Supplementary Information

Low-temperature synergistic effect of MA and Cl towards high-quality α -FAPbI₃ films for humid-air-processed perovskite solar cells

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Figure S1. Schematic of experimental procedures (left) and scanning electron microscopy (SEM) patterns (right) for CA and CAM films.



Figure S2. Photographs of CA, CAM, and MACI-CSA films exposed in humid air (RH: $60 \pm 10\%$) for different times.



Figure S3. UV-vis absorption spectra of CA film exposed in humid air for different times.



Figure S4. UV-vis absorption spectra of CAM film exposed in humid air for different times.



Figure S5. Top-view SEM image of the mesoporous TiO₂.



Figure S6. (a) UV-vis absorption spectra of MACI-CSA film exposed in humid air for different times. (b) XRD patterns of MACI-CSA film exposed in humid air for different times.



Figure S7. FTIR spectra of the CA, MAI-CSA, FACI-CSA, and MACI-CSA perovskite films, which is convenient to compare the intensity of the stretching vibration peak of C=N bond.



Figure S8. Schematic illustration for kinetic mechanism of MAI-CSA film and FACI-CSA film.



Figure S9. Grain size distribution of perovskite films annealed at 80 °C,100 °C and 120 °C, respectively.



Figure S10. J-V curves of the champion device (MACl-CSA-100) measured under AM1.5G 100 mW/cm^2 .



Figure S11. Time-resolved photoluminescence decay of gb-CA-150 film and MACI-CSA-100 film.



Figure S12. The corresponding IPCE spectra of the PSCs employing MACI-CSA-100 film.

gb-CA-150	$V_{\rm oc}$ [V]	$J_{\rm sc} [{\rm mA/cm^2}]$	FF [%]	PCE [%]
1	0.98	20.49	72.98	14.65
2	0.98	20.53	73.05	14.70
3	0.99	20.54	73.08	14.86
4	1.00	20.56	73.12	15.03
5	1.01	20.61	73.22	15.24
6	1.01	20.62	73.24	15.25
7	1.01	20.66	73.33	15.30
8	1.02	20.68	73.36	15.47
9	1.02	20.69	73.38	15.49
10	1.03	20.72	73.44	15.67
11	1.01	20.60	73.20	15.23
12	1.02	20.65	73.30	15.44
13	1.02	20.67	73.34	15.46
14	1.01	20.57	73.16	15.19
15	1.03	20.71	73.41	15.66
16	0.99	20.53	73.06	14.85
17	0.98	20.51	73.02	14.68
18	0.97	20.44	72.88	14.45
19	1.01	20.65	73.26	15.28
20	1.00	20.58	73.16	15.06

Table S1. The parameters of the perovskite solar cells employing the gb-CA-150 films.

MACI-CSA-100	$V_{\rm oc}$ [V]	$J_{\rm sc}$ [mA/cm ²]	FF [%]	PCE [%]
1	1.04	21.53	75.21	16.84
2	1.05	21.52	75.21	16.99
3	1.05	21.57	75.25	17.04
4	1.04	21.52	75.2	16.83
5	1.06	21.61	75.29	17.25
6	1.05	21.58	75.26	17.05
7	1.05	21.62	75.28	17.08
8	1.02	21.45	75.13	16.44
9	1.04	21.54	75.22	16.85
10	1.05	21.59	75.27	17.06
11	1.04	21.48	75.16	16.79
12	1.06	21.62	75.30	17.26
13	1.02	21.43	75.11	16.42
14	1.06	21.63	75.31	17.28
15	1.03	21.47	75.15	16.62
16	1.03	21.49	75.17	16.64
17	1.05	21.56	75.24	17.03
18	1.05	21.60	75.28	17.07
19	1.04	21.51	75.19	16.82
20	1.06	21.63	75.30	17.27

Table S2. The parameters of the perovskite solar cells employing the MACI-CSA-100 films.