

Quaternary ammonium halide-containing cellulose derivatives for defect passivation in MAPbI₃-based perovskite solar cells

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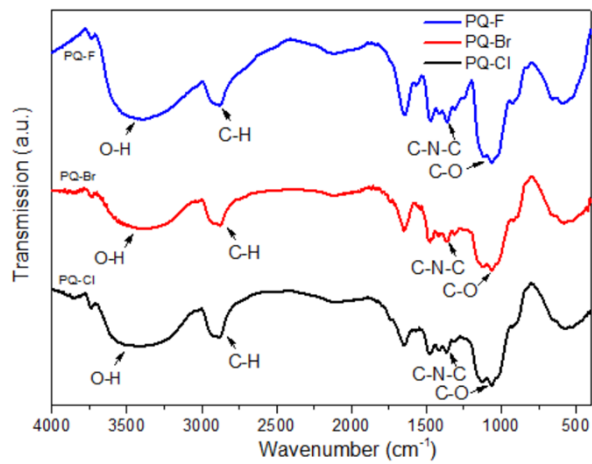


Figure S1. FTIR spectra of PQ-F, PQ-Cl, and PQ-Br.

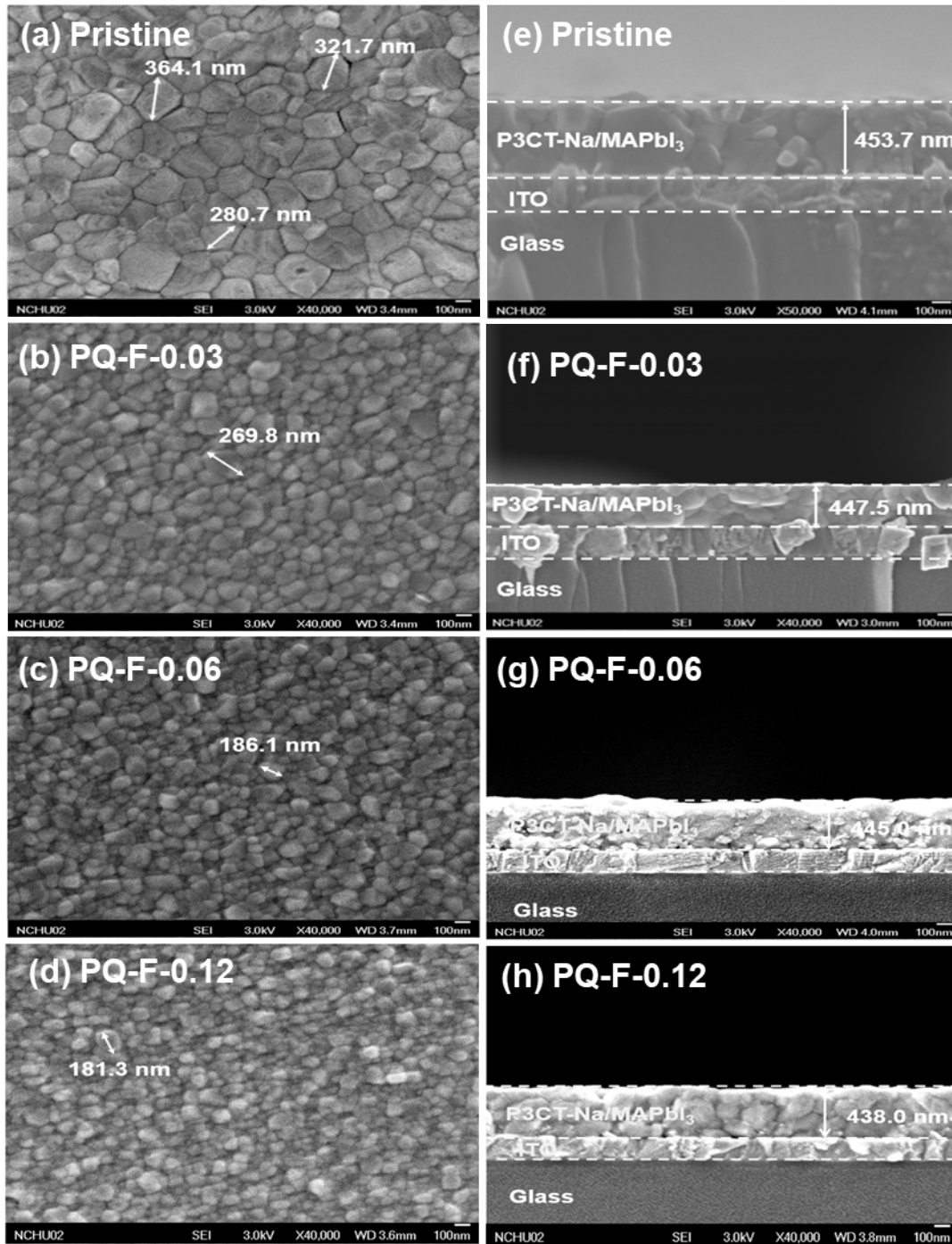


Figure S2. (a–d) Top-view and (e–h) cross-sectional SEM images of PQ-F–doped perovskite films prepared with various PQ-F contents, recorded after they had been subjected to annealing at 100 °C for 10 min.

