Supplementary Information

Solvent polishing engineering for high-performance quasi-two-dimensional perovskite blue light-emitting diodes

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Experimental Details :

Materials. PEABr (>99.5%), BABr (>99.5%), CsCl (>99.5%), $PbBr_2$ (>99.5%), CsBr (>99.5%), PEDOT:PSS (Clevios PVP AI 4083), and TPBi (>99%) were purchased from Xi'an Polymer Light Technology Corp. LiF (99.98%) was purchased from Sigma-Aldrich, Dimethyl sulfoxide (DMSO) (99.7+%, Extra Dry) and Anisole(99.9+%) were purchased from ACROS. Chlorobenzene (CB) (99.9+%) and Ethyl acetate (EA) were purchased from J&K. Toluene (99.9+%) was purchased from Aladdin. All the materials were used as received.

Preparation of Perovskite Precursor Solutions. 0.02 mmol CsBr, 0.18 mmol CsCl, 0.2 mmol $PbBr_2$, 0.12 mmol PEABr, 0.08 mmol BABr were dissolved in 1mL DMSO under continuous Stirring overnight at 50℃ and then filtered with a 0.22 μm polytetrafluoroethylene filter before spin coating (The ratio of bromo/chloride ratio and spacer cations have been optimized).

Device fabrication and characterization. The patterned ITO-coated glass substrates were washed with deionized water, ethanol, acetone, isopropanol and ethanol for 15 min, then dried with a nitrogen blow gun. The dried substrates were treated under oxygen plasma etching for 15 min. The PEDOT:PSS layers were spin-coated onto the substrates at 5000 rpm for 60 s and annealed at 150 °C for 20 min in air. After cooling, the samples were transferred into a nitrogen-filled glove box (O_2 <0.1 ppm, H_2O <0.1 ppm). The perovskite films were deposited in glovebox by spin-coating the precursor solution at 3000 rpm for 120 s and followed by annealing at 60℃ for 5 min. After cooling down, various solvents were dropped on the perovskite film by spin-coating at 5000rpm for 30s and annealed at 60 °C for 3 min. Finally, all samples were transferred to a thermal evaporator. TPBi (40 nm), LiF (1 nm), and Al (100 nm) were deposited under a high vacuum of ~ 10^{-5} torr. The active area of the devices was 8 mm^2. All devices were tested in atmospheric environments.

Perovskite film and device characterization. UV-visible (UV-vis) absorption spectra of the samples were recorded on a SHIMADZU UV-1800 spectrophotometer. The steady-state photoluminescence spectra and time-resolved PL (TRPL) were measured using a FLS 980 spectrofluorometer (Edinburgh Instruments Ltd). The excitation source is 365 nm from a Xe lamp for PL and the time-resolved luminescence decay were measured using time-correlated single-photon counting with a 375 nm laser. X-ray diffraction (XRD) patterns of the prepared samples were recorded on Bruker D8 advance X-ray diffractometer with graphite monochromatized Cu Kα (λ=1.5405 Å) radiation with a step of 0.02° at a scanning speed of 4° min^{-1} in 2θ ranging from 10° to 80°. Scanning electron microscopy (SEM) images were taken using a FEI Nova NanoSEM200 microscope. UPS was carried out using a Kratos AXIS ULTRA DALD UPS system. AFM and KPFM were conducted on Dimension iCon(Bruker), the work function of the needle tip is 5.1eV. Contact Angle was texted by DSA30. The current density-luminance-voltage (*J*-V-L), EL spectra, EQE and operating lifetime of the PeLED were recorded simultaneously by using a commercialized system (Shenzhen Spectrum Research Connected Technology Co., Ltd) equipped with Keithley 2400, a fiber integrating sphere and Ocean Optics QE65 Pro spectrometer.

