Supporting Information

High crystallization of multiple cation perovskite absorber for low-temperature stable ZnO solar cells with high-efficiency over 20%

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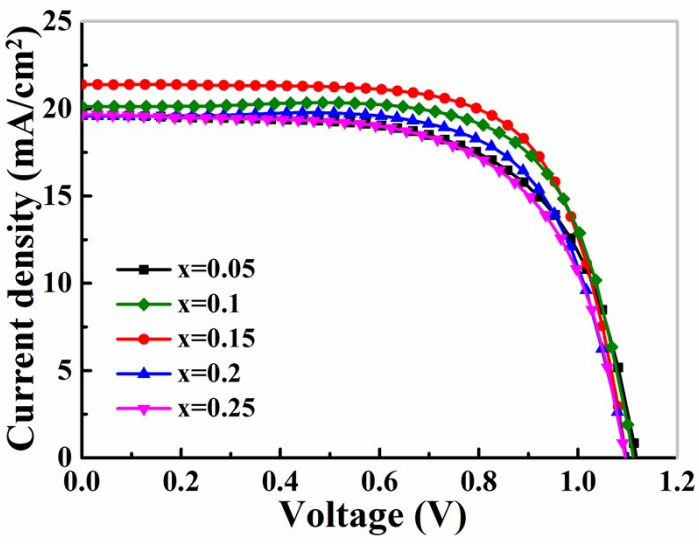

Figure S1 *J-V* curves of the $FA_{1-x}Cs_xPbI_3$ perovskite PSCs with different Cs contents (x values).

Table S1 Photovoltaic parameters of the $FA_{1-x}Cs_xPbI_3$ PSCs with different Cs contents (x values).

	V_{oc} (V)	J_{sc} (mA/cm^2)	*FF* (%)	PCE (%)
x=0.05	1.12	19.66	64.34	14.17
x=0.1	1.11	20.36	70.60	15.96
x=0.15	1.12	21.40	69.69	16.70
x=0.2	1.10	19.56	68.95	14.48
x=0.25	1.10	19.50	63.44	13.61

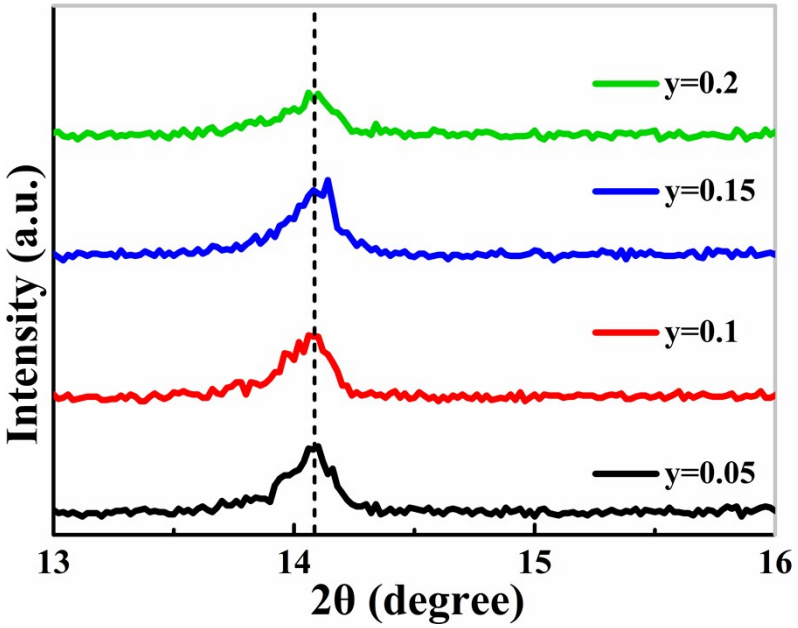

Figure S2 Magnified XRD patterns of the (101) lattice plane of $MA_yFA_{0.85-x}Cs_{0.15}PbI_3$ thin films with various MA contents (y values).