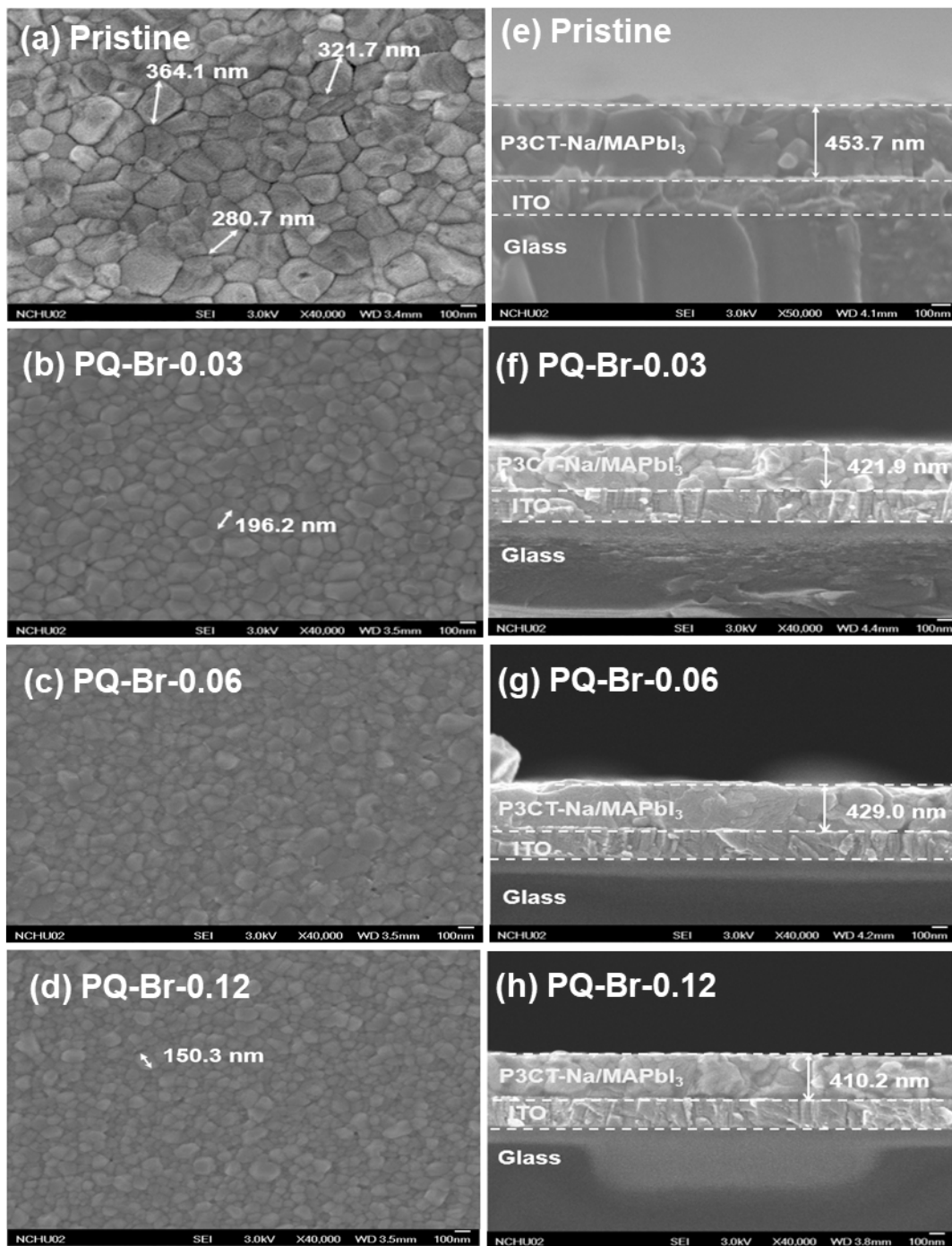


Figure S3. (a–d) Top-view and (e–h) cross-sectional SEM images of PQ-Br-doped perovskite films prepared with various PQ-Br contents, recorded after they had been subjected to annealing at 100 °C for 10 min.

