Supporting Information

The Effects of Annealing Time on Triple Cation Perovskite Films and Their Solar Cells

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Supplementary Figures and Tables:

Table S1. Fitting results of EIS parameters for the PSCs under dark condition.

Annealing time	R_s (Ω)	R_r (Ω)	C_d (F)
45 min	10.35	2729	1.66E-08
60 min	18.27	2500	1.74E-08
80 min	19.24	2183	1.71E-08
100 min	18.90	1808	1.70E-08

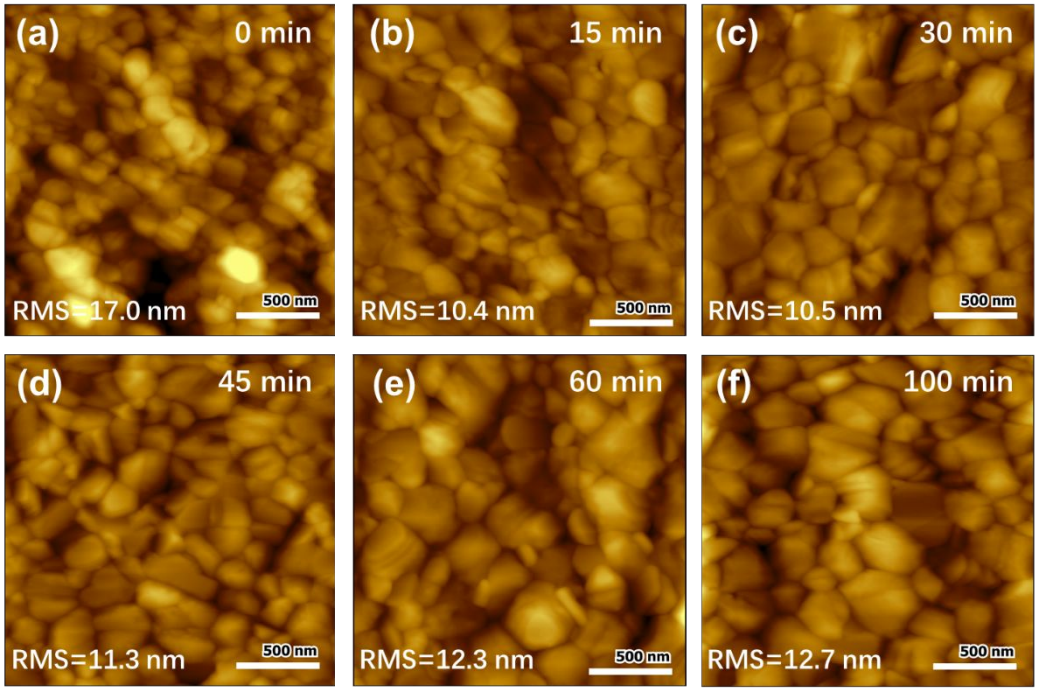


Figure S1. AFM images of CsFAMA perovskite films deposited on galss/ITO/SnO_2 substrates annealed for different periods.

