Supporting Information

Improving the performance of all-inorganic perovskite light-emitting diodes through using polymeric interlayers with a pendant design

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Synthesis of Monomers

Synthesis of 1-vinylpyrene. A solution of methyltriphenylphosphonium bromide (6.97 g, 19.5 mmol) in 20 mL of THF was cooled to 0 °C and dropwisely added with *n*-butyllithium (12.2 mL, 1.6 M in *n*-hexane). After stirring for 10 min at room temperature, the solution was slowly added with a solution of pyrene-1-carbaldehyde (3.00 g, 13.0 mmol) in 45 mL of THF. Afterwards, the reaction proceeded at room temperature for 16 h. The crude product was filtered, and the filtrate was diluted with DCM and extracted with water and brine sequentially. The organic phase was then dried by $MgSO_4$, filtered, and the solvent was removed under reduced pressure. Finally, the crude product was purified by column chromatography with hexane, and the pure fractions are combined and dried to give 2.09 g (70 % yield, yellow solids). ^{1}H-NMR (**Figure S1a**, 400 MHz, $CDCl_3$), δ (ppm): 8.36-8.39 (m, Ar-H, 1H), 8.08-8.20 (m, Ar-H, 5H), 7.96-8.03 (m, Ar-H, 3H), 7.74-7.81 (m, Ar-CH, 1H), 5.95-6.00 (dd, CH-CH_2, 1H), 5.58-5.61 (dd, CH-CH_2, 1H).

Synthesis of 4-(pyren-1-yl)butyl methacrylate. A solution of 4-(pyren-1-yl)butan-1-ol (1.00 g, 3.64 mmol) and 0.6 mL of triethylamine in 25 mL of THF was cooled to 0 °C and slowly added with a solution of methacryloyl chloride (0.4 mL) in 25 mL of THF. The reaction proceeded at room temperature for 16 h. The crude product was filtered, and the filtrate was diluted with DCM and extracted with water and brine

sequentially. The organic phase was then dried by $MgSO_4$, filtered, and the solvent was removed under reduced pressure. Finally, the crude product was purified by column chromatography with hexane/ethyl acetate, and the pure fractions were combined and dried to give 0.66 g (53% yield, yellow oil). ^{1}H-NMR (**Figure S1b**, 400 MHz, $CDCl_3$), δ (ppm): 8.23-8.26 (m, Ar-H, 1H), 8.07-8.16 (m, Ar-H, 4H), 7.97-8.01 (m, Ar-H, 3H), 7.84-7.86 (m, Ar-CH, 1H), 5.52-6.08 (d, C-CH_2, 2H), 4.20-4.23 (t, O-CH_2-CH_2, 2H), 3.36-3.40 (t, Ar-CH_2-CH_2, 2H), 1.83-1.99 (br, CH_2-CH_2-CH_2, -CH_3, 7H).

Synthesis of Polymers

Synthesis of poly(1-vinylpyrene) (P4a). 1-Vinylpyrene (2.00 g, 8.8 mmol) was dissolved in 18 mL of toluene and the solution was degassed with three freeze-pump cycles. Afterwards, AIBN (14 mg, 1 mol% respect to 1-vinylpyrene) was added and the polymerization proceeded at 80 ^{o}C for 24 h. Crude polymers were precipitated in MeOH to afford 0.89 g (44% yield, yellowish solids). Molecular weight evaluated by size exclusion chromatography (SEC): M_n = 8150, M_w = 13850, PDI = 1.70. ^{1}H-NMR (400 MHz, $CDCl_3$) of **P4a** is presented in **Figure S2**.

Synthesis of poly(pyren-1-ylmethyl methacrylate) (P4b). Pyren-1-ylmethyl methacrylate (2.00 g, 6.7 mmol) was dissolved in 13 mL of toluene and the solution was degassed with three freeze-pump cycles. Afterwards, AIBN (47 mg, 5 mol%

respect to pyren-1-ylmethyl methacrylate) was added and the polymerization proceeded at 80 ^{o}C for 24 h. Crude polymers were precipitated in MeOH to afford 1.73 g (77% yield, yellowish solids). Molecular weight evaluated by SEC: M_n = 8810, M_w = 13160, PDI = 1.49. ^{1}H-NMR (400 MHz, $CDCl_3$) of **P4b** is presented in **Figure S3**.

Synthesis of poly(4-(pyren-1-yl)butyl methacrylate) (P4c). 4-(Pyren-1-yl)butyl methacrylate (0.66 g, 2.0 mmol) was dissolved in 5 mL of toluene and the solution was degassed with three freeze-pump cycles. Afterwards, AIBN (10 mg, 3 mol% respect to 4-(pyren-1-yl)butyl methacrylate) was added and the polymerization proceeded at 80 ^{o}C for 24 h. Crude polymers were precipitated in MeOH to afford 0.55 g (83% yield, yellowish solids). Molecular weight evaluated by SEC: M_n = 6670, M_w = 13140, PDI = 1.97. ^{1}H-NMR (400 MHz, $CDCl_3$) of **P4c** is presented in **Figure S4**.

