Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Mitigating deep-level defects through a self-healing process for highly efficient wide-bandgap inorganic $CsPbI_{3-x}Br_x$ perovskite photovoltaics

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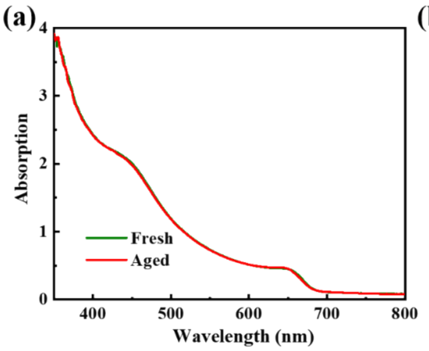

Figure S1 (a) UV-vis absorption spectra and (b) Energy bandgap of the fresh and aged perovskite films.

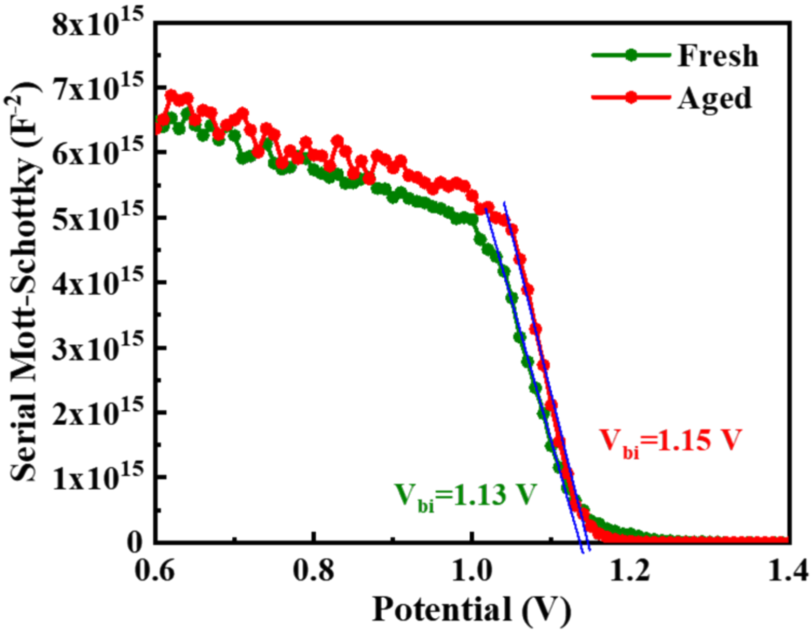

Figure S2 Mott−Schottky analysis for the fresh and aged devices.

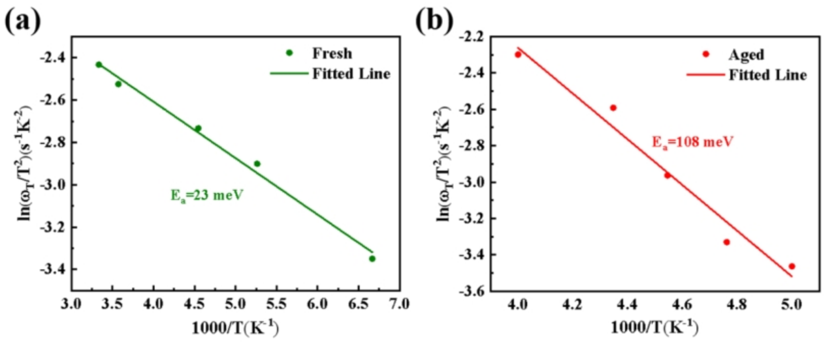

Figure S3 Arrhenius plot extracted from the derivative peak of ω(dC/dω) versus ω plot at low-frequency region for (a) Fresh; (b) Aged devices.

