

Supplementary Information

Bright and stable perovskite light-emitting diodes in the near-infrared range

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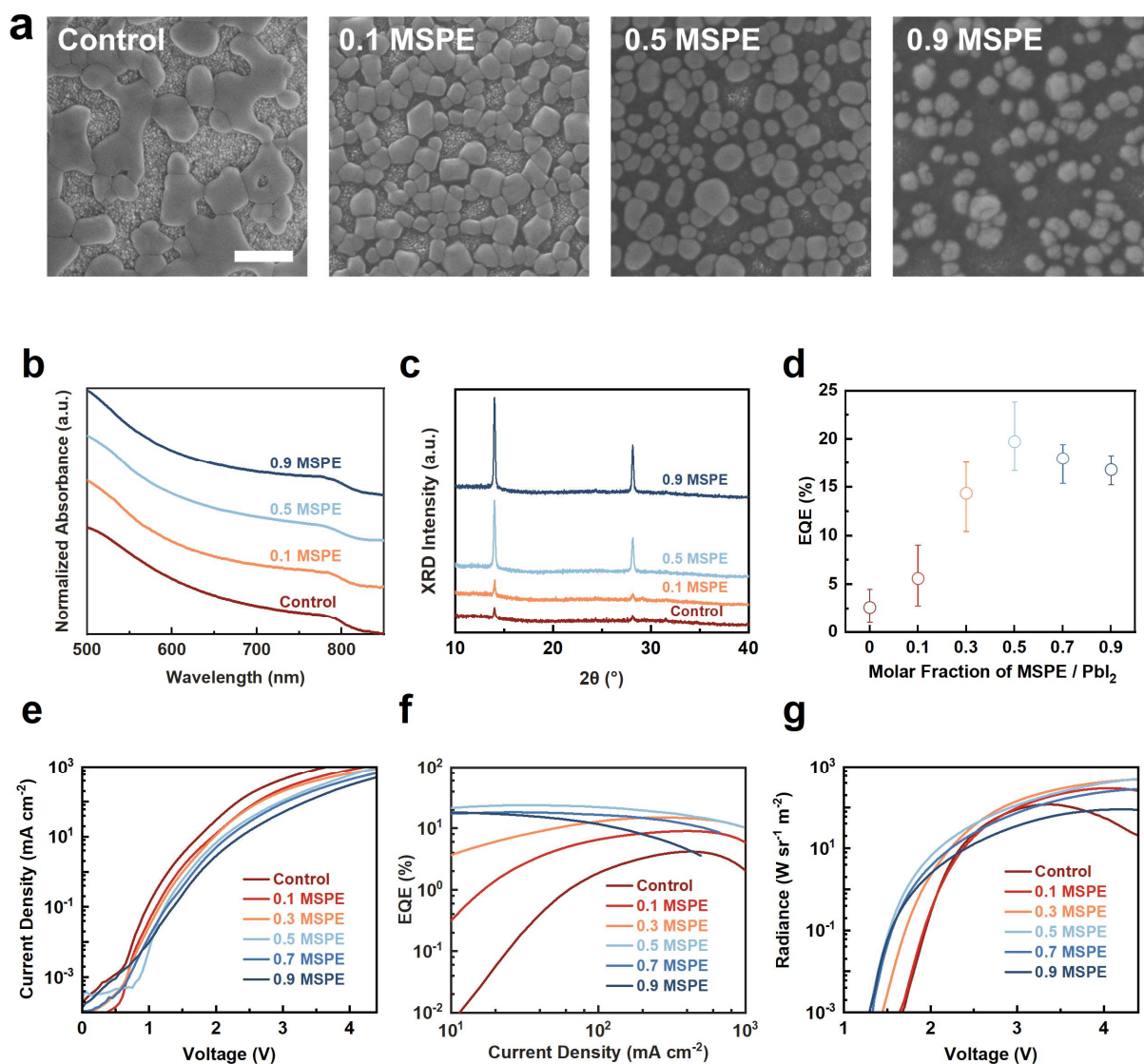
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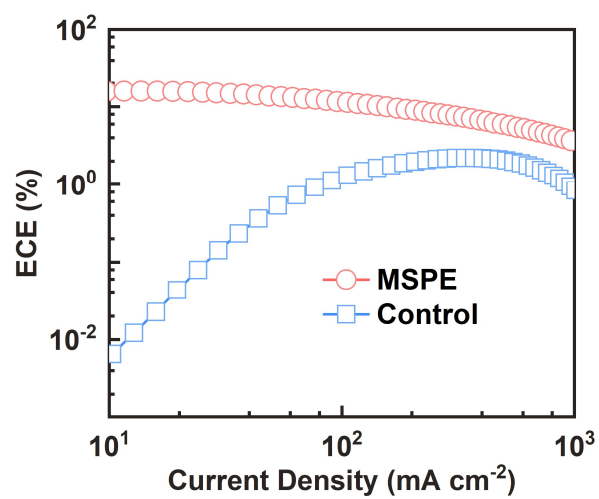
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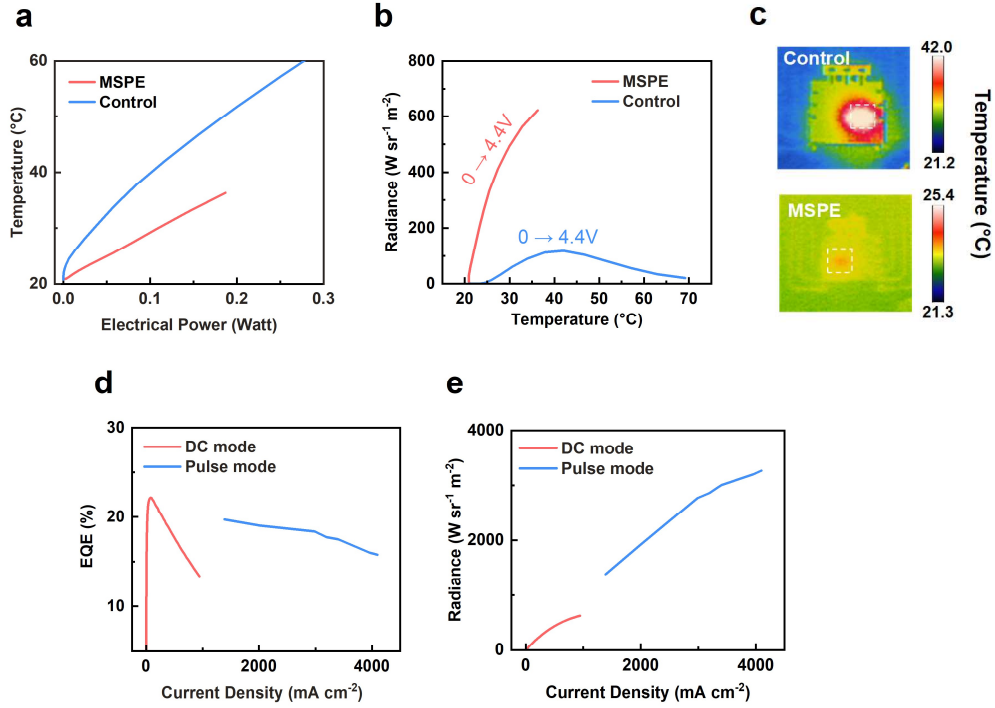
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Supplementary Fig. 1. Characteristics of perovskite films and LEDs with different molar ratio of MSPE. **a-c**, Characteristics of perovskite films with different molar fractions of MSPE. **(a)**, SEM images. The scale bar represents 200 nm. **(b)**, UV-Vis spectra. **(c)**, XRD patterns. **d-g**, The device performance using perovskite layers prepared by FAI, PbI_2 and MSPE precursors at an optimized molar ratio of 2.7: 1: x ($x = 0, 0.1, 0.3, 0.5, 0.7, 0.9$). **(d)**, Peak EQEs distribution. **(e)**, Current density versus voltage curves. **(f)**, EQE versus current density curves. **(g)**, Radiance versus voltage curves.