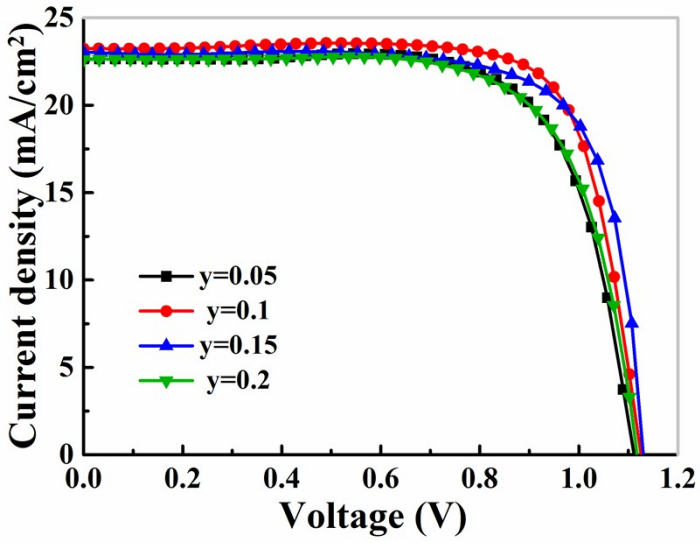

Figure S3 *J-V* curves of the $MA_yFA_{0.85-y}Cs_{0.15}PbI_3$ perovskite PSCs with different MA contents (y values).

Table S2 Photovoltaic parameters of the $MA_yFA_{0.85-y}Cs_{0.15}PbI_3$ PSCs with different MA contents (y values).

	V_{oc} (V)	J_{sc} (mA/cm^2)	*FF* (%)	PCE (%)
y=0.05	1.12	22.72	72.04	18.33
y=0.1	1.13	23.15	76.81	20.09
y=0.15	1.13	23.08	74.94	19.54
y=0.25	1.12	22.81	71.40	18.24

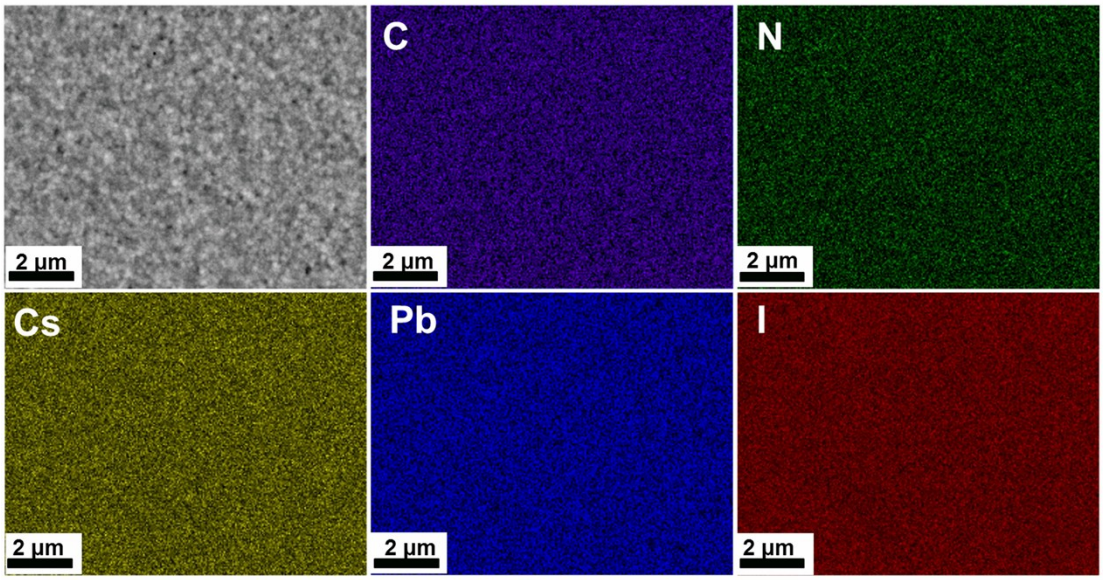

Figure S4 EDS mapping of $MA_{0.1}FA_{0.75}Cs_{0.15}PbI_3$ films on ZnO layer.

Table S3 Short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF) and power conversion efficiency (PCE) of 30 perovskite solar cells employing $FAPbI_3$. The data were obtained under AM 1.5G one sun illumination (100 mW/cm^2)

