Energy Materials

Supplementary Materials

Precursor aging homogenizes PbI_2 sol-clusters for humidity-tolerant fabrication of perovskite solar cells

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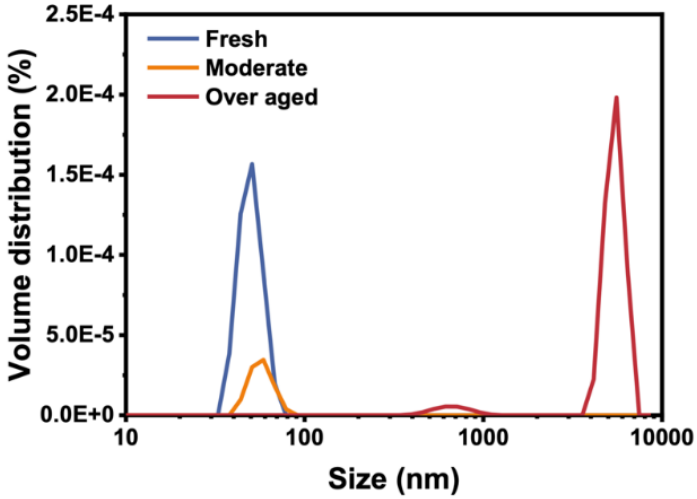
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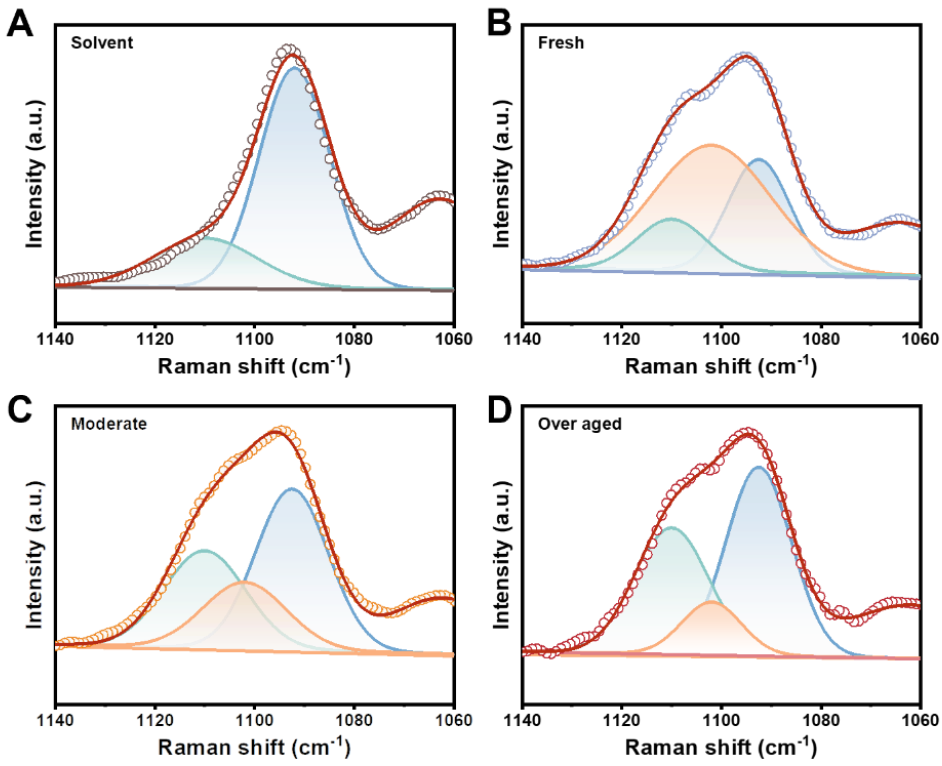