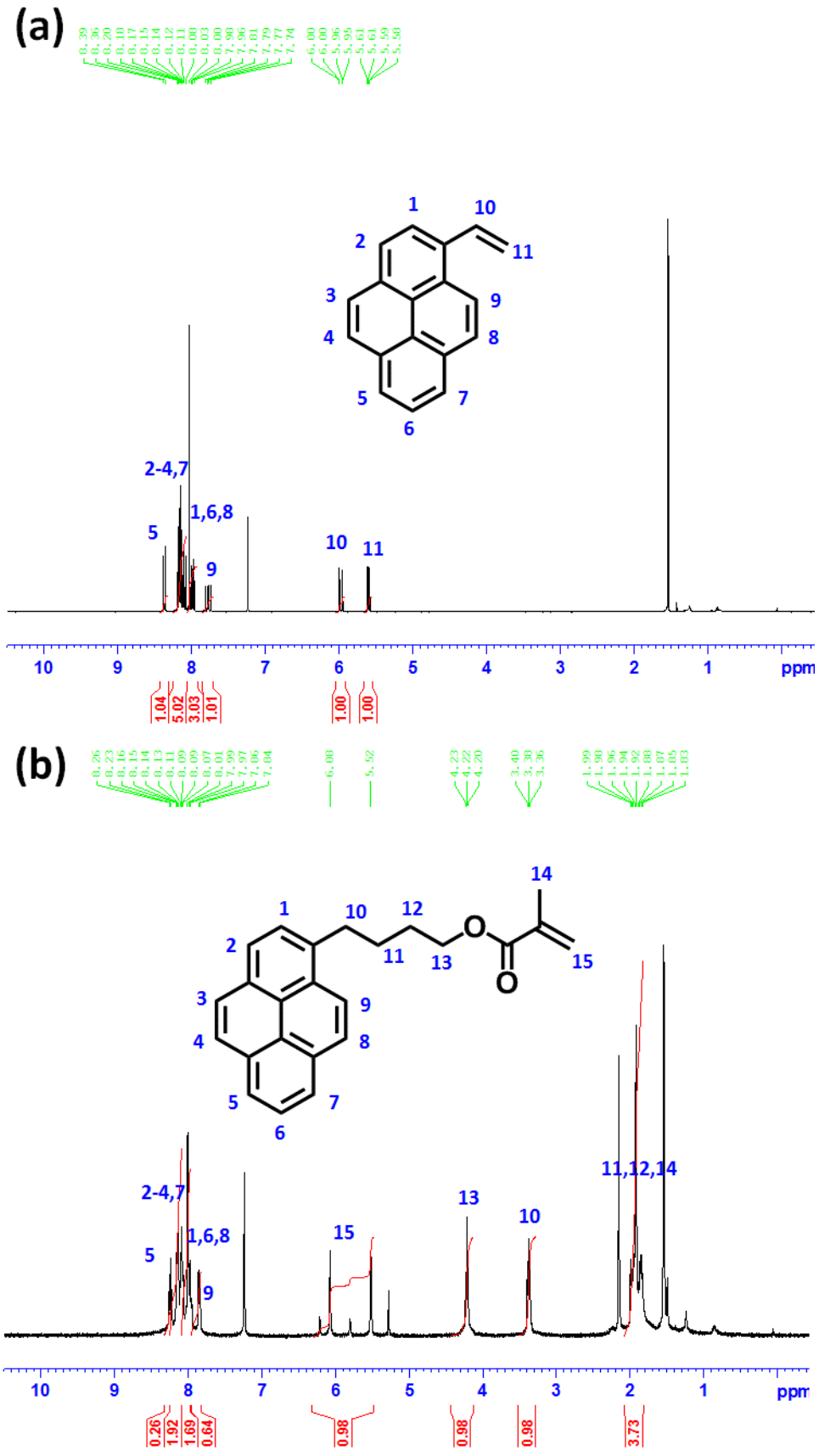


Figure S1. ^{1}H NMR spectra of (a) 1-vinylpyrene and (b) 4-(pyren-1-yl)butyl

methacrylate in $CDCl_3$.

