Supplementary Information (SI) for Energy & Environmental Science.

Flowing liquid Phase Induces the Crystallization Processes of Cesium Lead Triiodide for 21.85%-Efficiency Solar Cells and Low-Energy Loss

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Experimental procedures

Materials Section:

N, N -Dimethylformamide (DMF, 99.8%), Dimethyl sulfoxide (DMSO, ≥ 99.9%), Chlorobenzene (CB), Li-bis-(trifluoromethanesulfonyl) imide (Li-TFSI), and 4-tert-butylpyridine (TBP, 96%) were purchased from Alfa Aesar. Ammonium formate ($HCOOHNH_4$, 99.0%) was purchased from Aladdin. Cesium iodide (CsI, 99.5%), lead iodide (PbI_2, 99.5%), Dimethylammonium iodine (DMAI), Spiro-OMeTAD (2, 2', 7, 7'-Tetrakis (N, N-di-p-methoxyphenylamine)-9, 9-spirobifluorene) were purchased from Advanced Election Technology Co.,Ltd. All the chemicals were used without further purification.

Functional Layer Preparation Section:

Preparation of Compact TiO_2 layer: TiO_2 is deposited on a glass (25 * 25 mm) substrate coated with fluorine-doped tin oxide (FTO) by chemical bath method. Clean the FTO substrate separately with acetone/ isopropanol /ethanol for 30 minutes. The TiO_2 layer was deposited on FTO glass by immersing itself in 200 mL aqueous solution

containing 4.5 mL $TiCl_4$ at 70 °C. After sedimentation, clean TiO_2 with ultrapure water and then soak it in ethanol, followed by drying with N_2.

Preparation of Precursor Perovskite Solution: 0.6M CsI, 0.6M PbI_2, and 0.6M DMAI were dissolved in the mixture of DMSO and DMF (3:17, v/v). Then the mixture was stirred at 30 °C for more than 8 h to obtain a clear, light-yellow solution. The target $CsPbI_3$ perovskite solutions with additives were gained by adding $HCOOHNH_4$ (x=0, 3, 5, 8 mg) and then stirred at room temperature for 12 h. Finally, the solutions were filtering through a 0.46-μm pore PVDF (Polyvinylidene Fluoride) syringe filter before spin-coating.

Preparation of spiro-OMeTAD Solution: 90 mg Spiro-OMeTAD was dissolved in 1 mL chlorobenzene and mixed with 36 μL t-BP and 22 μL Li-TFSI (520 mg/ml in acetonitrile).

Device Assembly Section:

The prepared compact TiO_2 layer was annealed on a hot platform at 200 °C for 30 min and UV for 15 min, and the perovskite precursor solution with and without AFMS additive was spin-coated at 1000 rpm for 10 s and 3000 rpm for 30 s on the prepared compact TiO_2 substrate. Then the substrates were sequentially annealed at 200 °C for 10 min. After that, 40 μL spiro-OMeTAD solution was spin-coated onto the perovskite films at 5000 rpm for 30 s. Finally, an ~ 80-nm thick Au was deposited on the top of the spiro-OMeTAD layer via thermal evaporation method. The active area of each device is 0.09 cm^2.

Characterization Section

XRD: XRD patterns of the samples were obtained by X-ray Diffraction (XRD; DX-2700) using Cu-Kα radiation (λ =1.5416 Å).

Thermogravimetric analysis (TGA): The TGA were obtained on TA Discovery TGA 550, the temperature varied from 30 to 800 °C at a heating rate of 10 °C min^{-1} in nitrogen atmosphere.

Optical microscope: The nucleation process of perovskite films was observed in situ on a hot table at 200 °C by optical microscope with a lens of 50 times.

SEM and AFM: The top-view and cross-sectional SEM images of the perovskite films were investigated using a HITACHI SU-8020 microscope. The AFM images were acquired using a Veeco Nano Scope IV with a silicon cantilever.

Water contact angles: The Water contact angles were measured using a Data Physics OCA 20.

Absorbance, PL and TR-PL: Absorbance spectra were collected using a Shimadzu UV-3600. The PL (excitation at 510 nm) and TR-PL spectra were measured using a PicoQuant FluoTime 300.

XPS and UPS: The XPS measurements were performed in a VG ESCALAB MK2 system with monochromatized Al Kα radiation at a pressure of $5.0 * 10^{-7}$ Pa. The UPS measurements were carried out using a photoelectron spectrometer (Thermo Scientific, ESCA Lab 250Xi system) with helium gas admitted employing the He-I (21.22 eV) emission line.

NMR: The NMR date was carried using JNM-ECZ400S/L1 with a frequency of 400 MHz.

FTIR: The FTIR spectra were measured by FTIR (VERTEX70, Bruker, Germany).

J-V and EQE: The *J-V* characteristics of the PSCs were performed by using the solar simulator (SS-F5-3A, Enlitech) along with AM 1.5G spectra, which was calibrated by a NREL-traceable KG5-fltered silicon reference cell. All devices measured on a Keithley 2400 Source Meter under ambient conditions. The *J-V* reversed test was scanned from 2.5 to -0.1 V at a rate of 10 mV/s, and the forward test was scanned from -0.1 to 2.5 V at a rate of 10 mV/s. The EQE data were obtained by using the solar cell spectral-response measurement system (QE-R3011, Enlitech).

