Electronic Supplementary Information

Nanomorphology-driven two-stage hole mobility in blend films of regioregular and regiorandom polythiophenes

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Figure S1. Optical absorption coefficient (α) spectra for all pristine and blend films: (a) on quartz substrates, (b) glass/ITO/PI substrates.



Figure S2. CV diagrams of Rreg-P3HT (a,c) and Rran-P3HT (b,d): The photographs in (a) and (b) show slightly different color of each solution even though the concentration is same. The onset points of first oxidation are given in (c) and (d). If the first oxidation peaks are enlarged (not shown here), we can find the starting point: 0.307 V for Rreg-P3HT and 0.488 V for Rran-P3HT. References: (1) M. Al-Ibrahima, H. K. Rotha, U. Zhokhavetsb, G. Gobsch, S. Sensfuss, *Sol. Energy Mater. Sol. Cells.* **2005**, 85, 13; (2) D. M. Stevens, Y. Qin, M. A. Hillmyer, C. D. Frisbie, *J. Phys. Chem. C.* **2009**, 113, 11408.



Figure S3. Photoelectron (PE) yield spectra of Rreg-P3HT and Rran-P3HT thin films. The onset point difference was almost 0.05 eV, whereas the starting point was not clearly differentiated.



Figure S4. Output characteristics for OFETs fabricated in this work. The gate voltage was increased from -10 V to -80 V as indicated with arrow on the top panel. Note that the drain current unit was changed from μ A to nA at the bulk RR range between 87.5 % and 86.3 %.



Figure S5. Phase mode 2D AFM images (left: grey scale; right: full color scale) for the surface of pristine and blend films: (a) Pristine Rreg-P3HT film (RR = 92.2 %), (b) Rreg-P3HT:Rran-P3HT = 54:46 (by weight) (bulk RR = 65.0 %), and (c) pristine Rran-P3HT film (RR = 33.0 %). The scan size was $5\mu m \times 5\mu m$.



Figure S6. Enlarged HR-TEM images of (a) Rreg-P3HT (RR = 92.2 %) and (b) Rran-P3HT (RR = 33.0 %).



Figure S7. 2D GIXD images of ITO-glass substrate (left) and PMDA-ODA PI coated ITO-glass (right): The bottom image was taken from the image of PI coated ITO-glass to focus on the PI diffraction close to the (100) diffraction of P3HTs.



Figure S8. 1D GIXD profiles of pristine and blend films with azimuthal angle (α): (a) Pristine Rreg-P3HT film (RR = 92.2 %), (b) Rreg-P3HT:Rran-P3HT = 96:4 (by weight) (bulk RR = 89.8 %), (c) Rreg-P3HT:Rran-P3HT = 90:10 (by weight) (bulk RR = 86.3 %), and (d) pristine Rran-P3HT film (RR = 33.0 %).



Figure S9. 1D GIXD profiles of (a) ITO-glass substrate and (b) PMDA-ODA PI coated ITO-glass with azimuthal angle (α).



Figure S10. The (100) intensity change as a function of azimuthal angle (α): (a) Pristine Rreg-P3HT film (RR = 92.2 %), (b) Rreg-P3HT:Rran-P3HT = 96:4 (by weight) (bulk RR = 89.8 %), (c) Rreg-P3HT:Rran-P3HT = 90:10 (by weight) (bulk RR = 86.3 %), (d) pristine Rran-P3HT film (RR = 33.0 %), and (e) PMDA-ODA PI coated ITO-glass (the intensity here was taken at $2\theta = 4.3 \sim 4.5^{\circ}$).



Figure S11. Schematic illustration (ideal case) of P3HT chain stacking: The direction of each coordinate (a, b, c) is shown in the bottom right corner.