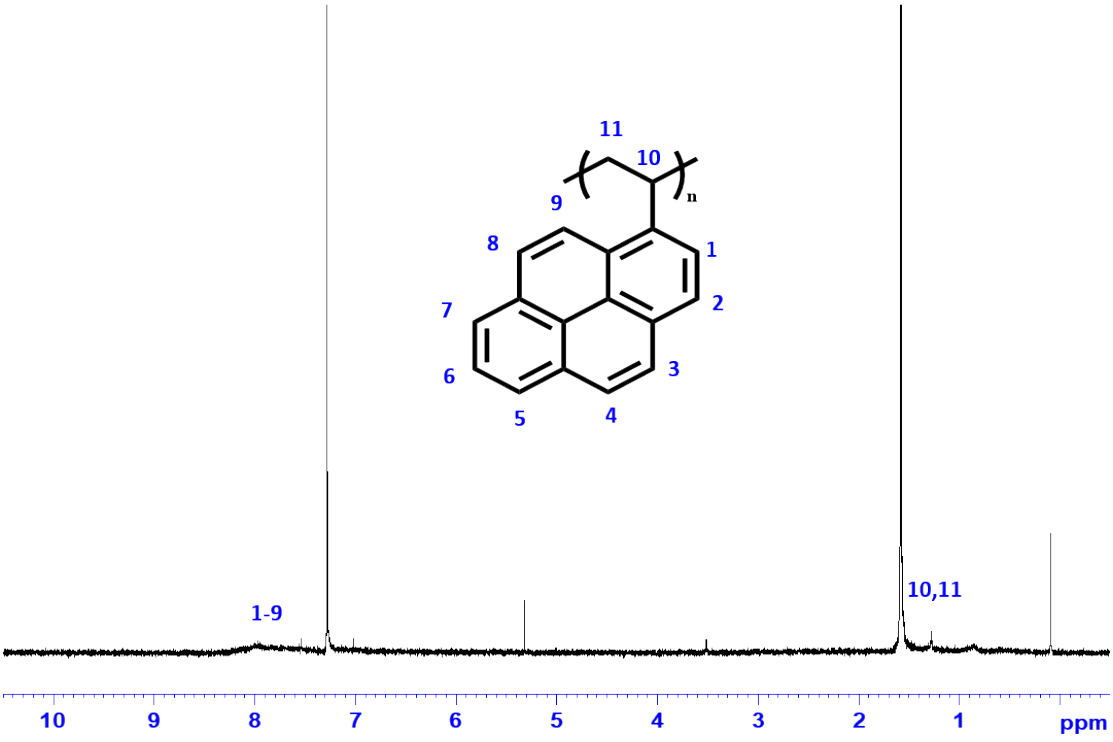


Figure S2. ^{1}H NMR spectrum of **P4a** in $CDCl_3$.

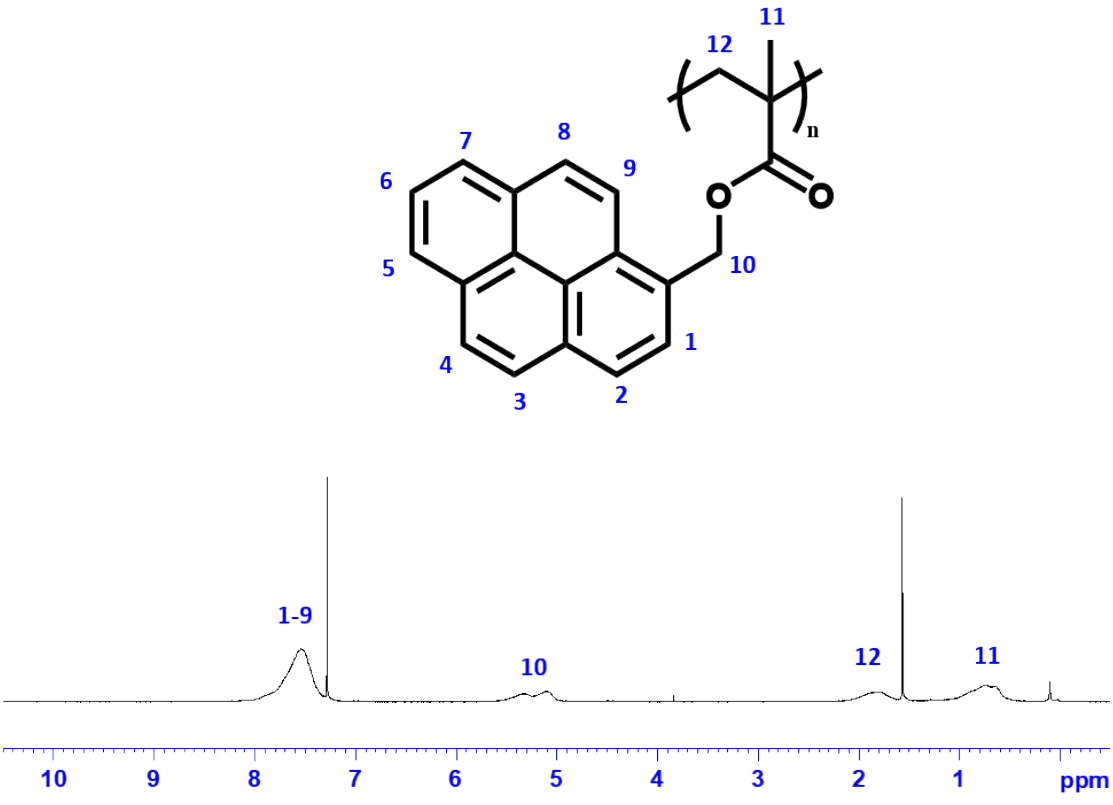


Figure S3. ^{1}H NMR spectrum of **P4b** in $CDCl_3$.