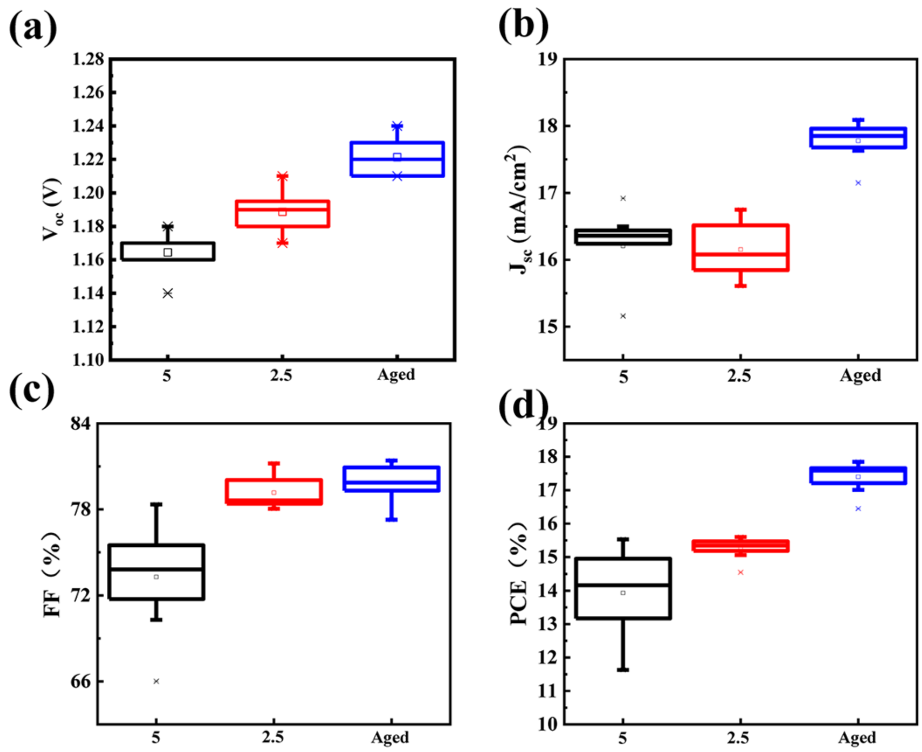

Figure S4. The statistical data of the $CsPbI_{3-x}Br_x$ PSCs with 5 µl, 2.5 µl water as an additive in the inorganic perovskite precursor. The data of the aged devices is given for comparison.(a) V_{oc}; (b) J_{sc}; (c) FF; (d) PCE

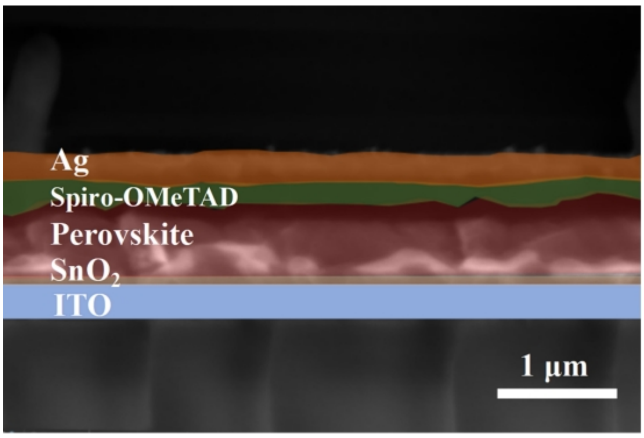

Figure S5 Cross-section SEM image of the $CsPbI_{3-x}Br_x$ PSC.

