Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Fluorine Modulated Bulk-Phase Heterojunction and Tolerance Factor for Enhanced Performance and Structure Stability of Cesium Lead Halide Perovskite Solar Cells

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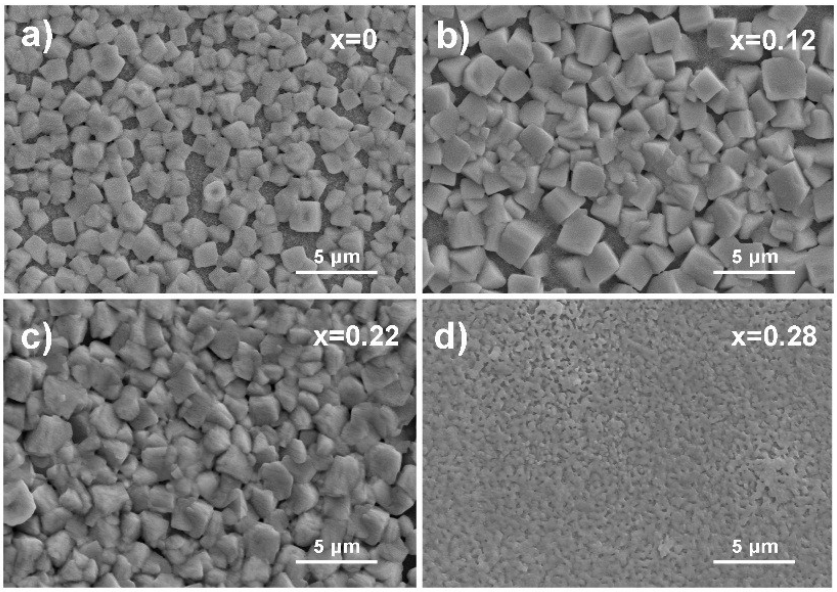

Fig. S1 Top view SEM images of $CsPbBrI_{2-x}F_x$ films.

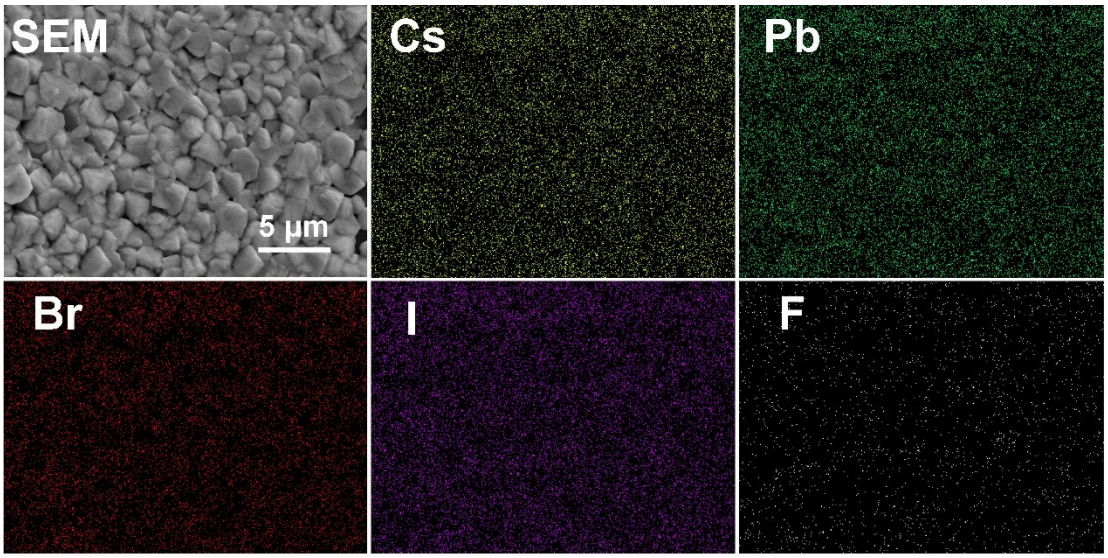

Fig. S2 EDS mapping images for each element of the $CsPbBrI_{1.78}F_{0.22}$ film in top view.