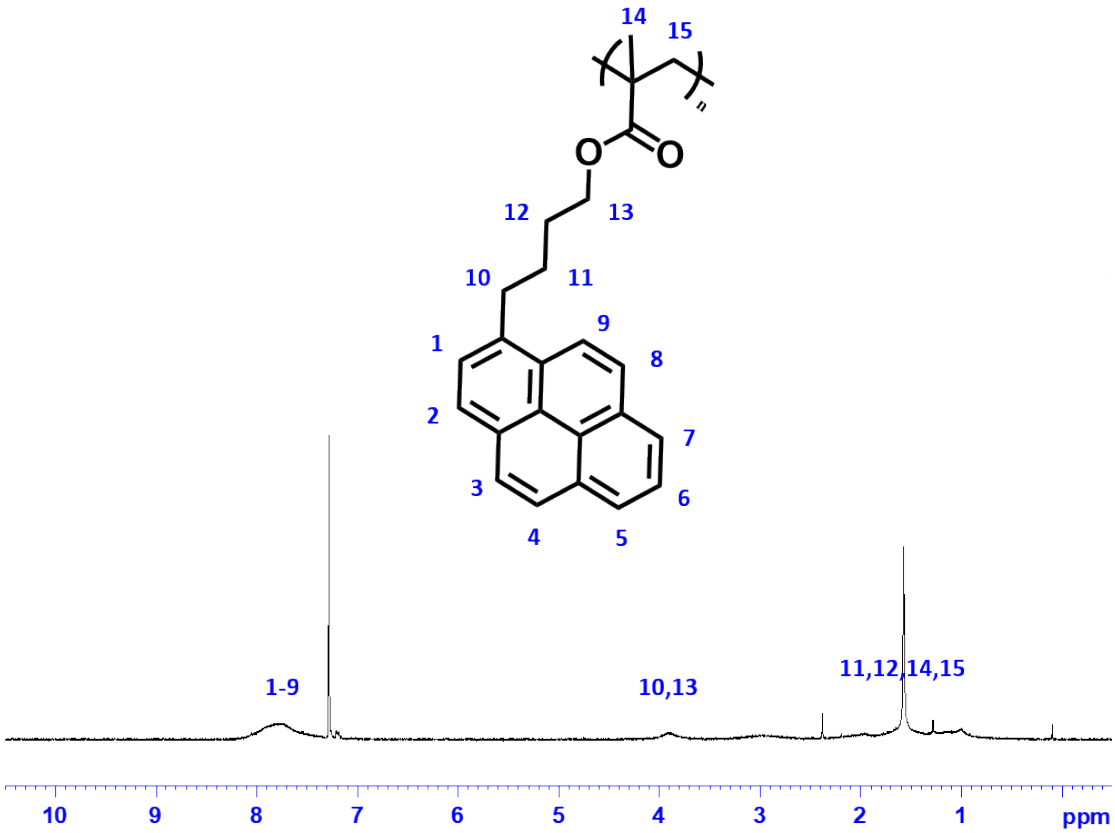
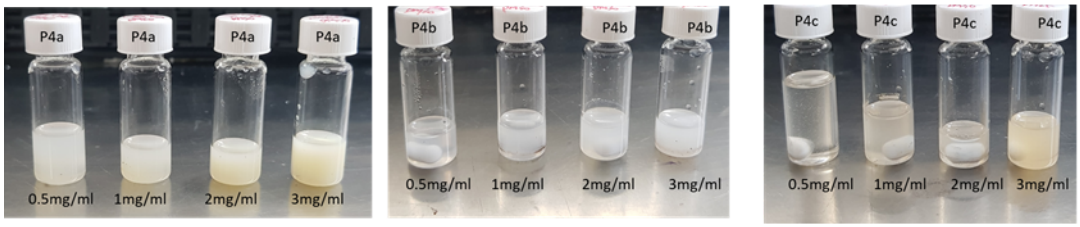


Figure S4. ^{1}H NMR spectrum of **P4c** in $CDCl_3$.

Solubility test of **P4a-c** in DMSO.



Solubility test of **P4a-c** in DMF.

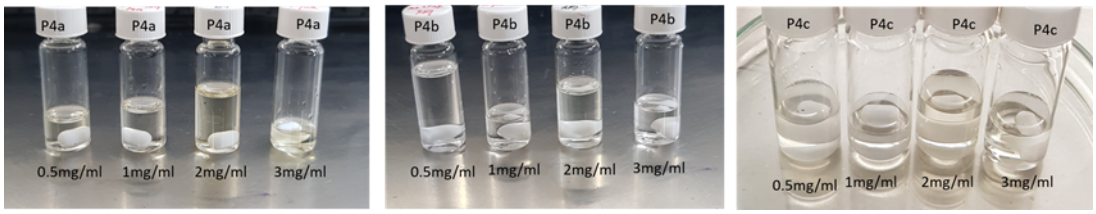


Figure S5. Solubility test of **P4a-c** in DMSO and DMF.