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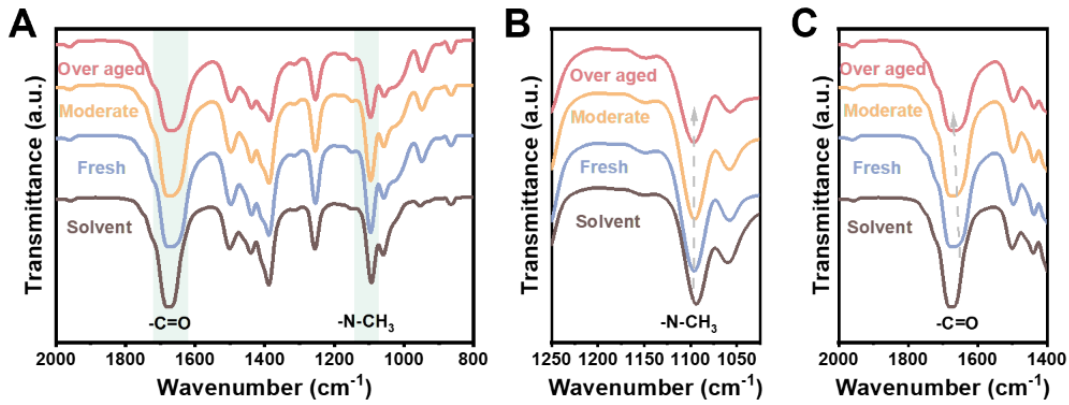
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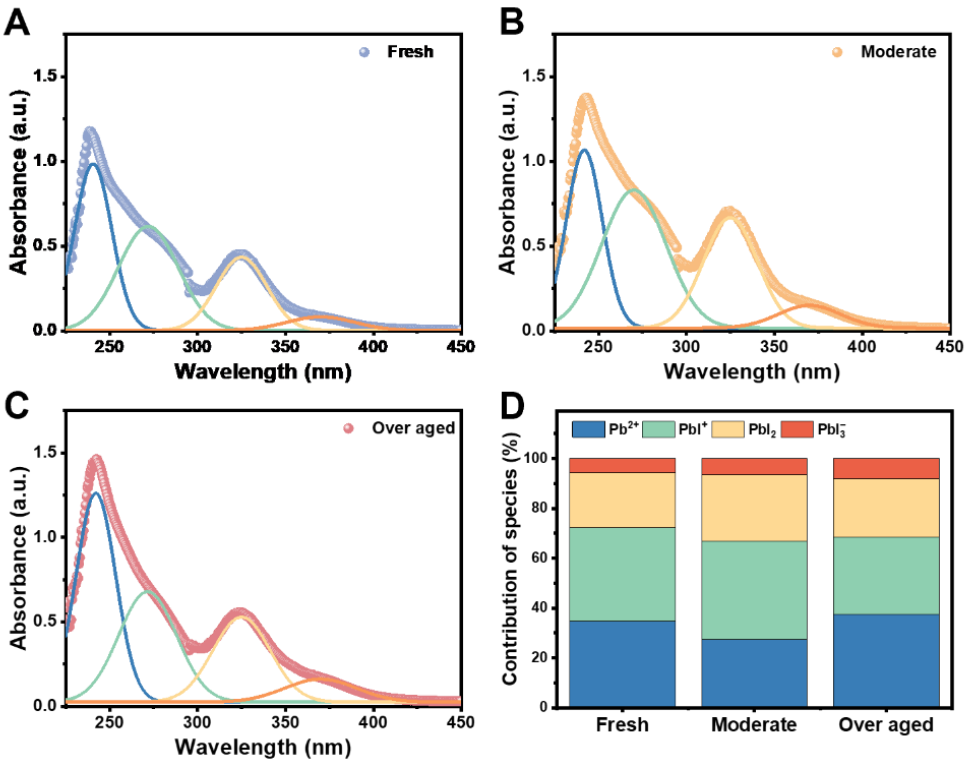
Supplementary Figure 1. Colloid size distributions for the PbI_2 precursor solution with different aging times.