EIS and C-V: The EIS measurements were performed using a Zahner Zennium electrochemical workstation. The C-V measurements were performed using a Zahner Zennium electrochemical workstation.

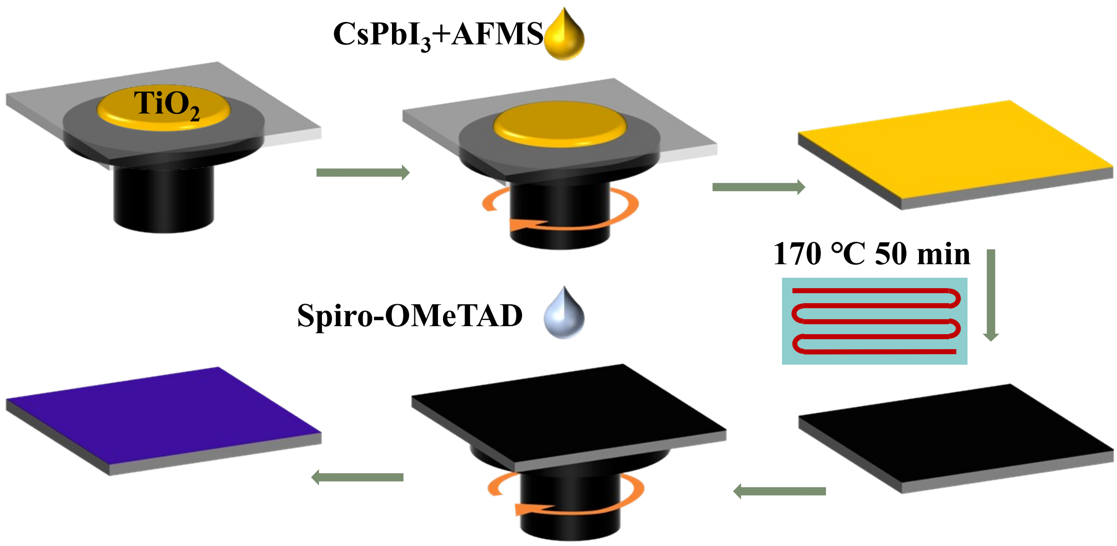

Figure S1. Illustration of the preparation of perovskite films.

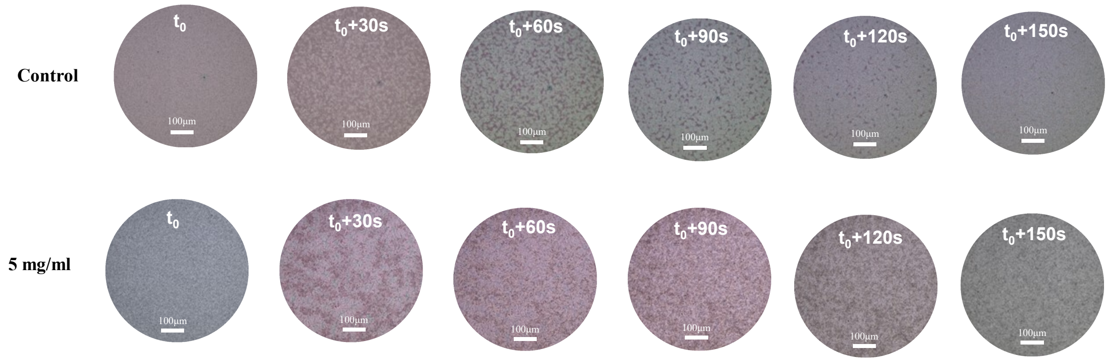

Figure S2. *In situ* microscopy images of the crystallization process of the perovskite system without and with AFMS.

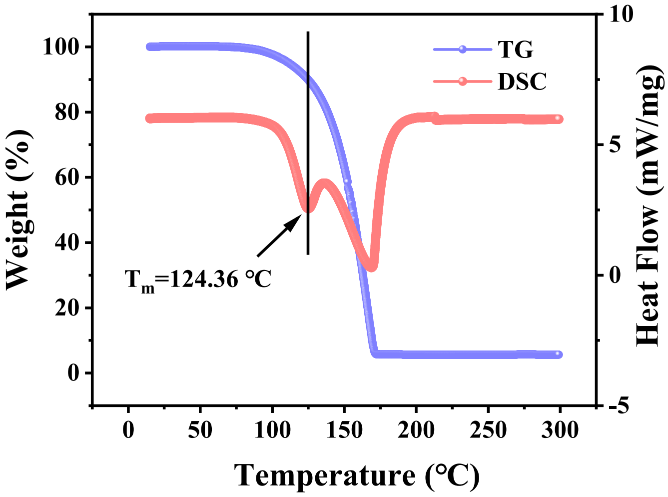

Figure S3. The TGA and DSC curves for AFMS.

