

Supporting Information

Interfacial dipole engineering in all-inorganic perovskite solar cells

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1. Experimental

2.1 Materials and Reagents

Stannous chloride anhydrous (SnCl_2 , Aladdin), thiourea ($\text{CH}_4\text{N}_2\text{S}$, Aladdin), lead bromide (PbBr_2 , Aladdin), cesium bromide (CsBr , Aladdin), N,N-dimethylformamide (DMF, Sinopharm), titanium tetrachloride (TiCl_4 , Sinopharm), methanol (CH_3OH , Sinopharm), isopropanol ($\text{C}_3\text{H}_8\text{O}$, Sinopharm), 5-nitro-7-azaindole ($\text{C}_7\text{H}_5\text{N}_3\text{O}_2$, Acmecc), 7-azaindole ($\text{C}_7\text{H}_5\text{N}_2$, Acmecc), 5-methoxy-7-azaindole ($\text{C}_8\text{H}_8\text{N}_2\text{O}$), [6,6]-phenyl- C_{61} -butyric acid methyl ester (PC_{61}BM , Polymer), FTO glass ($7 \Omega \text{ square}^{-1}$) and carbon paste (Shanghai Mater Win New Materials Co., Ltd) were used as supplied without further purification.

2.2 Solar Cell Fabrication

The etched FTO conductive glass was ultrasonically cleaned with detergent, deionized water, and ethanol. The freshly cleaned FTO substrate was further cleaned in a plasma cleaner for 5 min. The cleaned FTO substrate and SnO_2 QDs solution were preheated at $80 \text{ }^\circ\text{C}$ for 5 min. The preheated SnO_2 QDs solution was spin-coated on FTO substrate at 2000 rpm for 30 s, and annealed at $200 \text{ }^\circ\text{C}$ for 1 h to obtain a dense SnO_2 ETL. The $\text{TiO}_x\text{Cl}_{4-2x}$ modification was performed by soaking the SnO_2 coated FTO glass in 40 mM TiCl_4 aqueous solution at $75 \text{ }^\circ\text{C}$ for 30 min, and the substrates were rinsed with deionized water and ethanol, and then dried in air at $200 \text{ }^\circ\text{C}$ for 30 min. Next, DMF solution of 1 M PbBr_2 was spin-coated on the obtained substrate (2000 rpm, 30 s) and heated at $90 \text{ }^\circ\text{C}$ for 30 min to obtain PbBr_2 film. After that, the methanol solution of 0.07 M CsBr was spin-coated (2000 rpm, 30 s) on PbBr_2 film surface and annealed at $250 \text{ }^\circ\text{C}$ for 5 min. This process was repeated for seven times until a high quality CsPbBr_3 film was formed. For devices with the surface passivation treatment, when the annealed perovskite films were cooled to room temperature, 7-AI derivatives in isopropanol solution with different concentration was dropped

onto the perovskite films and spin-coated at 2000 rpm for 30 s and annealed at 90 °C for 15 min. At last, a conductive carbon paste was coated onto CsPbBr₃ layer and annealed at 90 °C for 30 min. For CsPbI₂Br devices, 1.2 M CsPbI₂Br precursor containing 312 mg of CsI, 277 mg of PbI₂ and 220 mg of PbBr₂ dissolved in 1 mL of DMSO was spin-coated onto FTO/SnO₂-TiO_xCl_{4-2x} substrate at 1000 rpm for 10 s and then at 5000 rpm for 50 s. Afterwards, the substrate was placed on a hotplate at 160 °C for 2 min and then at 270 °C for 8 min and cooled down to room temperature naturally. The remaining steps are identical to those involved in preparing CsPbBr₃ PSC.

2.3 Measurements and characterizations

The energy dispersive spectroscopy (EDS) mapping images were carried out using a scanning electron microscope (HORIBA EX-350). The X-ray diffraction (XRD) patterns were recorded by X-ray diffractometer (Bruker D8 Advance). Fourier transform infrared (FTIR) spectra were obtained on Fourier infrared spectrometer (Nicolet IS 10). Kelvin probe force microscopy (KPFM) images were obtained using Kelvin probe force microscopy (KPFM, CypherTM, Asylum Research). The ultraviolet photoelectron spectrometer (UPS) spectra were characterized by ultraviolet photoelectron spectroscopy (Thermo ESCALAB 250XI). A space charge limited current (SCLC) model was employed to characterize the trap density of PVK films for electron-only equipment with the structure of FTO/SnO₂-TiO_xCl_{4-2x}/CsPbBr₃/PCBM/carbon at the voltage range of 0-7 V under dark condition. Ultraviolet-visible (UV-vis) transmission and absorption spectra were characterized via Meipuda UV-3200 spectrophotometer. The photocurrent density-voltage (*J-V*) curves of various devices were measured by the sourcemeter of Keithley 2450 under irradiation of simulated sunlight (AM 1.5, 100 mW cm⁻² calibrated with a standard silicon solar cell, Newport, Oriel Class A, 91195 A) at a voltage range of 0-1.7 V. The external quantum efficiency (EQE) spectra were obtained using a monochromatic incident photon-to-electron

conversion efficiency (IPCE) kit from Enli Technology Co., Ltd equipped with a standard Si crystalline solar cell as a reference. Steady power output curves at the maximum power point (MPP) were recorded by electrochemical workstation of CHI660E at the bias voltage at maximal power output point. A confocal fluorescence microscope (ISS Q2 FLIM/FFS, ISS Inc.) was employed to measure the confocal PL mapping images. The steady state photoluminescence (PL) spectra were recorded via a fluorescence spectrometer (FLS1000/FS5) excited by a 350 nm laser and the time-resolved photoluminescence (TRPL) spectra were characterized using a Horiba spectrometer with an excitation wavelength of 500 nm. Capacitance-voltage ($C-V$) curves were measured at a frequency of 5 kHz with an amplitude of 5 mV under dark. The dark $J-V$ curves and the electrochemical impedance spectroscopy (EIS) of PSCs were measured on CHI660E electrochemical workstation in the dark. The open-circuit voltage (V_{oc}) decay curve was measured in open circuit mode by irradiating with 100 mW cm⁻² light intensity for several seconds and then instantaneously switching off the light.