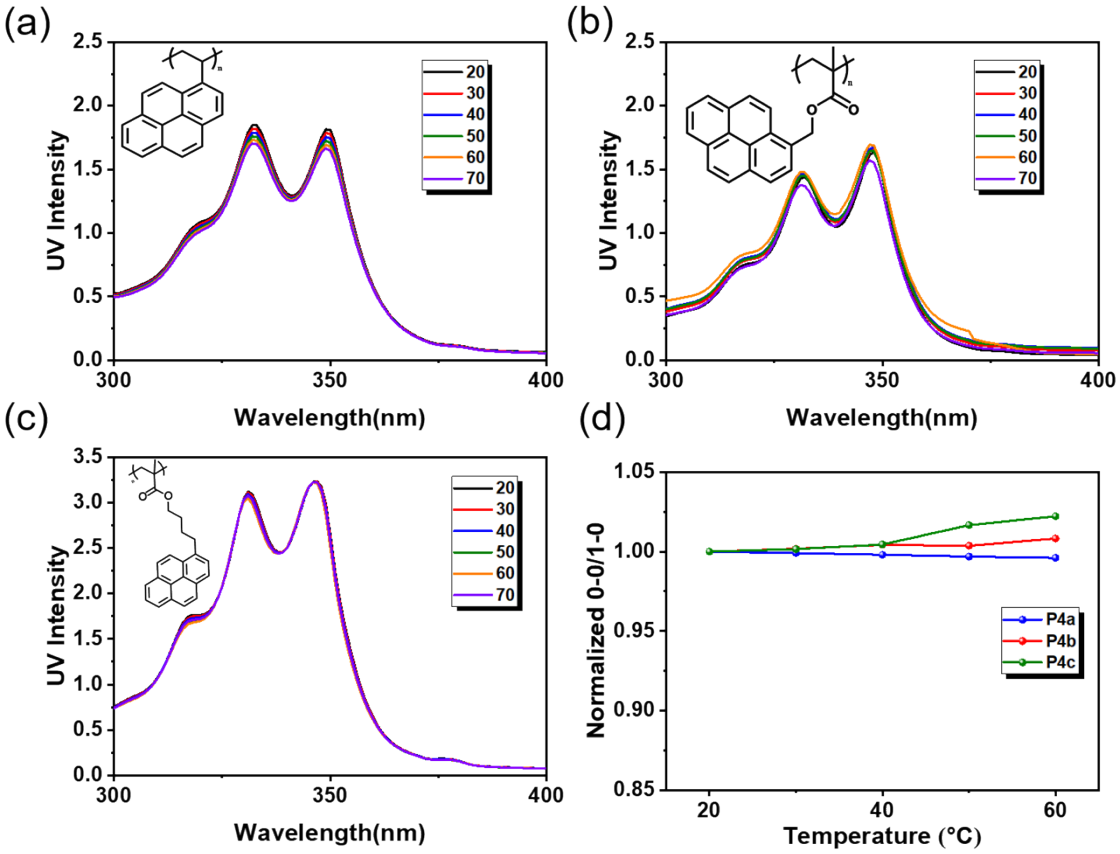


Figure S6. Temperature-dependent UV-vis absorption spectra of (a) **P4a**, (b) **P4b**, and (c) **P4c** solutions from 20−70 °C. (d) The calculated intensity ratio of the 0−0 and 0−1 characteristic absorption peaks of **P4a**, **P4b**, and **P4c**.

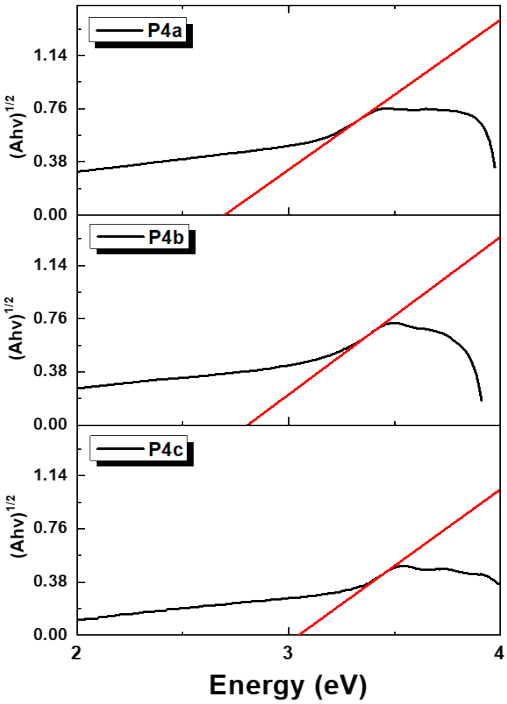


Figure S7. Tauc plots of the films of **P4a**–**c**.