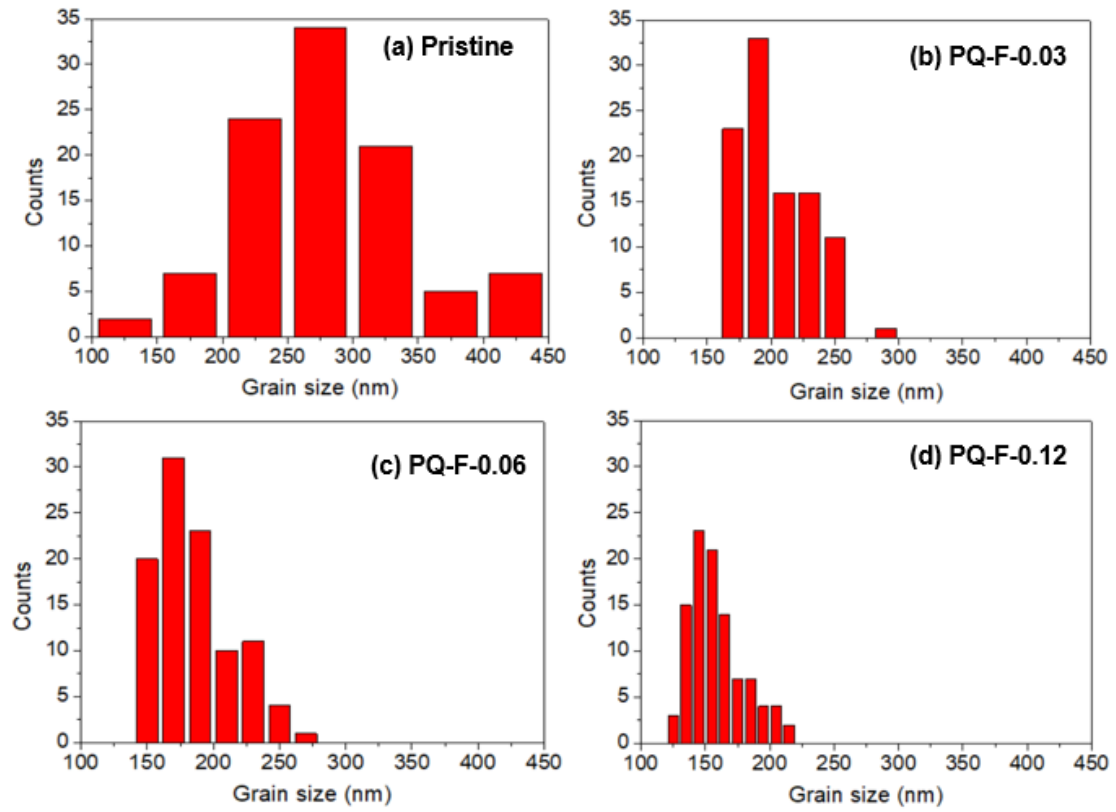


Figure S4. Crystal grain size distributions of PQ-F-doped MAPbI₃ films prepared with various PQ-F contents, measured after they had been subjected to annealing at 100 °C for 10 min.