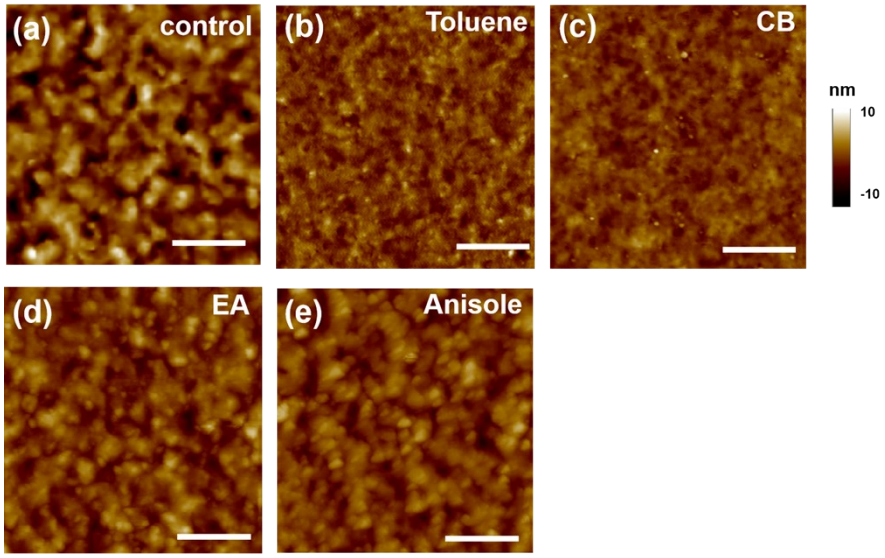

Figure S1. 2D AFM images of perovskite films before and after solvent-polishing treatment. (a) control, (b) toluene, (c) CB, (d) EA, (e) anisole.

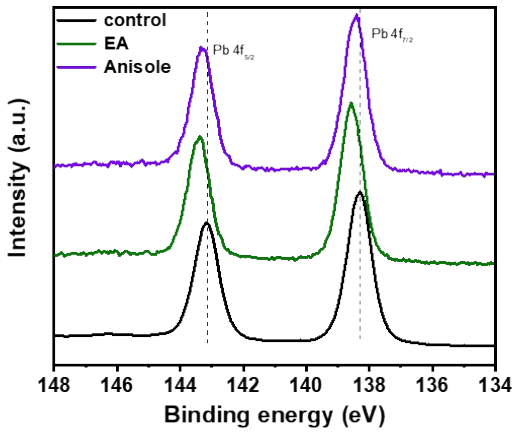

Figure S2. XPS of Pb 4f peaks with and without solvent-polishing treatment.

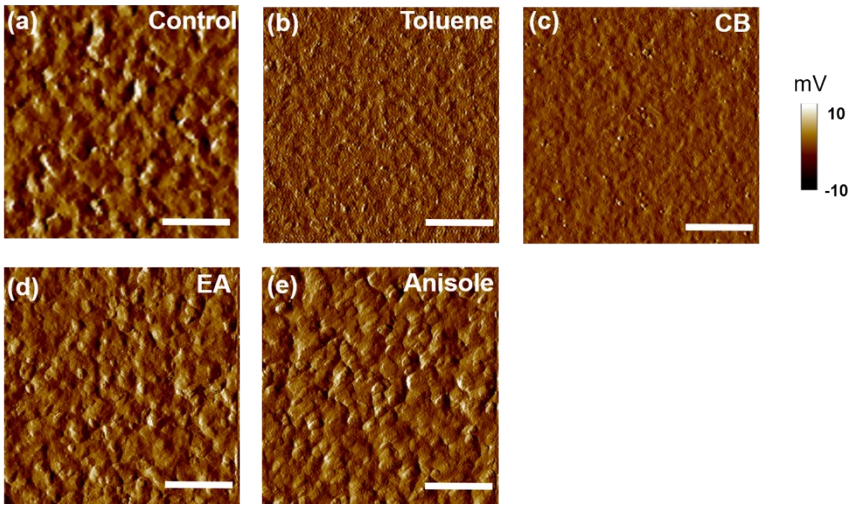

Figure S3. AFM images of Amplitude error before and after solvent-polishing treatment. (a) control, (b) toluene, (c) CB, (d) EA, (e) anisole.

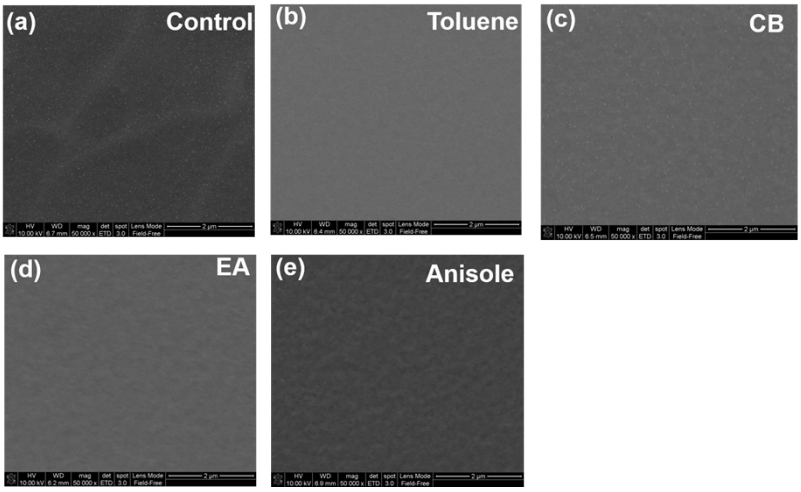

Figure S4. SEM images of perovskites before and after solvent-polishing treatment. (a) control, (b) EA, (c) toluene, (d) CB, (d) anisole.