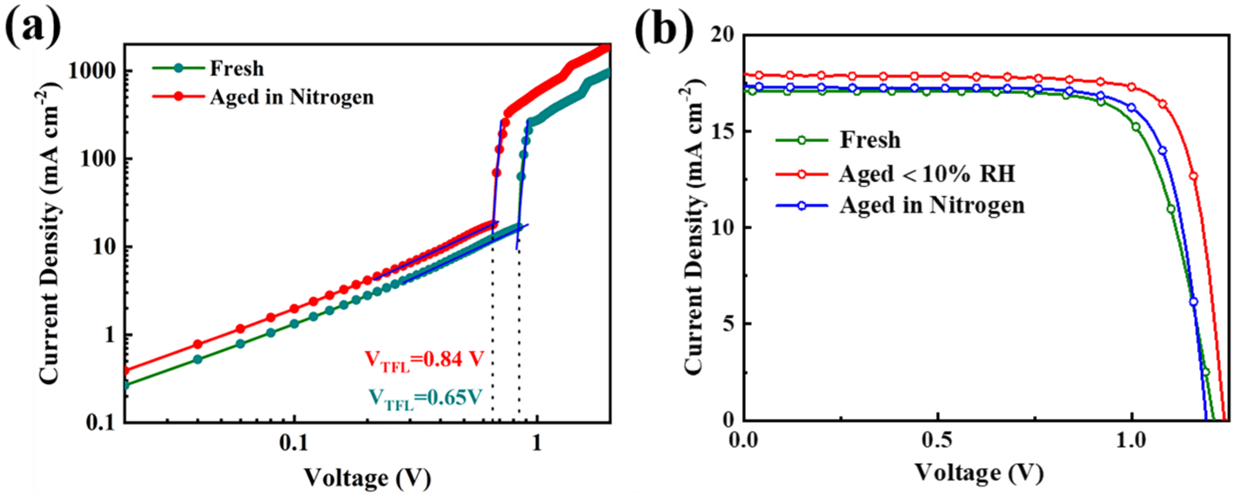

Figure S6. (a) J–V curves of electron-only devices for the fresh and aged film in an inert N_2 gas condition; (b) Typical J-V curves of $CsPbI_{3-x}Br_x$ PSCs aged in a low humidity condition and in an inert N_2 gas condition.

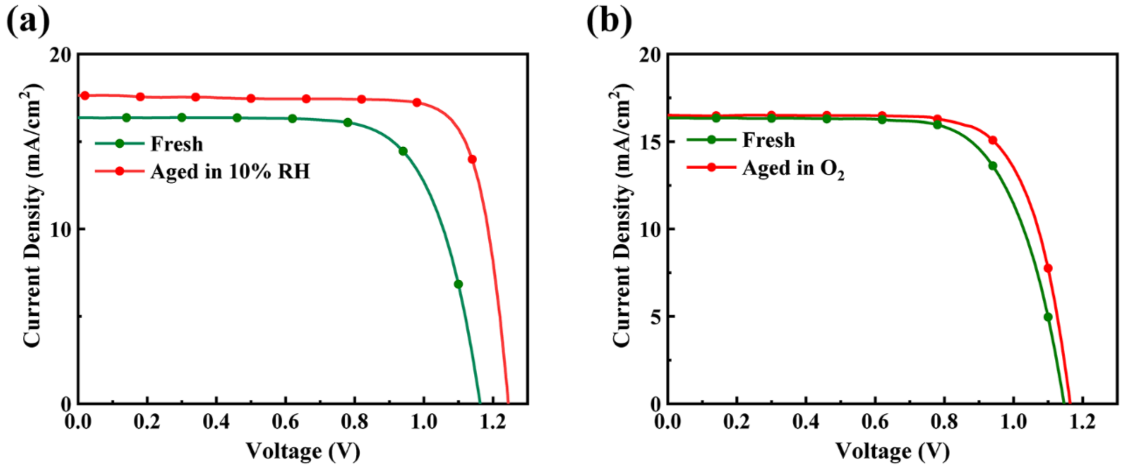

Figure S7. Comparison of the $CsPbI_{3-x}Br_x$ PSCs fabricated with the aging process conducted in different conditions. (a) Aged in 10% RH condition, (b) Aged in O_2 condition.