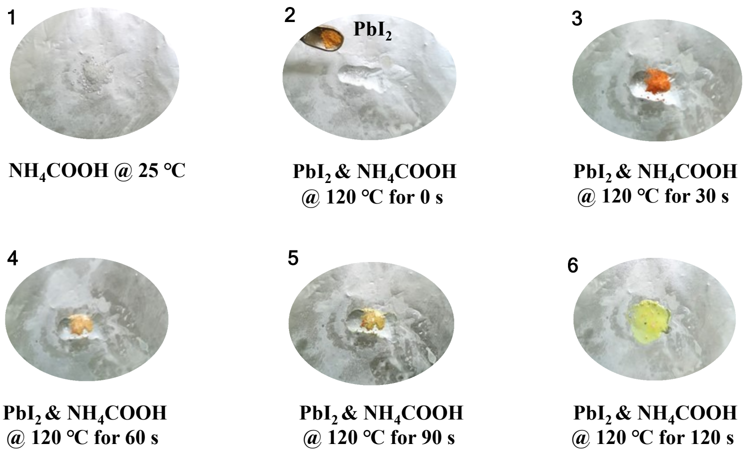

Figure S4. The solution of PbI_2 in NH_4COOH at 120 ℃ with time.

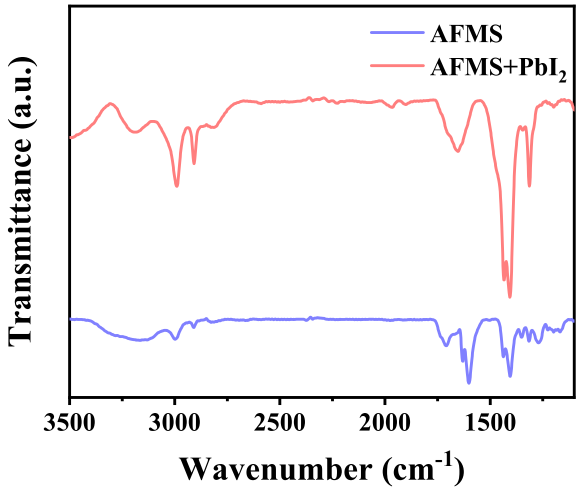

Figure S5. FTIR spectra of AFMS and AFMS with PbI_2.

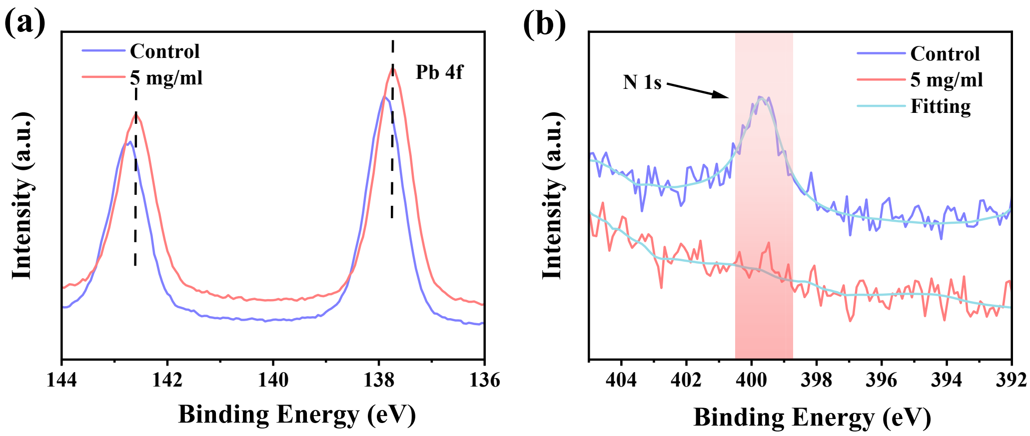

Figure S6. XPS spectra of Pb 4f (a) and N 1s (b) for the $CsPbI_3$ and AFMS-$CsPbI_3$ film.

