Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

Synergistic effects of thiocyanate additive and cesium cations on improving the performance and initial illumination stability of efficient perovskite solar cells

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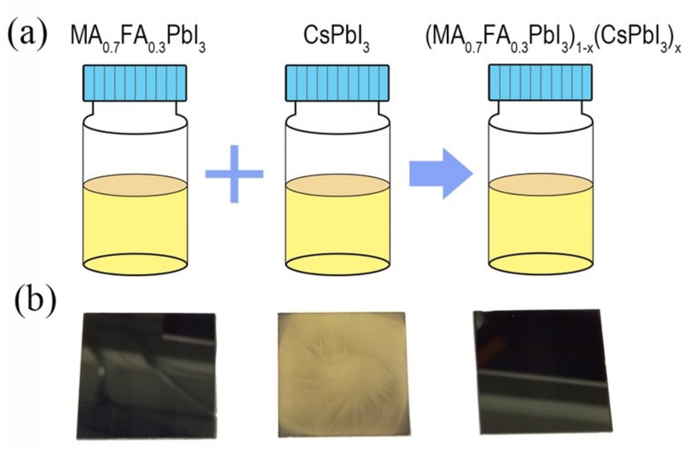

Fig. S1. (a) Scheme of the mixture of $(MA_{0.7}FA_{0.3}PbI_3)_{1-x}(CsPbI_3)_x$ precursor. (b) Perovskite films fabricated at 100 ^oC in a glovebox and then been brought out for taken picture. From left to right, x=0, x=1 and x=0.1, respectively.

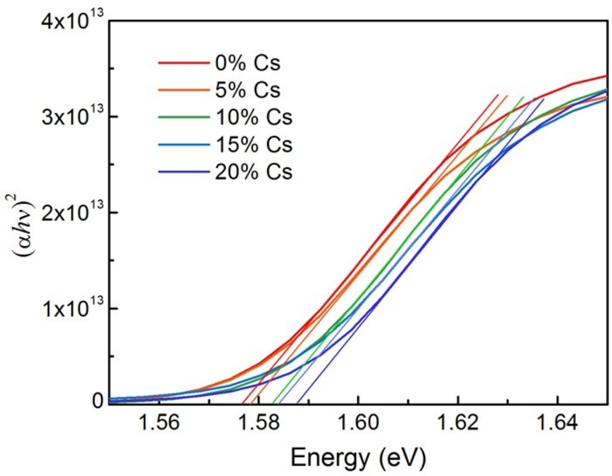

Fig. S2. Tacu plots derived from absorbance spectra of perovskite films with different Cs concentrations.

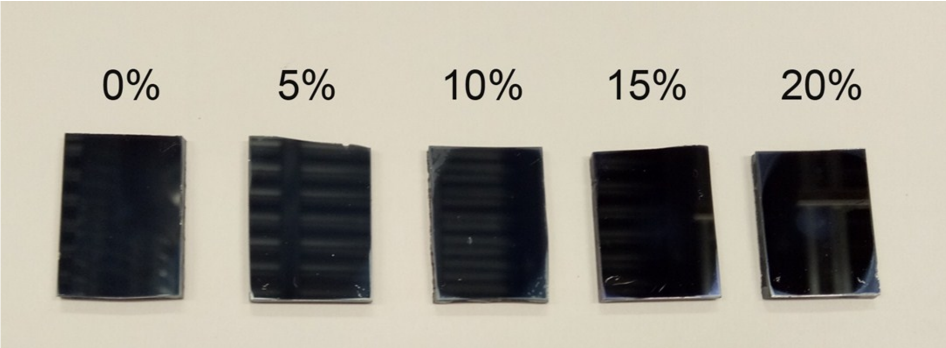

Fig. S3. Photos of perovskite films with different Cs concentrations.