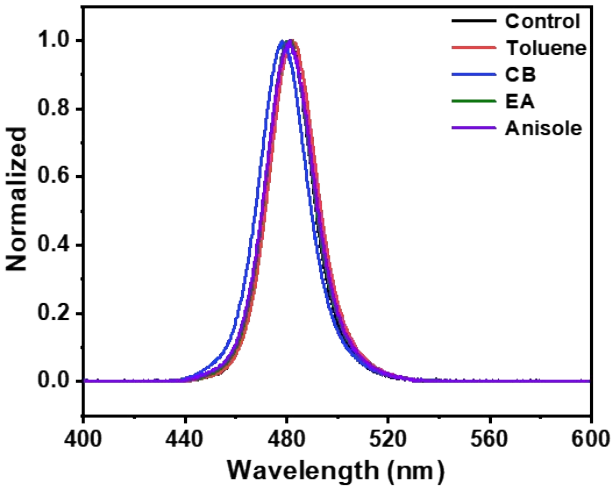

Figure S5. Normalized PL spectra of perovskite films with solvent-polishing treatment.

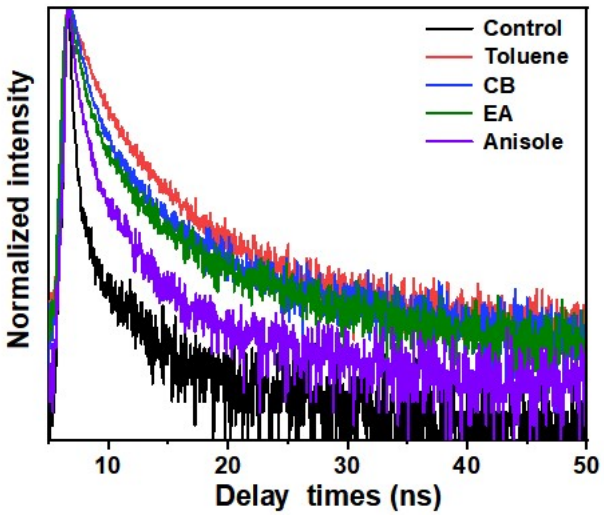

Figure S6. TRPL decay curves.

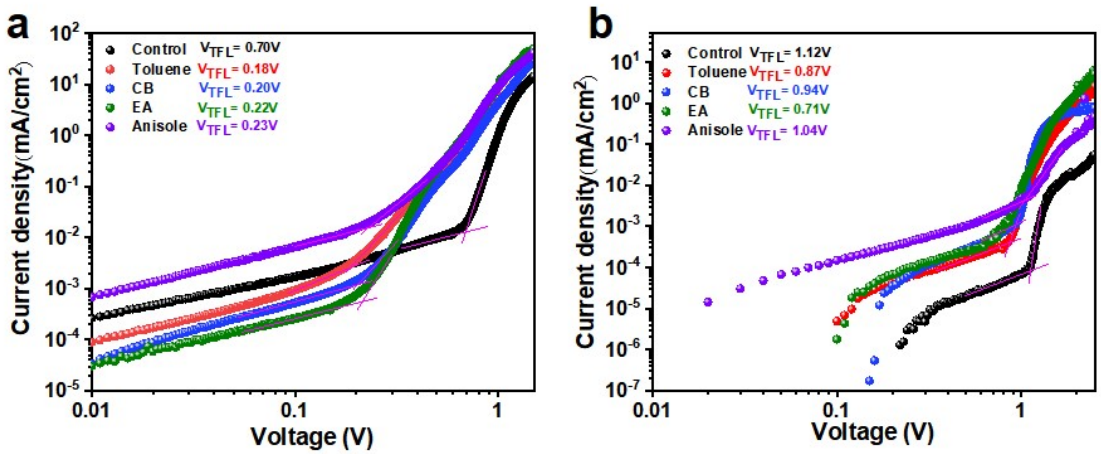

Figure S7. Current density-voltage characteristics of (a) hole-only devices based on ITO/PEDOT:PSS/pero/solvents/MoO_3/Ag structure, (b) electron-only devices based on ITO/TPBi/pero/solvents/TPBi/LiF/Al structure.