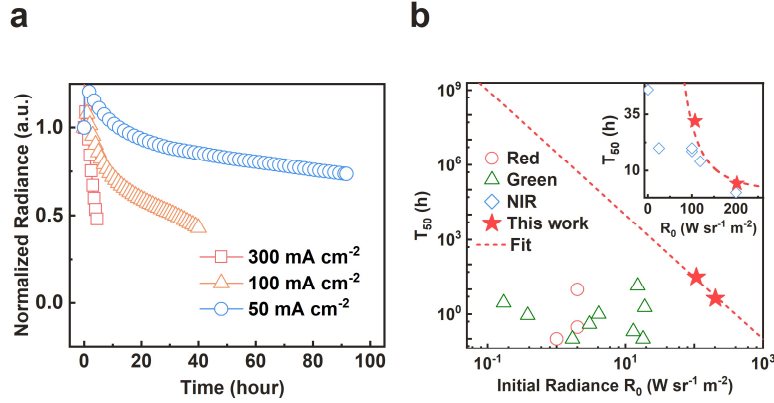


Supplementary Fig. 2. Energy conversion efficiency. Energy conversion efficiency (ECE) versus current density curves of the MSPE and control LEDs.

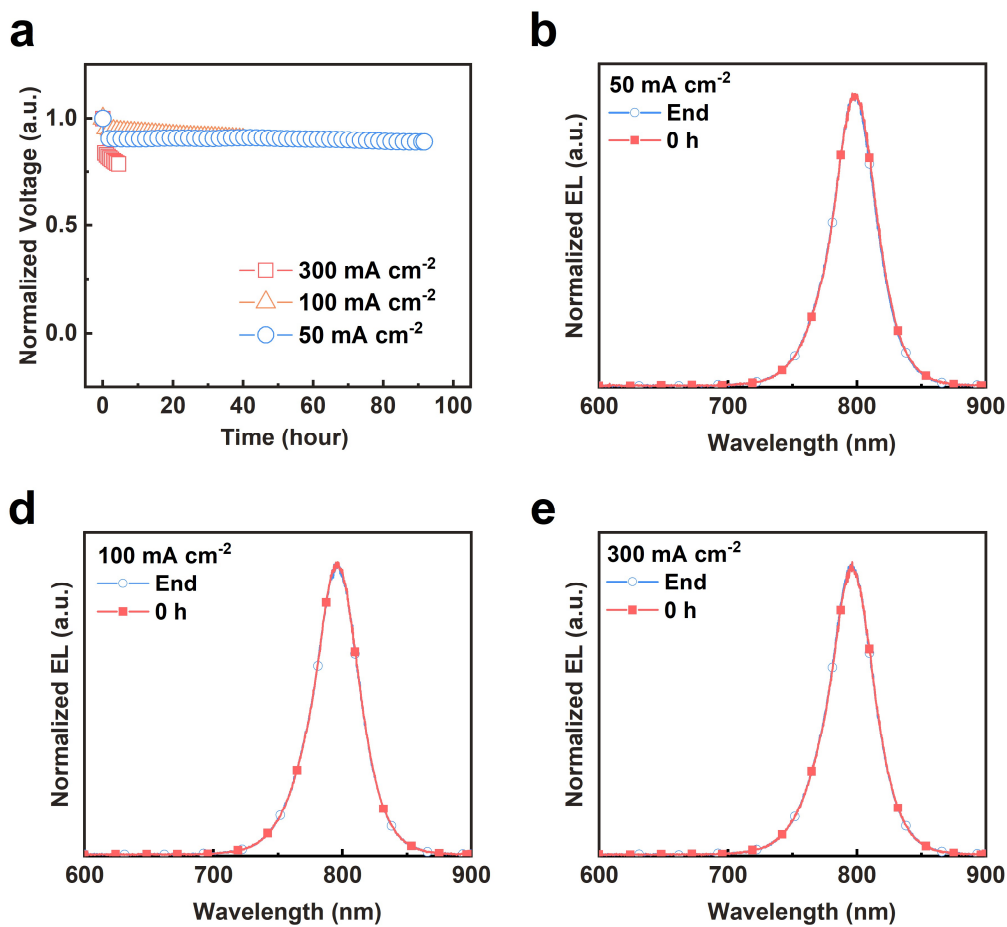


Supplementary Fig. 3. Device temperature and pulse mode operation. a-b, the real-time temperature of a MSPE LED and the control one at a voltage scan from zero to 4.4 V at a rate of 0.05 V s^{-1} . c, infrared images of the control device at $118 \text{ W sr}^{-1} \text{m}^{-2}$ and MSPE device at $335 \text{ W sr}^{-1} \text{m}^{-2}$. d-e, EQE versus current density and radiance versus current density characteristics of MSPE LEDs under direct current (DC) and pulse mode. Short voltage pulses with $200 \mu\text{s}$ width were applied to devices at a repetition rate of 50 Hz, corresponding to duty cycles of 1%.

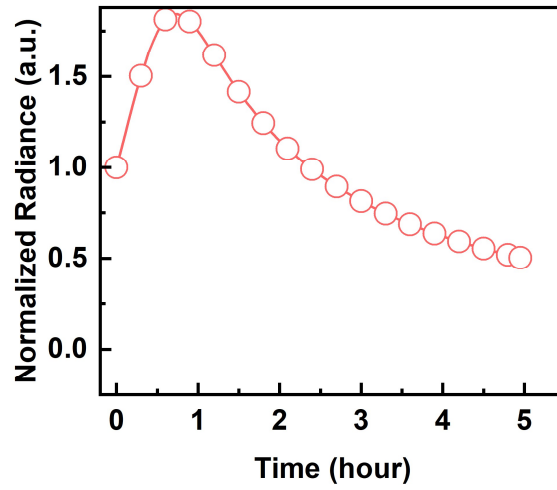
The temperature reaches $42 \text{ }^{\circ}\text{C}$ at the peak radiance of $118 \text{ W sr}^{-1} \text{m}^{-2}$ in the control devices. As the voltage is increased further the brightness decreases, indicating that thermal degradation occurs. We note that the temperature of the perovskite layer is likely to be much higher than the value we measured due to the low thermal conductivity of the glass substrate. MSPE LEDs show a smaller increase in device temperature during the voltage scan. Higher ECE of MSPE devices implies that less heating is produced during device operation (Supplementary Fig. 2).