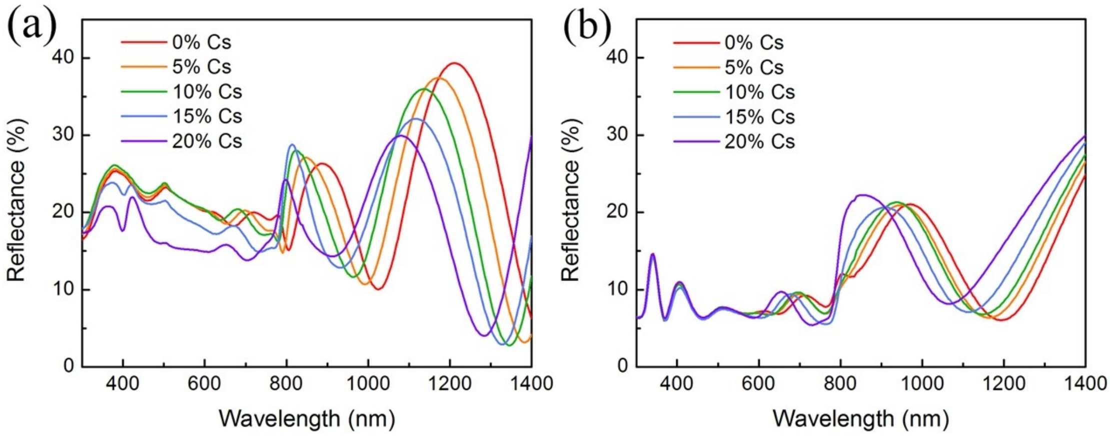

Fig. S4. The reflectance spectra from (a) film side and (b) glass side of perovskite films fabricated on FTO substrates with different amount of Cs.

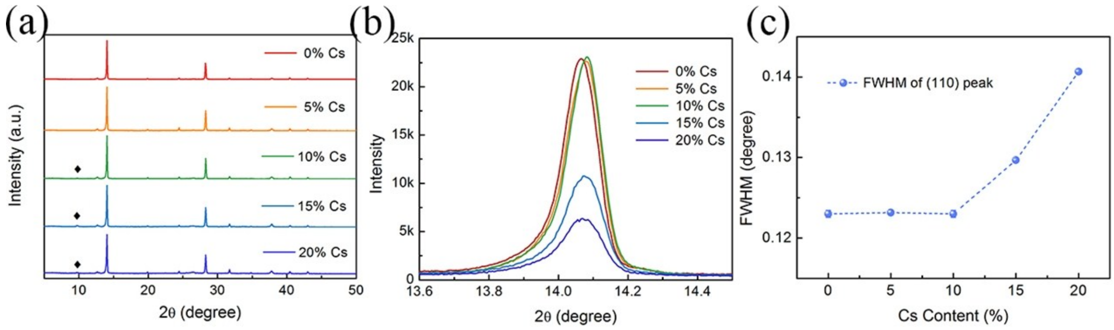

Fig. S5. (a) XRD patterns and (b) enlarged (110) peak of perovskite films fabricated on FTO substrates with different amount of Cs. (c) Full width half maximum (FWHM) of the (110) peak calculated from (b).