Samples	V_{oc} **(V)**	J_{sc} **(mA/cm^2)**	FF **(%)**	**PCE (%)**
1	1.07	20.01	59.00	12.63
2	1.11	19.53	61.23	13.27
3	1.11	18.65	65.10	13.48
4	1.12	18.76	62.98	13.23
5	1.11	19.59	64.32	13.99
6	1.12	19.13	63.18	13.54
7	1.12	19.33	63.69	13.79
8	1.11	19.63	63.54	13.84
9	1.12	18.79	63.84	13.44
10	1.10	18.66	63.75	13.09
11	1.11	18.92	64.11	13.46
12	1.13	17.65	64.32	12.83
13	1.11	18.46	63.21	12.95
14	1.12	18.76	60.11	12.63
15	1.10	18.32	67.11	13.52
16	1.10	17.68	65.31	12.70
17	1.11	17.31	64.77	12.44
18	1.12	18.76	63.98	13.44
19	1.12	19.24	63.78	13.74
20	1.10	17.11	54.00	10.16
21	1.10	19.34	63.82	13.58
22	1.08	19.63	63.11	13.38
23	1.11	18.35	64.01	13.04
24	1.09	19.65	62.75	13.44
25	1.11	18.91	63.11	13.25
26	1.10	18.74	63.33	13.05
27	1.12	18.26	64.10	13.11
28	1.14	19.42	60.55	13.41
29	1.11	18.79	63.21	13.18
30	1.09	19.22	56.00	11.73
Average	1.11±0.01	18.82±0.71	62.84±2.65	13.11±0.73

Table S4 Short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF) and power conversion efficiency (PCE) of 30 perovskite solar cells employing $FA_{0.85}Cs_{0.15}PbI_3$. The data were obtained under AM 1.5G one sun illumination (100 mW/cm^2)

Samples	V_{oc} **(V)**	J_{sc} **(mA/cm^2)**	FF **(%)**	**PCE (%)**
1	1.08	21.40	68.69	15.88
2	1.09	20.35	67.98	15.08
3	1.11	21.46	67.10	15.98
4	1.12	21.21	67.65	16.07
5	1.12	20.98	67.35	15.83
6	1.10	20.95	70.02	16.14
7	1.13	20.76	67.86	15.92
8	1.12	21.10	67.31	15.91
9	1.12	21.13	65.12	15.41
10	1.09	21.03	69.31	15.89
11	1.11	21.34	66.98	15.87
12	1.11	22.12	64.96	15.95
13	1.13	20.43	69.12	15.96
14	1.12	21.70	65.98	16.04
15	1.12	21.50	64.65	15.57
16	1.10	21.01	70.10	16.20
17	1.12	20.69	63.99	14.83
18	1.11	21.50	69.89	16.68
19	1.10	21.03	60.32	13.95
20	1.10	22.03	66.98	16.23
21	1.11	21.30	66.98	15.84
22	1.12	21.87	67.12	16.44
23	1.11	20.97	68.11	15.85
24	1.13	20.34	67.67	15.55
25	1.12	20.65	70.01	16.19
26	1.09	21.89	67.10	16.01
27	1.11	21.94	65.13	15.86
28	1.12	21.30	69.32	16.54
29	1.12	21.10	67.99	16.07
30	1.09	20.97	63.78	14.58
Average	1.11±0.01	21.2±0.48	67.15±2.20	15.81±0.57

Table S5 Short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF) and power conversion efficiency (PCE) of 30 perovskite solar cells employing $MA_{0.1}FA_{0.75}Cs_{0.15}PbI_3$. The data were obtained under AM 1.5G one sun illumination (100 mW/cm^2)

Samples	V_{oc} **(V)**	J_{sc} **(mA/cm^2)**	FF **(%)**	**PCE (%)**
1	1.12	22.74	74.81	19.05
2	1.12	22.83	74.65	19.09
3	1.12	22.78	75.10	19.16
4	1.13	22.89	76.81	19.87
5	1.12	23.40	76.51	20.05
6	1.12	22.89	75.64	19.39
7	1.11	22.76	75.11	18.98
8	1.13	23.31	75.63	19.92
9	1.13	23.24	75.92	19.94
10	1.12	22.73	75.43	19.20
11	1.12	22.57	74.86	18.92
12	1.12	22.78	75.25	19.20
13	1.12	22.45	75.57	19.00
14	1.13	22.54	74.16	18.89
15	1.13	23.62	76.20	20.34
16	1.12	23.59	75.58	19.97
17	1.14	22.73	76.22	19.75
18	1.12	22.70	72.36	18.40
19	1.15	22.72	75.52	19.73
20	1.13	23.11	74.43	19.44
21	1.13	23.35	74.34	19.61
22	1.13	22.98	76.12	19.77
23	1.13	23.05	76.09	19.82
24	1.14	22.78	75.89	19.71
25	1.14	22.80	75.86	19.72
26	1.13	23.13	75.67	19.78
27	1.13	22.79	75.98	19.57
28	1.13	22.95	75.95	19.70
29	1.13	22.89	76.10	19.68
30	1.13	22.87	72.05	18.62
Average	1.13±0.01	22.93±0.29	75.33±1.07	19.48±0.46

Table S6 Comprehensive comparison of the solvent of perovskite precursor and reproducibility between our work and the other reported mixed-cation perovskite solar cell.