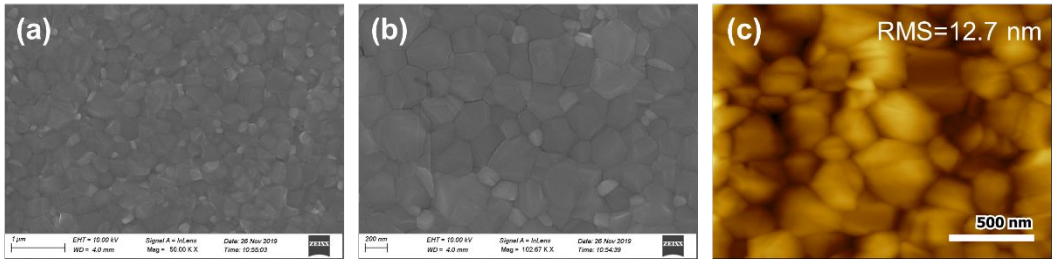
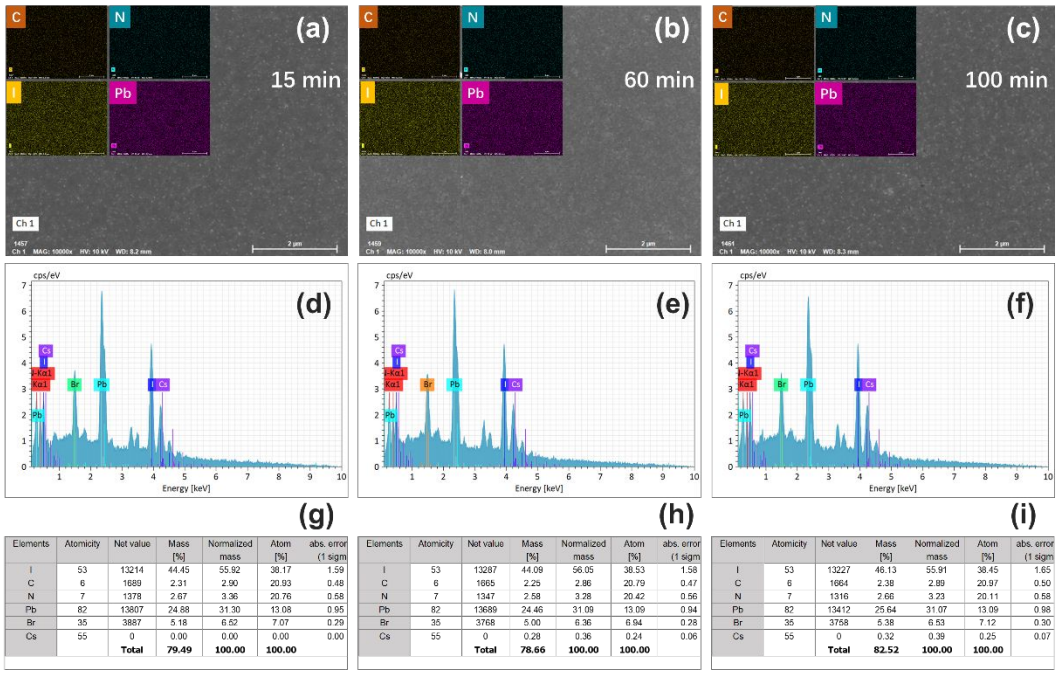


Figure S2. SEM (a-b) and AFM (c) images of 100 min annealed CsFAMA perovskite films. Note that these off-white grains are PbI_2 phase.



(g)

Elements	Atomicity	Net value	Mass [%]	Normalized mass	Atom [%]	abs. error (1 sigm
I	53	13214	44.45	55.92	38.17	1.59
C	6	1689	2.31	2.90	20.93	0.48
N	7	1378	2.67	3.36	20.76	0.58
Pb	82	13807	24.88	31.30	13.08	0.95
Br	35	3887	5.18	6.52	7.07	0.29
Cs	55	0	0.00	0.00	0.00	0.00
		Total	**79.49**	**100.00**	**100.00**	

(h)

Elements	Atomicity	Net value	Mass [%]	Normalized mass	Atom [%]	abs. error (1 sigm
I	53	13287	44.09	56.05	38.53	1.58
C	6	1665	2.25	2.86	20.79	0.47
N	7	1347	2.58	3.28	20.42	0.56
Pb	82	13689	24.46	31.09	13.09	0.94
Br	35	3768	5.00	6.36	6.94	0.28
Cs	55	0	0.28	0.36	0.24	0.06
		Total	**78.66**	**100.00**	**100.00**	

(i)

Elements	Atomicity	Net value	Mass [%]	Normalized mass	Atom [%]	abs. error (1 sigm
I	53	13227	46.13	55.91	38.45	1.65
C	6	1664	2.38	2.89	20.97	0.50
N	7	1316	2.66	3.23	20.11	0.58
Pb	82	13412	25.64	31.07	13.09	0.98
Br	35	3758	5.38	6.53	7.12	0.30
Cs	55	0	0.32	0.39	0.25	0.07
		Total	**82.52**	**100.00**	**100.00**	

Figure S3. Surface elements analysis of CsFAMA films with different annealing time at 100 ℃. (a-c) SEM images (a) and corresponding elemental mappings. (d-f) Energy spectra of corresponding films. (g-i) Element content analysis results of corresponding films.