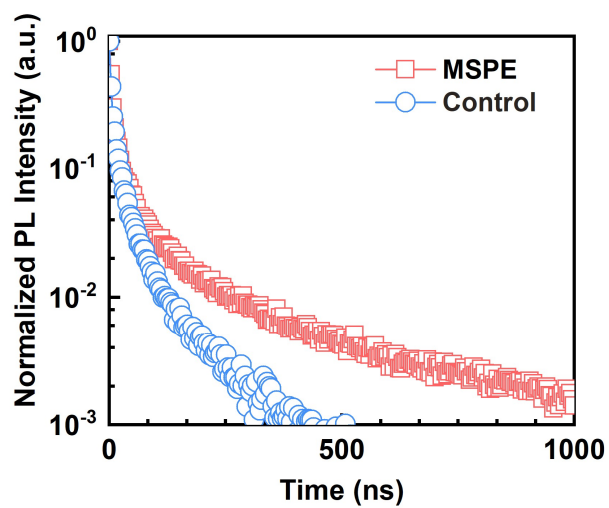
Supplementary Fig. 4. Lifetime measurement of MSPE LEDs. **a**, Measured lifetimes of MSPE LEDs at constant current densities of 50 mA cm⁻² ($R_0 = 45 \text{ W sr}^{-1} \text{ m}^{-2}$; $R_{\text{max}} = 55 \text{ W sr}^{-1} \text{ m}^{-2}$; $T_{80} = 62 \text{ h}$), 100 mA cm⁻² ($R_0 = 107 \text{ W sr}^{-1} \text{ m}^{-2}$; $R_{\text{max}} = 117 \text{ W sr}^{-1} \text{ m}^{-2}$; $T_{50} = 32 \text{ h}$) and 300 mA cm⁻² ($R_0 = 202 \text{ W sr}^{-1} \text{ m}^{-2}$; $R_{\text{max}} = 253 \text{ W sr}^{-1} \text{ m}^{-2}$; $T_{50} = 4 \text{ h}$), where R_0 is the radiance at time = 0 h, R_{max} is the maximum radiance during stability measurement and T_x is defined as the time to reaches x % of R_0 . **b**, Reported T_{50} and the corresponding initial radiance R_0 of the reported perovskite LEDs having EQEs exceeding 20%, according to the data provided in Supplementary Table 1-2. An empirical formula $T_{50}(R_1) = T_{50}(R_0) \times (R_0/R_1)^n$ was used to predict the T_{50} at lower radiance, where R is the initial radiance and the acceleration factor n is fitted to be ~ 2.5 .



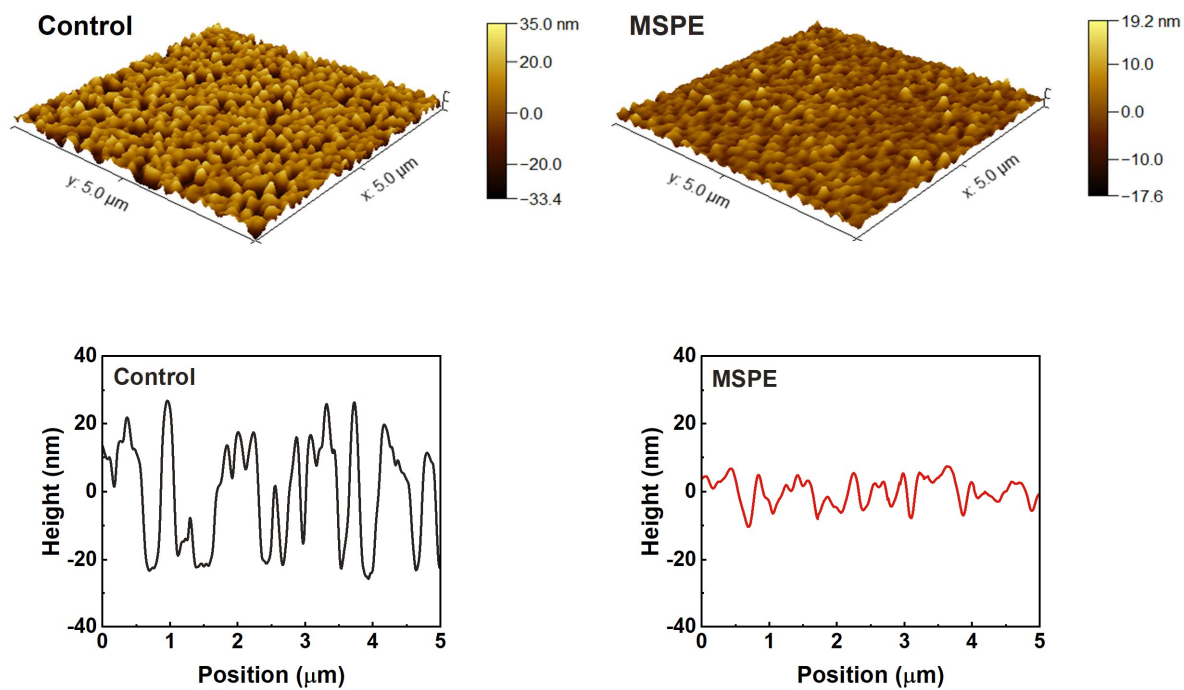
Supplementary Fig. 5. Voltages and EL during lifetime measurement. **a**, Voltages during operational lifetime measurement. **b-d**, EL spectra at the beginning and the end of the operational lifetime measurement.



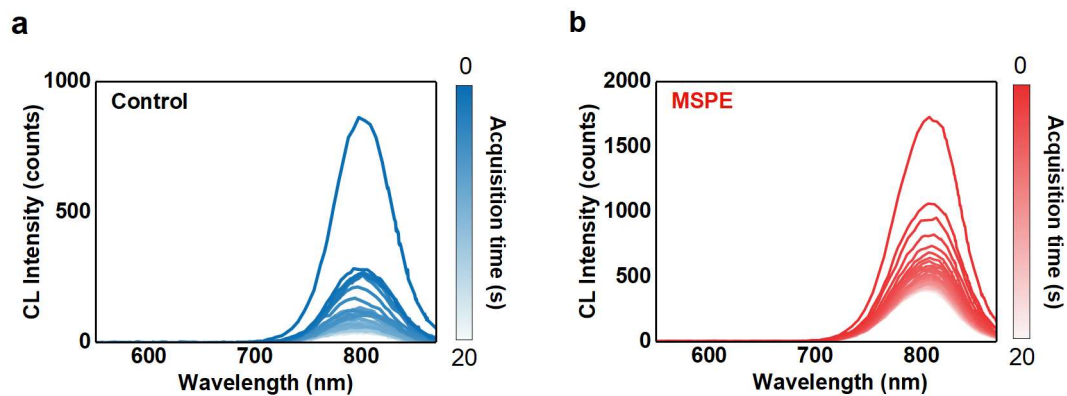
Supplementary Fig. 6. Lifetime measurement of control LEDs. Operational stability measurement of control LEDs at $R_0 = 10 \text{ W sr}^{-1} \text{ m}^{-2}$ at a constant current density of 100 mA cm^{-2} .



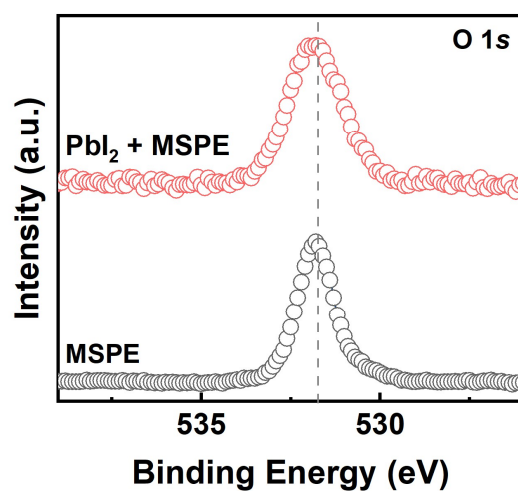
Supplementary Fig. 7. Time-resolved PL measurement. Time-resolved PL spectroscopy measurements of perovskite films. Samples were excited by 560 nm laser at a fluence of $6 \mu\text{J cm}^{-2}$.