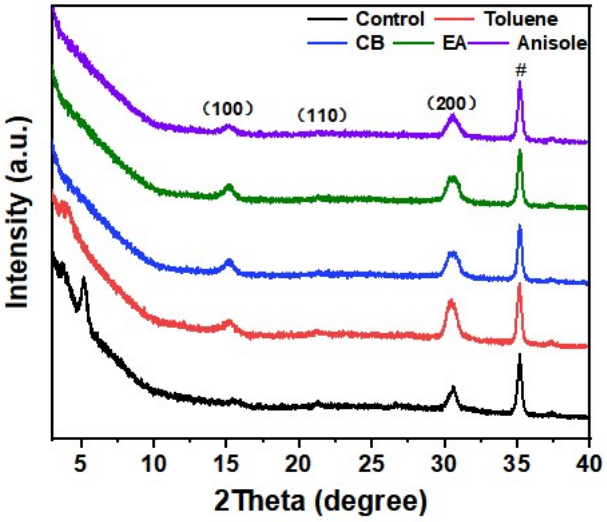

Figure S8. XRD patterns (# represent to ITO).

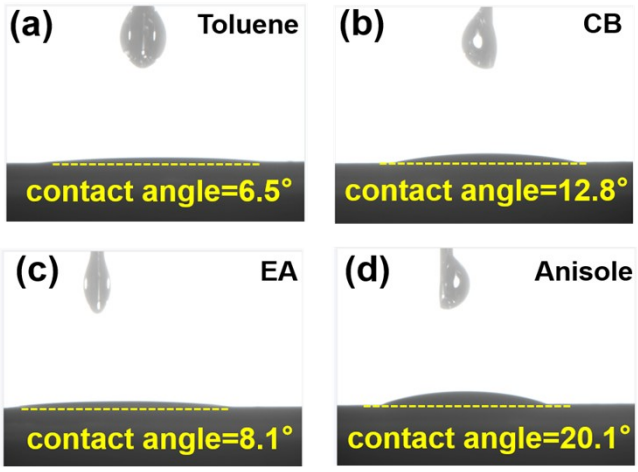

Figure S9. Contact angle of the various solvents droplets on the perovskite film.

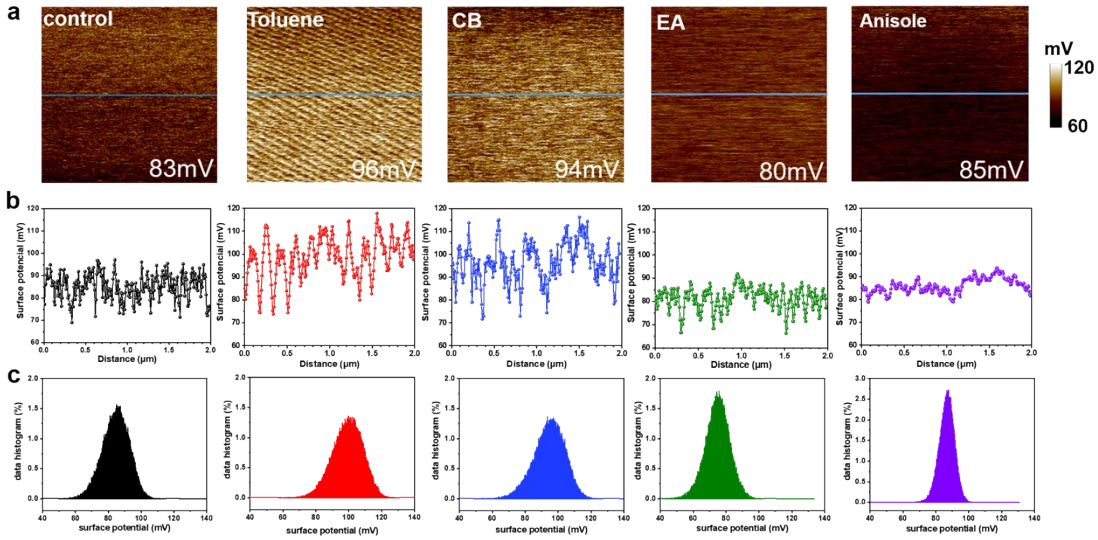

Figure S10. (a) Surface potential mapping, (b) surface potential line profiles of the perovskite films, and (c) Statistical distribution of surface potential.