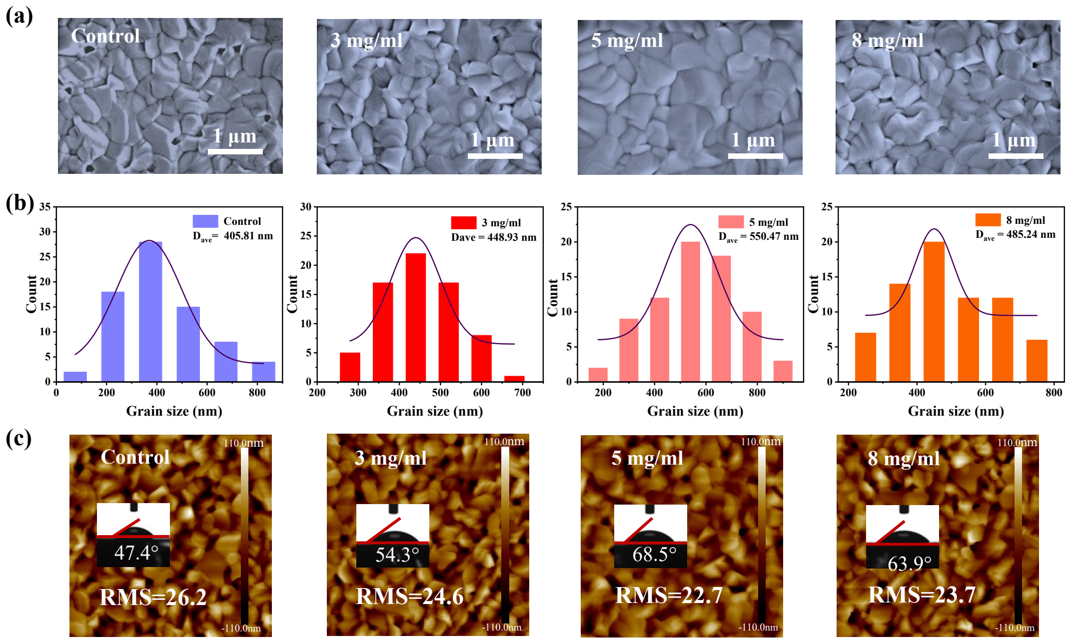

Figure S7. (a)Top view SEM images, (b) Grain size distribution, (c) AFM and Water contact angle image of $CsPbI_3$ films via different concentration of AFMS process.

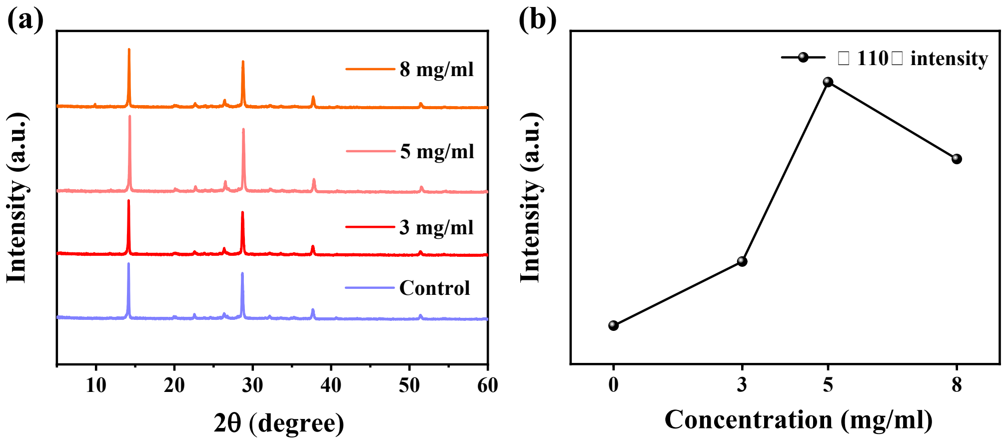

Figure S8. (a). XRD spectra of perovskite films without and with treatment by different concentrations of AFMS (b). The variation of the (110) peak intensity of perovskite as a function of AFMS concentration.

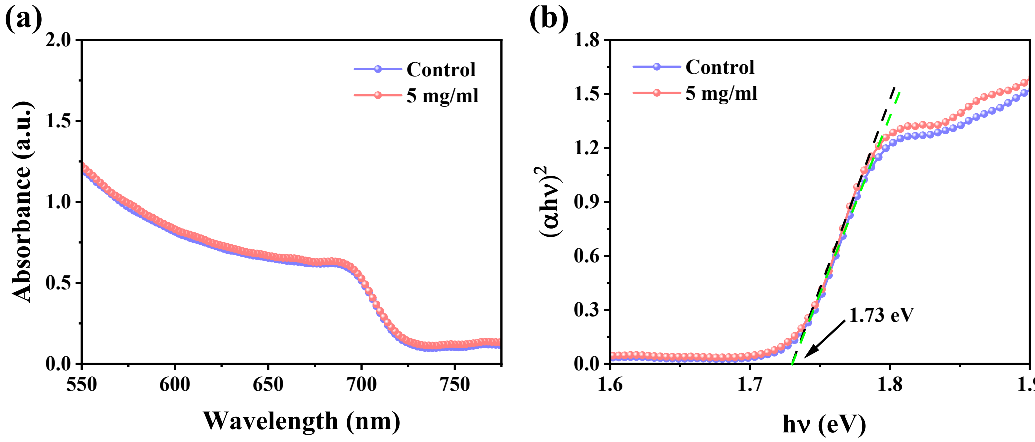

Figure S9. UV-vis absorption spectra (a) and Tauc plots (b) of $CsPbI_3$ films via 5 mg/ml AFMS.

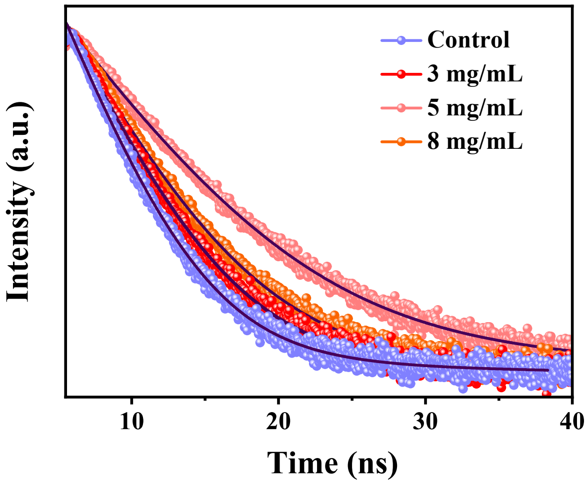

Figure S10. TRPL spectra of $CsPbI_3$ films via different AFMS concentration.