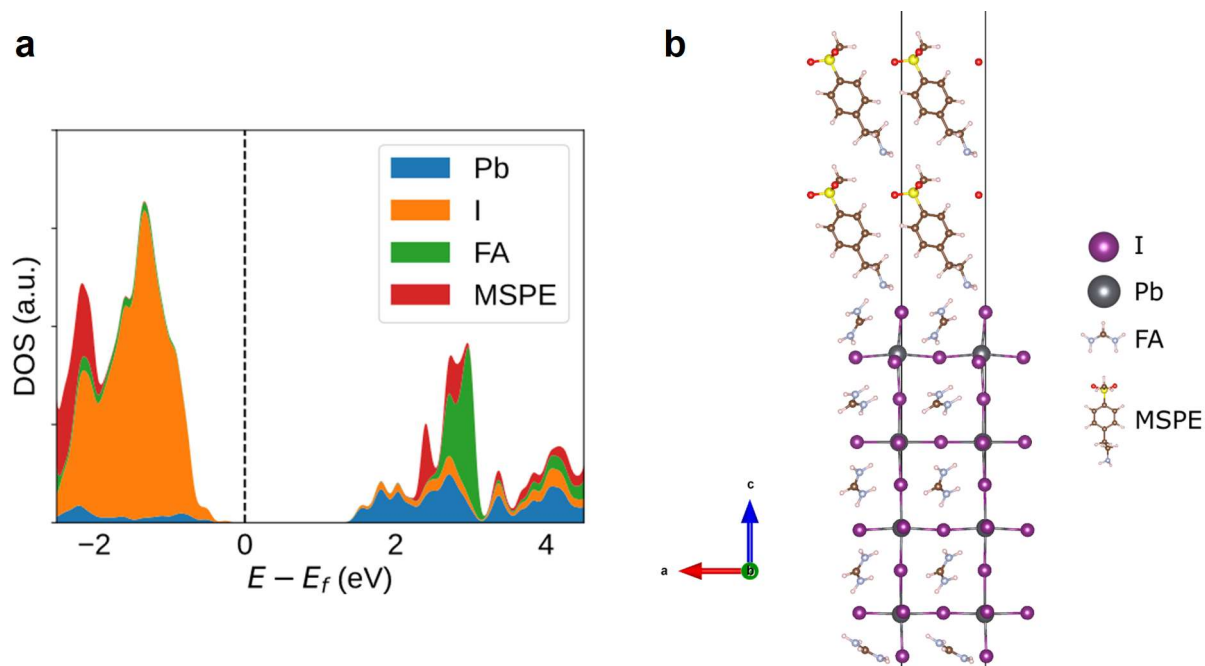
Supplementary Fig. 8. AFM measurement. AFM images and line analysis of height distribution of perovskite films. MSPE films exhibit a smaller roughness than the control one.



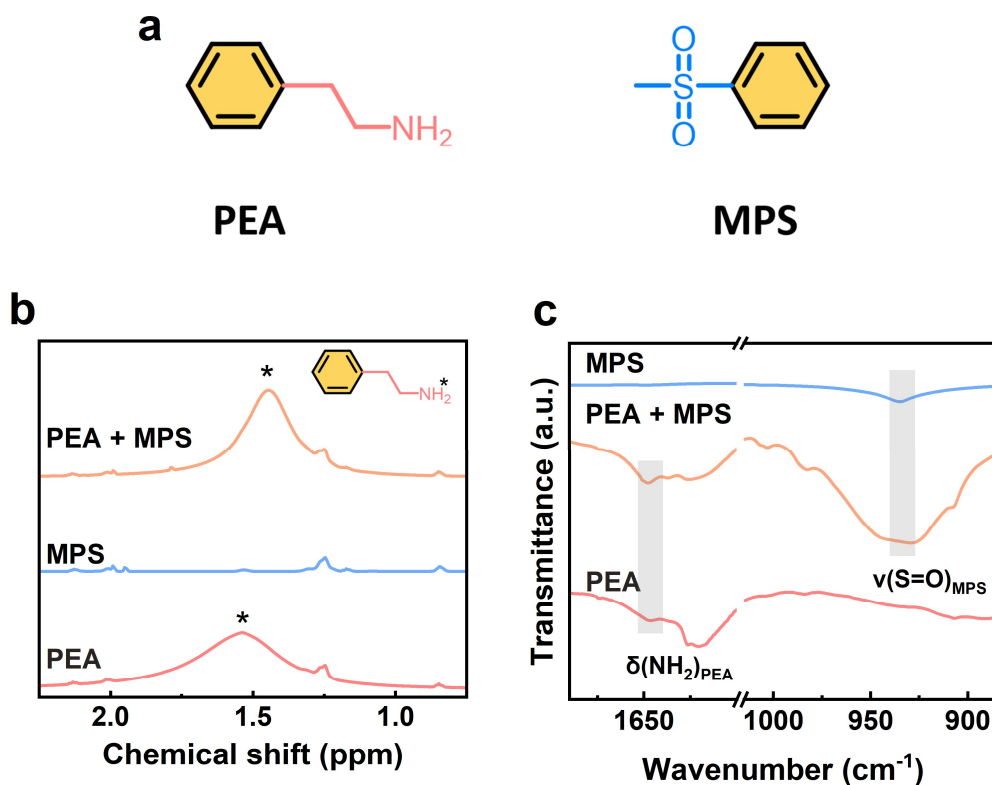
Supplementary Fig. 9. CL emission evolution. Evolution (acquisition time from 0 s to 20 s) of CL emission profile of perovskite films. CL intensity degrades slower with MSPE passivation.



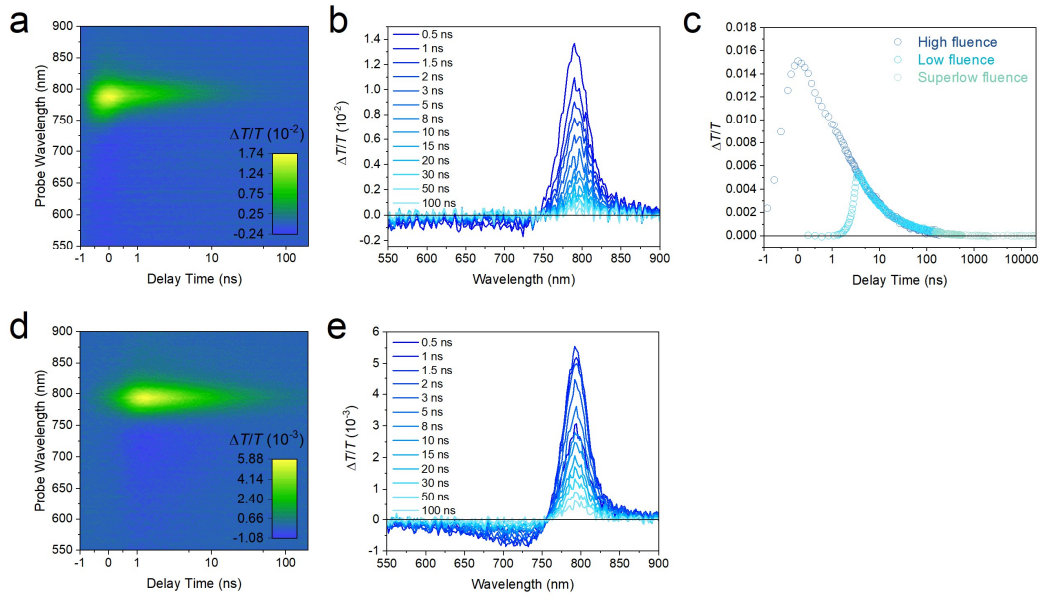
Supplementary Fig. 10. XPS spectra of O 1s of MSPE.