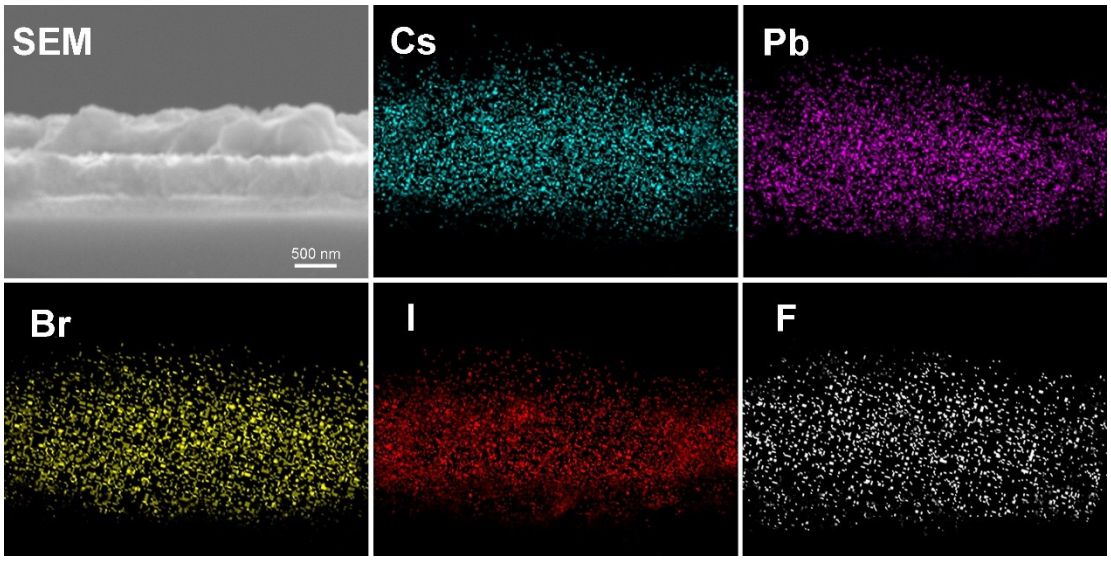

Fig. S3 Cross-section EDS mapping images for each element of the $CsPbBrI_{1.78}F_{0.22}$ film on FTO.

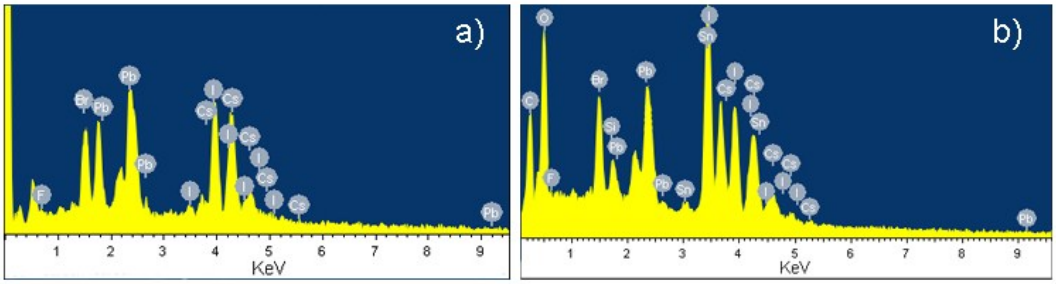

Fig. S4 EDS spectra of $CsPbBrI_{1.78}F_{0.22}$ film on FTO in a) top view and b) sectional view.

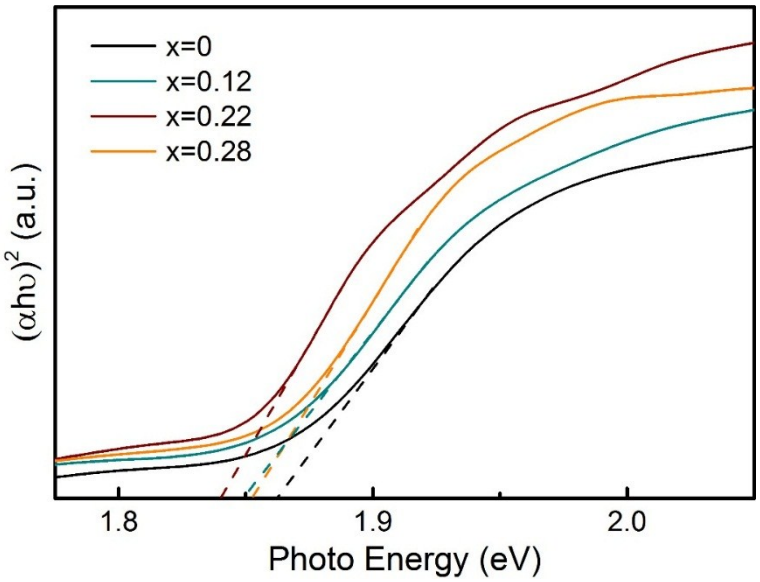

Fig. S5 Tauc plots of $CsPbBrI_{2-x}F_x$ films calculated from the absorption spectra for the band gaps.

