Suppolementary information

Experimental section

Materials

Fluorine-doped tin oxide (FTO) glass substrates and commercially available conductive carbon paste were obtained from Youxuan Tech. ZnO colloidal precursor consisting of 2.5 wt.% Zinc (II) oxide in n-butanol was obtained from Xi'an Polymer Light Technology Corp. Tin chloride dihydrate (SnCl₂·2H₂O, 99.5%) and 4-(2-thioureido)benzoic acid (TB, 99.99%) were supplied by Shanghai Aladdin Biochemical Technology Co., Ltd. Cesium iodide (Csl, 99.9%), lead bromide (PbBr₂, 99.99%), lead iodide (Pbl₂, 99.99%), and dimethyl sulfoxide (DMSO, 99.9%) were received from Tokyo Chemical Industry Co., Ltd. Ultrapure water was obtained from Suzhou Industrial Distilled Water. Thiourea (TR, 99.9%) was sourced from Adamas, while thioglycolic acid (TGA, 98.0%) was obtained from J&K Scientific Ltd. Hydrochloric acid (HCl, 37.0 wt.%) was supplied by Guangzhou Chemical Reagent Factory.

Device fabrication

FTO glass substrates were sequentially cleaned using ultrasonic agitation in detergent, deionized water (DI water), acetone, and isopropanol for 20 min per cycle, with the process repeated twice. Afterwards, the substrates were dried using a N₂ air gun and subjected to ultraviolet ozone (UV-O₃) treatment for 15 min before use. SnO₂ electron transport layer was fabricated using a chemical bath deposition method¹. Tin chloride dihydrate (SnCl₂·2H₂O, 687.5 mg), TR (3125.0 mg), TGA (62.5 μ L), HCl (3.125 mL), and ultrapure water (250 mL) were combined in a plastic dyeing tank and stirred thoroughly until completely dissolved and homogeneously mixed. FTO substrates were secured onto slide carriers using high-temperature polyimide tape and positioned in a holder for reaction at 90 °C for 2.5 h. Following the reaction, the SnO₂-coated FTO substrates were retrieved and subjected to ultrasonic cleaning in deionized water and isopropanol for 5 min per cycle, repeated three times. Finally, the FTO/SnO₂ substrates were annealed at 170 °C for 60 min to complete the process.

ZnO colloidal solution was spin-coated onto UV-O₃-treated FTO/SnO₂ substrates at 3500 rpm with an acceleration of 1000 rpm for 45 s. This was followed by annealing at 150 °C for 30 min. The prepared FTO/SnO₂/ZnO substrates were then transferred to a nitrogen-filled glove box for subsequent processing. A 1.2 M CsPbl₂Br precursor solution was prepared by dissolving CsI (312 mg), Pbl₂ (277 mg), and PbBr₂ (220 mg) in 1 mL of DMSO. Modified CsPbl₂Br precursor solutions containing varying concentrations of TB were prepared (denoted as X-TB-CsPbl₂Br, where X = 0, 0.08 mmol%, 0.10 mmol%, 0.12 mmol%, and 0.14 mmol%) by incorporating TB into the perovskite precursor solution at the specified molar concentrations. The precursor solutions and FTO/SnO₂/ZnO substrates were preheated to 45 °C for 10 min. Subsequently, precursor solutions containing different TB concentrations were deposited onto the substrates and spin-coated in two steps: 500 rpm for 10 s, followed by 3000 rpm for 40 s. After spin-coating, the samples underwent a two-step annealing process: first at 45 °C for 1 min, followed by 200 °C for 6 min. Commercial carbon paste was blade-coated onto the perovskite films to prepare the carbon electrodes and then dried at 120 °C for 15 min, resulting in the formation of the carbon electrodes. **Characterization**

Morphology was examined using scanning electron microscopy (SEM, SU8220, Japan). X-ray diffraction (XRD, Ultima-IV, Japan) with Cu K α radiation (λ = 0.15418 nm) was employed to analyze the crystal structure. Absorption spectra were measured using a spectrophotometer (UV-3600 Plus, China). Photoluminescence (PL) spectra were recorded using a Hitachi F-7000 PL spectrophotometer. PL decay curve was measured with an Edinburgh FLS920 luminescence spectrophotometer. Scanning Kelvin probe microscopy (SKPM) measurements were performed using an atomic force microscope (Dimension FastScan, USA). X-ray photoelectron spectroscopy (XPS) was conducted using an Escalab 250Xi system equipped with an Al anode, operating at 15 kV and 15 mA (250Xi, England). The binding energy of the C 1s peak at 284.8 eV was utilized as an internal standard. Under AM 1.5G solar illumination at 100 mW cm⁻², the current density-voltage (J-V) and capacitance-voltage (C-V) characteristics were measured using a solar simulator (Oriel 94043A, American) and an electrochemical workstation (CHI660, China), respectively. The active cell area was defined using a black metal mask with an area of 0.09 cm². The incident photo-to-electron conversion efficiency (IPCE) spectra of the various devices were measured using an IPCE test system (SCS 100, China). ¹H NMR spectra were recorded on a Bruker AVANCE 400 M spectrometer, with tetramethylsilane as the internal reference. Contact angle analysis was conducted with an optical goniometer (JC2000D1, POWEREACH, China). Fourier transform infrared (FT-IR) spectra were obtained as KBr pellets on a Nicolet iS10 FT-IR spectrometer.



Figure S1. XPS spectra of (a)Cs 3d; (b) I 3d; (c) Br 3d; and (d) FT-IR spectra spectra of pure TB and TB-CsPbl₂Br.