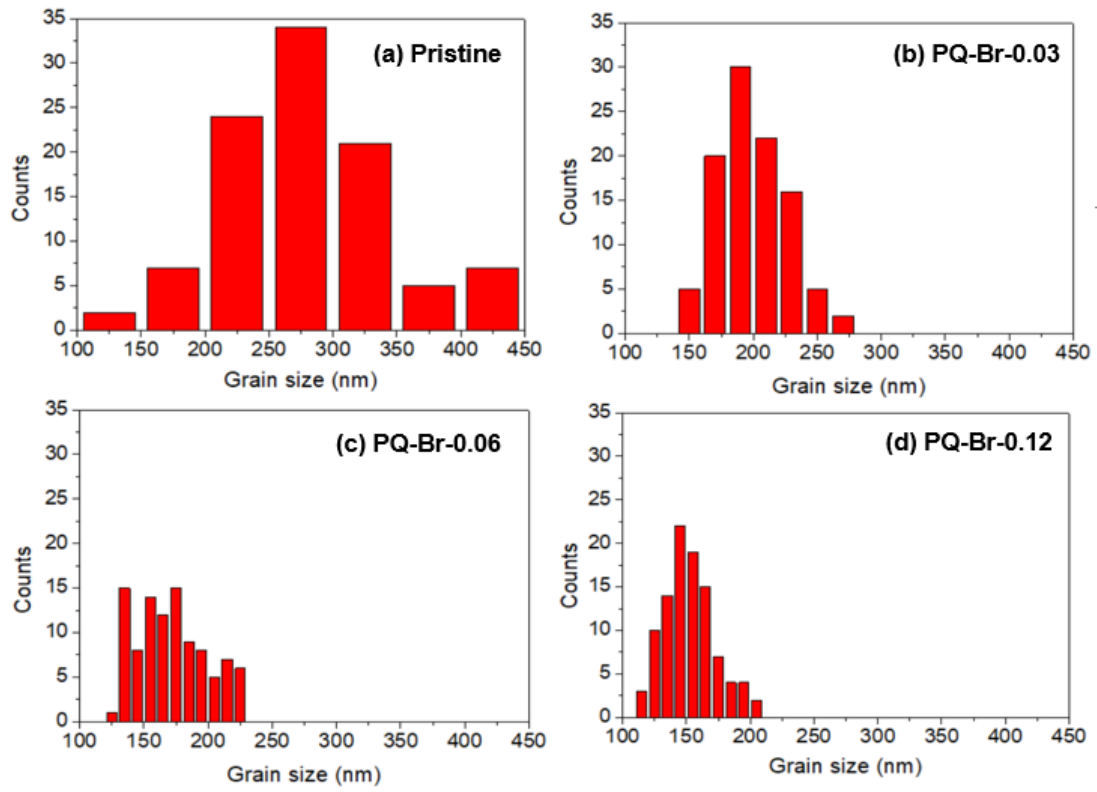


Figure S5. Crystal grain size distributions of PQ-Br-doped MAPbI₃ films prepared with various PQ-Br contents, measured after they had been subjected to annealing at 100 °C for 10 min.

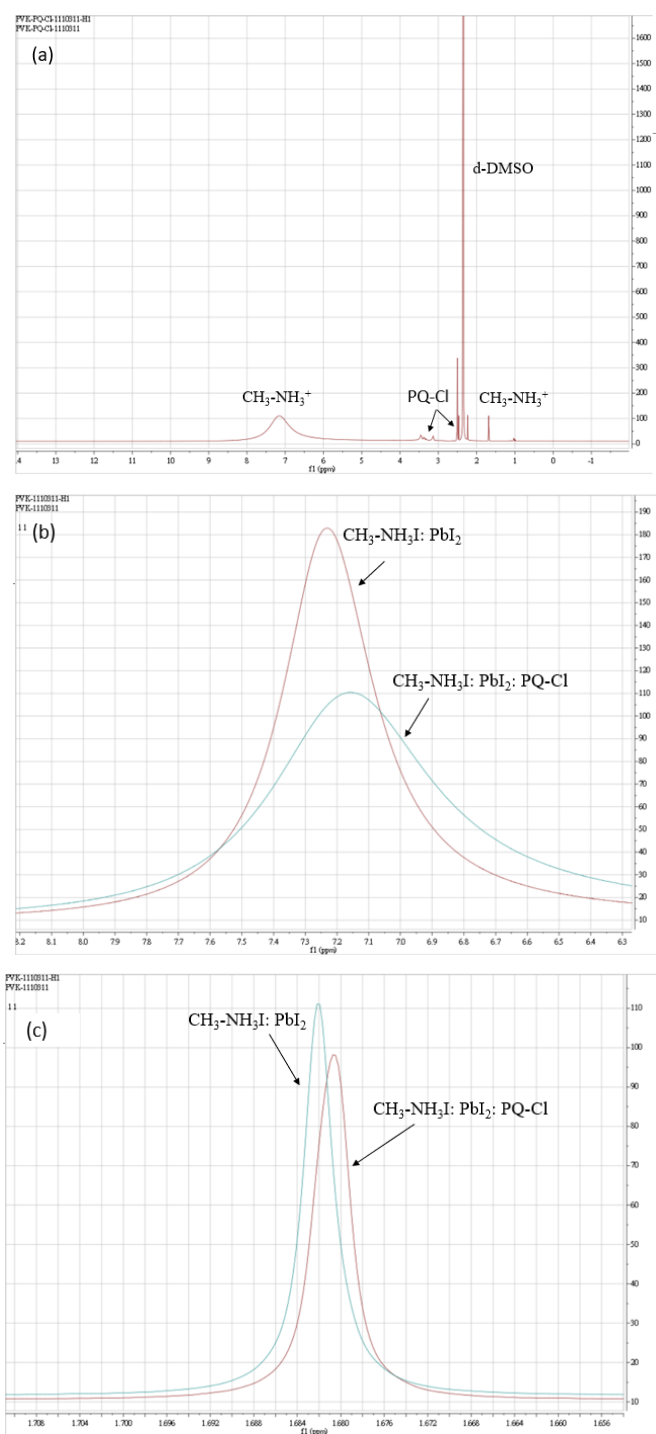


Figure S6. ^1H NMR spectra (solutions in $\text{DMSO-}d_6$) of (a) $\text{CH}_3\text{NH}_3\text{I:PbI}_2\text{:PQ-Cl}$ (PQ-Cl-0.06) and (b, c) $\text{CH}_3\text{NH}_3\text{I:PbI}_2$ (1:1, molar ratio) and PQ-Cl-0.06.