Table S2. Fitting results of TRPL decay of Figure 5(c) and (d) via a bi-exponential decay function of the form as $R(t)=B_1\exp(-t/\tau_1)+B_2\exp(-t/\tau_2)$

PbI_2 content	Annealing time	τ_1 (ns)	Fraction 1 (%)	τ_2 (ns)	Fraction 2 (%)	χ^2
5 mol% excess	0 min	5.7	0.21	924.5	99.79	1.072
	5 min	3.6	0.88	405.9	99.12	1.016
	10 min	5.3	1.78	238.6	98.22	1.075
	15 min	3.5	2.98	166.7	97.02	1.099
	20 min	3.3	4.34	106.0	95.66	1.005
	30 min	4.5	5.54	103.1	94.46	1.026
	45 min	3.5	7.41	79.9	92.59	1.063
	60 min	2.8	8.47	64.4	91.53	1.026
Without excess	0 min	24.0	0.77	709.0	99.23	1.002
	5 min	21.6	48.93	138.0	51.07	1.268
	10 min	10.7	54.56	55.5	45.44	1.250
	15 min	10.3	64.54	53.4	35.46	1.282
	20 min	7.4	60.74	32.5	39.26	1.236
	30 min	3.7	18.11	22.3	81.89	1.185
	45 min	2.4	20.25	58.8	79.75	1.179
	60 min	3.2	10.00	68.8	90.00	1.195

Figure S4. Photographs of FAMA and CsFAMA perovskite films with respect to annealing time at 100 ℃.

