig. S8** Dark J-V characteristics of polymer solar cells with 0 (a), 0.25 (b), 0.5 (c), 1.0 (d), 3.0 (e), 5.0 (f) and 10 (g) % of F-ADD.



**Fig. S9** Devices data of polymer solar cells 0 (a), 0.5 (b) wt % of F-ADD in various annealing conditions.