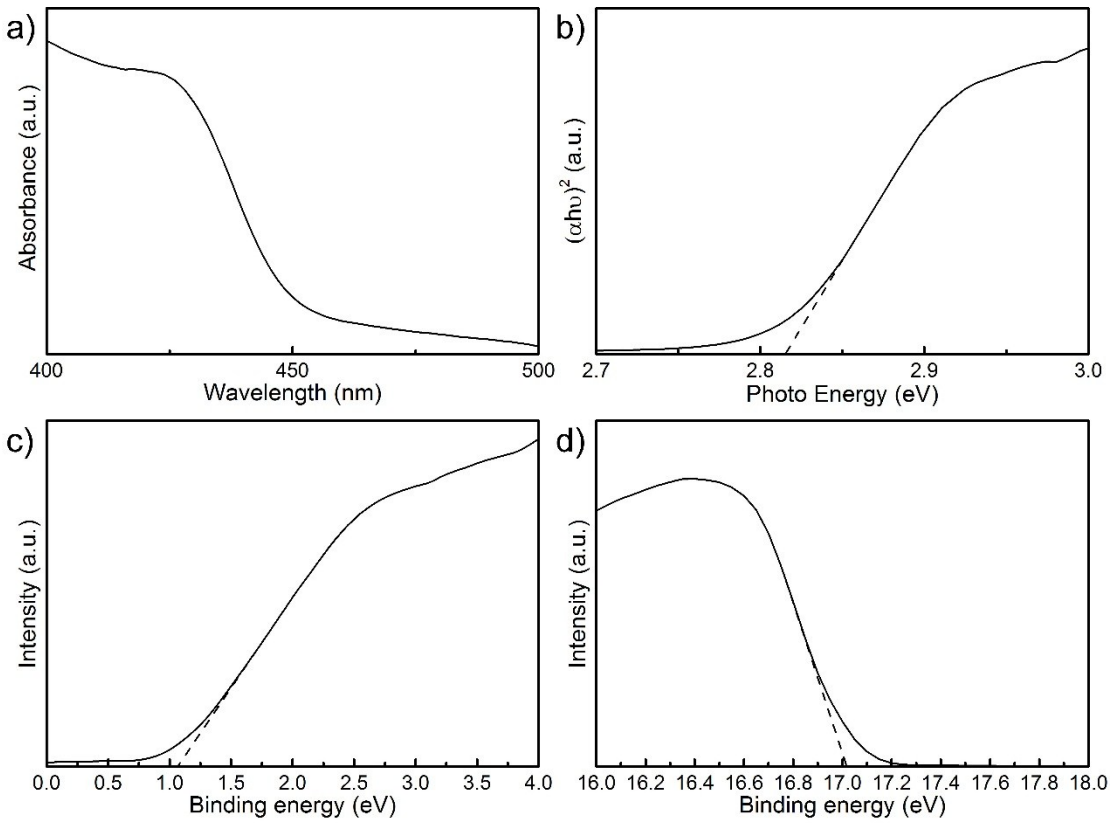

Fig. S6 a) Absorption spectra of the $CsPbBrI_2$ film in δ-phase. b) Tauc plots calculated from the absorption spectra for the band gap of the δ-$CsPbBrI_2$. c-d) UPS spectra of the $CsPbBrI_2$ film in δ-phase.