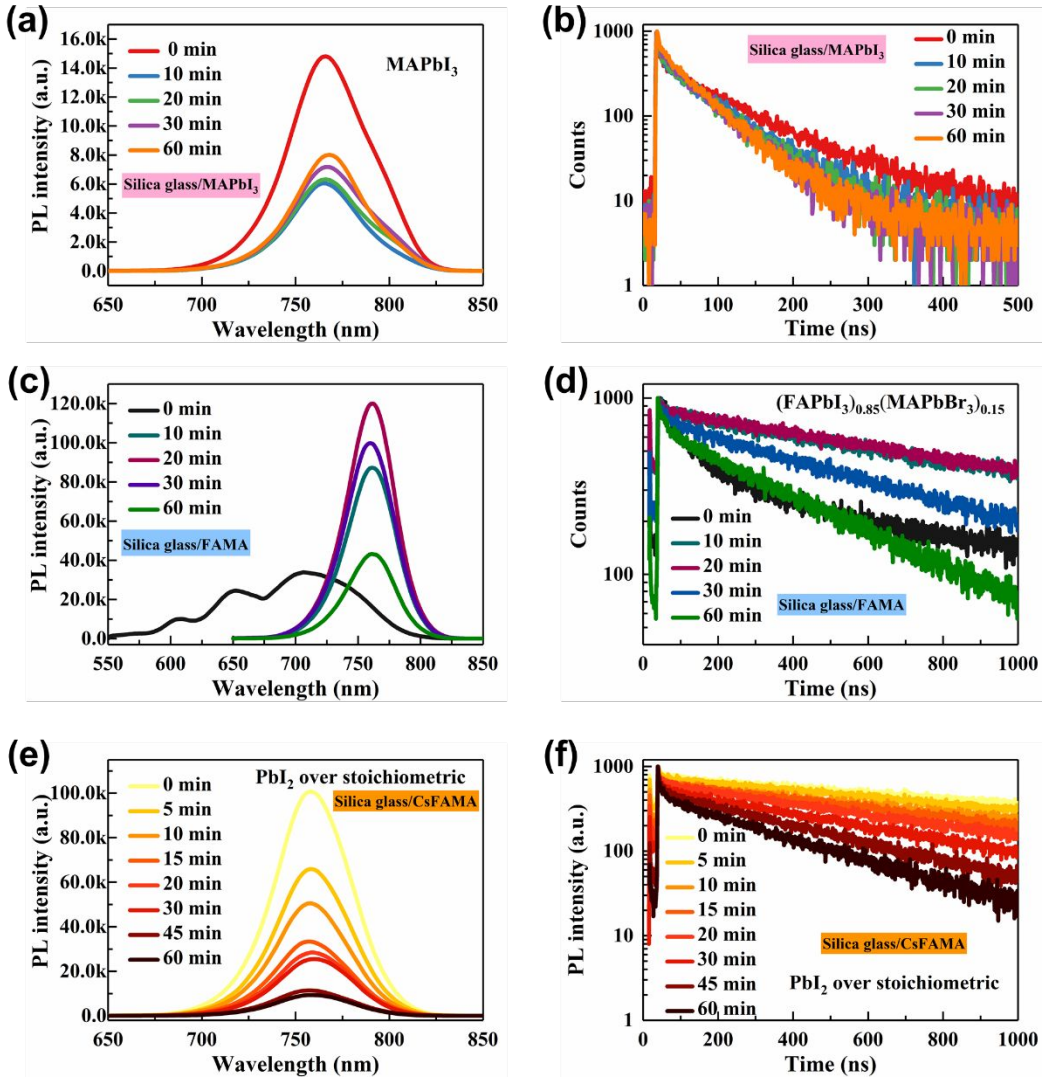


Figure S5. PL spectra and TRPL decay of $MAPbI_3$ (a-b), FAMA (c-d), CsFAMA (e-f) perovskite films deposited on silica glass substrates as a function of annealing time at 100 ℃.

Table S3. Fitting results of TRPL decay of Figure S4(b), (d) and (f) via a bi-exponential decay function of the form as $R(t)=B_1\exp(-t/\tau_1)+B_2\exp(-t/\tau_2)$

Perovskite material	Annealing time	τ_1 (ns)	Fraction 1 (%)	τ_2 (ns)	Fraction 2 (%)	χ^2
$MAPbI_3$	0 min	4.1	4.43	92.8	95.57	1.054
	5 min	4.8	7.23	70.1	92.77	1.179
	10 min	4.0	6.12	64.2	93.88	1.291
	20 min	4.3	6.72	55.4	93.28	1.152
	30 min	5.3	5.61	53.3	94.39	1.123
FAMA	0 min	57.3	15.46	434.0	84.54	1.061
	10 min	17.6	0.43	856.5	99.57	1.056
	20 min	17.6	0.22	1026.9	99.78	1.030
	30 min	15.8	0.97	561.	99.01	1.021
	60 min	18.6	2.12	400.3	97.88	1.035
CsFAMA	0 min	18.8	0.25	1274.0	99.75	1.041
	5 min	6.4	0.27	1006.2	99.73	0.989
	10 min	7.8	0.36	883.2	99.64	1.093
	15 min	7.9	0.44	800.2	99.56	0.976
	20 min	12.4	0.62	610.0	99.38	1.080
	30 min	5.0	0.94	505.1	99.06	1.099
	45 min	5.8	1.61	377.1	98.39	1.013
	60 min	7.6	2.35	302.2	97.65	1.103