As shown in Figure S1d, for pure TB, the peak at 3342 cm⁻¹ corresponds to the N-H stretching vibration in the amino group (-NH₂), while the peak at 3172 cm⁻¹ corresponds to the O-H stretching vibration in the carboxyl group. The peak at 1301 cm⁻¹ is characteristic of C-O stretching vibrations, and the peak at 1682 cm⁻¹ corresponds to the C=O stretching vibration in the carboxyl group. The peak at 1060 cm⁻¹ is attributed to the C=S stretching vibration in the thiourea group. Upon the addition of CsPbl₂Br, the -OH and C=O stretching vibration peaks shift to 3282 cm⁻¹ and 1272 cm⁻¹, respectively. Additionally, the intensity of the O-H stretching vibration decreases, and the C-O peak broadens, further supporting the hydrogen bonding interaction between -OH and Br⁻, which effectively reduces Br⁻ vacancies. Meanwhile, the C=S and C=O stretching vibration peaks shift to 1694 cm⁻¹ and 1010 cm⁻¹, with notable intensity changes, indicating that the C=S and C=O groups interact with CsPbl₂Br through coordination, which effectively passivates Pb²⁺ defects.



Figure S2. XRD spectra of CsPbl₂Br with different TB concentrations: (a) 10-50 °; (b) 25-35 °.



Figure S3. The trend of the film changes with annealing time after spin-coating the precursor solutions of Pure CsPbl₂Br and TB-CsPbl₂Br



Figure S4. SEM images of TB-CsPbl₂Br with varying TB concentrations: (a) 0.08 mmol% TB; (b) 0.12 mmol% TB; (c) 0.14 mmol% TB.



Figure S5. pure CsPbl₂Br and TB- CsPbl₂Br with different TB concentrations of (a)UV-visible absorption spectra; (b) PL spectra on an FTO/SnO₂/ZnO substrate; and (c) on a glass substrate.



Figure S6 (a) J-V curves of CsPbl₂Br solar cells device at varying TB concentrations; (b) EQE spectra of the Pure CsPbl₂Br and TB -CsPbl₂Br solar cells; (c) the dependence of J_{SC} on light intensity for and pure CsPbl₂Br and TB -CsPbl₂Br devices; (d) the dependence of Voc on light intensity for and pure CsPbl₂Br and TB -CsPbl₂Br devices.



Figure S7. XPS spectra of S 2p for pure $CsPbl_2Br$ and 0.10 mmol%-TB-CsPbl_2Br



Figure S8 water contact angle analysis for (a) pure CsPbl₂Br, (b) TB-TPT-CsPbl₂Br.



Figure S9. Normalized PCE decay curves under 365 nm UV-light for the pure CsPbl₂Br and TB-CsPbl₂Br devices.



Figure S10. Boxplot distribution of 30 devices for the pure CsPbl₂Br and TB-CsPbl₂Br solar cells device, illustrating: (a) V_{oc}, (b) J_{sc}, (c) FF, (d) PCE.

The data were analyzed using a bi-exponential fitting model to calculate the average lifetime (τ_{ave}) based on the following Eq. (S1)²:

$$\tau_{ave} = \frac{A_1 \tau_1^2 + A_2 \tau_2^2}{A_1 \tau_1 + A_2 \tau_2} # # # # # # # # # (S1)$$

In this equation, A_1 and A_2 denote the decay amplitudes, while τ_1 and τ_2 correspond to the short and long lifetimes, which are attributed to grain boundary trap-assisted recombination and intra-grain radiative recombination, respectively.

The exponent factors (α) were subsequently calculated using the defect density Eq. (S2)²:

 $J_{sc} = a \times I_{light}^{\alpha} #$ (S2)

In this equation, J_{sc} is the photo-current of the CsPbl₂Br device at the corresponding light intensity, a is the slope, I_{light} is the light intensity relative to the 1.5 AM standard, and α is the ideal factor.

The ideal diode ideality factors (n_{id}) were subsequently calculated using the defect density Eq. $(S3)^2$:

$$n_{id} = \frac{q}{kt} \times \frac{dV_{oc}}{dln(L)} \# \# (S3)$$

In this equation, L is the normalized light intensity, K is the Boltzmann constant, T is the temperature, and q is the elementary charge.

The defect density was subsequently calculated using the defect density Eq. (S4)³:

$$N_{t} = \frac{2\varepsilon_{r}\varepsilon_{0}V_{FTL}}{eL^{2}} # # # # # # (S4)$$

In this equation, ε_r is the relative dielectric constant ($\varepsilon_r = 8.5$), ε_0 is the vacuum permittivity, V_{FTL} is the voltage trapping-limited voltage ($\varepsilon_0 = 8.85 \times 10^{-14} \text{ F/m}$), e is the elementary charge (e = $1.60 \times 10^{-19} \text{ C}$), and L is the thickness of the film (~500 nm, Figure S11), with N_t representing the electronic defect density.



Figure S11. Cross-sectional SEM images of (a) CsPbI₂Br and (b) TB-CsPbI₂Br.

Additive ratio (mmol%)	V _{oc} (V)	J _{sc} (mA/cm²)	Fill factor	PCE (%)					
					0	1.18	14.33	71.95	12.18
					0.08	1.19	14.81	74.75	13.19
0.10	1.22	14.96	76.66	13.96					
0.12	1.21	14.68	75.35	13.40					
0.14	1.16	14.52	74.45	12.58					

Table 1. Performance parameters of CsPbl₂Br devices with different TB concentrations, derived from J-V curves

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