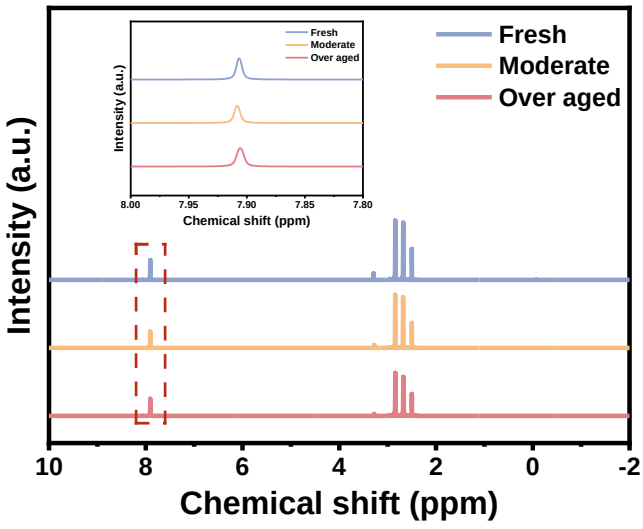
Supplementary Figure 2. Raman spectra of solutions. (A) solvent; (B) fresh, (C) moderate aging, and (D) over aged PbI_2 precursor.



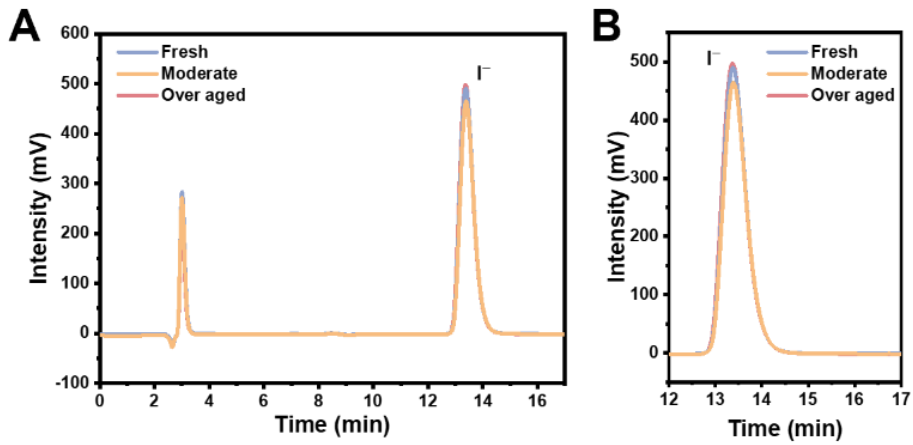
Supplementary Figure 3. FTIR spectra of PbI_2 precursor solutions: (A) full-range spectra and (B, C) enlarged views of selected regions.



Supplementary Figure 4. UV-vis spectra of PbI_2 precursor solutions. The fitting curves of (A) fresh, (B) moderate aging, and (C) over aged PbI_2 precursor; (D) relative contribution distributions of iodoplumbates.



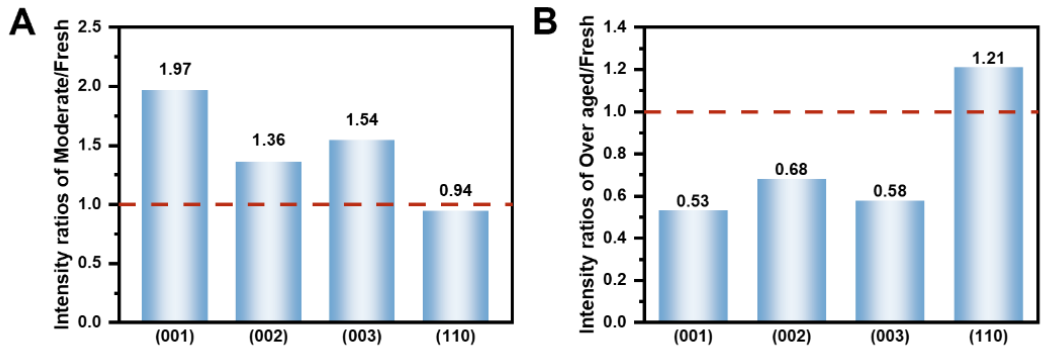
Supplementary Figure 5. ^{1}H NMR spectra of PbI_2 precursor solutions.



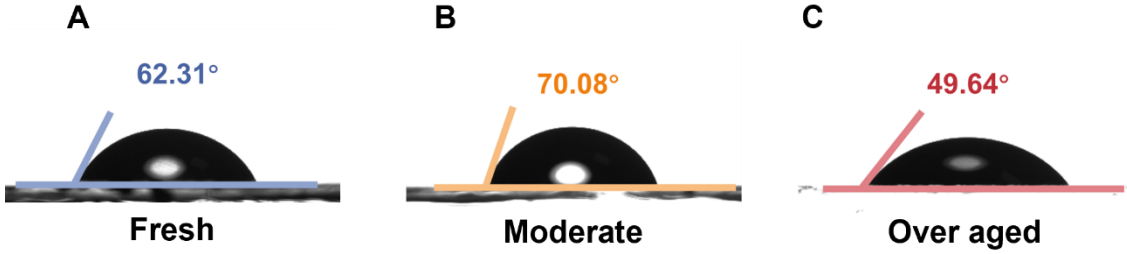
Supplementary Figure 6. Potential signals in iodide ion chromatography. (A) Original potential signals of iodide ion chromatography and (B) partially magnified signals of I^- peak.