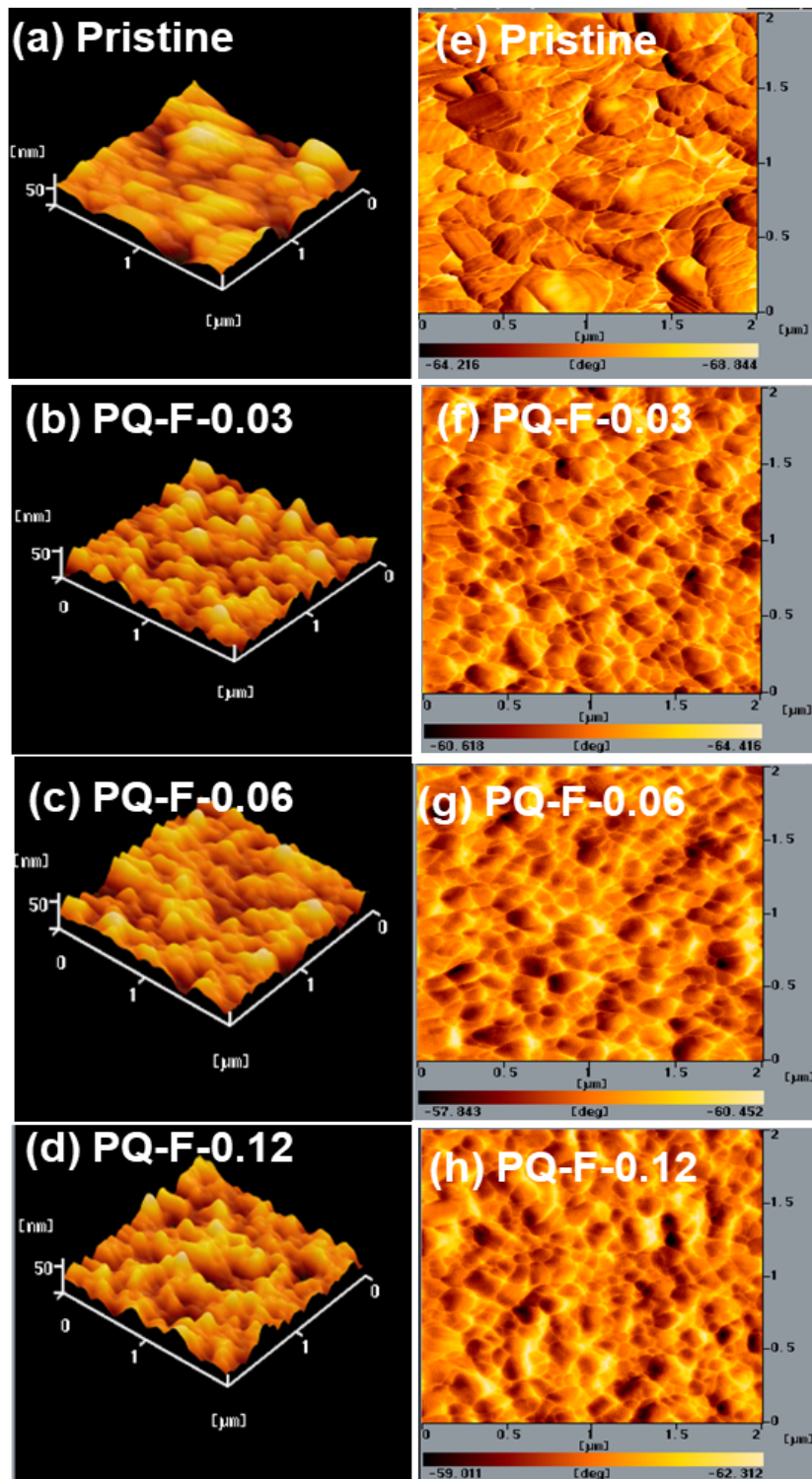


Figure S7. (a–d) Topographic and (e–h) phase AFM images of (a, e) pristine, (b, f) PQ-F-0.03, (c, g) PQ-F-0.03, and (d, h) PQ-F-0.12 perovskite films, recorded after they had been subjected to annealing at 100 °C for 10 min.