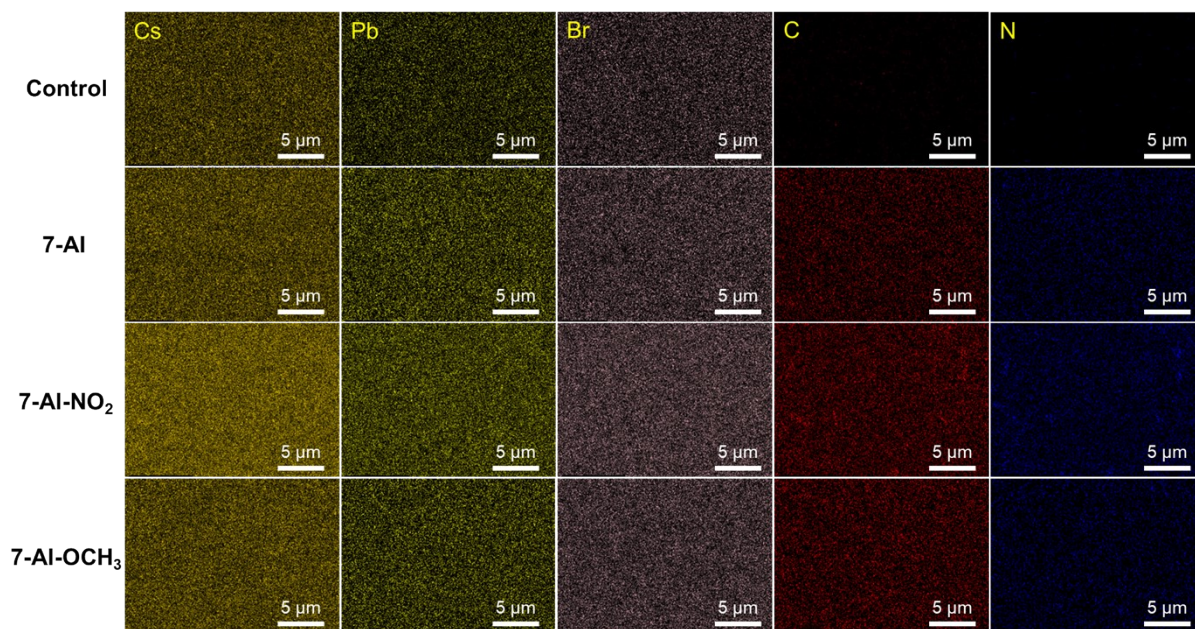


Fig. S1. The corresponding EDS mapping images of various CsPbBr₃ films.

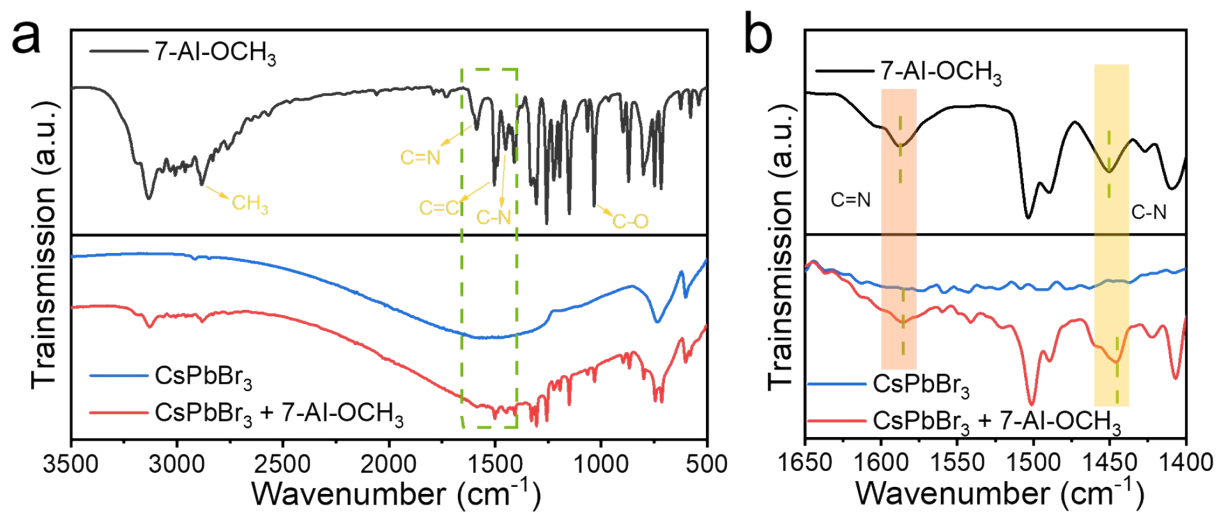


Fig. S2. (a, b) FTIR spectra of pure 7-AI-OCH₃, pure CsPbBr₃ perovskite, and 7-AI-OCH₃ treated CsPbBr₃ perovskite film.

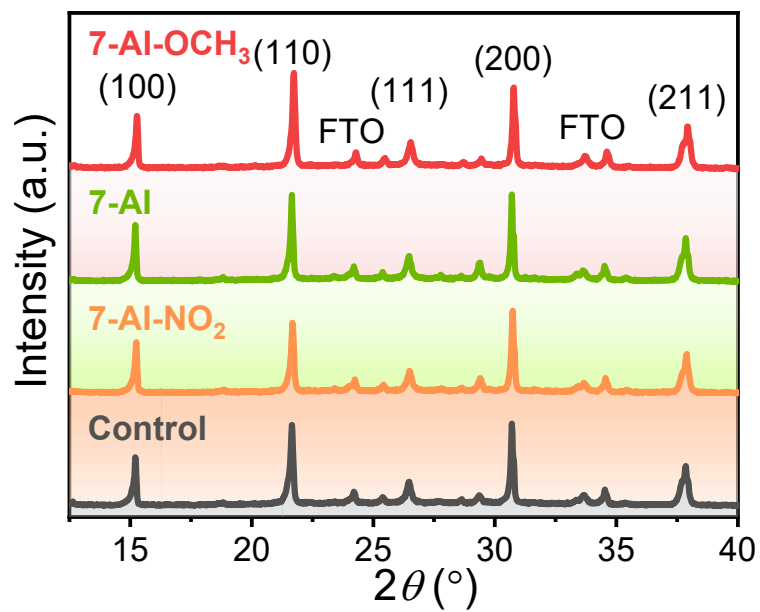


Fig. S3. XRD patterns of perovskite films with and without 7-AI derivatives treatment.