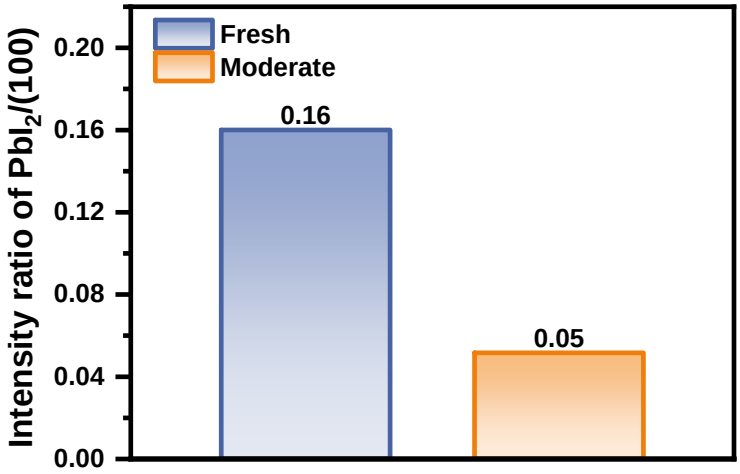
Supplementary Figure 7. Contact angle between PbI_2 precursor solutions and SnO_2 substrates. (A): fresh, (B) moderate aging and (C) over aged PbI_2 precursor.



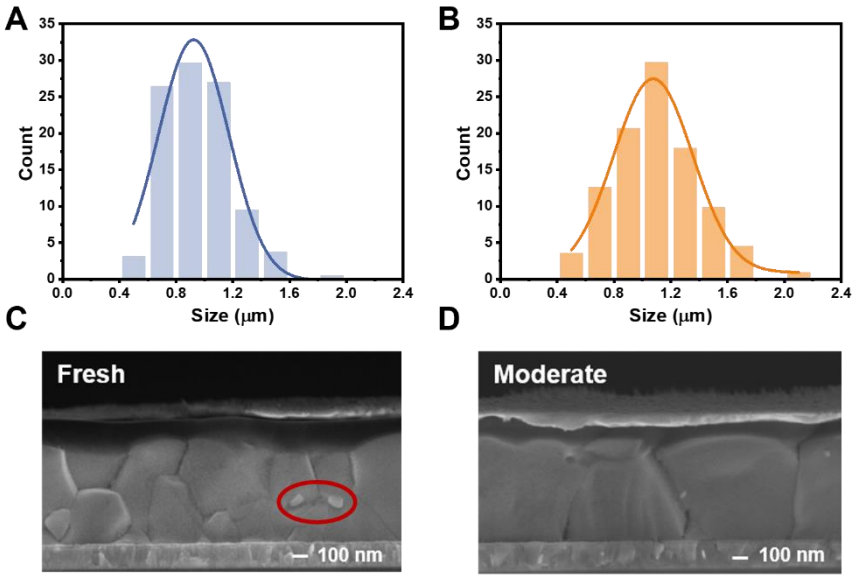
Supplementary Figure 8. The intensity ratios of (001), (002), (003), (004) and (110) peaks of (A) Moderate PbI_2 films to those peaks of Fresh PbI_2 films and (B) Over aged PbI_2 films to those peaks of Fresh PbI_2 films.



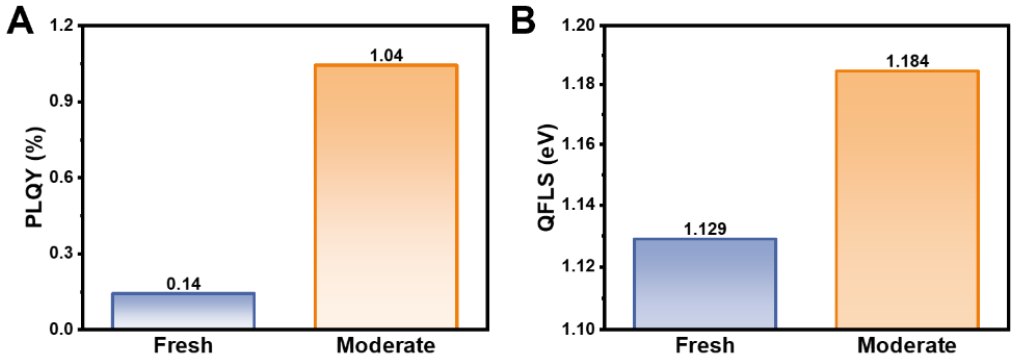
Supplementary Figure 9. Contact angle between PbI_2 films and water drop. (A) fresh, (B) moderate aging and (C) over aged PbI_2 precursor.