Solvent of perovskite precursor	**V_{oc} (V)**	**J_{sc} (mA/cm^2)**	***FF* (%)**	**PCE (%)**	**Ref.**
DMF: DMSO (4:1)	1.13±0.25	22.69±0.75	74.8±0.018	19.20±0.91	10
DMSO	**1.13±0.01**	**22.93±0.29**	**75.33±1.07**	**19.48±0.46**	**Our work**

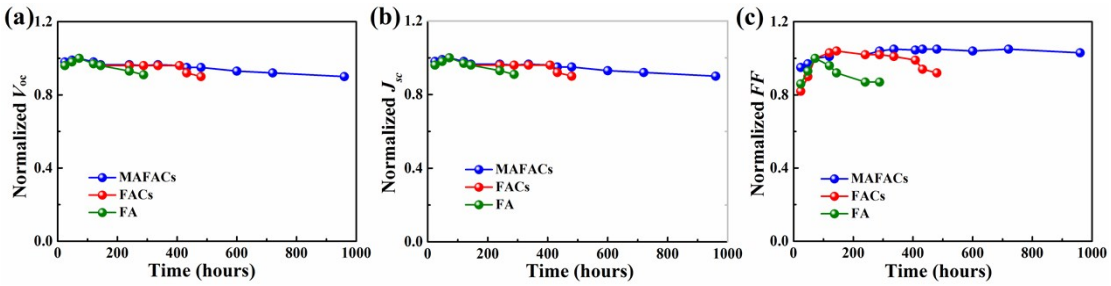

Figure S5 The long-term (a) V_{oc} (b) J_{sc} (c) *FF* of $FAPbI_3$, $FA_{0.85}Cs_{0.15}PbI_3$ and $MA_{0.1}FA_{0.75}Cs_{0.15}PbI_3$ devices without encapsulation stored in the ambient environment with relative humidity of 40−60% at room temperature.

Table S7 Fit results of the TR-PL spectra in Figure 8a. The decay curves were fitted with a tri-exponential function as PL intensity = A + $\Sigma B_i \exp(-t/\tau_i)$, and the relative content (A_i) of each lifetime constant (τ_i) was calculated by $A_i = B_i\tau_i/(\Sigma B_i\tau_i)$. The average PL lifetime constants ($\bar{\tau}$) are calculated by $\bar{\tau} = A_1\tau_1 + A_2\tau_2 + A_3\tau_3$.

Sample	**τ_1 (ns)**	**A_1 (%)**	**τ_2 (ns)**	**A_2 (%)**	**τ_3 (ns)**	**A_3 (%)**	**τ_{avg} (ns)**
$FAPbI_3$	6.29	3.27	47.43	23.46	110.30	73.27	92.15
$FA_{0.85}Cs_{0.15}PbI_3$	7.11	8.68	40.74	42.13	107.08	49.19	70.45
$MA_{0.1}FA_{0.75}Cs_{0.15}PbI_3$	8.74	11.35	46.15	63.80	105.36	24.85	56.62

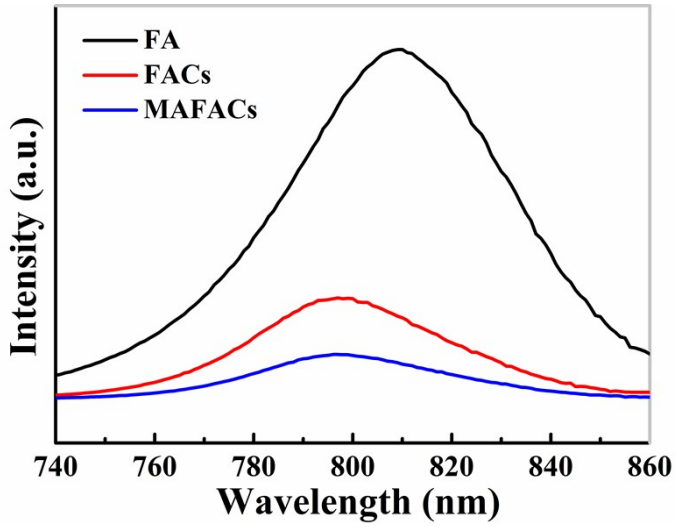

Figure S6 SS-PL of $FAPbI_3$, $FA_{0.85}Cs_{0.15}PbI_3$ and $MA_{0.1}FA_{0.75}Cs_{0.15}PbI_3$ perovskite films.