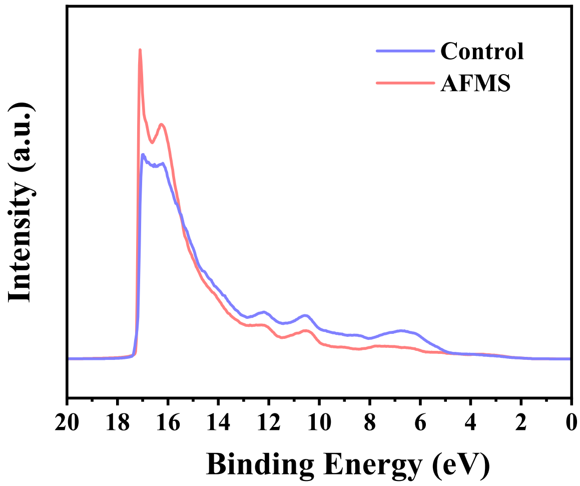

Figure S11. The full UPS spectra of the $CsPbI_3$ and AFMS-$CsPbI_3$ film.

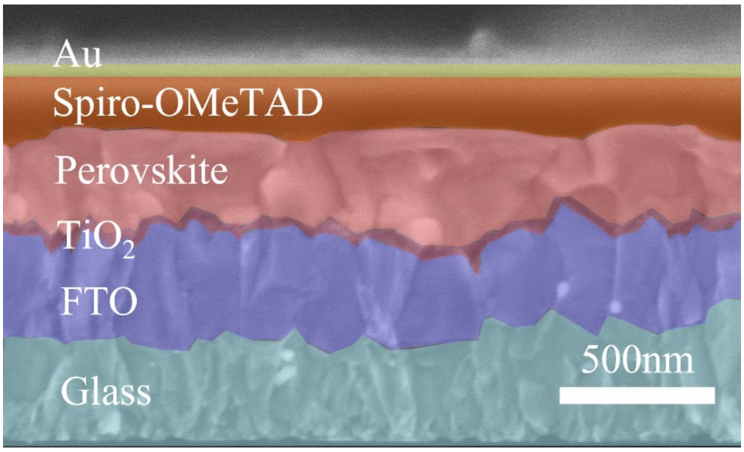

Figure S12. Cross-sectional SEM images of the $CsPbI_3$ PSCs.

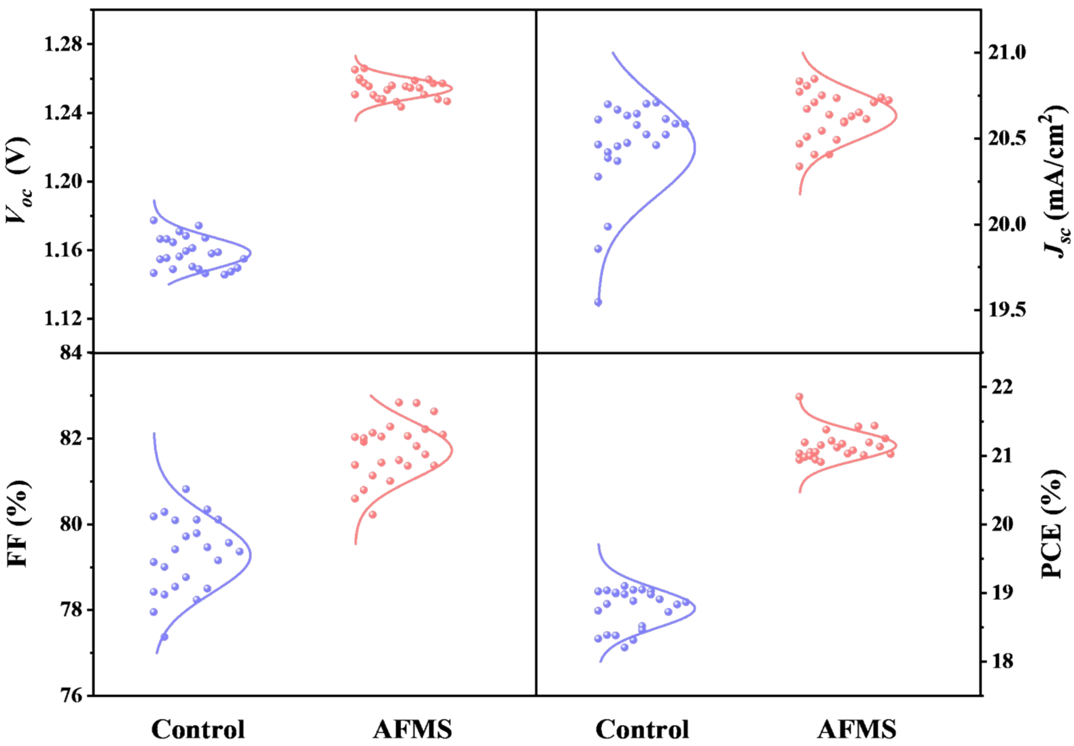

Figure S13. Statistical comparison of *J*-*V* parameters of $CsPbI_3$ perovskite solar cells prepared without or with AFMS process with different content.

