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Supporting Information

Systematic Investigation of Self-Organization Behavior in Supramolecular π-Conjugated Polymer for Multi-color Electroluminescence

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EXPERIMENTAL SECTION

Materials: All solvents were purchased from Aldrich or J&K Chemicals without further purification. Anhydrous THF (HPLC grade) was obtained from Solvent Purification Systems (Innovative Technology, Inc.). Anhydrous chloroform was pre-dried over molecular sieves. DMF was dried over calcium hydride (CaH) and distilled under a dry nitrogen atmosphere immediately prior to use. The other solvents were distilled under a dry nitrogen atmosphere immediately prior to use.

Characterizations: Gel permeation chromatography (GPC) analysis was performed on a HP1100 HPLC system equipped with 7911GP-502 and GP NXC columns using polystyrenes as the standard and tetrahydrofuran (THF) as a eluent at a flow rate of 1.0 mL/min. Absorption spectra were measured with a Shimadzu UV-3600 spectrometer at 25 °C, and emission spectra were recorded on a Shimadzu RF-5301(PC)S luminescence spectrometer. The quartz cells of 10 mm thickness were used to measure the spectra. Dynamic light scattering (DLS, Zetasizer Nano-ZS) measurements were carried out at a wavelength of 633 nm laser as the light source at room temperature. The time-dependent autocorrelation function of the scattered light intensity was measured at an angle of 90 °. The DLS measurements were usually repeated at least three times, and the average values were used for an analysis. Atomic force microscopy (AFM) measurements were obtained with a NanoScope IIIa AFM. Film thicknesses were measured using a Bruker Dektak XT stylus profiler. A DI water contact angle method was adopted to explore the surface energy (γ_s) of polymer films on SiO₂ substrate. Recorded FTIR spectra are obtained from IR Prestige-21 (Fouere Transform Infrared Spectrophotoometer, Shimadzu.).

The experimental parameters of fluorescence lifetime measurements are show as follows. The incident 390-nm, 150-fs laser pulses were generated from a Coherent TOPAS-C optical parametric amplifier; pumped by a 1 kHz Coherent Legend regenerative amplifier that is seeded

by a Coherent Vitesse oscillator. These input laser pulses were focused by a lens (f = 20 cm) on the sample solutions in a 1-mm-thick quartz cell (beam spot ~1 mm inside the cell). The emission from the samples was collected at a back scattering angle of 150° by a pair of lenses and directed to an Optronis OptoscopeTM streak camera system which has an ultimate temporal resolution of 6 ps.

AFM and NanoIR measurements. For all the nanoscale infrared measurements, we used a nanoIR platform (Anasys Instruments Corp., Santa Barbara, CA, USA). The nanoIR2TM AFM-IR instrument used to collect the spatially resolved IR spectra is a new reconfiguration of an earlier version. For nanoIR study, samples were directly spin-coated on the Si substrates that are transparent in the mid-infrared and the tip of the AFM remained in contact with the object. When a sample absorbs the IR laser pulse, it warms via the photothermal effect, resulting in a rapid thermal expansion of the absorbing region of the sample. The thermal expansion pulse impacts the tip of the AFM cantilever and causes it to oscillate. As the amplitude of oscillations is proportional to the absorption, we were able to record infrared absorption spectra at a given point and to make chemical maps by scanning the surface at a given wavelength. The IR source was tuned from wavelength 3600 cm⁻¹ to 3200 cm⁻¹ in order to identify further the formation of hydrogen-bonding interaction (Free -OH and associated -OH, respectively.).

Preparations of solutions and films of PPFOH for optical analysis. The preparation of PPFOH solutions was carried out by dissolving polymers in the organic solvents, such as chloroform (CHCl₃), toluene and 1,2-dichloroethane (DCE), tetrahydrofuran (THF), 1, 4-dioxane (DOX) and N,N-dimethyl formamide (DMF), spontaneously overnight. Pristine films of PPFOH were spin-coated on quartz substrates from its solution (all concentrations are about 5 mg/ml) using the KW-4A spin-coater from the institute of micro-electronics of Chinese Academy of Science) at different spin-coated speed for 30 s to remove solvent molecules and control film thickness. For the chloroform (CHCl₃) and THF solutions, the spin-coated speed is about 500 rpm/min 8 s in the beginning and 2000 rpm/min 60 s later. And for the high solvent

boil point of DMF, we used a higher spin-coated speed, about 800 rpm/min 10 s first and 3000 rpm/min 120 s later. Besides, the spin-coated speed is 500 rpm/min 5 s in the beginning and 1500 rpm/min 90 s later for the toluene, 1, 4-dioxane (DOX) and 1,2-dichloroethane (DCE) solutions. And all spin-coated films are vacuum drying at 55 °C. The reported absorbance of the samples has been corrected for the solvent background. The excitation wavelength was 380 nm. PPFOH films for FTIR are prepared from THF and CHCl₃ solution on the KBr plates and vacuum drying at 55 °C. The spin-coated film on the quartz or Si substrate from THF and CHCl₃ solution (5 mg/ml) were prepared for decay time (quartz substrate) and GIXD analysis (Si substrate).

Preparations of PPFOH-based PLEDs and device characterization. The prototype device structure employed is indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene): poly(styrene sulfonicacid) (PEDOT:PSS)/PPFOH/TPBi/LiF/Al. A thin PEDOT: PSS hole-injection layer of 20 nm was coated on the transparent conductive ITO at a spin speed of 2000 rpm for 78 s. After baking at 100 °C for 15 min in an oven, a polymer layer (~50 nm) was spin-coated on top of the PEDOT: PSS layer from its solutions. Finally, a 20 nm electron transport layer (TPBi) and LiF (0.8 nm)/Al (100 nm) were deposited by thermal evaporation in vacuum. The current-voltage luminescence characteristics of the devices were recorded using a Keithley source meter (model 2602) and a luminance meter. The EL spectra and CIE coordinates of the devices were analyzed with a spectra-scan PR655 spectrophotometer. All the devices were characterized without encapsulation and carried out in the ambient condition at room temperature (RT). The emission area of each device is 12 mm².

Supported experimental results are shown as follow:



Figure S1. GPC curve of PPFO8, PPFOH-L, PPFOH and PPFOH-H.



Figure S2. Infrared characterization (IR) of polymer powder: PPFOH (Bottom) and PPFOH-H (Top).



Figure S3. DSC curve of PPFOH.



Figure S4. TGA curve of PPFOH.



Figure S5. CV curve of PPFOH.



Figure S6. Absorption spectra of PPFOH films spin-coated from various solutions with the concentration of 5 mg/ml.



Figure S7. Absorption and PL spectra of PPFO8 films spin-coating from CHCl₃, toluene and THF (5 mg/ml).



Figure S8. Tyndall effect of (a) PPFOH in CHCl₃ (Left) and THF (Right) solution with the same concentration of 5 mg/ml, (b) blank CHCl₃ and (c) THF.



Figure S9. Infrared characterization (IR) of polymer powder: PPFOH and PPFOH-H film coated from THF and CHCl₃.



Figure S10. Photographs of water droplets sitting on the surfaces of corresponding spin-coated films from THF (a) and CHCl₃ (b) (5 mg/ml).



Figure S11. Current density-voltage curves of PPFOHs-based PLEDs.



Figure S12. GIXD pattern images of PPFOH spin-coated films from CHCl₃ (a) and THF (b).