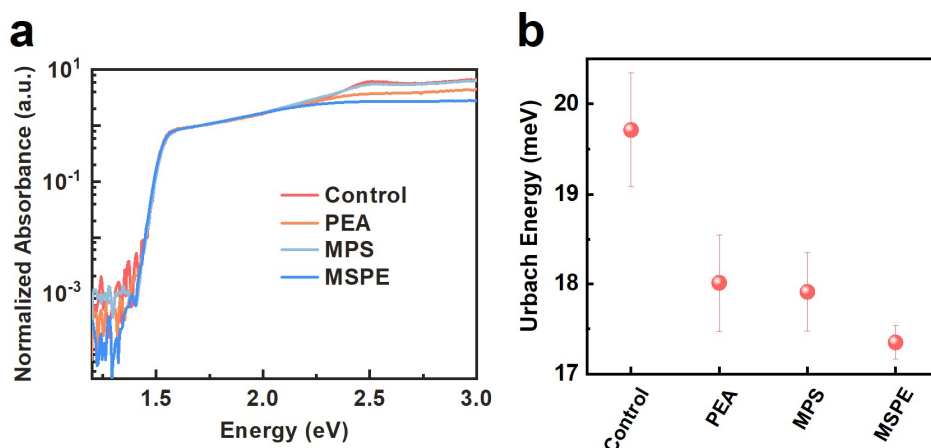
Supplementary Fig. 11. DFT calculations. DFT calculations reveal trap-free bandgap of perovskites with MSPE passivation, as shown in (a) the projected density of states (PDOS) plot. DFT further unveils that the amino and sulfonyl groups of MSPE can bind to the FAI surface plane of perovskites with binding affinities being 2.3 eV and 2.0 eV. We show a possible configuration in (b), where MSPE binds to the perovskite surface through hydrogen bonding between amino group and FAI at an angle of approximately 64 degrees. An additional layer of MSPE can bind to MSPE at a similar angle through the hydrogen bonding between the amino and sulfonyl groups.



Supplementary Fig. 12. NMR and FTIR measurements of PEA and MPS. **a**, Organic compounds with different functional groups. Left: phenethylamine (PEA). Right: methyl phenyl sulfone (MPS). **b**, ¹H NMR spectra of PEA, MPS and PEA + MPS samples. The N-H protons of PEA (indicated with *) shift to lower frequency with the addition of MPS. **c**, FTIR spectra of PEA, MPS and PEA + MPS samples. Both the sulfonyl group of MPS and the amino group of PEA are broadened in the mixture sample.

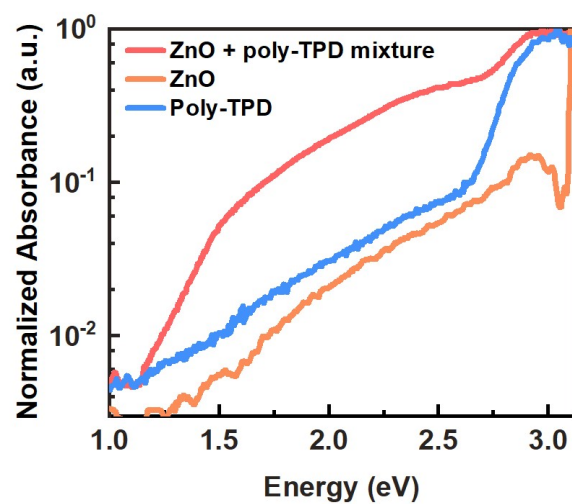


Supplementary Fig. 13. Transient absorption (TA) spectroscopy measurements. TA maps and corresponding TA spectra of the MSPE films under **(a, b)** high fluence pump and **(d, e)** low fluence pump, respectively (pump wavelength, 400 nm). **c**, The kinetics of the bleach under these excitation conditions. The overlapping kinetics indicate that the recombination rate only depends on carrier density, i.e., it shows the history-independent behaviour. The peak $\Delta T/T$ corresponds to an injected carrier density of $7.1 \times 10^{18} \text{ cm}^{-3}$.

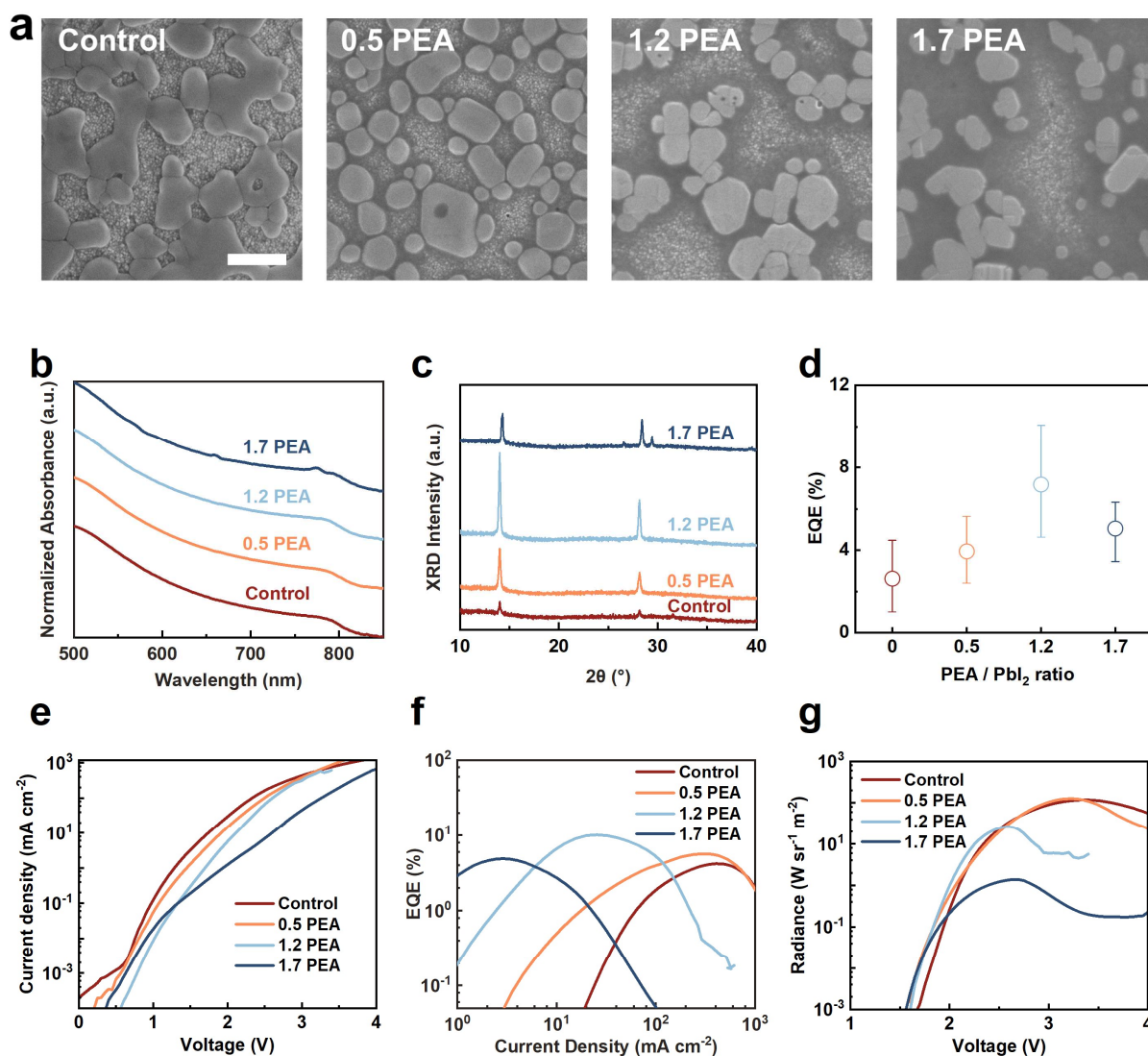


Supplementary Fig. 14. PDS measurement of perovskite films. **a**, PDS spectra of perovskite films. **b**, Urbach energy of perovskite films.