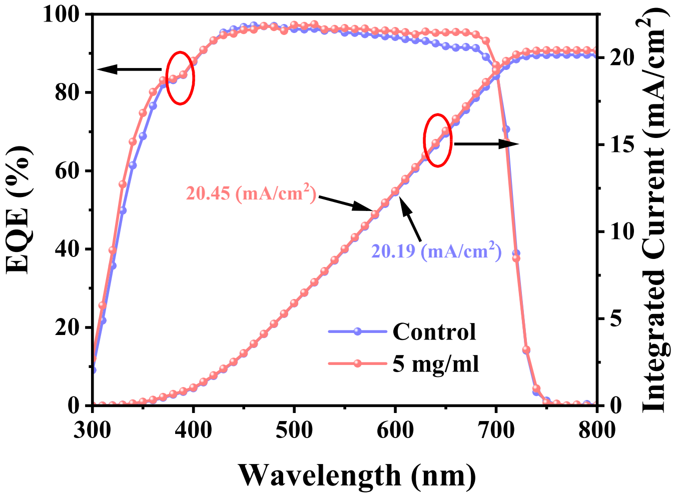

Figure S14. Wavelength-dependent EQE spectra, and integrated current density.

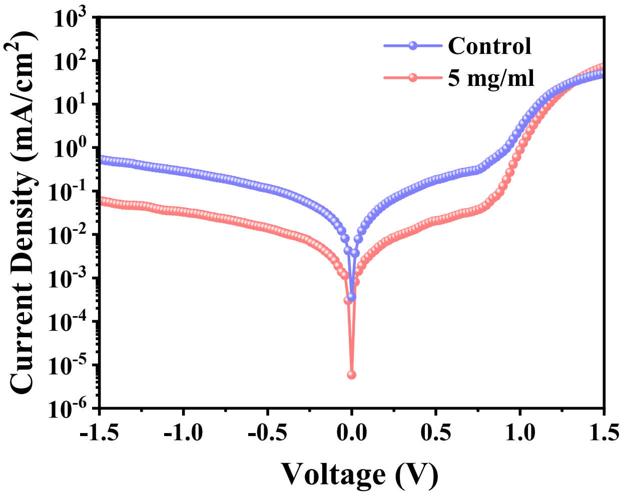

Figure S15. Dark *J-V* curves of the $CsPbI_3$ and AFMS-$CsPbI_3$ perovskite solar cells.

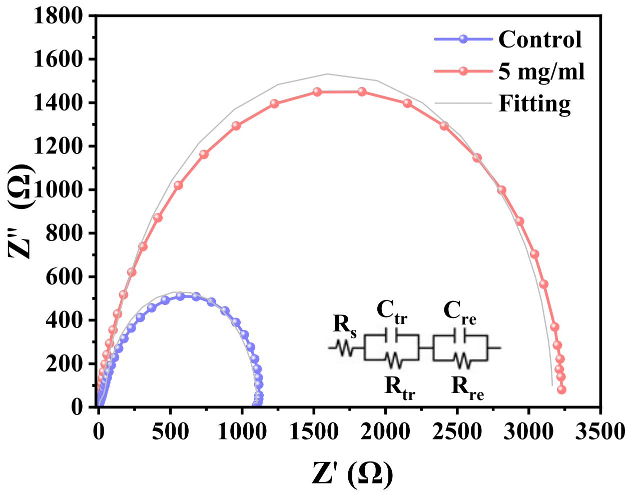

Figure S16. Nyquist plots of the $CsPbI_3$ and AFMS-$CsPbI_3$ perovskite solar cells.

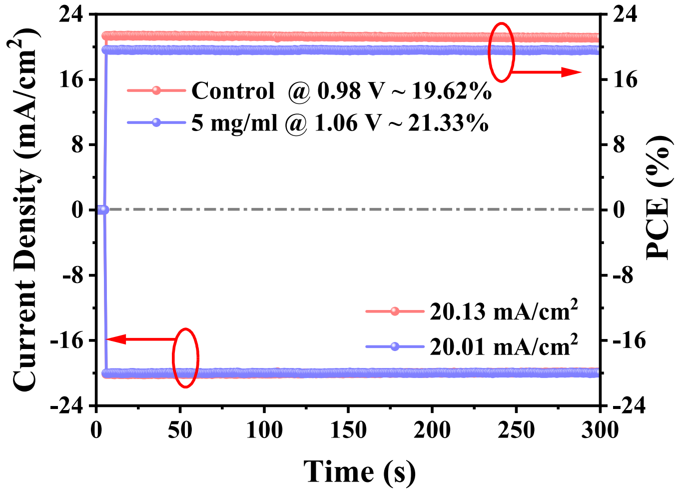

Figure S17. Stabilized photocurrent and SPO at the MPP voltage.

Table S1. Parameters of the TRPL spectra for pristine and AFMS $CsPbI_3$ film.