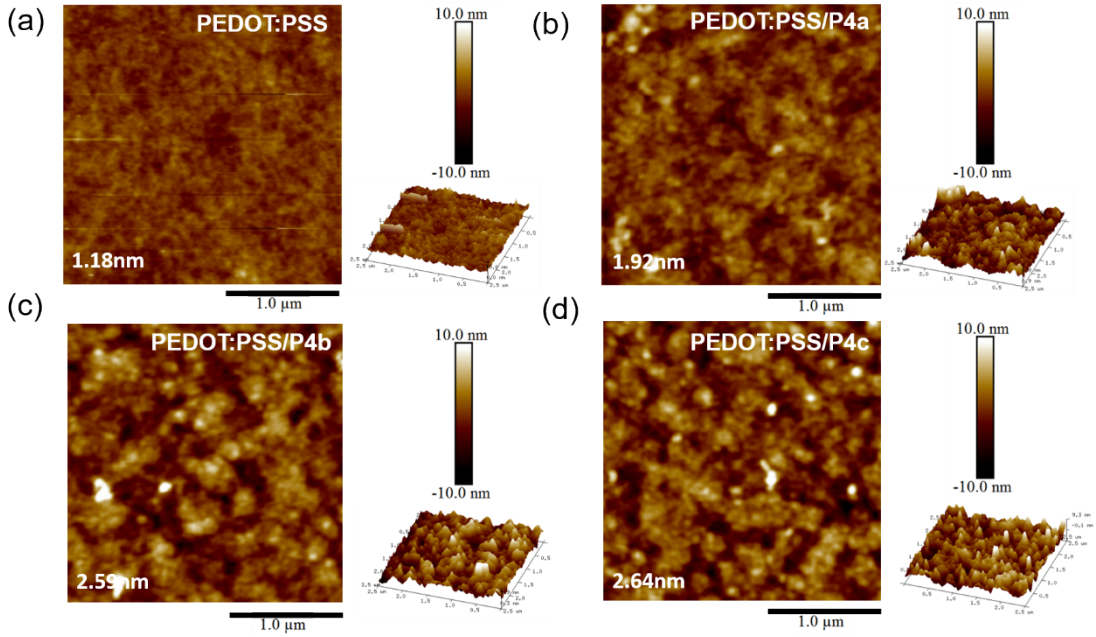
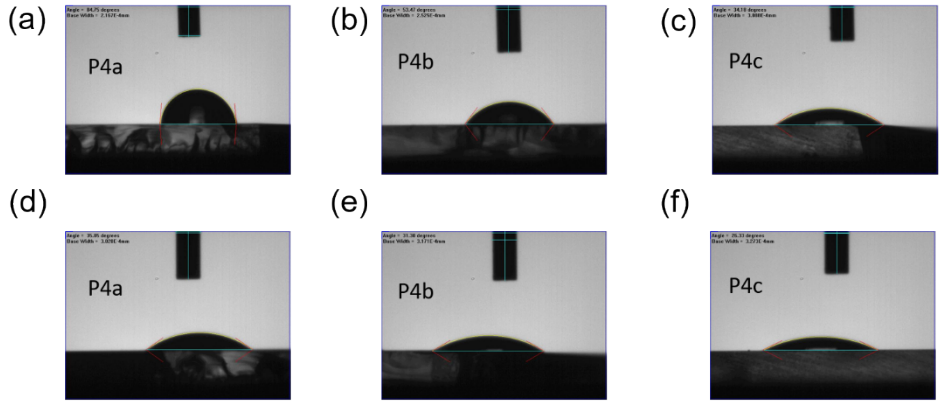


Figure S8. 2D and 3D AFM topographies of the films of (a) PEDOT:PSS, (b) PEDOT:PSS/**P4a**, (c) PEDOT:PSS/**P4b**, and (c) PEDOT:PSS/**P4c**. The surface roughness of the films is labeled on the images.



	θ_{water}(°) [a]	$\theta_{glycerol}$(°) [a]	σ_s(mN/m) [b]
P4a	84.75	35.85	43.8
P4b	53.47	31.30	54.9
P4c	34.18	26.33	61.3

[a] Contact angle between the film and water or glycerol. [b] Surface energy fitted by Wu method.

Figure S9. Contact angle measurement of (a,d) **P4a**, (b,e) **P4b**, (c,f) and **P4c** films using (a-c) water and (d-f) glycerol. The respective contact angle values for **P4a**, **P4b**, and **P4c** are presented below and the surface energy was calculated using Wu model.