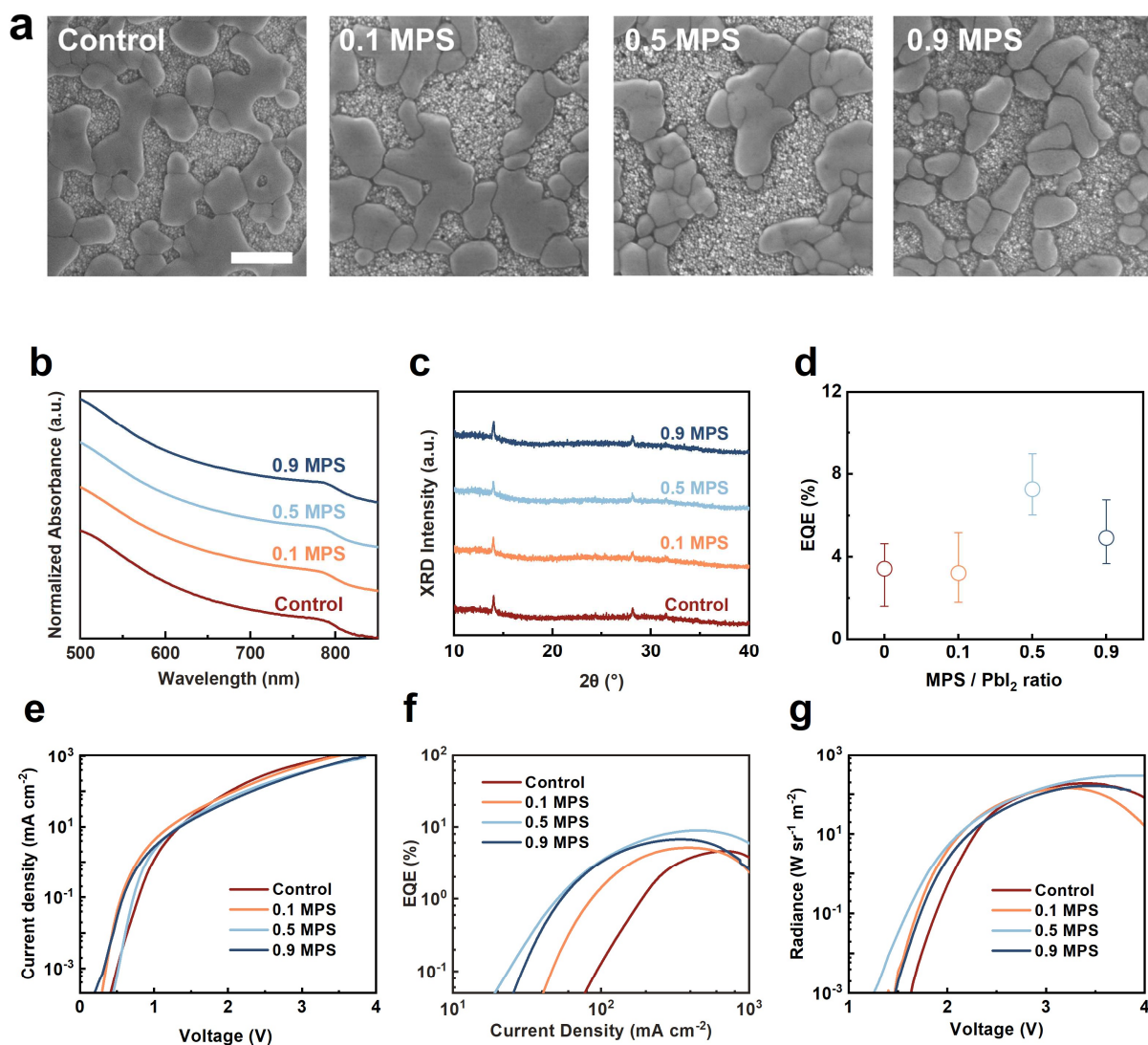
The energetic disorder and defect density in these films can be evaluated by PDS, which shows the optical absorption of these perovskite films with sharp band edges (i.e., Urbach tail). The slope of Urbach tail can be used to estimate the defect density in terms of Urbach energy¹. The Urbach energies of the control, PEA, MPS and MSPE were fitted to be 19.7 meV, 18.0 meV, 17.9 meV, and 17.3 meV, respectively, indicating that PEA and MPS have lower defect density compared to the control ones and MSPE has the lowest defect density. Note that the control film shows a state at ~ 2.5 eV, which could be attributed to inactive hexagonal phase FAPbI_3 (δ -phase). The concentration of this state is slightly reduced by MPS, greatly reduced by PEA and almost completely removed by MSPE. These observations confirm the effects on reduction of defects and energetic disorder by the amino and sulfonyl groups.



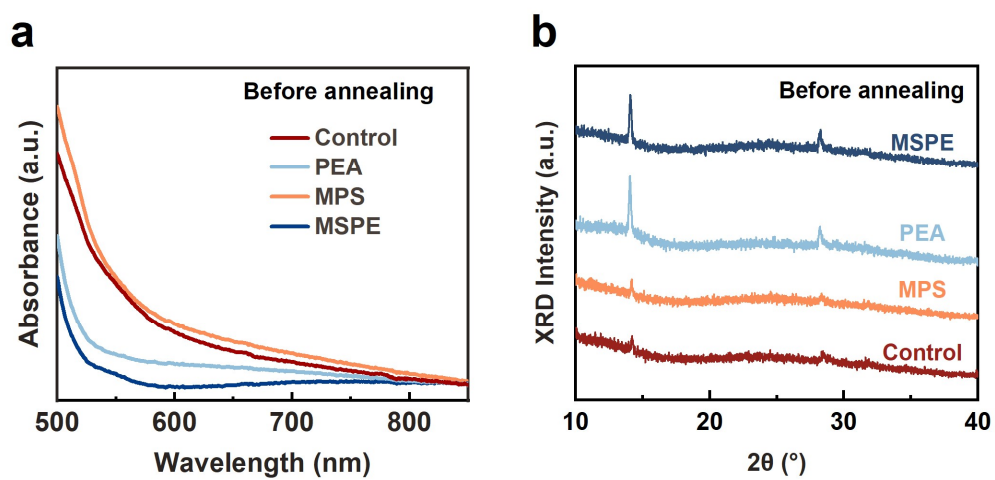
Supplementary Fig. 15. PDS spectra of ZnO, poly-TPD and the mixture samples.



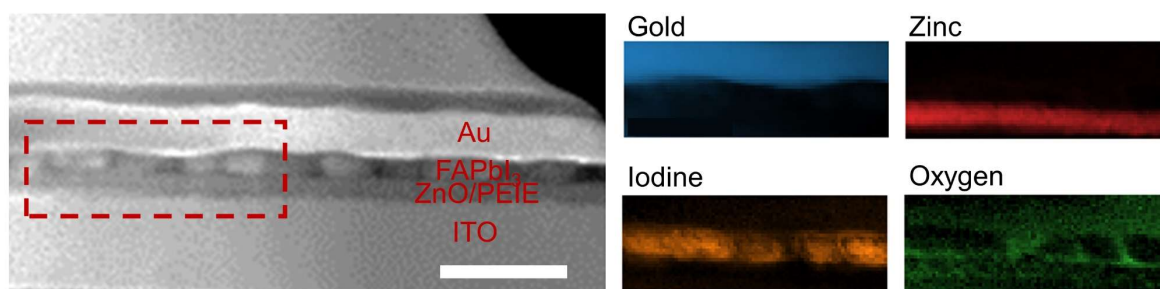
Supplementary Fig. 16. Characteristics of perovskite films and LEDs with different molar ratio of PEA. **a-c**, Characteristics of perovskite films with different molar fractions of PEA. **(a)**, SEM images. The scale bar represents 200 nm. **(b)**, UV-Vis spectra. **(c)**, XRD patterns. **d-g**, The device performance using perovskite layers prepared by FAI, PbI_2 and PEA precursors at an optimized molar ratio of 2.7: 1: x ($x = 0, 0.5, 1.2, 1.7$). **(d)**, Peak EQEs distribution. **(e)**, Current density versus voltage curves. **(f)**, EQE versus current density curves. **(g)**, Radiance versus voltage curves.