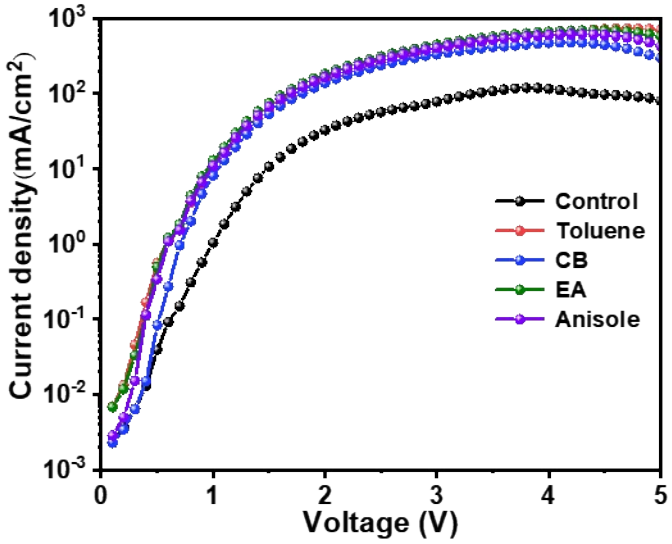

Figure S11. Current density-voltage curves of the hole-only devices based on following structure: ITO/PEDOT: PSS (30 nm)/Pero (20 nm) /MoO_3 (10 nm)/Ag.

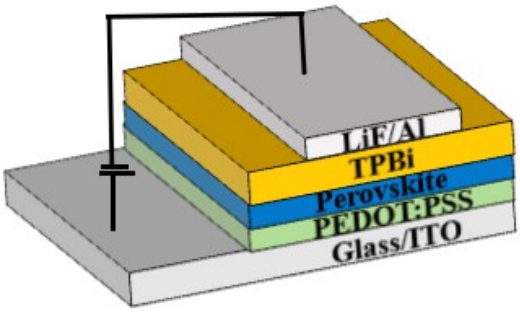

Figure S12. The architecture of the PeLED.

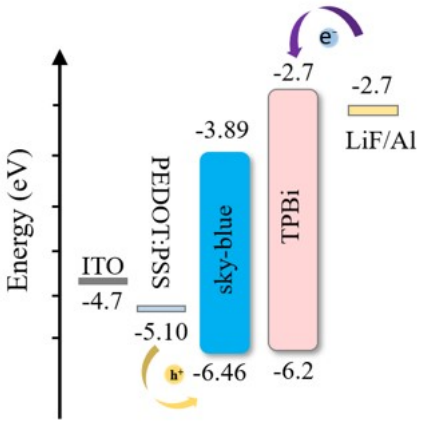

Figure S13. Energy level diagram of the devices.

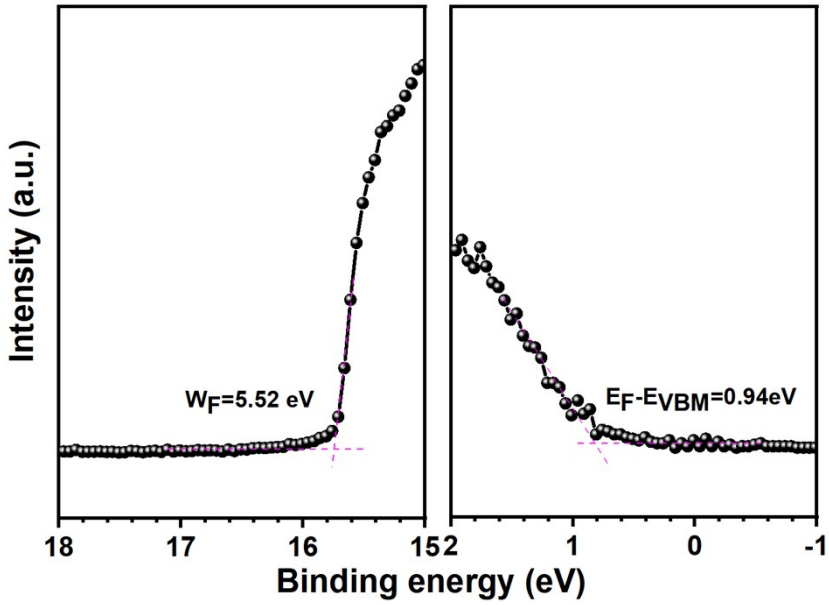

Figure S14. The VB-edge region (left), secondary-electron cutoff region (right) of the unpolished perovskite films.