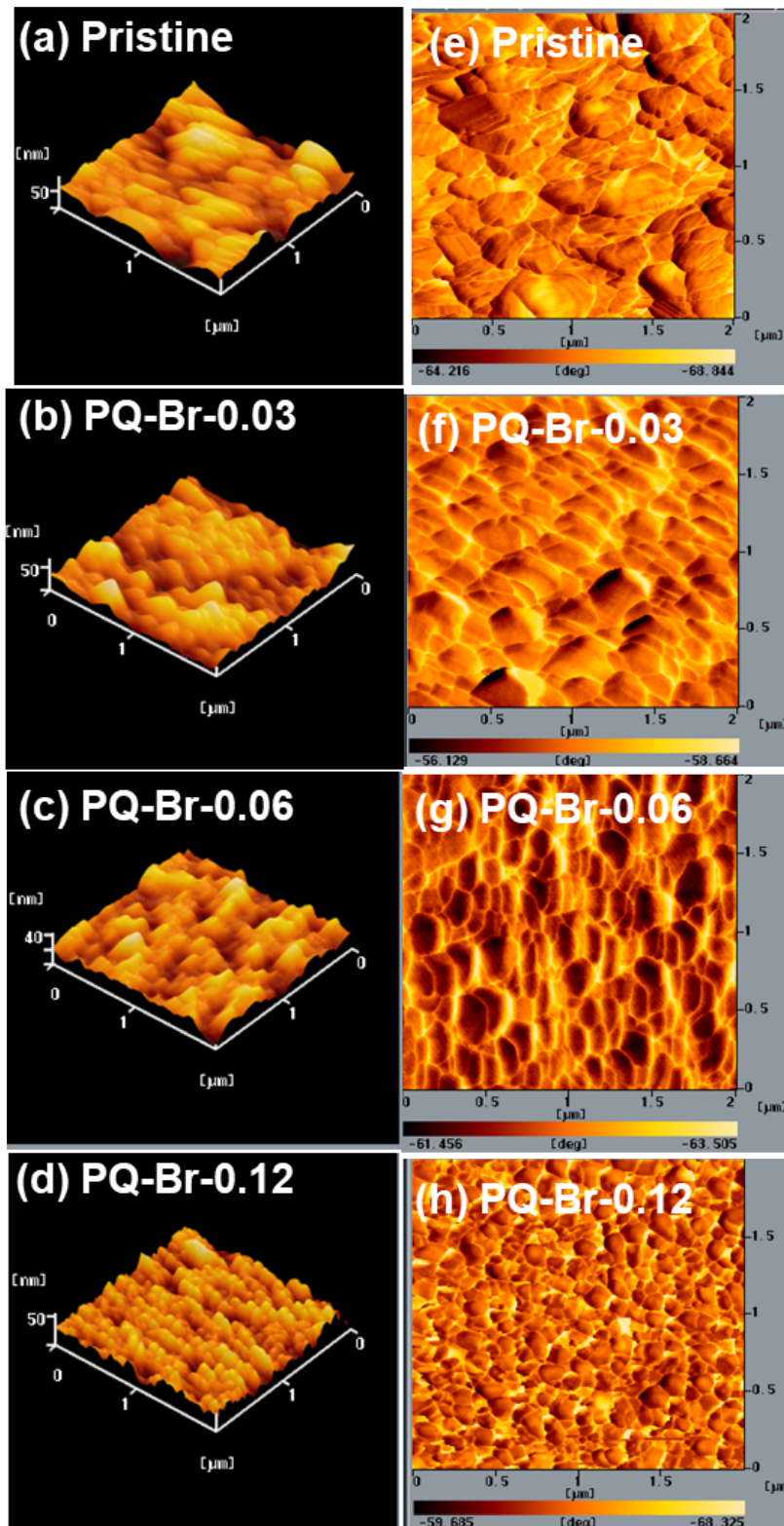


Figure S8. (a–d) Topographic and (e–h) phase AFM images of (a, e) pristine, (b, f) PQ-Br-0.03, (c, g) PQ-Br-0.03, and (d, h) PQ-Br-0.12 perovskite films, recorded after they had been subjected to annealing at 100 °C for 10 min.

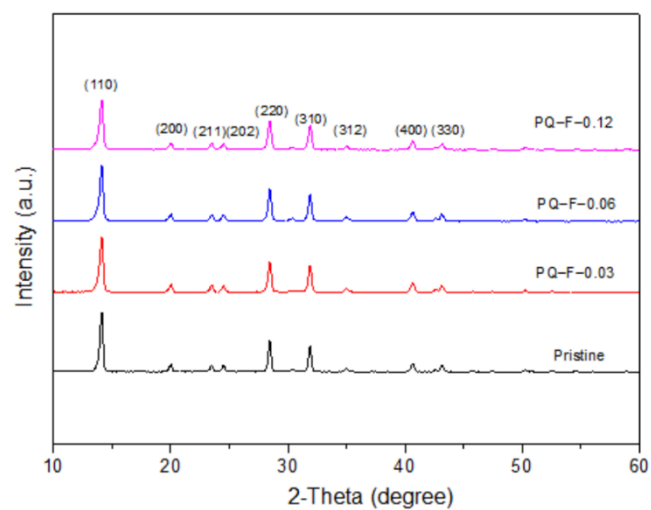


Figure S9. XRD patterns of MAPbI₃ perovskite films doped with various contents of PQ-F, recorded after they had been subjected to annealing at 100 °C for 10 min.