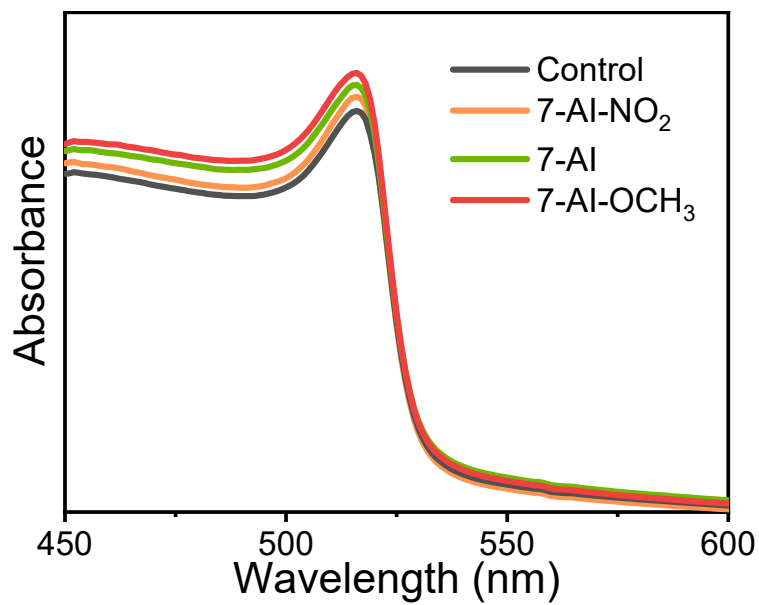


Fig. S4. UV-vis absorption spectra of various CsPbBr₃ films.

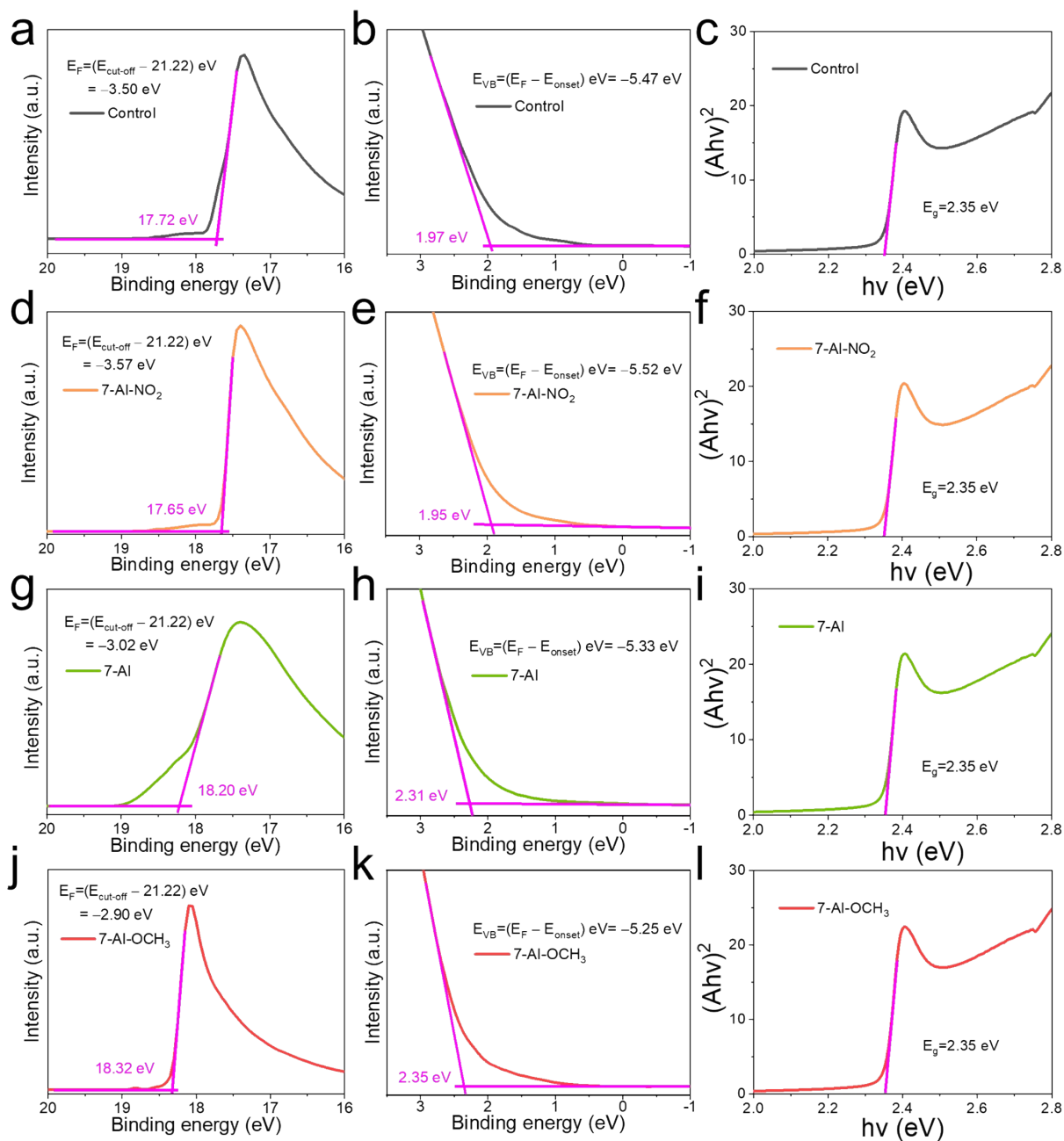


Fig. S5. UPS spectra of the (a-b) control perovskite film and the perovskite films treated with (d-e) 7-Al-NO₂, (g-h) 7-Al, (j-k) 7-Al-OCH₃. The calculation of bandgap of (c) control perovskite film and the perovskite films treated with (f) 7-Al-NO₂, (i) 7-Al, (l) 7-Al-OCH₃ dipoles.