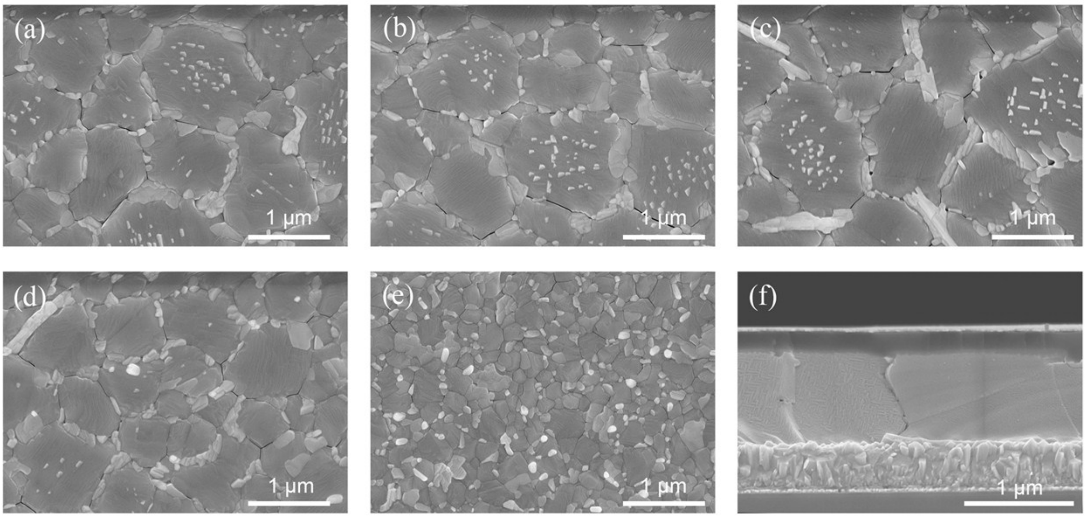

Fig. S6. Top-view of SEM images of SCN-incorporated $(MA_{0.7}FA_{0.3})_{1-x}Cs_xPbI_3$ films with the concentration of Cs varying from 0 to 20%. (a) 0%, (b) 5%, (c) 10%, (d) 15%, (e) 20%. (f) Cross-sectional SEM of 10% Cs incorporated perovskite device.

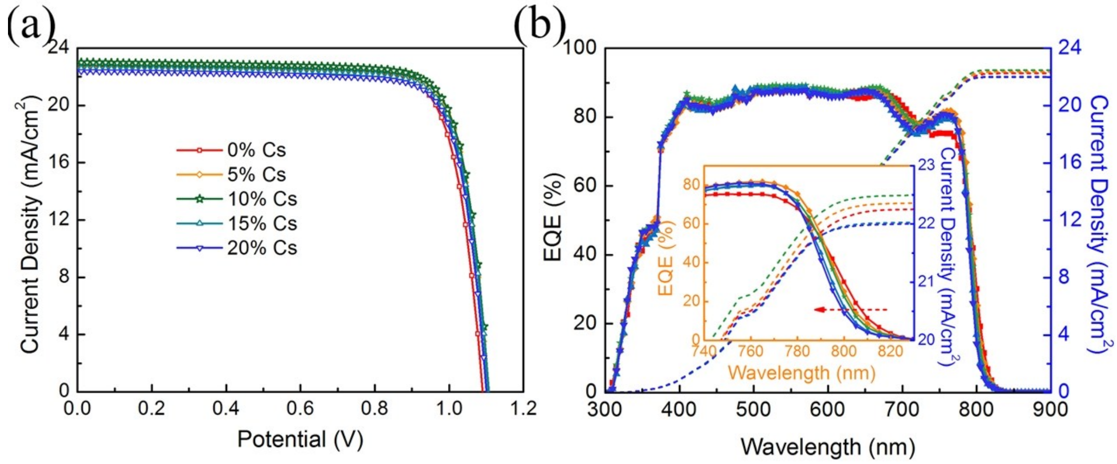

Fig. S7. (a) *J-V* curves measured under reverse voltage scans and (b) EQE plots of PVSCs with SCN and different contents of Cs. Inset shows that the onset of EQE gradually blue-shifted while increasing the ratio of Cs.

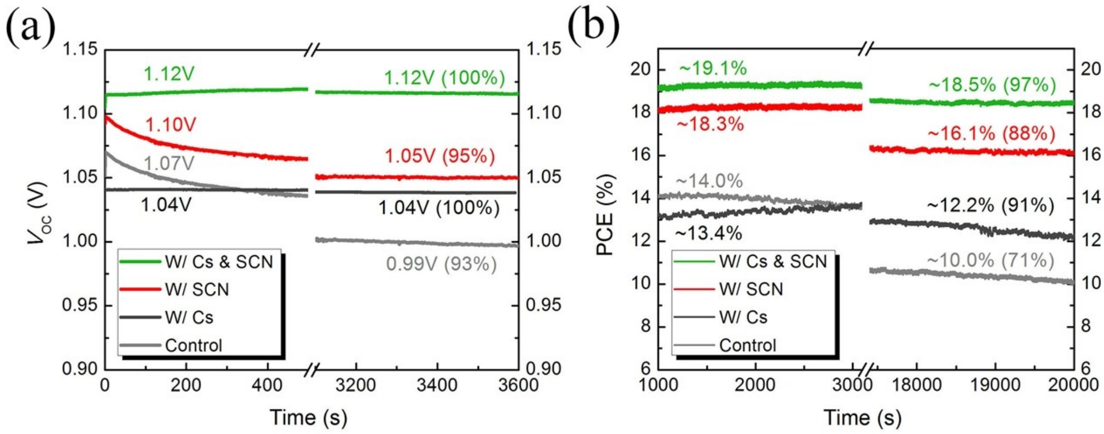

Fig. S8. The (a) V_{OC} tracking and (b) maximum power point tracking of PVSCs of pure $MA_{0.7}FA_{0.3}PbI_3$ (control), incorporated with only Cs (W/ Cs), incorporated with only $Pb(SCN)_2$ (W/ SCN) and with both (W/ Cs & SCN).