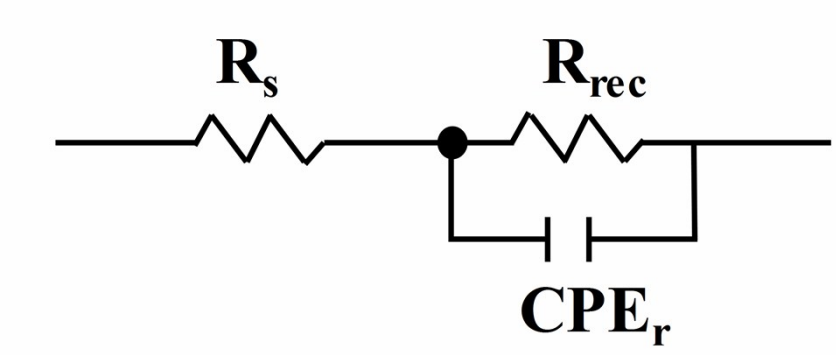

Figure S7 The equivalent circuit model for PSCs in EIS under dark condition.

Table S8 EIS parameters for the PSCs under dark condition.

Sample	R_s (Ω)	R_{rec} (Ω)
$FAPbI_3$	28.78	282.7
$FA_{0.85}Cs_{0.15}PbI_3$	26.39	499.8
$MA_{0.1}FA_{0.75}Cs_{0.15}PbI_3$	23.01	1116.0

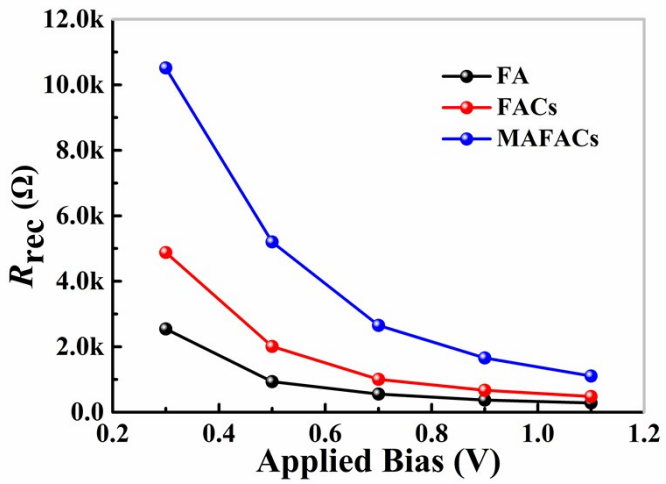

Figure S8 Plots of R_{rec} at different applied biases extracted from impedance measurements of the three devices under dark conditions.

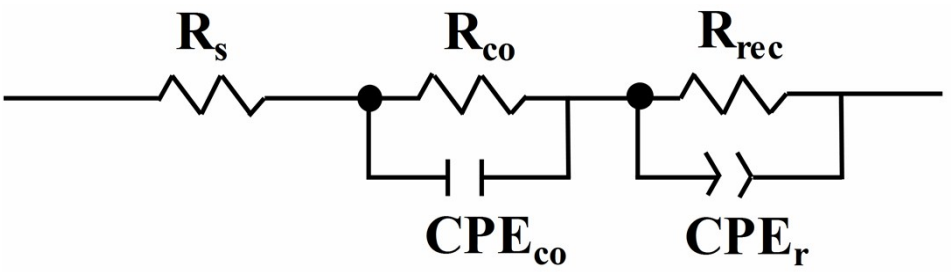

Figure S9 The equivalent circuit model for PSCs in EIS under AM 1.5G illumination.

Table S9 EIS parameters for the PSCs under AM 1.5G illumination.

Sample	R_s **(Ω)**	R_{co} **(Ω)**	R_{rec} **(Ω)**
$FAPbI_3$	23.55	214.9	1551
$FA_{0.85}Cs_{0.15}PbI_3$	23.80	305.0	1877
$MA_{0.1}FA_{0.75}Cs_{0.15}PbI_3$	32.67	331.8	5361