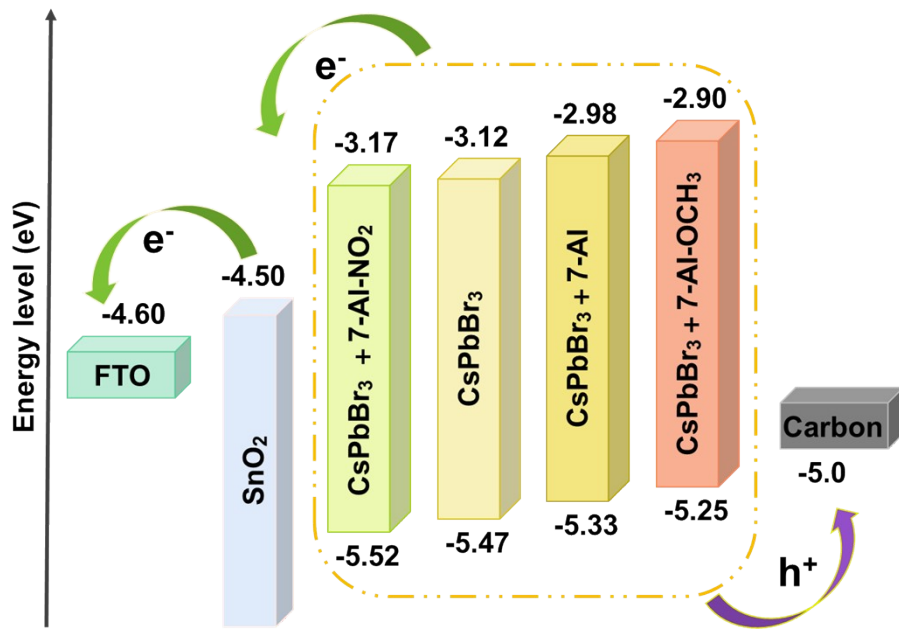


Fig. S6. The energy level schematic of the CsPbBr₃ PSC after treatment.

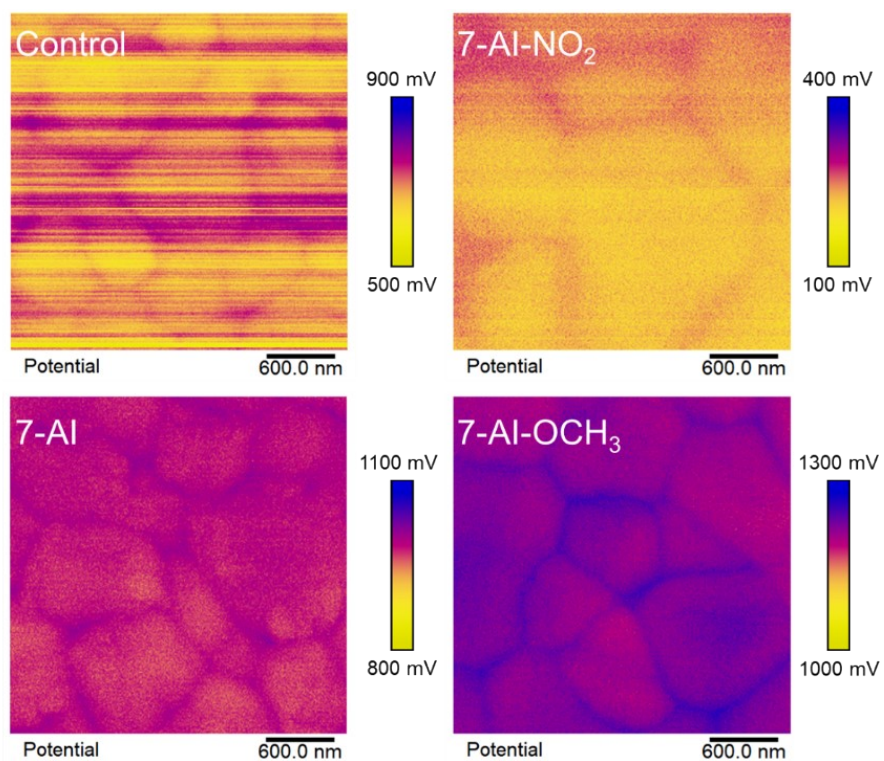


Fig. S7. The surface potential spectra for the control perovskite film and the films treated with 7-AI-NO₂, 7-AI, and 7-AI-OCH₃.

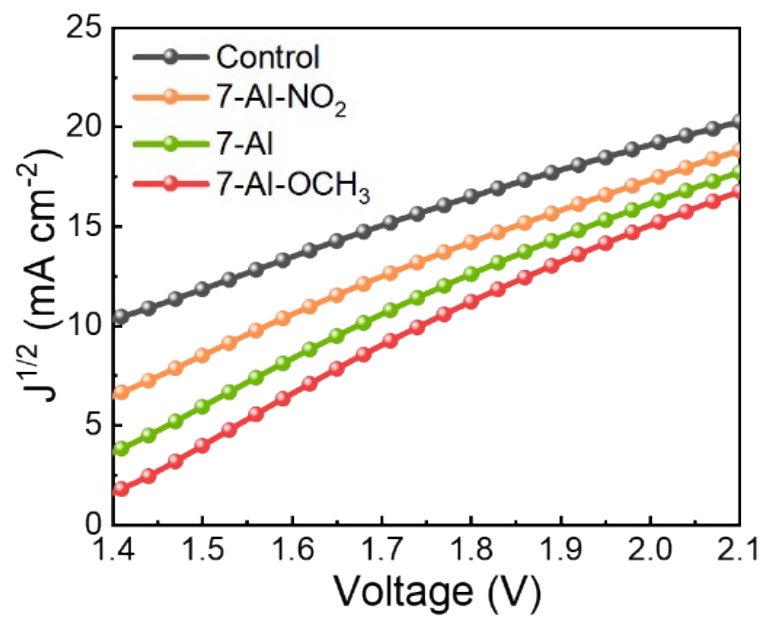


Fig. S8. The $J^{1/2}$ - V curves of various devices.