Supplementary Fig. 17. Characteristics of perovskite films and LEDs with different molar ratio of MPS. **a-c**, Characteristics of perovskite films with different molar fractions of MSPE. **(a)**, SEM images. The scale bar represents 200 nm. **(b)**, UV-Vis spectra. **(c)**, XRD patterns. **d-g**, The device performance using perovskite layers prepared by FAI, PbI_2 and MPS precursors at an optimized molar ratio of 2.4: 1: x ($x = 0, 0.1, 0.5, 0.9$). **(d)**, Peak EQEs distribution. **(e)**, Current density versus voltage curves. **(f)**, EQE versus current density curves. **(g)**, Radiance versus voltage curves.



Supplementary Fig. 18. Characteristics of perovskite films before annealing. a. UV-Vis spectra. **b.** XRD patterns.



Supplementary Fig. 19. STEM-HAADF and EDX. STEM-HAADF image and associated energy-dispersive X-ray (EDX) analysis of a half device sample with structure of ITO/ ZnO (35 nm)/ PEIE (~ 0.5 nm) / MSPE-modified FAPbI₃ (30 nm)/ Au (60 nm). STEM-HAADF image shows that dark regions fill in the gaps between perovskite grains. EDX further unveils that MSPE molecules (oxygen) exist between perovskite islands (iodine). The scale bar represents 200 nm.

Supplementary Note 1:

Radiance ($\text{W sr}^{-1} \text{m}^{-2}$) corresponds to the power from photons emitted per unit area per unit solid angle and is a general parameter to evaluate the brightness of an LED regardless of emission colours. Luminance (cd m^{-2}) can be converted from radiance using luminous efficacy (i.e., the photopic response of human eyes, standardized by the Commission Internationale de l'Éclairage) and thus only applies to visible emission². As human eyes are insensitive to 800 nm, luminance is not suitable for NIR emission. Radiance can be used as the parameter to compare the brightness of visible and NIR LEDs.

Supplementary Note 2:

Due to the small size of the perovskite grains, CL was used as a complementary technique to the optical microscopies to further understand the emission properties at higher spatial resolution, in parallel to SEM. The intensity of CL signal from a SEM is proportional to the generation rate of electron-hole pairs per second, the internal luminescence efficiency of the absorber, and a function to account for losses from absorption, reflection, and surface recombination. The two CL maps were acquired under the same conditions, keeping most of these factors comparable across samples. For instance, the rate of the charge carrier production, proportional to the electron beam current and the acceleration voltage (which affects the penetration depth) can be assumed to be the same. Also, the thickness of two films is nearly the same. Therefore, the stronger CL intensity in the MSPE sample can be attributed to the promotion of internal luminescence efficiency.

Supplementary Note 3:

The growth of perovskites can be slowed down during film formation due to the strong interactions between FAI and amino group (Fig. 2f). Slower growth can contribute to high crystallinity and regular the shape of grains³. Consequently, both MSPE and PEA molecules with amino group can induce perovskite films with high crystallinity and regular island-shape grains (Supplementary Fig. 1, Supplementary Fig. 16), while MPS with only a sulfonyl group has minor effects on crystallinity and shape of grains (Supplementary Fig. 17). Further insights were gained by characterizing perovskite films with different additives before annealing (after spin-coating). As shown by UV-Vis (Supplementary Fig. 18a), before annealing, MSPE- and PEA-based films show weaker absorption of perovskites than the control and MPS-based film, indicating that the crystallization process is slowed down by

additives with amino group. XRD further unveils that MSPE and PEA films before annealing show higher crystallinity than the MPS and control ones (Supplementary Fig. 18b). These investigations suggest that crystallinity and shape of grains are mainly controlled by the amino group, rather than the sulfonyl group.

Supplementary Note 4:

The recombination processes in perovskite can be described by the rate equation of charge carriers,

$$-\frac{dn(t)}{dt} = k_1 \cdot n + k_2 \cdot n^2 + k_3 \cdot n^3$$

where $n(t)$ is the carrier density, k_1 is the monomolecular recombination rate of geminate exciton recombination or the non-radiative recombination rate, k_2 is the bimolecular recombination rate, and k_3 is the Auger recombination rate. In 3D bulk perovskites, the radiative recombination has been confirmed to be the electron-hole bimolecular recombination and therefore k_1 here is only related to defect-mediated non-radiative recombination⁴⁻⁶. **Fig. 3b** and **d** show the rate equation plots of $dn(t)/dt$ against $n(t)$ extracted from transient absorption (TA) measurements of the control and MSPE films, respectively. We also checked the broadband TA data and found the absence of low-dimensional features in any of our perovskite films, consistent with other data in the manuscript.

Supplementary Note 5:

ZnO substrates beneath the perovskite grains are partially covered by darker regions in 0.1 MSPE film and nearly fully covered in 0.5 MSPE and 0.9 MSPE films (Supplementary Fig. 1a). These darker regions are composed of organic materials, which have lower atomic numbers and thus scatter less electrons and look darker than perovskites and ZnO. Consistent with this, increasing molar ratio of MSPE leads to increasing areal density of dark regions and decreasing areal density of the perovskite grains, suggesting that MSPE exists between perovskite grains. Note that non-perovskite regions are barely covered in the PEA- or MPS-based films (Supplementary Fig. 16-17). PEA or MPS with only one functional group can barely form a layer between discontinuous perovskite grains. In contrast, MSPE can assemble between perovskite grains through hydrogen bonding between amino and sulfonyl group.

Supplementary Table. 1. Features of the reported NIR perovskite LEDs (without additional light-outcoupling structures).

Year	Type of devices	Emitters	EL peak / nm	Peak EQE / %	Peak radiance / W sr ⁻¹ m ⁻²	Stability			Device structure
						Lifetime	Initial Radiance / W sr ⁻¹ m ⁻²	Current density / mA cm ⁻²	
2022	This work	FAPbI ₃ -MSPE	800	23.8	497	T ₅₀ = 4 h	202	300	ITO/ZnO/PEIE/FAPbI ₃ -MSPE/ poly-TPD/MoO _x /Au
						T ₅₀ = 32 h	107	100	
						T ₈₀ = 62 h	45	50	
2022	PeLED ⁷	FAPbI ₃ -SFB10	800	22.8	278.9	T ₅₀ = 14.1 h †	118.8	200	ITO/ZnO/PEIE/FAPbI ₃ -SFB10/ TFB/MoO _x /Au
2019	PeLED ⁸	FAPbI ₃ -ODEA	800	21.6	308	T ₅₀ = 0.3 h	200 ††	200	ITO/ZnO/PEIE/FAPbI ₃ -ODEA/ TFB/MoO _x /Au
						T ₅₀ = 20 h	25 ††	25	
2021	PeLED ³	FAPbI ₃ -AEAA	800	22.2	250 †	T ₅₀ = 18.6 h	100 ††	100	ITO/ZnO/PEIE/FAPbI ₃ -AEAA/ TFB/MoO _x /Au
2019	PeLED ⁹	FAPbI ₃ -5AVA	799	20.2	170	T ₈₀ = 20 h	57	57	ITO/ZnO/PEIE/FAPbI ₃ -5AVA/ poly-TPD/MoO _x /Au
2018	PeLED ¹⁰	FAPbI ₃ -5AVA	803	20.7	390	T ₅₀ = 20 h	100 ††	100	ITO/ZnO/PEIE/FAPbI ₃ -5AVA/ TFB/MoO _x /Au
2018	PeLED ¹¹	(NMA) ₂ (FA)Pb ₂ I ₇ :poly-HEMA	795	20.1	5 †	T ₅₀ = 46 h	0.1 ††	0.1	ITO/MZO/PEIE/(NMA) ₂ (FA)Pb ₂ I ₇ :poly-HEMA/TFB:PFO/MoO _x /Au
2020	PeLED ¹²	DDS:FAPbI ₃	800 †	17.3	260 †	T ₅₀ = 0.3 h	200 ††	300	ITO/ZnO/PEIE/DDS:FAPbI ₃ / TFB/MoO _x /Au
						T ₅₀ = 100 h	15	20	