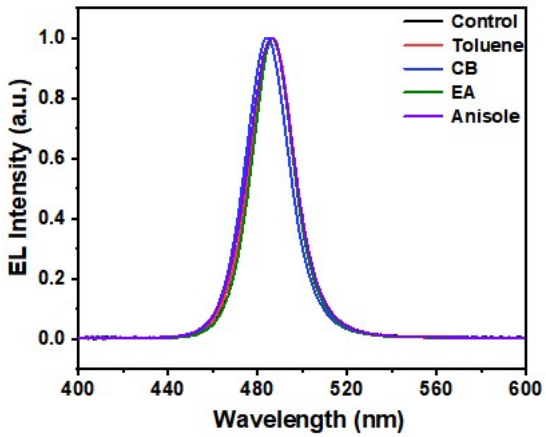

Figure S15. Normalized EL spectra.

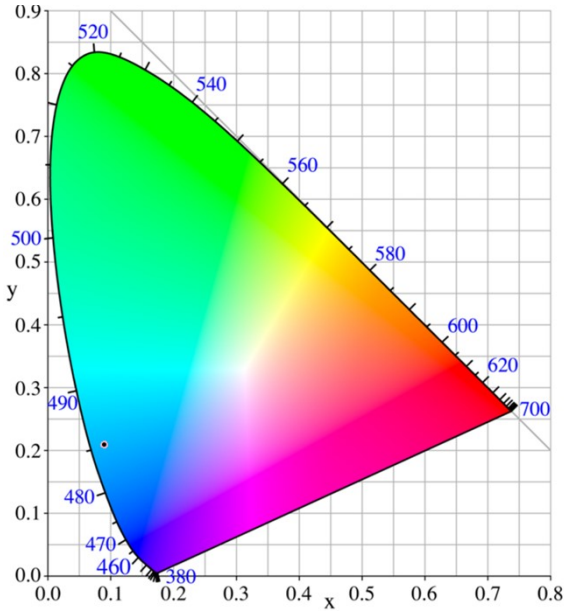

Figure S16. CIE coordinates of PeLEDs based on toluene-polished perovskite film.

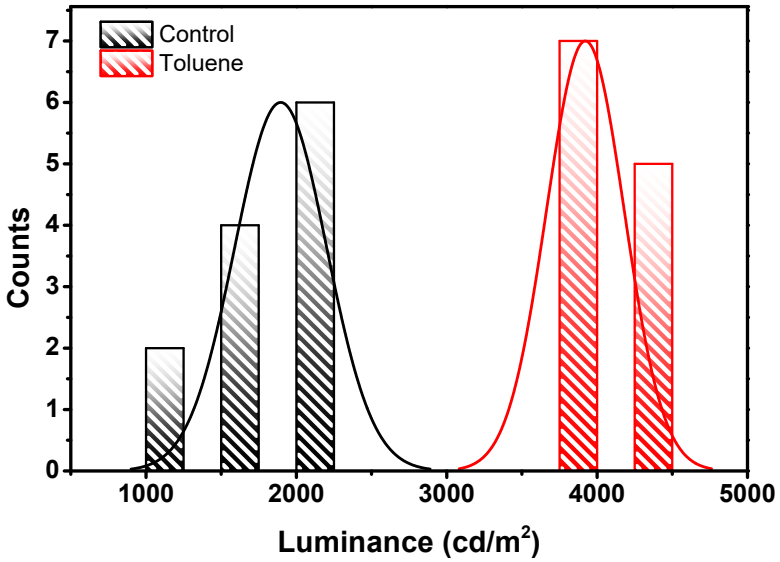

Figure S17. Luminance histogram of the devices.

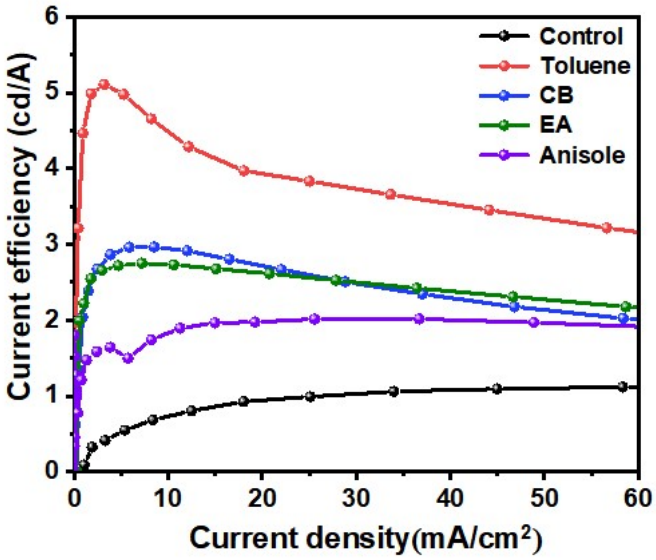

Figure S18. CE-J curves.