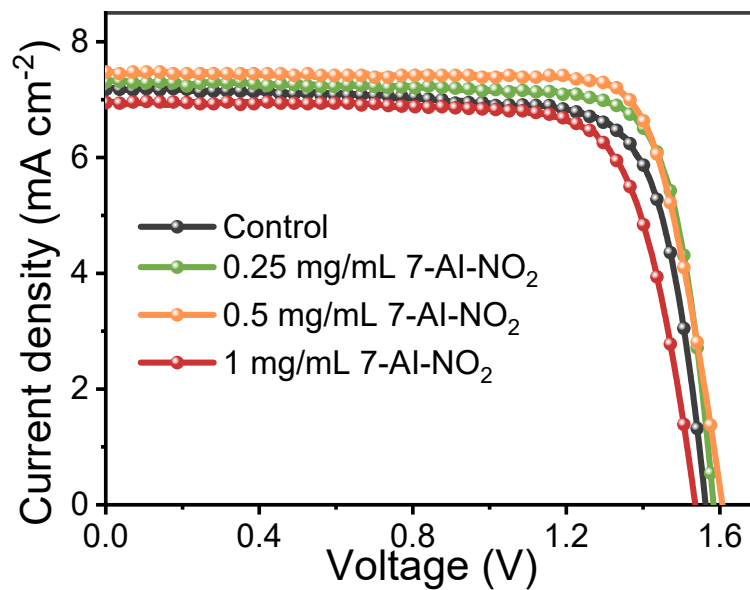


Fig. S9. *J-V* curves of the CsPbBr₃ PSCs fabricated with different concentrations of 7-AI-NO₂ molecules.

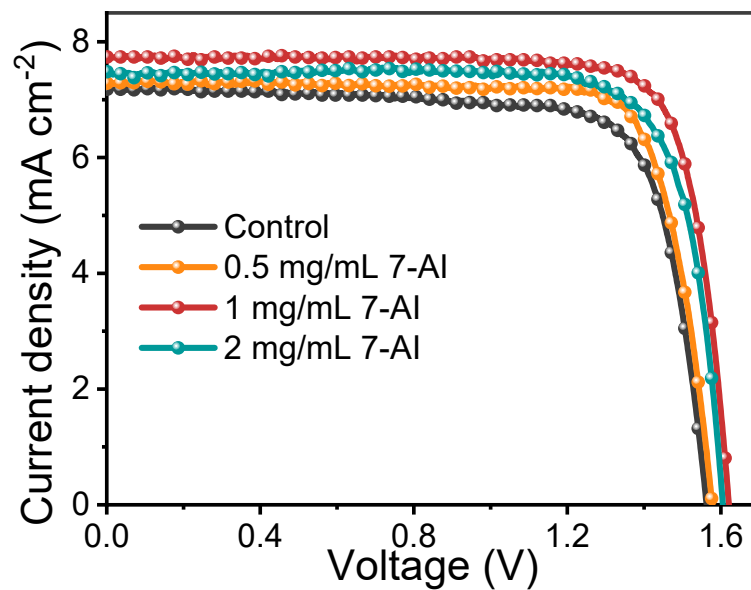


Fig. S10. J - V curves of the CsPbBr₃ PSCs fabricated with different concentrations of 7-AI molecules.

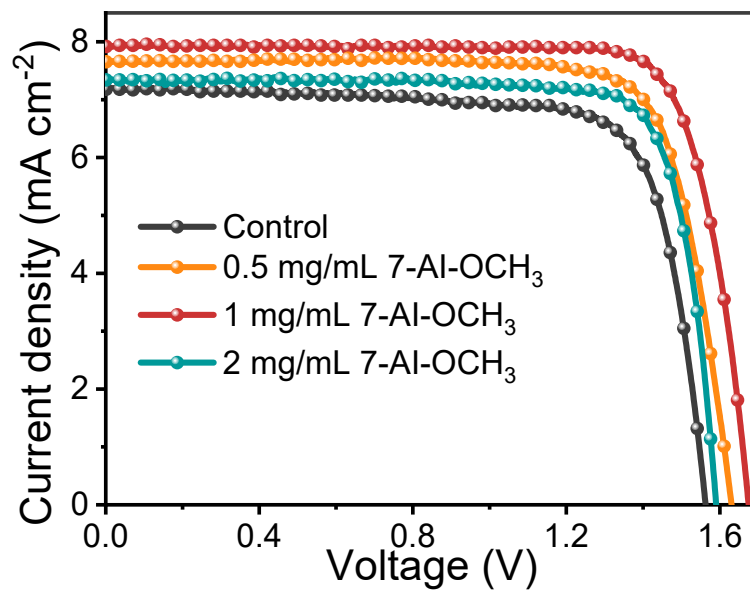


Fig. S11. J - V curves of the CsPbBr₃ PSCs fabricated with different concentrations of 7-AI-OCH₃ molecules.

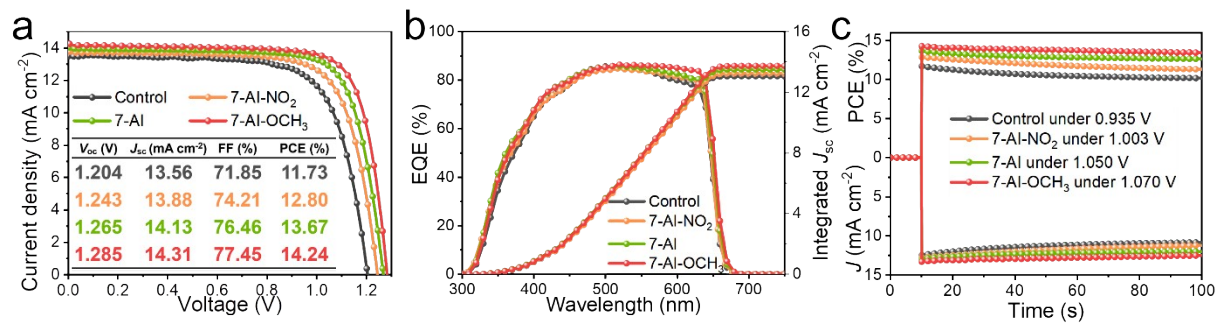


Figure S12. (a) J - V curves, (b) EQE spectra and (c) steady power outputs of various CsPbI₂Br PSCs.