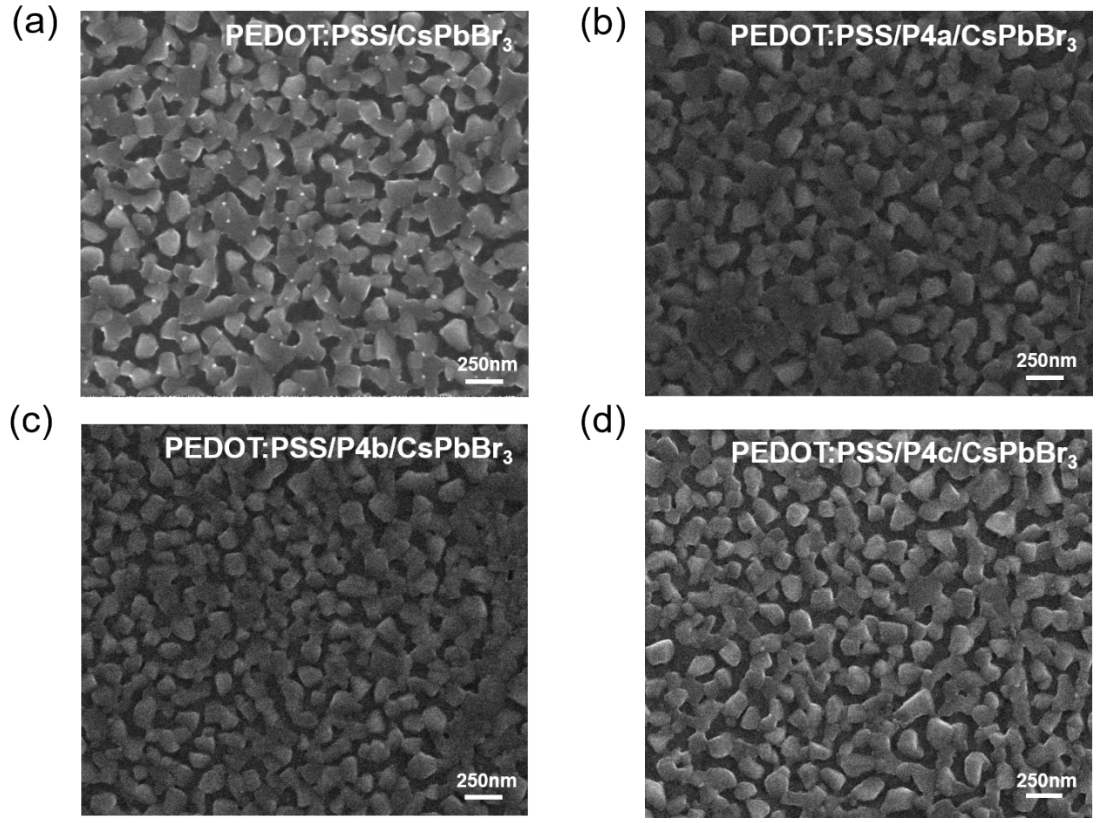


Figure S10. SEM images of $CsPbBr_3$ films grown on (a) PEDOT:PSS, (b) PEDOT:PSS/**P4a**, (c) PEDOT:PSS/**P4b**, and (c) PEDOT:PSS/**P4c**.

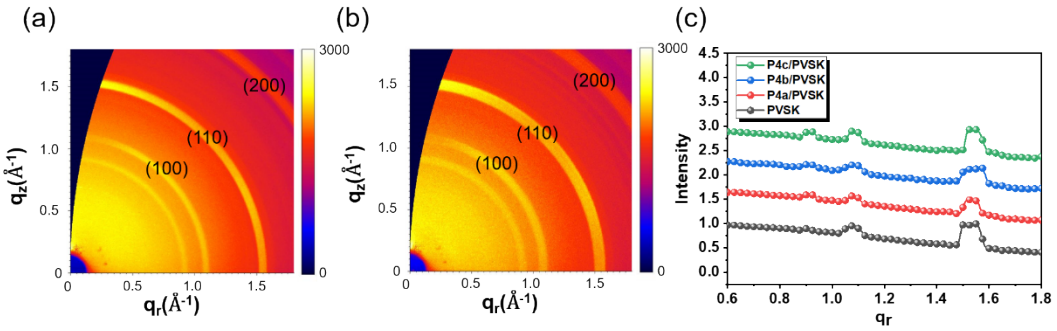


Figure S11. GIWAXS patterns of the samples of (a) PEDOT:PSS/**P4a**/$CsPbBr_3$ and (b) PEDOT:PSS/**P4b**/$CsPbBr_3$. (c) The corresponding 1-D incident characteristics of $CsPbBr_3$ films grown on the pristine and **P4a-c**-modified HTLs.

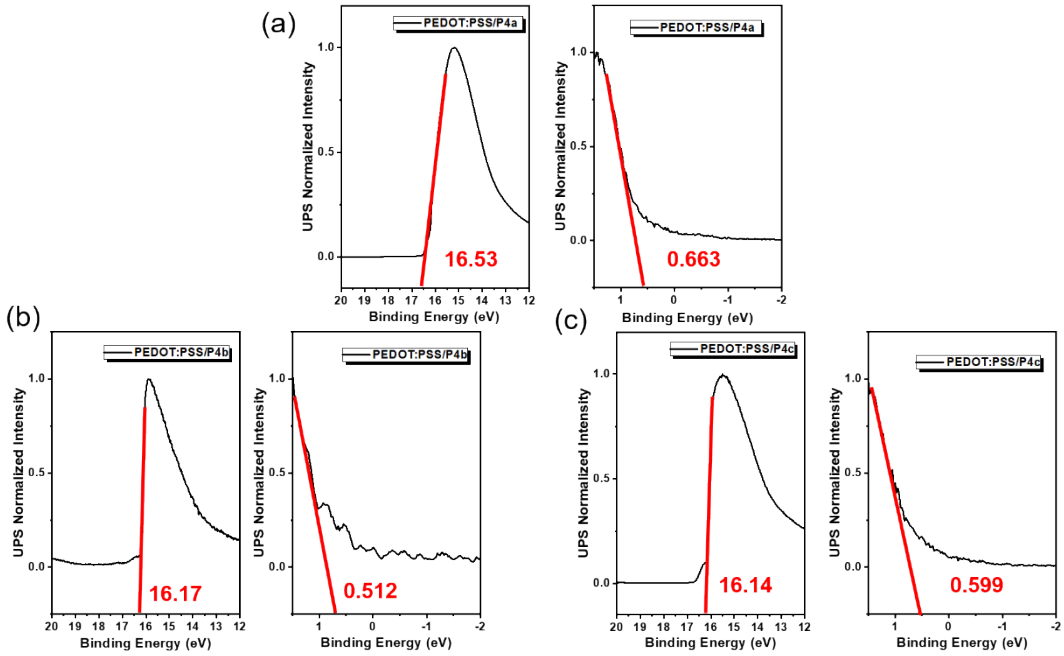


Figure S12. UPS measurements of the films of (a) PEDOT:PSS/**P4a,** (b) PEDOT:PSS/**P4b**, and (c) PEDOT:PSS/**P4c**.