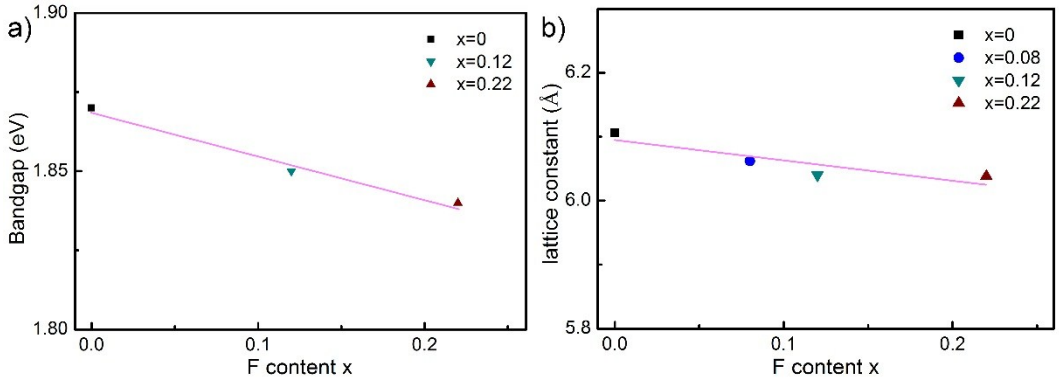

Fig. S7 a) Optical bandgap and b) lattice constant of $CsPbBrI_{2-x}F_x$, showing a linear relationship of the F content in the mixed halide perovskites.

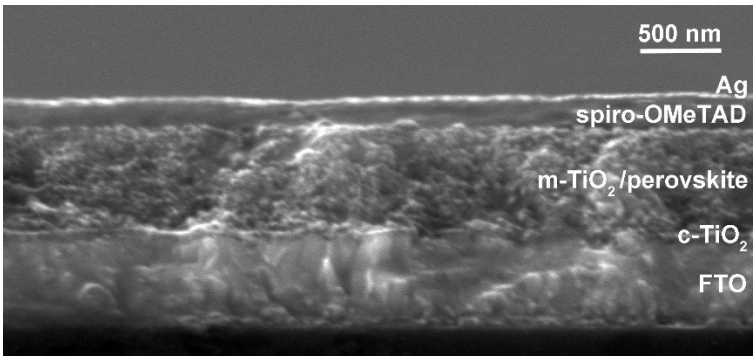

Fig. S8 Cross-section SEM image of $CsPbBrI_{1.78}F_{0.22}$ PSC.

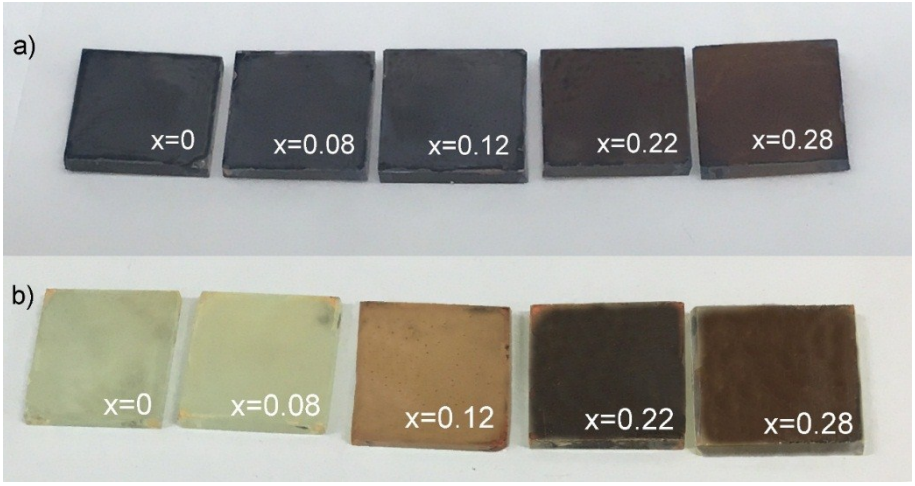

Fig. S9 Photos of $CsPbBrI_{2-x}F_x$ films prepared on glass/FTO/c-TiO_2/m-TiO_2 substrates a) immediately after annealing and b) under the humidity of RH 70% at room temperature after one day.

Table S1. Lattice constants of (110) peaks identified from XRD according to the Bragg equation.

x	2θ (degree)	a (Å)
0	20.57	6.106
0.08	20.72	6.062
0.12	20.80	6.039
0.22	20.80	6.039
0.28	20.67	6.077

Table S2. Photoluminescence lifetimes fitted by the biexponential formula for $CsPbBrI_{2-x}F_x$ films.

x	τ_1 (ns)	a_1 (%)	τ_2 (ns)	a_2 (%)	Ave (ns)
0	0.75	83.43	3.24	16.57	1.16
0.22	1.69	74.49	19.01	25.51	6.11

Table S3. Photovoltaic parameters of short-circuit photocurrent density (J_{SC}), open-circuit voltage (V_{OC}), fill factor (FF) and power conversion efficiency (PCE) of $CsPbBrI_2$ PSCs.