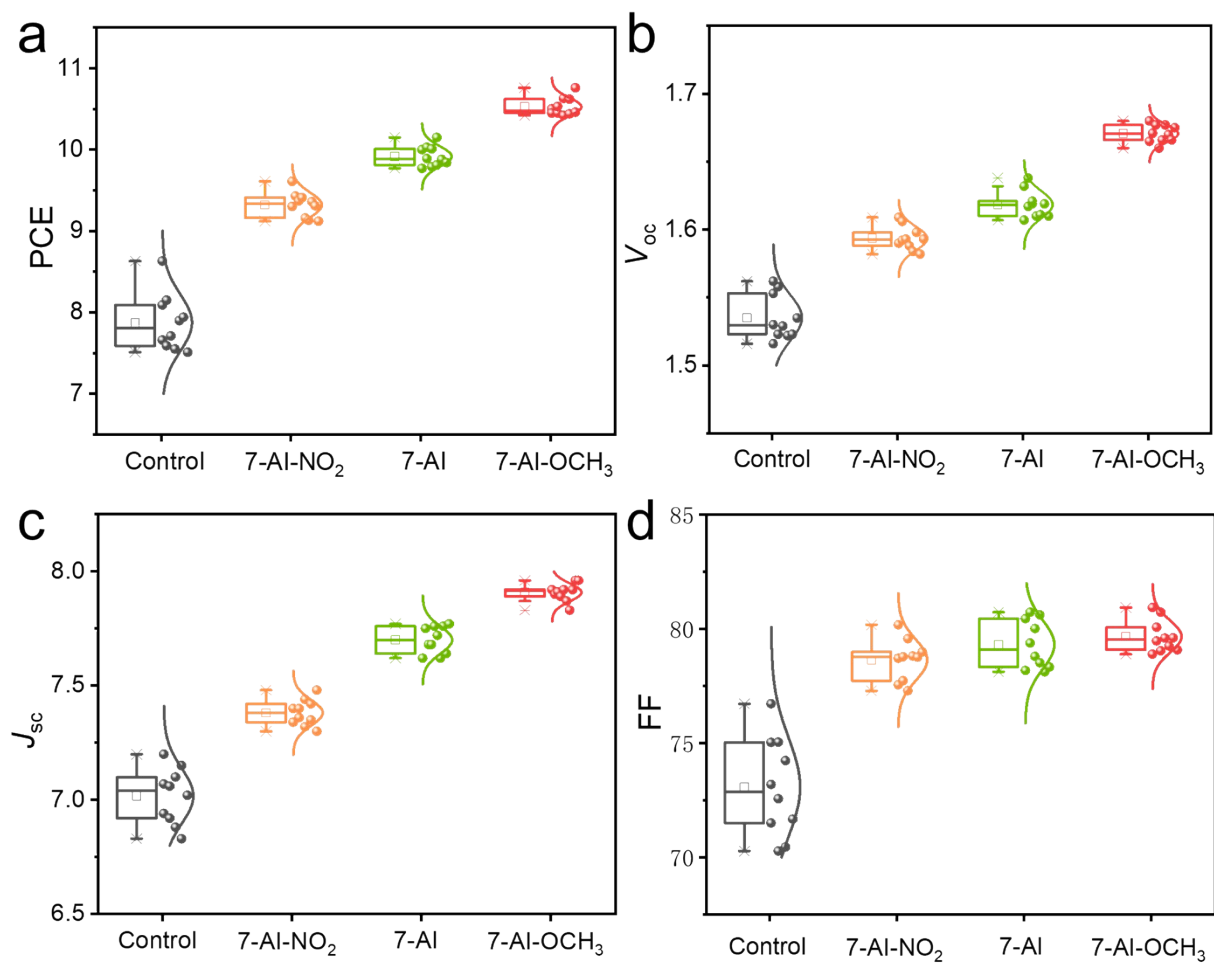


Fig. S13. (a-d) The distribution statistics of photovoltaic data (PCE, V_{oc} , J_{sc} and FF) for inorganic CsPbBr₃ PSCs.

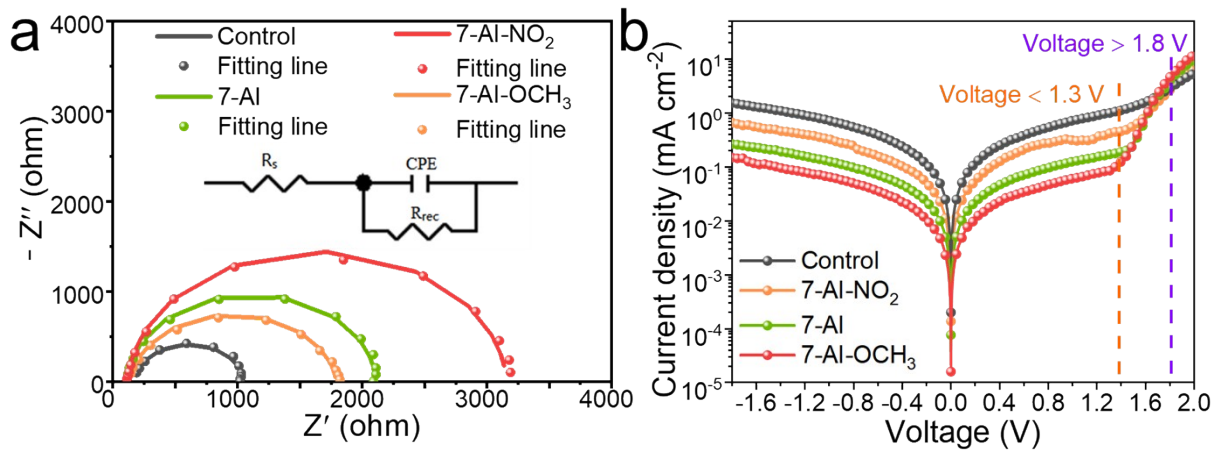


Fig. S14. (a) EIS plots and (b) the dark $J-V$ curves of inorganic PSCs with control and dipole molecule tailored perovskite films.

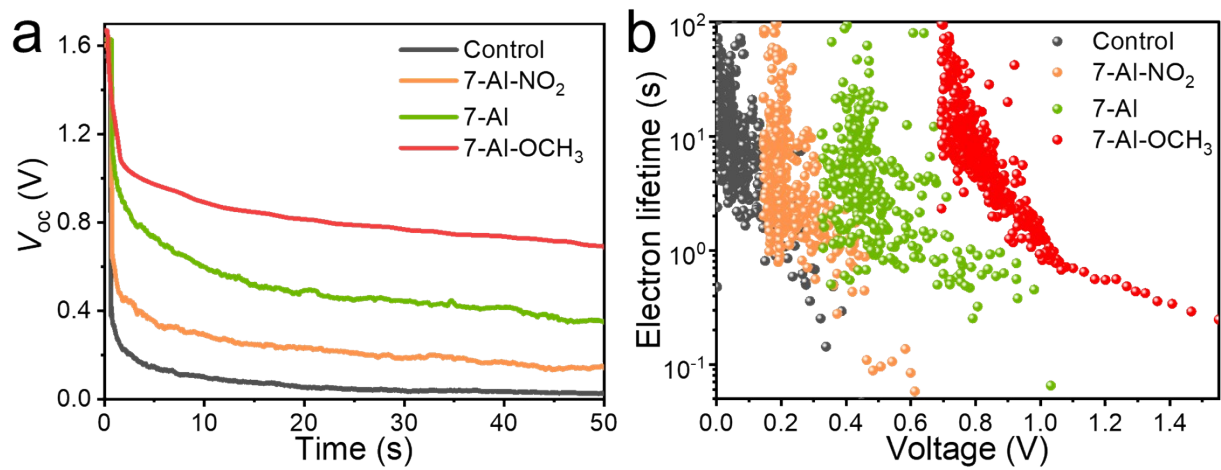


Fig. S15. (a) V_{oc} decay curves and (b) calculated electron lifetime from V_{oc} decay curves of various devices.