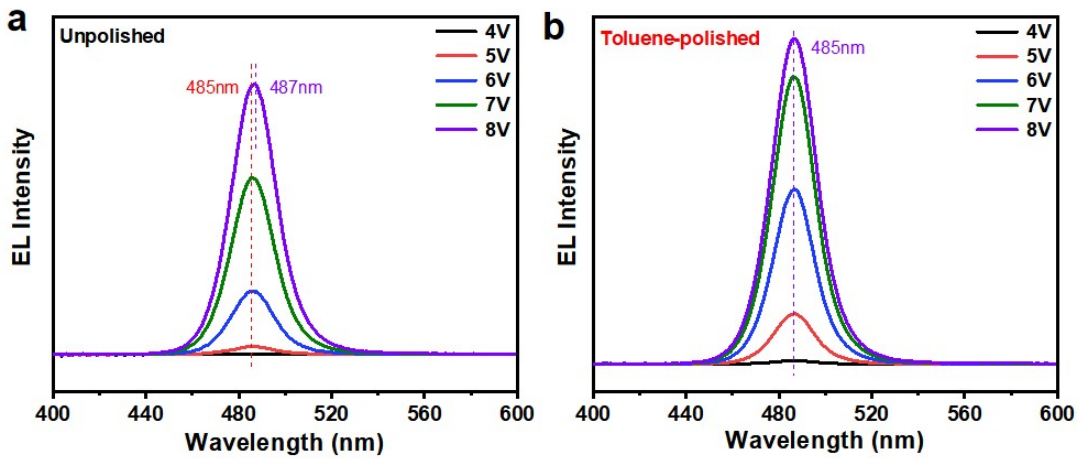

Figure S19. EL spectra of PeLEDs (g) without and (h) with toluene-polishing treatment.

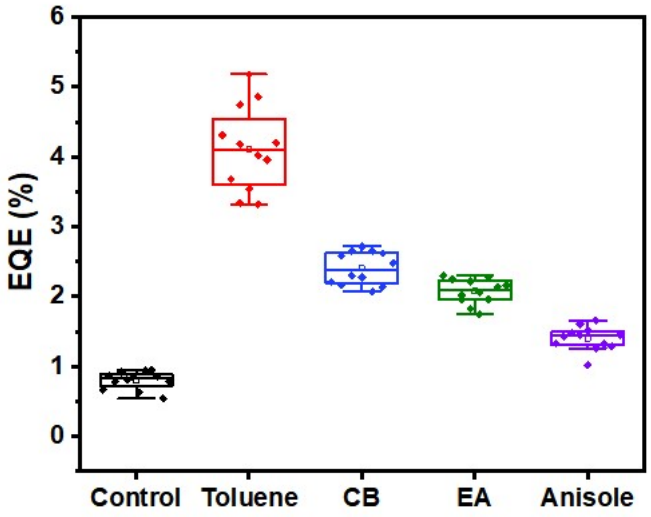

Figure S20. EQE histogram of the devices.

Table S1. The physiochemical properties of solvents.

Solvent	Density (g/mL)	Boiling point (°C)	Dynamic viscosity (mpa·s)	Dipole moment (D)	Polarity (P)
CB	1.11	132	0.80	1.69	2.7
Toluene	0.87	111	0.59	0.36	2.4
EA	0.90	78	0.46	1.78	4.3
Anisole	1.00	154	1.20	2.30	
DMSO	1.10	189	2.20	3.96	7.2

Table S2. PL lifetimes of the perovskite films with and without polishing treatment by various solvents.

Samples No.	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)	τ_3 (ns)	A_3 (%)	τ_{ave} (ns)	χ^2
Control	0.50	42	2.21	39	9.98	19	7.09	1.156
EA	0.80	34	2.87	37	12.70	29	9.98	1.104
Toluene	0.90	34	3.40	39	16.31	27	12.68	1.199
Anisole	0.48	15	2.26	40	10.50	45	9.08	1.071
CB	0.48	29	2.20	38	12.36	33	10.39	1.140

All TRPL decay curves are fitted according to the three-exponential decay formula, the average life of the sample can be calculated according to the following formula:

$$\tau_{ave} = \sum A_i\tau_i^2/\sum A_i\tau i$$

A_i (i = 1, 2 and 3) and τ_i(i = 1 2 and 3) are the fractions and lifetimes of the three decay components, respectively.