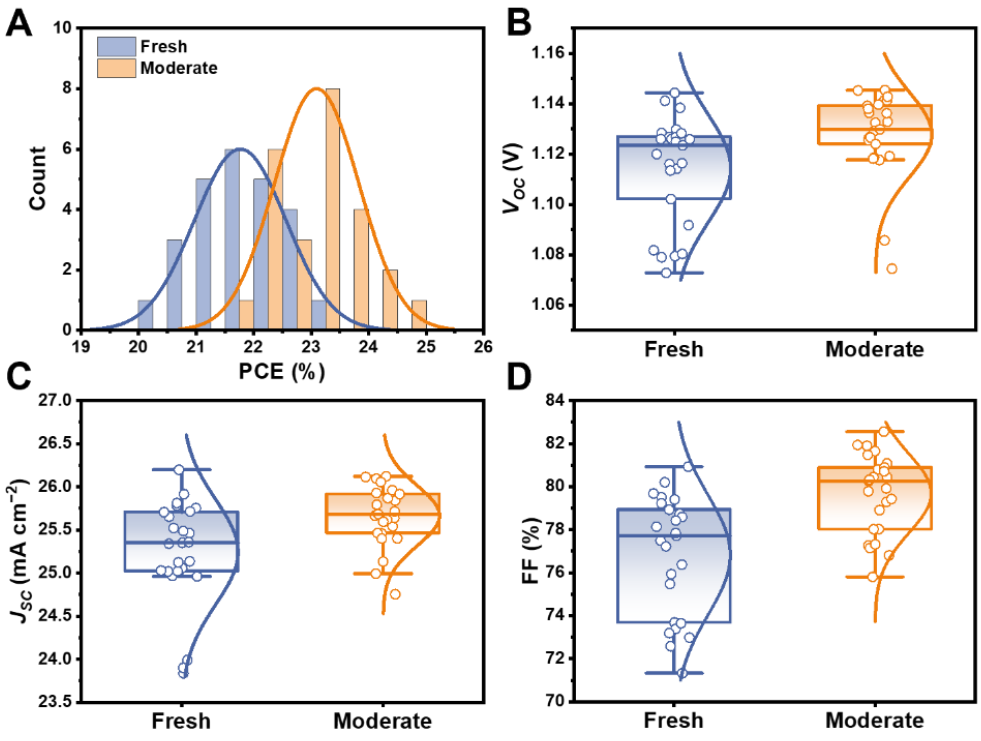
Supplementary Figure 10. The intensity ratio of residual PbI_2 peak to (100) peak of perovskite films.



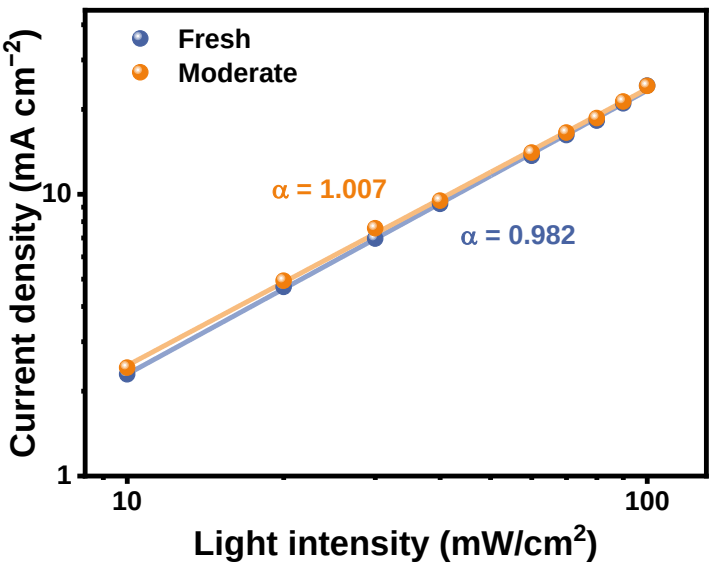
Supplementary Figure 11. SEM analysis of perovskite films. The grain size distribution of perovskite films for the (A) fresh and (B) moderately aged sample; Cross-sectional SEM images of PSCs fabricated by (C) fresh and (D) moderately aged PbI_2. The red circle highlights small grains in the fresh perovskite film, which are tentatively attributed to residual PbI_2.