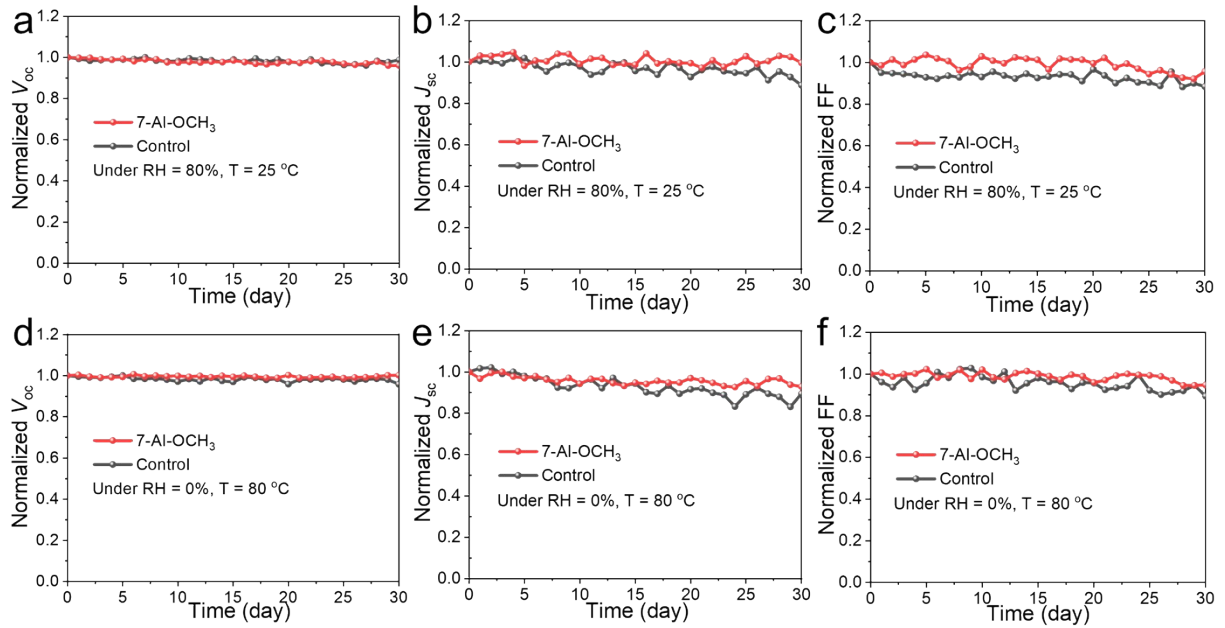


Fig. S16. The normalized parameters including V_{oc} , J_{sc} and FF of CsPbBr₃ PSCs at (a, b, c) RH=80%, T=25 °C and (d, e, f) RH=0%, T=80 °C.

Table S1. Photovoltaic data for state-of-the-art CsPbBr₃ PSCs are compared.

Devices	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)	Ref.
Based on CsPbBr₃					
FTO/SnO₂-TiO_xCl_{4-2x}/CsPbBr₃/7-Al-OCH₃/carbon	7.96	1.675	80.73	10.76	This work
FTO/TiO ₂ /DTPT/CsPbBr ₃ /DTPT/carbon	8.52	1.574	83.67	11.21	[1]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /(WS ₂ /AgIn ₅ S ₈) QDs HTM/carbon	7.49	1.627	84.03	10.24	[2]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /Br-CQDs/CsPbBr ₃ -Br-CQDs/carbon	7.84	1.651	83.36	10.79	[3]
FTO/TiO ₂ /CsPbBr ₃ /ReSe ₂ /carbon	7.92	1.622	83.06	10.67	[4]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /DCC/carbon	7.79	1.611	80.96	10.16	[5]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /ASF/CsPbBr ₃ /carbon	7.47	1.615	83.56	10.08	[6]
FTO/TiO ₂ /GQDs /CsPbBr ₃ /P3HT/carbon	9.36	1.420	73.00	9.74	[7]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ -choline bromide/carbon	7.82	1.532	75.6	9.06	[8]
FTO/SnO ₂ -TiO _x Cl _{4-2x} /CsPbBr ₃ +Ti ₃ C ₂ Cl _x /Ti ₃ C ₂ Cl _x /carbon	7.87	1.702	82.7	11.08	[9]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /Br-GO/carbon	7.88	1.602	80.01	10.10	[10]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /HMTA/carbon	7.51	1.605	83.63	10.08	[11]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ :g-C ₃ N ₄ /carbon	7.80	1.278	80.25	8.00	[12]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /carbon	7.18	1.320	75.90	7.19	[13]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /CuInS ₂ /ZnS QDs/LPP-C	7.73	1.626	86.30	10.85	[14]
FTO/SnO ₂ /CsPbBr ₃ /N-CQDs/carbon	7.87	1.622	80.10	10.71	[15]
FTO/SnO ₂ -TiO _x Cl _{4-2x} /WS ₂ /CsPbBr ₃ /carbon	7.95	1.700	79.00	10.65	[16]
FTO/SnO ₂ /TiO _x Cl _{4-2x} /Cs _{0.91} Rb _{0.09} PbBr ₃ /carbon	7.96	1.629	80.50	10.44	[17]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /AC/CsPbBr ₃ /ZnPc/carbon	7.64	1.606	82.47	10.12	[18]