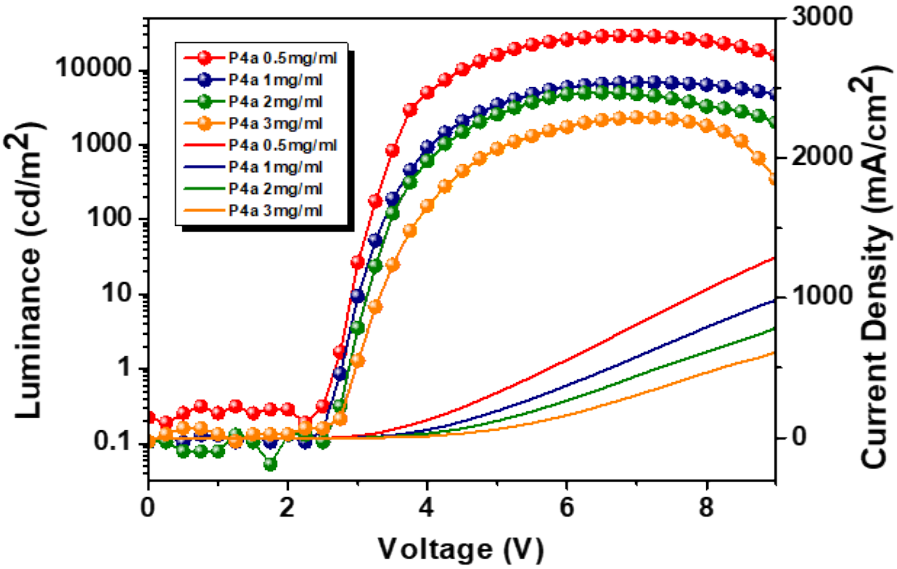


Figure S13. *J-V-L* curves of the PeLEDs made from **P4a** solutions with different concentration for optimization.

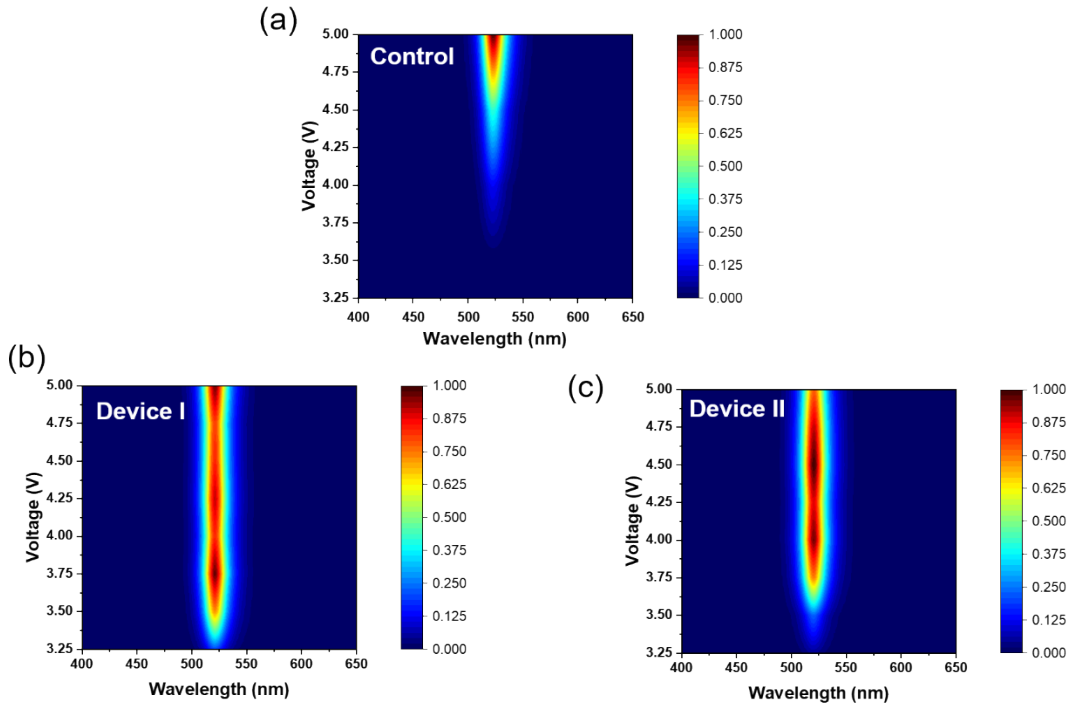


Figure S14. Contour plots of EL spectra of (a) control device, (b) Device I, and (c) Device II.