Sample	τ_{ave} (ns)	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)
Pristine	17.45	53.59	22.71	6.839	77.28
5 mg/mL	54.12	104.38	47.75	8.182	52.25

Table S2. Parameters of the UPS spectra for pristine and AFMS $CsPbI_3$ film.

Sample	$E_{cut\text{-}off}$ (eV)	E_V-E_F (eV)	E_F (eV)	E_V (eV)	E_C (eV)
Pristine	17.22	1.46	4.01	5.47	3.77
5 mg/mL	17.25	1.40	3.97	5.37	3.67

Table S3. Statistical average values of various parameters for 25 devices with and without added AFMS.

Average	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
Pristine	1.15	20.44	79.27	18.77
AFMS	1.25	20.63	81.72	21.14

Table S4. EIS fitting parameters for the $CsPbI_3$ films with and without AFMS process.

Sample	R_s (Ω)	R_{tr} (Ω)	C_{tr} (F)	R_{rec} (Ω)	C_{rec} (F)
Pristine	2.512	39.93	1.21×10^{-7}	1061	2.23×10^{-7}
5 mg/mL	1.833	113.9	1.35×10^{-7}	3189	1.98×10^{-7}

Table S5. Summaries of V_{oc} deficits of $CsPbX_3$: perovskite solar cells fabricated with diffierent configuration and defect passivation method in recent years.

Configuration	Eg (eV)	PCE (%)	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	Eloss (eV)	Ref.
FTO/TiO_2/$CsPbI_3$/Carbon	1.67	15.23	1.121	18.30	74.2	0.549	1
ITO/P3HT-N/ $CsPbI_3$/PCBM/C_{60}/BCP/Ag	1.68	18.93	1.176	20.10	80.04	0.504	2
FTO/c-TiO_2/ $CsPbI_3$/spiro/Au	1.73	20.04	1.171	20.48	83.40	0.559	3
ITO/PTAA/ $CsPbI_3$/PCBM/BCP/Ag	1.745	15.00	1.09	17.33	79.41	0.655	4
FTO/TiO_2/ $CsPbI_3$/spiro/Au	1.70	20.06	1.197	20.64	81.19	0.503	5
FTO/TiO_2/ $CsPbI_xBr_{3-x}$ /spiro/Au	1.72	20.06	1.192	20.32	82.8	0.528	6
FTO/TiO_2/ $CsPbI_3$/spiro/Au	1.68	20.10	1.18	20.80	82.0	0.5	7
FTO/TiO_2/ $CsPbI_3$/spiro/Au	1.68	20.1	1.18	20.4	83.5	0.5	8
FTO/TiO_2/ $CsPbI_3$/ Carbon	1.68	16.642	1.117	18.81	79.2	0.563	9
FTO/TiO_2/ $CsPbI_{2.80}Br_{0.2}$/spiro/Au	1.73	20.8	1.21	20.64	83.2	0.52	10
FTO/TiO_2/ $CsPbI_3$/Carbon	1.7	15.35	1.075	18.7	76.4	0.625	11
FTO/SnO_2/TBAAc/ $CsPbI_3$/ Carbon	1.73	12.79	1.08	17.48	67.8	0.65	12
FTO/c-TiO_2/ $CsPbI_3$/spiro/Au	1.72	20.01	1.17	20.6	83.0	0.55	13
FTO/c-TiO_2/ $CsPbI_3$/spiro/Au	1.7	21.0	1.195	20.86	84.1	0.505	14
FTO/TiO_2/ $CsPbI_3$/spiro/Au	1.7	19.18	1.118	20.77	82.58	0.582	15
ITO/TiO_2/$CsPbI_{2.81}Br_{0.19}$/spiro/Au	1.788	14.25	1.09	18.47	70.67	0.698	16
FTO/MX-TiO_2/ $CsPbI_2Br$/carbon	1.90	11.223	1.313	11.818	72.32	0.587	17
FTO/TiO_2/$CsPbI_{2.85}Br_{0.15}$/TFA/spiro/Au	1.742	21.35	1.239	20.74	83.07	0.503	18
FTO/TiO_2/ $CsPbI_3$/quasi-2D/2D/spiro /Au	1.71	19.06	1.13	20.28	82.65	0.58	19
FTO/TiO_2/ $CsPbI_3$-AC/spiro/Au	1.672	19.04	1.13	20.69	80.79	0.542	20
FTO/TiO_2/ $CsPbI_3$/spiro/Au	1.73	20.26	1.22	20.51	81.0	0.51	21

FTO/TiO_2/ BTABr-$CsPbI_3$/spiro/Au	1.725	21.31	1.20	20.81	85.4	0.525	22
FTO/TiO_2/PFPA/$CsPbI_2Br$ / Carbon	1.92	14.15	1.21	14.80	79.07	0.71	23
ITO/SnO_2/$CsPbI_3$/MD/MoO_x/Ag	1.73	10.33	1.12	14.97	61.30	0.61	24
FTO/ TiO_2/ $CsPbI_3$/spiro-OMeTAD/MoO_3/Ag	1.73	17.47	1.06	20.01	82.2	0.67	25
FTO/TiO_2/ $CsPbI_3$/spiro/Au	**1.73**	**21.85**	**1.26**	**20.84**	**82.82**	**0.47**	**Our work**

Table S6. Summary of some key photovoltaic parameters of the PSCs with high-efficiency in recent years.