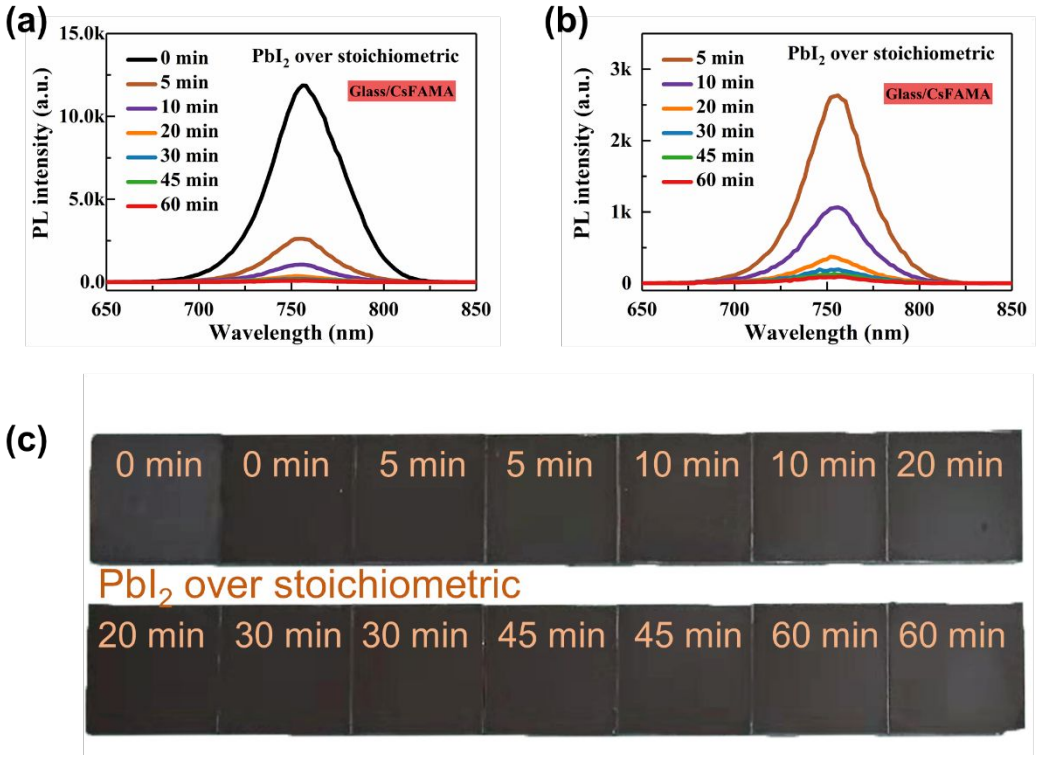


Figure S6. (a-b) PL spectra of CsFAMA perovskite films with different heating time at 100 ℃ with PbI_2 excessively added (5 mol% excess) in precursor solution. (c) Photographs of CsFAMA perovskite films deposited on glass substrates as a function of annealing time at 100 ℃.

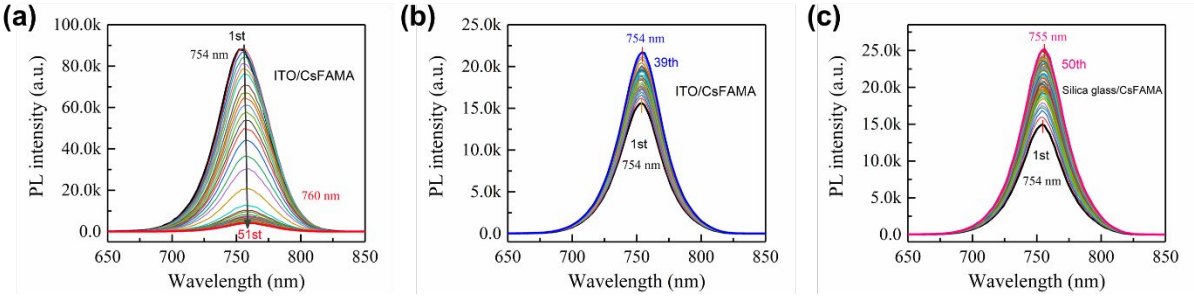


Figure S7. Two-dimensional PL spectra over time of CsFAMA perovskite films without (a) or with annealing for 10 min (b) and 45 min (c), respectively. They are corresponding to the three-dimensional PL spectra in Figure 6(a), (b) and (c), respectively.