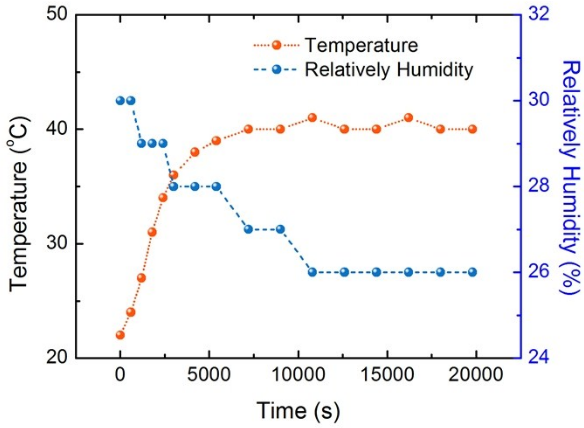

Fig. S9. The evolution of surface temperature of the device and relatively humidity of the atmosphere during the MPPT measurement of one PVSC.

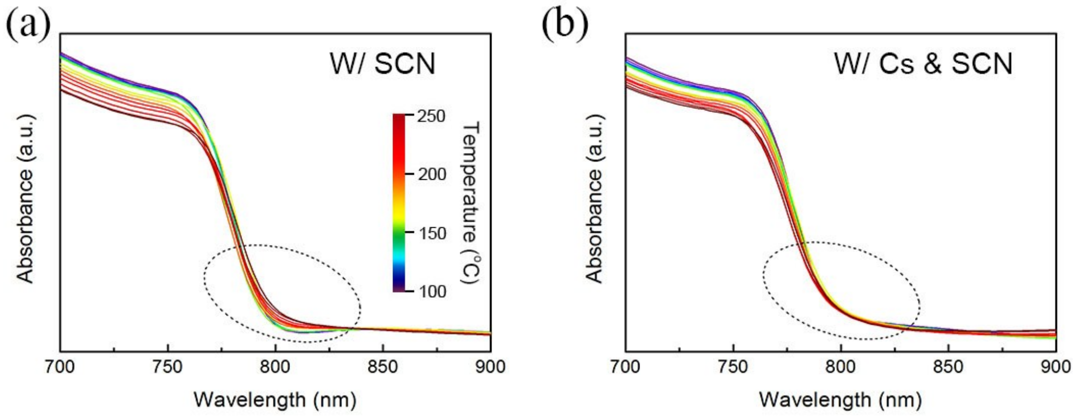

Fig. S10. Absorbance spectra of perovskite films W/ SCN and W/ Cs & SCN heated at different temperatures for 1 minute at each. The temperature changes from 100 to 250 ^{o}C.