2021	PeLED ¹³	PPAI- FA _{0.83} CS _{0.17} PbI ₃	789	17.5	1282.8	T ₅₀ = 130 h	70 ††	100	ITO/ZnO/PEIE/ PPAI-FA _{0.83} CS _{0.17} PbI ₃ /TFB/Au
2018	PeLED ¹⁴	FPMAl- MAPb _{0.6} Sn _{0.4} I ₃	917	5.0	2.7	T ₈₀ = 0.1 h	1 ††	5	ITO/poly-TPD/FPMAl- MAPb _{0.6} Sn _{0.4} I ₃ /TPBi/LiF/Al
2019	PeLED ¹⁵	FAPbI ₃	804	14.2	241	T ₅₀ = 23.7 h	67	100	ITO/ZnO/PEIE/FAPbI ₃ / TFB/MoO _x /Au
2016	PeLED ¹⁶	(NMA) ₂ (FA)Pb ₂ I ₆ Br	763	11.7	82	T ₅₀ = 2 h	5 ††	10	ITO/ZnO/PEIE/(NMA) ₂ (FA)Pb ₂ I ₆ Br /TFB/MoO _x /Au
2016	PeLED ¹⁷	PEA ₂ MA _{n-1} Pb _n I _{3n+1}	760 †	8.8	80	Not reported			ITO/TiO ₂ /PEA ₂ MA _{n-1} Pb _n I _{3n+1} / F8/MoO _x /Au
2015	PeLED ¹⁸	MAPb(I/Cl) ₃	768	3.5	28	Not reported			ITO/ZnO/PEI/MAPb(I/Cl) ₃ / TFB/MoO _x /Au

† Average lifetime as reported in the reference paper.

†† These numbers were estimated from the data or figures in the reference papers.

Supplementary Table. 2. Features of the reported perovskite LEDs with EQE exceeding 20% (NIR excluded, without additional light-outcoupling structures).

Year	Type of devices	EL peak / nm	Peak EQE / %	Stability			
				Lifetime	Initial Luminance / cd m^{-2}	Initial Radiance / $\text{W sr}^{-1} \text{m}^{-2}$	Current Density / mA cm^{-2}
2022	This work	800	23.8	$T_{50} = 4 \text{ h}$	N/A	202	300
				$T_{50} = 32 \text{ h}$		107	100
				$T_{80} = 62 \text{ h}$		45	50
2021	PeLED ³²	640	23	$T_{50} = 10 \text{ h}$	200	1.5 †	Not reported
2021	PeLED ³³	620	20.3	$T_{50} = 0.3 \text{ h}$	585	2.0 †	10
2018	PeLED ³⁴	649	21.3	$T_{50} < 0.1 \text{ h}$ ††	100	1.2 †	1.25
2022	PeLED ³⁵	540	28.9	$T_{50} = 14 \text{ h}$	10,000	15 †	Not reported
2021	PeLED ³⁶	512 ††	25.6	$T_{50} = 1.9 \text{ h}$	7,200	19 †	8
2021	PeLED ³⁷	525 ††	20.36	$T_{50} = 0.1 \text{ h}$	10,000	18 †	Not reported
2021	PeLED ³⁸	512	20.5	$T_{50} = 0.4 \text{ h}$	1,000	2.6 †	Not reported
2020	PeLED ³⁹	531	20.1	$T_{80} = 2.9 \text{ h}$	100	0.17 †	Not reported
2020	PeLED ⁴⁰	505 ††	22	$T_{50} = 1 \text{ h}$	1,200	4.1 †	Not reported
2021	PeLED ⁴¹	512	22.49	$T_{50} = 0.9 \text{ h}$	144	0.38 †	0.3
2021	PeLED ⁴²	530 ††	23.4	$T_{50} = 0.1 \text{ h}$ †	1,000	1.7 †	0.053 mA
2018	PeLED ⁴³	525 ††	20.3	$T_{50} = 0.2 \text{ h}$	7,130	13 †	166.67

† For LEDs emitting visible light, initial radiance ($\text{W sr}^{-1} \text{m}^{-2}$) was estimated from the initial luminance (cd m^{-2}) provided in the reference papers using luminous efficacy at their peak EL wavelengths (Supplementary Note 1)². For example, 100 cd m^{-2} of Ir(ppy)_3 OLEDs (EL peak of 516 nm) corresponds to a radiance of $\sim 0.2 \text{ W sr}^{-1} \text{m}^{-2}$. †† These numbers were estimated from the data or figures in the reference papers.

Supplementary Table. 3. Features of the reported NIR organic and quantum-dot LEDs (without additional light-outcoupling structures).

Year	Type of devices	Emitters	EL peak / nm	Peak EQE / %	Peak Radiance / W sr ⁻¹ m ⁻²	Stability		
						Lifetime	Initial Radiance / W sr ⁻¹ m ⁻²	Current density / mA cm ⁻²
2017	OLED ¹⁹	Pt(fprpz) ₂	740	24	360	Not reported		
2018	OLED ²⁰	TTM-3NCz	710	27.0	1 †	Not reported		
2019	OLED ²¹	F-Pt	724	16.7	1.164	Not reported		
		tBu-Pt	708	13.9	10.38			
2020	OLED ²²	Pt(fprpz) ₂	794	10.61	97.8	Not reported		
2019	OLED ²³	Os(ftrmpz) ₂ (PPhMe ₂) ₂	710	11.5	73 †	Not reported		
2007	OLED ²⁴	Pt(tpbp)	769	6.3	0.6 †	T ₉₀ = 1000 h	0.6 †	40
2021	OLED ²⁵	DPSF:CN-T2T:TTDSF	774	5.3	5 †	T ₉₅ = 295.7 h	0.5 †	10
2016	QDLED ²⁶	PbS	1391	5.2	2.6	Not reported		
2019	QDLED ²⁷	PbS	1400 †	7.9	9	T ₉₅ = 48 h	0.16	1.75
2020	QDLED ²⁸	PbS	1400 †	8.1	35	T ₅₀ = 532 h	7	Not reported
2020	QDLED ²⁹	PbS in Perovskites	980	8.1	7.4	T ₅₀ = 1.1 h	4 †	10
2020	QDLED ³⁰	Ag ₂ S@SiO ₂ in Perovskites	1397	16.98	83.93	T ₈₅ = 480 h	83 †	Not reported
2015	QDLED ³¹	PbS-CdS	1242	4.3	1 †	Not reported		

† These numbers were estimated from the data or figures in the reference papers.

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