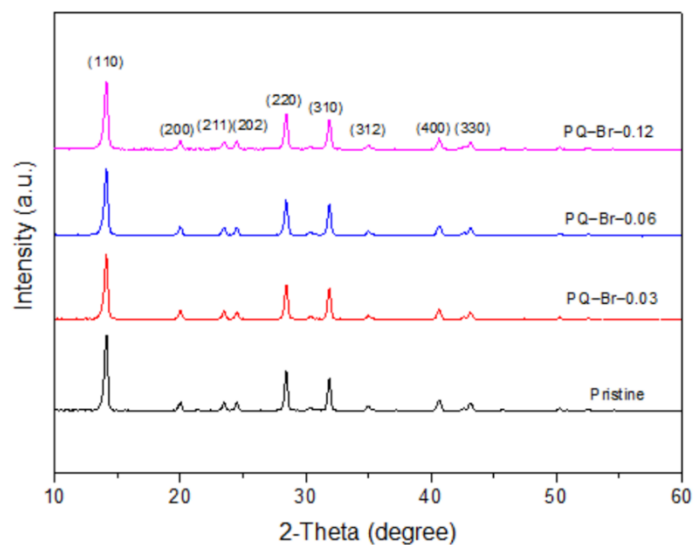


Figure S10. XRD patterns of MAPbI₃ perovskite films doped with various contents of PQ-Br, recorded after they had been subjected to annealing at 100 °C for 10 min.

Table S1. Fitted parameters of TRPL spectra of pristine MAPbI₃ and PQ-F/MAPbI₃, PQ-Cl/MAPbI₃, and PQ-Br/MAPbI₃ composite films coated on P3CT-Na-deposited ITO glass.

Perovskite layer	A_1 (%)	τ_1 (ns)	A_2 (%)	τ_2 (ns)	τ_{avg} (ns)
MAPbI ₃	59.14	4.02	40.86	87.09	81.89
PQ-Cl-0.03	33.73	9.99	66.27	74.19	70.07
PQ-Cl-0.06	46.99	4.34	53.01	69.72	66.31
PQ-Cl-0.12	31.73	3.92	68.27	87.45	85.75
PQ-F-0.03	43.13	5.05	56.87	89.79	82.53
PQ-F-0.06	29.40	2.41	70.60	102.30	101.33
PQ-F-0.12	30.05	2.63	69.95	111.34	110.25
PQ-Br-0.03	38.38	4.23	61.62	87.62	85.19
PQ-Br-0.06	24.79	3.58	75.21	117.17	116.04
PQ-Br-0.12	16.26	4.59	83.74	119.64	118.79

Table S2. PV parameters of pristine MAPbI₃-based PVSC I without encapsulation, recorded after various storage times in air.

Storage time (h)	Photoactive layer	V_{OC} (V)	J_{SC} (mA cm ⁻²)	FF (%)	PCE (%)
0	MAPbI ₃	1.02	18.12	73.1	13.51
24	MAPbI ₃	1.02	18.07	73.0	13.46
48	MAPbI ₃	1.02	18.09	73.0	13.48
72	MAPbI ₃	1.02	18.10	73.1	13.49
102	MAPbI ₃	1.01	18.15	73.2	13.42
126	MAPbI ₃	1.01	18.00	72.1	13.11
146	MAPbI ₃	1.01	17.93	72.0	13.04
240	MAPbI ₃	1.00	17.61	71.0	12.49
266	MAPbI ₃	1.00	17.23	71.0	12.23
314	MAPbI ₃	1.00	17.01	70.8	12.00
338	MAPbI ₃	1.00	16.82	70.4	11.84
362	MAPbI ₃	1.00	16.64	70.2	11.68
386	MAPbI ₃	0.99	16.56	69.6	11.41
510	MAPbI ₃	0.99	16.45	69.5	11.32
434	MAPbI ₃	0.99	16.41	69.5	11.29
458	MAPbI ₃	0.99	16.33	69.3	11.20
482	MAPbI ₃	0.98	15.96	69.2	10.82
506	MAPbI ₃	0.98	15.85	69.2	10.75
530	MAPbI ₃	0.98	15.61	68.9	10.54
554	MAPbI ₃	0.97	15.44	68.5	10.26
578	MAPbI ₃	0.97	15.40	68.5	10.23
602	MAPbI ₃	0.96	15.10	68.0	9.86
626	MAPbI ₃	0.96	14.97	67.7	9.73
650	MAPbI ₃	0.94	14.80	67.0	9.32
674	MAPbI ₃	0.92	14.07	66.8	8.65

Table S3. PV parameters of PQ-C1-0.06–based PVSC without encapsulation, recorded after various storage times in air.