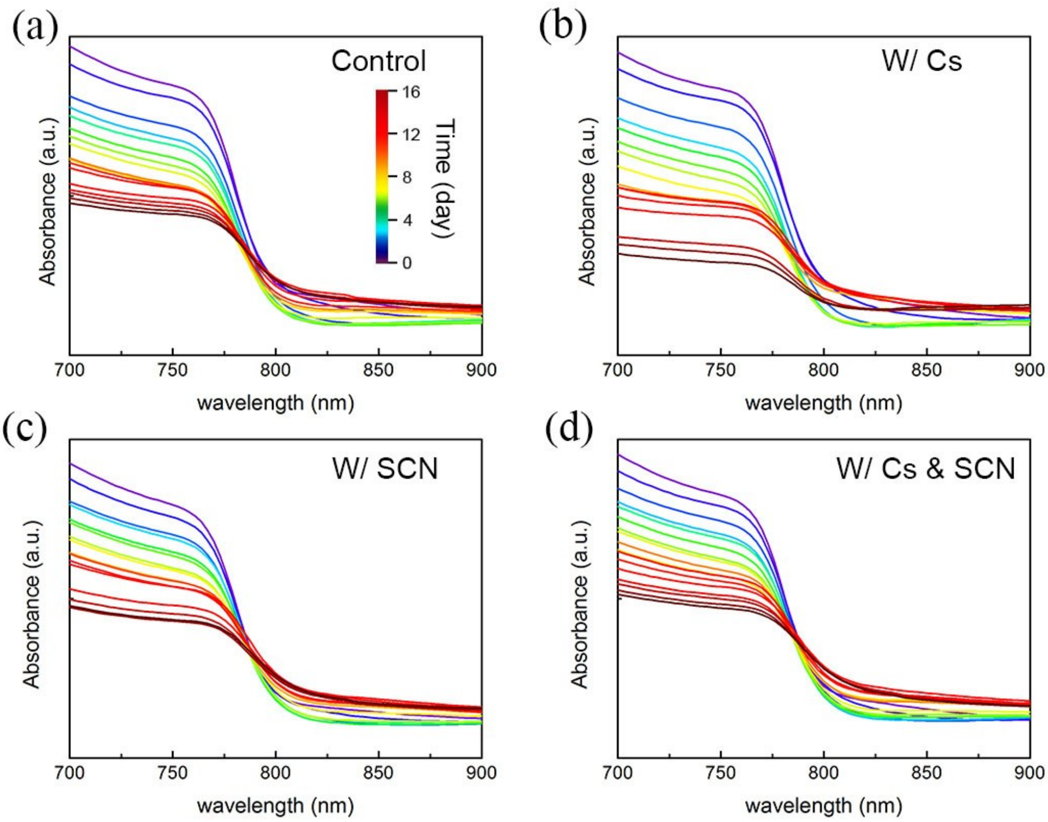

Fig. S11. Absorbance spectra of perovskite films exposed in ~60% relatively humidity air for different days.

Table S1. Photovoltaic parameters of control, W/ Cs, W/ SCN, and W/ Cs & SCN PVSCs.

samples	V_{OC} (V)	J_{SC} (mA/cm^2)	FF (%)	PCE (%)
Control	1.07	21.92	73.15	17.16
W/ Cs	1.04	21.71	71.86	16.22
W/ SCN	1.10	22.20	77.09	18.83
W/ Cs & SCN	1.12	22.61	78.68	19.92

Table S2. Averaged photovoltaic parameters calculated from 135 PVSCs based on $(MA_{0.7}FA_{0.3})_{1-x}Cs_x PbI_3$. Error values represent the standard deviations.

x (%)	Scan directions	V_{OC} (V)	J_{SC} (mA/cm^2)	FF (%)	PCE (%)
0	Reverse	1.092±0.003	22.74±0.15	78.44±0.70	19.47±0.22
	Forward	1.071±0.013	22.74±0.15	73.34±0.70	17.86±0.60
5	Reverse	1.093±0.012	22.74±0.03	78.79±0.51	19.57±0.25
	Forward	1.075±0.011	22.74±0.03	73.34±0.70	18.10±0.37
10	Reverse	1.098±0.008	22.82±0.06	79.75±0.58	19.98±0.29
	Forward	1.081±0.010	22.82±0.06	75.55±0.66	18.63±0.53
15	Reverse	1.094±0.009	22.38±0.13	78.95±1.03	19.33±0.43
	Forward	1.075±0.009	22.38±0.13	74.85±1.01	18.01±0.54
20	Reverse	1.089±0.008	22.32±0.26	78.56±1.15	19.09±0.35
	Forward	1.067±0.008	22.32±0.26	74.26±1.10	17.68±0.61

Table S3. Photovoltaic performance of PVSCs shown in Fig. S7 with different amount of Cs measured under reverse voltage scan with a scan rate of 1V/s.

x (%)	V_{OC} (V)	J_{SC} (mA/cm^2)	FF (%)	PCE (%)	Integrated J_{SC} (mA/cm^2)
0	1.090	22.75	79.37	19.68	22.29
5	1.104	22.81	80.05	20.16	22.36
10	1.107	23.00	80.08	20.39	22.50
15	1.106	22.59	79.65	19.90	22.14
20	1.100	22.39	78.89	19.43	22.02