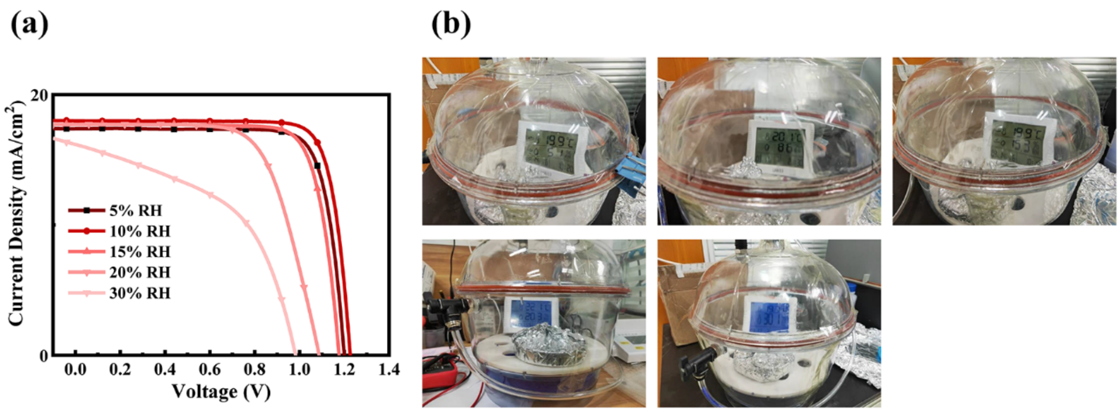

Figure S8. (a) J–V characteristics of the PSCs fabricated from the aging process with different humidity conditions; (b) Photographs of the storage boxes used for controlling the humidity.