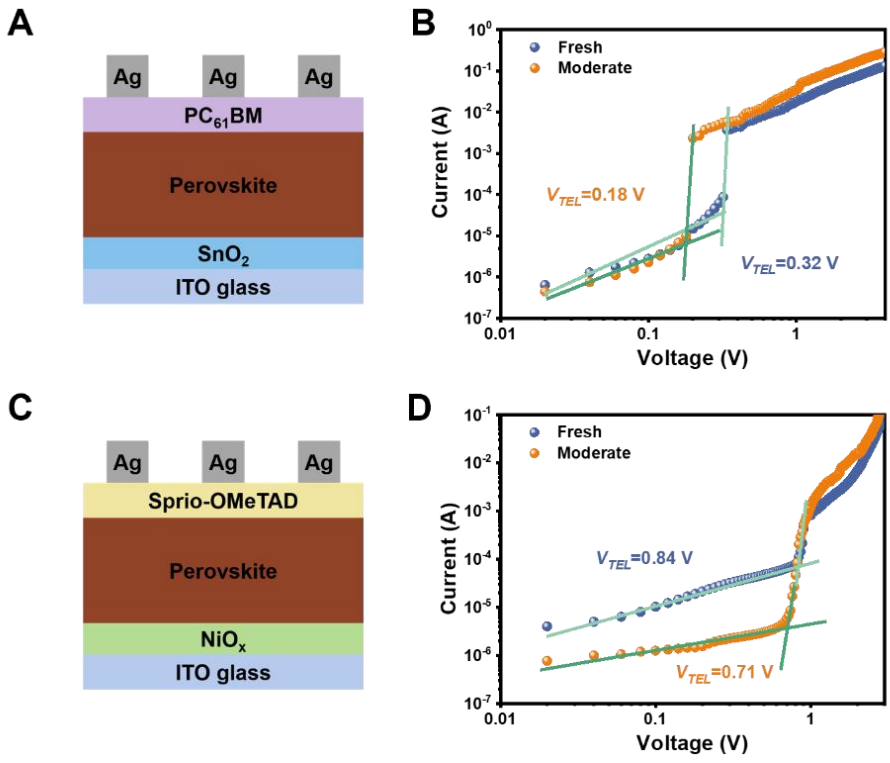
Supplementary Figure 12. (A) PLQY and (B) corresponding QFLS of perovskite films.



Supplementary Figure 13. The distribution of (A) PCE, (B) V_{OC}, (C) J_{SC} and (D) FF for 25 PSCs based on fresh and moderately aged PbI_2 solution.



Supplementary Figure 14. Evolution of J_{SC} as a function of light intensity. The dependency of J_{SC} on light intensity for the control and target devices.



Supplementary Figure 15. Device architectures and SCLC measurements. Schematic structures of (A) the electron-only and (C) hole-only devices; corresponding SCLC curves of (B) the electron-only and (D) hole-only devices.

Supplementary Table 1. xyY chromaticity coordinate parameters of different PbI_2 precursor solutions

Coordinates	Fresh	Moderate	Over aged
x	0.4238	0.4308	0.4408
y	0.522	0.5146	0.5042
Y	94.3	92.6	89.5

Supplementary Table 2. Relative integrated peak areas of the fitted UV–vis absorption features assigned to different iodoplumbate species in PbI_2 precursor solutions under different aging conditions

Aging time	Pb^{2+} (%)	PbI^{+} (%)	PbI_2 (%)	PbI_3^{-} (%)
Fresh	34.78	37.38	22.25	5.59
Moderate	27.32	39.58	26.71	6.39
Over aged	37.51	30.83	23.64	8.02

Supplementary Table 3. FWHM of α-phase crystal plane diffraction peaks in perovskite thin films

Perovskite films	**(100)**	**(200)**
Fresh	0.124	0.176
Moderate	0.091	0.149

Supplementary Table 4. Photovoltaic parameters of champion PSCs

Device	V_{OC} **(V)**	J_{SC} **(mA cm^{-2})**	**FF (%)**	**PCE (%)**	**HI (%)**
Fresh-forward	1.118	25.62	78.02	22.38	2.91%
Fresh-reverse	1.128	25.65	79.50	23.05	
Moderate-forward	1.140	26.12	80.80	24.07	1.84%
Moderate-reverse	1.145	26.12	81.94	24.52	

Supplementary Table 5. Statistical analysis of the photovoltaic parameters

Device	V_{OC} **(V)**	J_{SC} **(mA cm^{-2})**	**FF (%)**	**PCE (%)**
Fresh	1.114±0.021	25.24±0.60	76.82±2.78	21.63±0.81
Moderate	1.128±0.017	25.67±0.35	79.69±1.83	23.10±0.74