Table S3. The key parameters of blue PeLEDs with and without polishing treatment by various solvents.

Samples No	V_{on} (V)	L_{max} (cd/m^2)	EQE_{max} (%)	CE_{max} (cd/A)	EL (nm)
Control	4.2V	2309	0.95	1.11	485 nm
EA	3.8V	2327	2.25	2.75	485 nm
Toluene	3.2V	3737	5.18	5.11	485 nm
Anisole	3.6V	847	1.65	2.02	487 nm
CB	3.8V	2120	2.72	2.96	483 nm

Table S4. Summary of key EL parameters for sky-blue PeLEDs.

Composition	Type	L_{max} (cd m^{-2})	EQE (%)	EL (nm)	FWHM (nm)	Year	Ref
IPA-$PEA_2(MACs)_{1.5}Pb_{2.5}Br_{8.5}$	Q-2D	2480	1.5	490	20	2018	[1]
$(PBA)Br_2(Cs_{0.7}FA_{0.3}PbBr_3)$	Q-2D	700	9.5	483	26	2019	[2]
$(Cs/Rb/FA/PEA/K)Pb(Cl/Br)_3$	Q-2D	4015	2.01	484	-	2020	[3]
$CsPbBr_xCl_{3-x}$	QDs	182	1.8	489	18	2020	[4]
PEABr:GABr:$CsPbBr_{3-x}Cl_x$	Q-2D	1003	8.2	492	18	2020	[5]
PEABr:P-F-PEABr:$CsPbBr_{3-x}Cl_x$	Q-2D	1390	12.8	486	20	2021	[6]
$PEA2(Cs_{1-x}EA_xPbBr_3)_2PbBr_4$	Q-2D	2191	12.1	488	-	2020	[7]
PEABr:P-F-PEABr:$CsPbBr_{3-x}Cl_x$	Q-2D	1775	9.2	490	-	2021	[8]
$PEA_{0.6}BA_{0.4}$:$CsPbBr_{2.1}Cl_{0.9}$	Q-2D	3737	5.18	485	18		This wok

1. J. Xing, Y. Zhao, M. Askerka, L. N. Quan, X. Gong, W. Zhao, J. Zhao, H. Tan, G. Long, L. Gao, Z. Yang, O. Voznyy, J. Tang, Z. H. Lu, Q. Xiong and E. H. Sargent, *Nat. Commun.*, 2018, **9**, 3541.
2. Y. Liu, J. Cui, K. Du, H. Tian, Z. He, Q. Zhou, Z. Yang, Y. Deng, D. Chen, X. Zuo, Y. Ren, L. Wang, H. Zhu, B. Zhao, D. Di, J. Wang, R. H. Friend and Y. Jin, *Nat. Photon.*, 2019, **13**, 760-764.
3. F. Yuan, C. Ran, L. Zhang, H. Dong, B. Jiao, X. Hou, J. Li and Z. Wu, *ACS Energy Lett.*, 2020, **5**, 1062-1069.
4. F. Ye, H. Zhang, P. Wang, J. Cai, L. Wang, D. Liu and T. Wang, *Chem. Mater.*, 2020, **32**, 3211-3218.
5. F. Zhang, B. Cai, J. Song, B. Han, B. Zhang and H. Zeng, *Adv. Funct. Mater.*, 2020, **30**.
6. Y. Shen, H. Y. Wu, Y. Q. Li, K. C. Shen, X. Gao, F. Song and J. X. Tang, *Adv. Funct. Mater.*, 2021, **31**.
7. Z. Chu, Y. Zhao, F. Ma, C. X. Zhang, H. Deng, F. Gao, Q. Ye, J. Meng, Z. Yin, X. Zhang and J. You, *Nat. Commun.*, 2020, **11**, 4165.
8. Y. Shen, J. K. Wang, Y. Q. Li, K. C. Shen, Z. H. Su, L. Chen, M. L. Guo, X. Y. Cai, F. M. Xie, X. Y. Qian, X. Gao, I. S. Zhidkov and J. X. Tang, *Adv Sci*, 2021, **8**, e2102213.