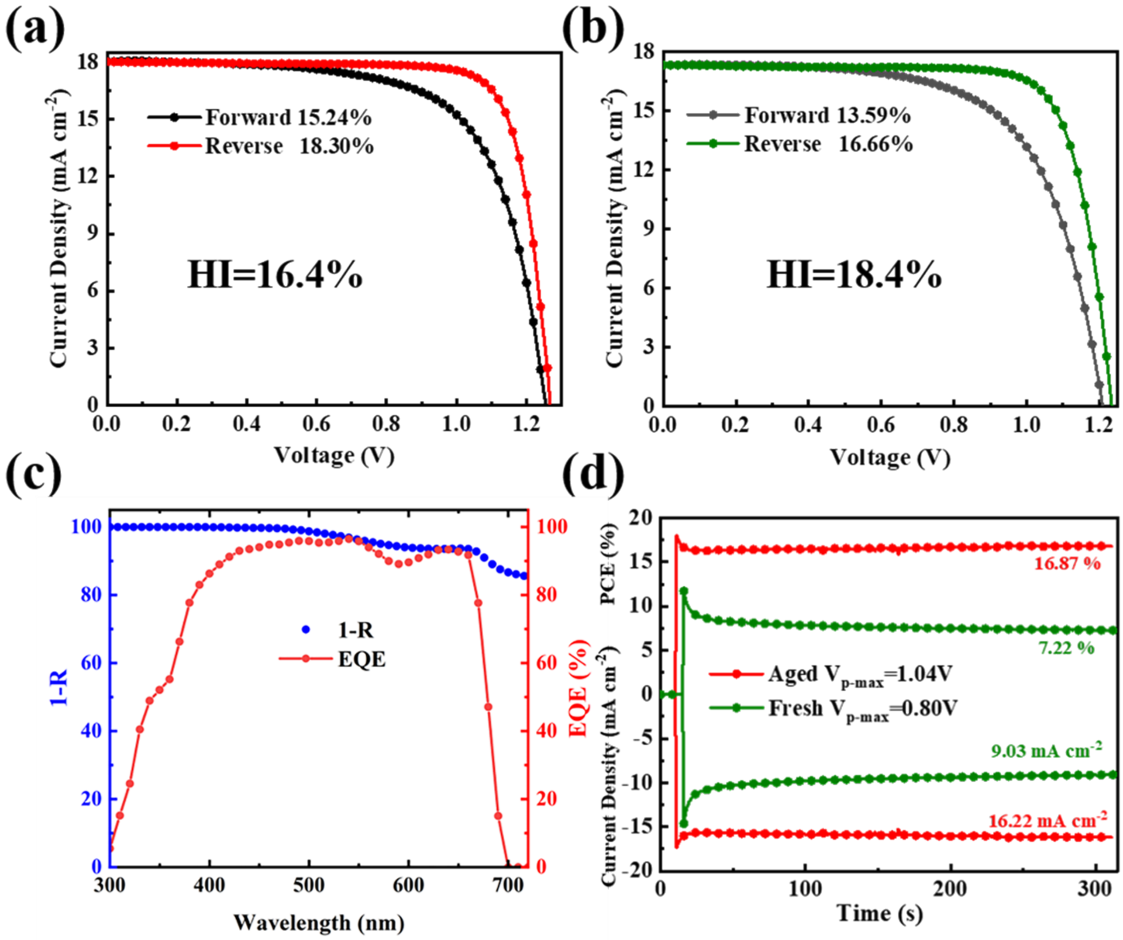

Figure S9 J-V curves of the (a) aged and (b) fresh PSCs measured with different scan directions. HI is defined according to the equation: $HI = (PCE_{Reverse} - PCE_{Forward})/PCE_{Reverse} \times 100\%$; (c) EQE and 1-R curves of the aged PSC. (d) Steady-state output of the fresh and aged PSCs under the voltages at maximum power points of 0.80 V and 1.04 V, respectively.

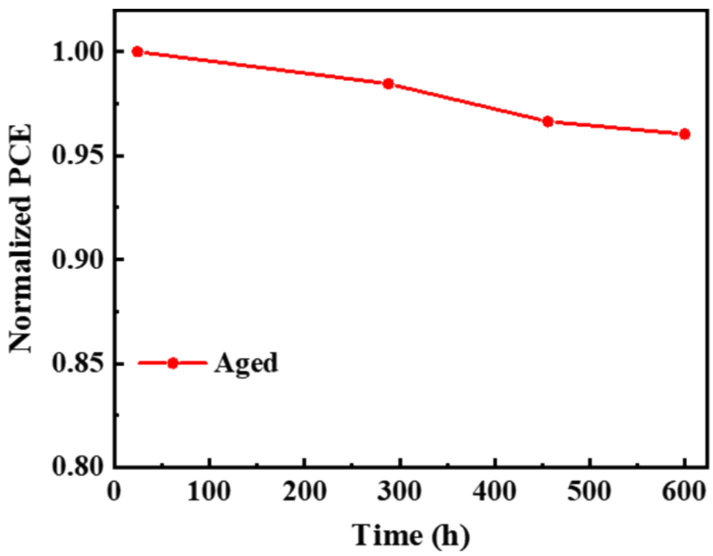

Figure S10 PCE evolution of the unencapsulated device under ~30% RH at room temperature in ambient condition.