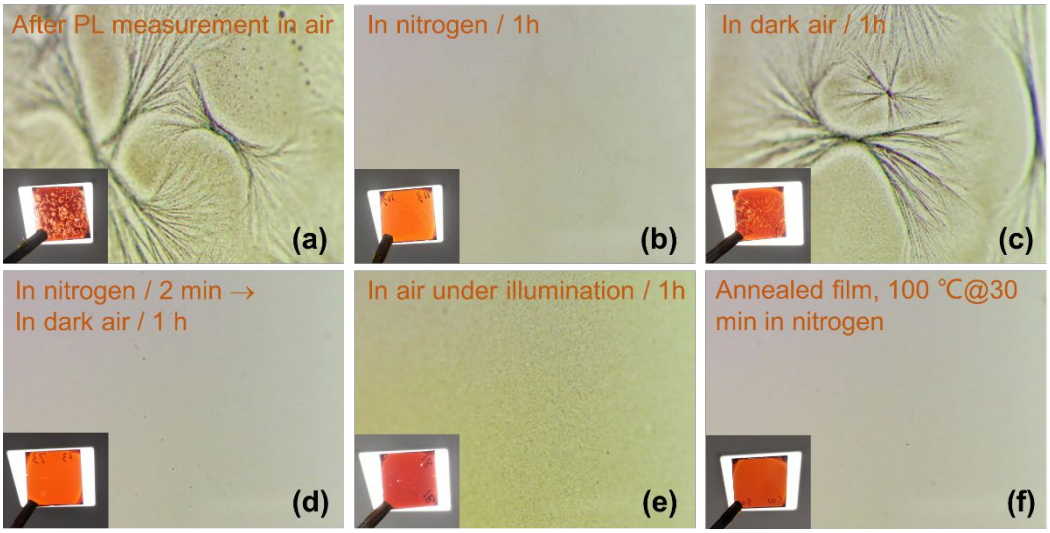


Figure S8. Mesoscopic morphologies and photographs of CsFAMA perovskite films placed in different environments as soon as anti-solvent dripping. (a) After PL measurement. (b) In nitrogen for 1 h. (c) In dark air for 1 h. (d) In nitrogen for 2 min, and then in dark air for 1 h. (e) In air under one sun illumination for 1 h. (f) Annealed at 100 ℃ for 30 min in glovebox. Insets are photographs of corresponding perovskite films.

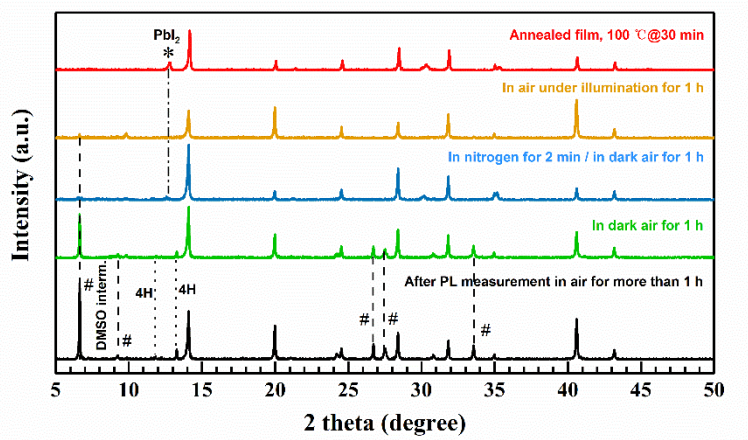


Figure S9. XRD patterns of perovskite films placed in different environments for certain periods as soon as anti-solvent dripping.

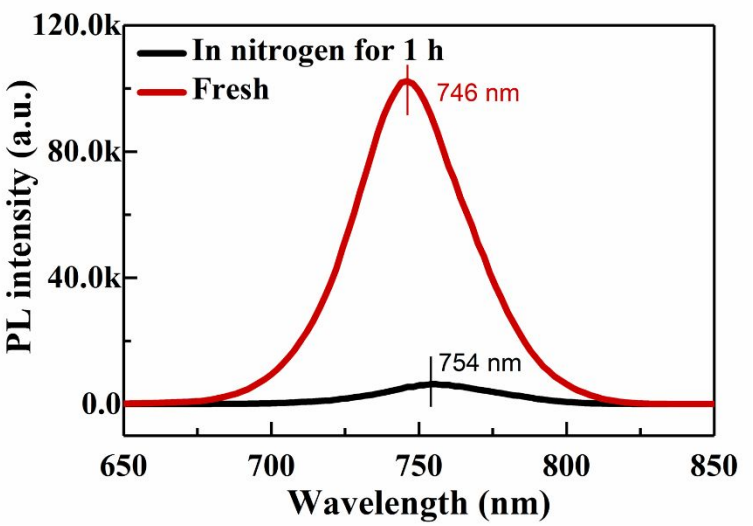


Figure S10. A comparison of PL spectra of fresh film after anti-solvent dripping and film placed in glovebox for 1 h. The PL intensity of fresh film is much stronger than that of film stored in glovebox for 1 h after CB dripping without annealing. The PL emission peak of fresh film exhibits blue shift.