Configuration	PCE(%)	V_{OC} (V)	J_{SC} (mA/cm^2)	FF(%)	Year[Ref.]
FTO/ TiO_2/MAPyA-$CsPbI_3$ /Spiro/Au	16.67	1.074	20.08	77.32	2020[26]
FTO/ TiO_2/ $CsPbI_3$/ PTAA/Au	16.52	1.10	19.53	77.0	2020[27]
FTO/ SnO_2/ $CsPbI_3$-$Zn(C_6F_5)_2$/ Spiro/Ag	19.00	1.12	20.67	81.98	2020[28]
FTO/ TiO_2/ $CsPbI_3$/ Spiro/Au	16.04	1.09	18.29	80.50	2020[29]
FTO/ c-TiO_2/ $CsPbI_3$/ Spiro/MoO_3/Au	18.27	1.121	20.25	80.5	2020[30]
FTO/ TiO_2/ $CsPbI_3$/ PTAA/Au	18.02	1.125	20.30	78.9	2020[31]
FTO/ TiO_2/ $CsPbI_3$/ PTAA/Au	17.30	1.098	21.15	76.96	2020[32]
FTO/c-TiO_2/ $CsPbI_3$/spiro/Au	20.04	1.171	20.48	83.40	2021[3]
FTO/ TiO_2/ $CsPbI_3$/ Spiro/Au	19.83	1.20	20.20	81.74	2021[33]
FTO/ c-TiO_2/ $CsPbI_3$/ Spiro/Au	20.37	1.198	20.59	82.5	2021[34]
FTO/ c-TiO_2/ $CsPbI_3$/ Spiro/Au	20.04	1.171	20.52	83.4	2021[35]
FTO/ TiO_2/ $CsPbI_3$/ Spiro/Au	20.08	1.148	20.76	84.3	2021[36]
FTO/ TiO_2/ $CsPbI_3$/ Spiro/Au	18.40	1.10	20.9	80.2	2021[37]
FTO/ TiO_2/ $CsPbI_{3-x}Br_x$/ MOPEABr/Spiro/Au	20.31	1.226	20.12	82.35	2021[38]
FTO/ c-TiO_2/ PCBA/$CsPbI_3$/ Spiro/Au	18.71	1.10	20.60	83.0	2021[39]
FTO/TiO_2/ $CsPbI_3$/spiro/Au	20.06	1.197	20.64	81.19	2022[5]
FTO/TiO_2/ $CsPbI_xBr_{3-x}$ /spiro/Au	20.06	1.192	20.32	82.8	2022[6]
FTO/TiO_2/ $CsPbI_3$/spiro/Au	20.10	1.18	20.80	82.0	2022[7]
FTO/ TiO_2/ $CsPbI_3$/ Spiro/Au	20.06	1.197	20.64	81.19	2022[40]
FTO/ c-TiO_2/ $CsPbI_3$/ Spiro/Au	21.14	1.25	20.76	81.6	2022[41]
FTO/ c-TiO_2/ PCBA/$Cs(Pb,Ge)I_3$/ Spiro/Au	19.52	1.165	20.33	82.4	2022[42]
FTO/ TiO_2/ PSM-$CsPbI_3$/ Spiro/Au	20.50	1.20	20.58	82.87	2022[43]
FTO/ SnO_2/ $CsPbI_3$/ Spiro/Ag	20.1	1.18	20.4	83.5	2022[44]
FTO/ c-TiO_2/ $CsPbI_3$/ Spiro/Au	20.01	1.17	20.60	83.0	2022[45]

FTO/ TiO_2/ $CsPbI_3$/ Spiro/Au	19.18	1.118	20.77	82.58	2022[15]
FTO/TiO_2/ $CsPbI_3$/quasi-2D/2D/spiro /Au	19.06	1.13	20.28	82.65	2023[19]
FTO/TiO_2/ $CsPbI_3$/spiro/Au	20.26	1.22	20.51	81.0	2023[21]
FTO/TiO_2/ BTABr-$CsPbI_3$/spiro/Au	21.31	1.20	20.81	85.4	2023[22]
FTO/ TiO_2/ RBITC-$CsPbI_3$ / Spiro/Au	20.95	1.23	20.65	82.44	2023[46]
FTO/ TiO_2/ $CsPbI_3$/ Spiro/Au	20.72	1.221	20.49	82.8	2023[47]
FTO/ TiO_2/ $CsPbI_3$/ Spiro/Au	21.35	1.239	20.74	83.07	2023[18]
ITO/ SnO_2/ $CsPbI_xBr_{3-x}$/ Spiro/Au	21.02	1.27	19.40	85.31	2023[48]
FTO/TiO_2/ $CsPbI_3$-AC/spiro/Au	20.69	1.13	19.04	80.79	2024[20]
FTO/TiO_2/ $CsPbI_3$ /spiro/Au	21.86	1.26	20.71	83.80	2024[49]
FTO/TiO_2/ $CsPbI_3$/spiro/Au	**21.85**	**1.26**	**20.84**	**82.82**	**Our work**

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