Storage time (h)	Photoactive layer	V_{OC} (V)	J_{SC} (mA cm ⁻²)	FF (%)	PCE (%)
0	PQ-C1-0.06	1.00	19.46	76.6	14.91
25	PQ-C1-0.06	1.00	19.45	76.6	14.89
40	PQ-C1-0.06	1.00	19.44	76.5	14.87
49	PQ-C1-0.06	1.00	19.42	76.5	14.86
97	PQ-C1-0.06	1.00	19.41	76.5	14.85
168	PQ-C1-0.06	1.00	19.40	76.4	14.83
216	PQ-C1-0.06	1.00	19.32	76.4	14.76
360	PQ-C1-0.06	1.00	19.31	76.4	14.75
504	PQ-C1-0.06	0.99	19.25	76.3	14.52
528	PQ-C1-0.06	0.99	19.02	76.0	14.31
574	PQ-C1-0.06	0.99	18.83	75.6	14.08
598	PQ-C1-0.06	0.98	18.76	75.4	13.86
622	PQ-C1-0.06	0.98	18.46	75.0	13.57
676	PQ-C1-0.06	0.98	18.34	74.9	13.46
816	PQ-C1-0.06	0.97	18.09	74.6	13.09
864	PQ-C1-0.06	0.97	17.86	74.5	12.91
888	PQ-C1-0.06	0.97	17.69	74.1	12.72
912	PQ-C1-0.06	0.97	17.39	73.8	12.45
936	PQ-C1-0.06	0.96	17.32	73.7	12.26
960	PQ-C1-0.06	0.96	17.20	73.4	12.12
984	PQ-C1-0.06	0.96	17.14	73.4	12.08
1008	PQ-C1-0.06	0.96	17.02	73.3	12.00

Table S4. PV parameters of PQ-F-0.03–based PVSCs without encapsulation, recorded after various storage times in air.

Storage time (h)	Photoactive layer	V_{OC} (V)	J_{SC} (mA cm ⁻²)	FF (%)	PCE (%)
0	PQ-F-0.03	0.99	14.90	75.4	11.12
24	PQ-F-0.03	0.99	14.89	75.4	11.12
48	PQ-F-0.03	0.99	14.88	75.4	11.11
72	PQ-F-0.03	0.99	14.87	75.4	11.11
102	PQ-F-0.03	0.99	14.87	75.4	11.10
126	PQ-F-0.03	0.99	14.86	75.4	11.09
146	PQ-F-0.03	0.99	14.85	75.3	11.07
240	PQ-F-0.03	0.99	14.76	75.3	11.01
266	PQ-F-0.03	0.98	14.57	75.0	10.71
314	PQ-F-0.03	0.96	14.42	74.6	10.33
338	PQ-F-0.03	0.96	14.32	74.1	10.19
362	PQ-F-0.03	0.95	13.53	73.6	9.46
410	PQ-F-0.03	0.94	12.43	73.2	8.55
434	PQ-F-0.03	0.94	12.10	72.7	8.27
458	PQ-F-0.03	0.94	11.91	72.2	8.08
482	PQ-F-0.03	0.93	10.90	71.1	7.21
506	PQ-F-0.03	0.93	10.25	70.7	6.74

Table S5. PV parameters of PQ-Br-0.03–based PVSCs without encapsulation, recorded after various storage times in air.

Storage time (h)	Photoactive layer	V_{OC} (V)	J_{SC} (mA cm ⁻²)	FF (%)	PCE (%)
0	PQ-Br-0.03	0.99	14.10	75.9	10.59
24	PQ-Br-0.03	0.99	14.09	75.9	10.59
48	PQ-Br-0.03	0.99	14.08	75.9	10.58
72	PQ-Br-0.03	0.99	14.07	75.9	10.57
102	PQ-Br-0.03	0.99	14.07	75.9	10.57
126	PQ-Br-0.03	0.99	14.06	75.9	10.56
146	PQ-Br-0.03	0.99	14.05	75.8	10.55
240	PQ-Br-0.03	0.98	13.65	75.0	10.03
266	PQ-Br-0.03	0.98	13.56	74.9	9.95
314	PQ-Br-0.03	0.98	13.48	74.7	9.87
338	PQ-Br-0.03	0.98	13.39	74.6	9.79
362	PQ-Br-0.03	0.98	13.27	74.3	9.66
410	PQ-Br-0.03	0.97	13.03	74.2	9.38
434	PQ-Br-0.03	0.97	12.90	74.0	9.26
458	PQ-Br-0.03	0.97	12.65	73.8	9.06
482	PQ-Br-0.03	0.97	12.14	73.7	8.68
506	PQ-Br-0.03	0.97	11.99	73.6	8.56
530	PQ-Br-0.03	0.96	11.38	73.3	8.00
554	PQ-Br-0.03	0.96	10.66	72.7	7.44
578	PQ-Br-0.03	0.95	9.53	71.4	6.46
602	PQ-Br-0.03	0.95	8.79	70.2	5.86