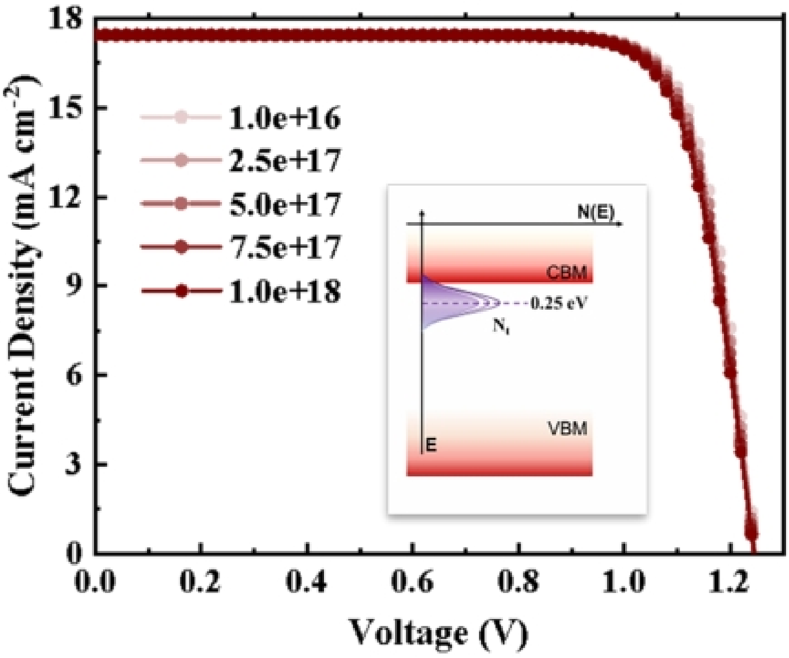

Figure S11 Device simulation using SCAPS for the $CsPbI_{3-x}Br_x$ PSC with different shallow-level defect densities. The E_a of the shallow-level defect is 0.25 eV.

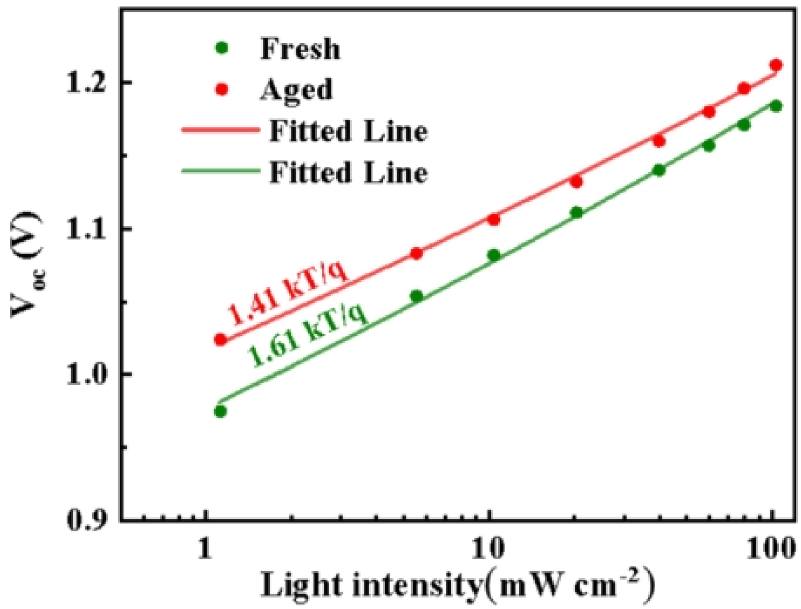

Figure S12 Light-dependent V_{oc} measurment.

Table S1 The device parameters of $CsPbI_{3-x}Br_x$ PSCs fabricated from the aging process with different RH conditions.

Aging condition	Voc(V)	Jsc(mA/cm2)	FF(%)	PCE(%)
RH=5%	1.18	17.38	79.37	16.3
RH=10%	1.23	17.98	80.96	17.85
RH=15%	1.17	17.20	68.09	13.80
RH=20%	1.08	17.70	67.12	12.92
RH=30%	0.98	6.17	49.45	7.86

Table S2 The details of the device structure and semiconductor parameters used for the SCAPS simulation.

Parameters	ITO	SnO_2	$CsPbI_{3-x}Br_x$	Spiro-OMeTAD
Thickness (nm)	60	25	500	100
Bandgap Energy (Eg) eV	3.2	3.4	1.82	3.2
Electron affinity (γ) eV	4	4.2	3.85	2.15
Relative Dielectric Permittivity (ε_r)	9	9	3.6	3
Effective Conduction Band Density (N_c) (cm^{-3})	1×10^{20}	1×10^{20}	8×10^{18}	1×10^{20}
Effective Valence Band Density (N_v) (cm^{-3})	1×10^{20}	1×10^{20}	8×10^{18}	1×10^{20}
Electron thermal velocity (cm s^{-1})	1×10^{7}	1×10^{7}	1×10^{4}	1×10^{7}
Hole thermal velocity (cm s^{-1})	1×10^{7}	1×10^{7}	1×10^{4}	1×10^{7}
Electron mobility (cm2 V^{-1} s^{-1})	0.1	0.1	2	0.1
Hole mobility (cm2 V^{-1} s^{-1})	0.1	0.1	2	0.1