Sample	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
1	0.97	13.16	67	8.55
2	0.89	12.89	68	7.80
3	0.96	12.56	66	7.96
4	0.98	12.63	65	8.04
5	0.99	12.15	65	7.82
6	0.96	11.96	64	7.35
7	0.99	12.69	64	8.04
8	0.95	12.80	69	8.39
9	1.02	13.00	64	8.48
10	0.99	13.51	63	8.43
11	0.93	13.21	63	7.74
12	0.95	13.10	62	7.71
13	0.98	12.66	62	7.69
14	0.92	12.95	61	7.27
15	0.90	12.86	60	6.74
16	0.96	12.03	58	6.70
Ave	0.96±0.04	12.76±0.43	63.81±2.90	7.79±0.56

Table S4. Photovoltaic parameters of short-circuit photocurrent density (J_{SC}), open-circuit voltage (V_{OC}), fill factor (FF) and power conversion efficiency (PCE) of $CsPbBrI_{1.88}F_{0.12}$ PSCs.

Sample	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
1	1.00	13.71	67	9.18
2	0.93	13.35	72	8.94
3	0.96	11.65	74	8.28
4	0.94	11.87	71	7.92
5	0.98	13.24	68	8.82
6	0.96	13.43	65	8.38
7	0.99	12.54	65	8.07
8	0.97	13.35	64	8.29
9	0.97	13.84	63	8.46
10	0.95	13.94	60	7.95
11	0.94	14.28	59	7.92
12	0.98	14.54	61	8.69
13	0.94	13.87	62	8.08
14	0.92	13.95	58	7.44
15	0.91	14.21	63	8.15
16	0.94	14.36	62	8.37
Ave	0.96±0.03	13.51±0.85	64.63±4.69	8.31±0.44

Table S5. Photovoltaic parameters of short-circuit photocurrent density (J_{SC}), open-circuit voltage (V_{OC}), fill factor (FF) and power conversion efficiency (PCE) of $CsPbBrI_{1.78}F_{0.22}$ PSCs.

Sample	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
1	1.01	14.94	68	10.26
2	0.96	14.69	67	9.45
3	0.94	14.91	67	9.39
4	0.96	14.45	68	9.53
5	0.95	14.77	66	9.26
6	1.00	13.22	66	8.73
7	1.01	15.18	65	9.97
8	0.98	14.59	65	9.29
9	0.96	13.75	65	8.58
10	0.97	15.63	65	9.85
11	0.95	15.08	64	9.17
12	0.99	15.29	63	9.54
13	1.00	15.54	63	9.79
14	0.90	13.54	67	8.16
15	0.98	14.92	62	9.06
16	1.02	13.86	61	8.62
Ave	0.97±0.03	14.65±0.71	65.13±2.09	9.29±0.56

Table S6. Photovoltaic parameters of short-circuit photocurrent density (J_{SC}), open-circuit voltage (V_{OC}), fill factor (FF) and power conversion efficiency (PCE) of $CsPbBrI_{1.72}F_{0.28}$ PSCs.

Sample	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
1	1.02	13.69	68	9.50
2	0.98	13.13	67	8.62
3	0.97	13.16	67	8.55
4	1.01	12.36	66	8.24
5	0.98	12.90	66	8.34
6	1.01	13.72	67	9.28
7	0.99	12.57	68	8.46
8	0.96	13.36	62	7.95
9	0.98	12.95	69	8.76
10	0.99	12.79	66	8.36
11	0.98	13.78	69	9.32
12	0.99	13.77	68	9.27
13	0.96	13.52	64	8.31
14	1.02	13.67	63	8.78
15	1.00	13.73	65	8.92
16	0.99	12.86	66	8.40
Ave	0.99±0.02	13.25±0.47	66.31±2.02	8.69±0.45

Table S7. Impedance characteristics of $CsPbBrI_{2-x}F_x$ PSCs extracted from the Nyquist plots.

x	R_{SH} (Ω)	R_{CT} (Ω)
0	37.04	58.50
0.12	35.98	57.01
0.22	36.88	42.12
0.28	37.43	92.5