FTO/TiO ₂ /CsPbBr ₃ /[BMMIm]Cl/carbon	7.45	1.610	83.00	9.92	[19]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /(MoO ₂ /NC)/carbon	7.20	1.532	85.20	9.40	[20]
FTO/TiO ₂ /CsPbBr ₃ /BHJ/carbon	7.82	1.500	76.20	8.94	[21]
FTO/ <i>c</i> -TiO ₂ /CsBr/CsPb _{1-x} Co _x Br ₃ /carbon	7.48	1.380	84.00	8.67	[22]
FTO/TiO ₂ /CsPbBr ₃ /Spiro-OMeTAD/MoO _x /ITO	7.56	1.410	73.00	7.80	[23]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /Cu(Cr,Ba)O ₂ /carbon	7.81	1.615	85.50	10.79	[24]
FTO/SnO ₂ /CsPbBr ₃ /CsSnBr ₃ /carbon	7.80	1.610	84.4	10.60	[25]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /GQDs/CsPbBr ₃ /MnS/carbon	8.28	1.520	83.00	10.45	[26]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /Cs _{0.91} Rb _{0.09} PbBr ₃ /J ₆₁ -ITIC/carbon	8.18	1.580	80.00	10.34	[27]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /PIZ1/carbon	7.652	1.578	83.06	10.03	[28]
FTO/TiO ₂ /CsPbBr ₃ /Ti ₃ C ₂ -MXene/carbon	8.54	1.444	73.08	9.01	[29]
FTO/ <i>c</i> -TiO ₂ /SnO ₂ /CsPbBr ₃ /CuPc/carbon	8.24	1.310	81.40	8.79	[30]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPb _{0.97} Tb _{0.03} Br ₃ /SnS:ZnS/NiO _x /carbon	8.21	1.570	79.60	10.26	[31]
FTO/TiO ₂ /GQDs CsPbBr ₃ /CISZ-QDs/carbon	7.35	1.522	84.30	9.43	[32]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /GQDs/CsPbBr ₃ CdZnSe@ZnSe/carbon	7.25	1.498	79.60	8.65	[33]
FTO/ <i>c</i> -TiO ₂ / <i>m</i> -TiO ₂ /CsPbBr ₃ /MoS ₂ QDs/carbon	6.55	1.307	79.40	6.80	[34]

Table S2. The carrier lifetimes obtained from TRRL spectra of various CsPbBr₃ devices.

Samples	τ_{ave} (ns)	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)
Control	0.649	0.649	90.81	0.649	9.19
7-AI-NO ₂	0.790	0.492	67.12	1.070	32.88
7-AI	0.919	0.476	60.26	1.188	39.74
7-AI-OCH ₃	1.268	0.403	69.66	1.731	30.34

The TRPL decay curves are fitted using a biexponential function of $I = Ae^{-(\tau-\tau_0)/\tau_1} + Be^{-(\tau-\tau_0)/\tau_2}$, where I is the PL intensity, τ_1 and τ_2 correspond to the fast decay time of the defect-induced non-radiative recombination and the low decay time of radiative recombination, respectively, and A_1 and A_2 are the corresponding decay constants. The average decay time (τ_{ave}) is calculated to evaluate the electron extraction ability according to $\tau_{\text{ave}} = (A_1\tau_1^2 + A_2\tau_2^2)/(A_1\tau_1 + A_2\tau_2)$, where τ_1 is the faster component of trap-mediated nonradiative recombination and τ_2 is the slower component correlated to radiative recombination.

Table S3. The V_{TFL} , N_{t} , and μ_{h} values of various CsPbBr₃ devices.

Samples	V_{TFL} (V)	N_{t} (cm ⁻³)	μ_{h} (cm ² V ⁻¹ s ⁻¹)
Control	1.402	9.478×10^{14}	2.05×10^{-3}
7-AI-NO ₂	1.337	9.039×10^{14}	3.38×10^{-3}
7-AI	1.275	8.620×10^{14}	1.01×10^{-2}
7-AI-OCH ₃	1.181	7.984×10^{14}	1.65×10^{-2}

The trap density (N_{t}) can be calculated by the following formula: $N_{\text{t}} = 2\varepsilon_{\text{r}}\varepsilon_0 V_{\text{TFL}}/qL^2$, where the ε_{r} value of 22 corresponds to the relative dielectric constant of the CsPbBr₃ film and the ε_0 value of 8.85×10^{-12} F m⁻¹ represents the vacuum permittivity, q is the electron charge, L is the thickness of perovskite film, and V_{TFL} , an important factor to measure the trap states density, is the onset voltage of the trap-filled limit region.

Table S4. Energy band structure data of control, 7-AI-NO₂, 7-AI and 7-AI-OCH₃ perovskite films.

Samples	$E_{\text{cut-off}}$ (eV)	E_{onset} (eV)	E_{F} (eV)	VB (eV)	E_{g} (eV)	CB (eV)
Control	17.72	1.97	-3.50	-5.47	2.35	-3.12
7-AI-NO ₂	17.65	1.95	-3.57	-5.52	2.35	-3.17
7-AI	18.20	2.31	-3.02	-5.33	2.35	-2.98
7-AI-OCH ₃	18.32	2.35	-2.90	-5.25	2.35	-2.90

Table S5. The photovoltaic data for the CsPbBr₃ PSCs fabricated with different concentrations of 7-AI-NO₂ molecules from Fig. S9.

Devices	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
Control	1.562	7.20	76.72	8.63
0.25 mg/mL	1.583	7.33	79.61	9.24
0.5 mg/mL	1.606	7.48	79.99	9.61
1 mg/mL	1.536	6.97	76.43	8.18

Table S6. The photovoltaic data for the CsPbBr₃ PSCs fabricated with different concentrations of 7-AI molecules from Fig. S10.

Devices	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
Control	1.562	7.20	76.72	8.63
0.5 mg/mL	1.578	7.33	80.17	9.27
1 mg/mL	1.621	7.77	80.61	10.15
2 mg/mL	1.605	7.50	78.86	9.49

Table S7. The photovoltaic data for the CsPbBr₃ PSCs fabricated with different concentrations of 7-AI-OCH₃ molecules from Fig. S11.

Devices	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
Control	1.562	7.20	76.72	8.63
0.5 mg/mL	1.630	7.73	78.40	9.88
1 mg/mL	1.675	7.96	80.73	10.76
2 mg/mL	1.590	7.39	80.16	9.47

Reference

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