Supplementary Information

Improved Light Absorption and Charge Transport for Perovskite Solar Cells with Rough Interfaces by Sequential Deposition

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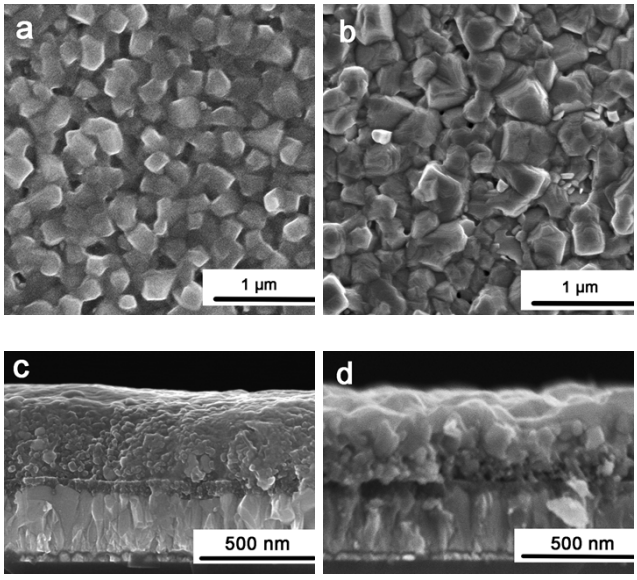
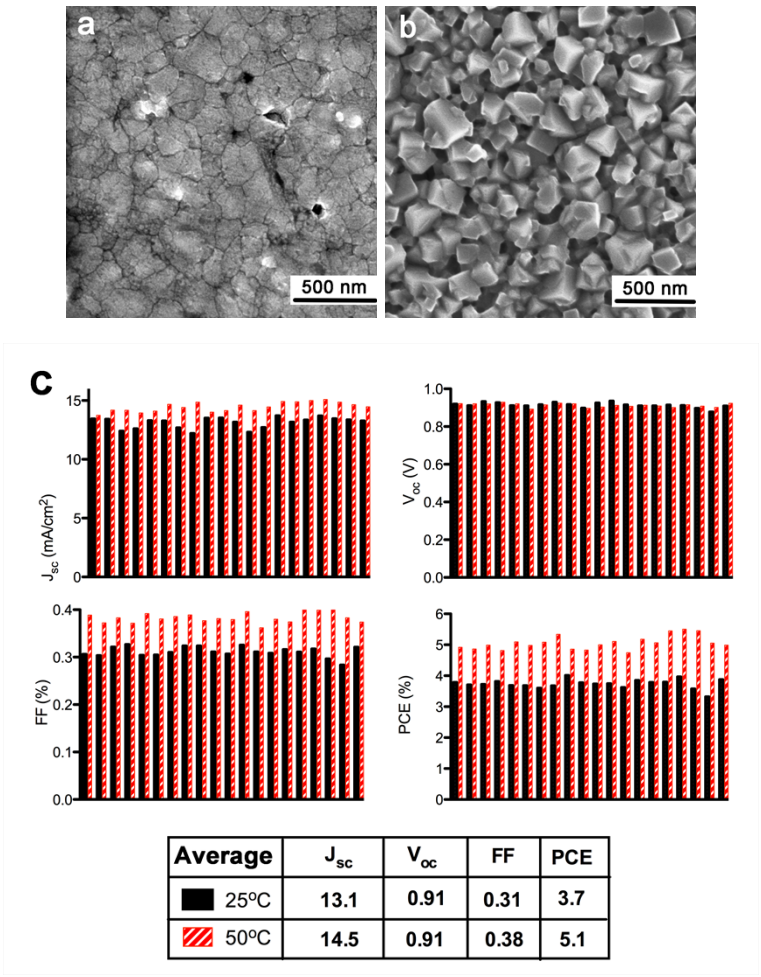
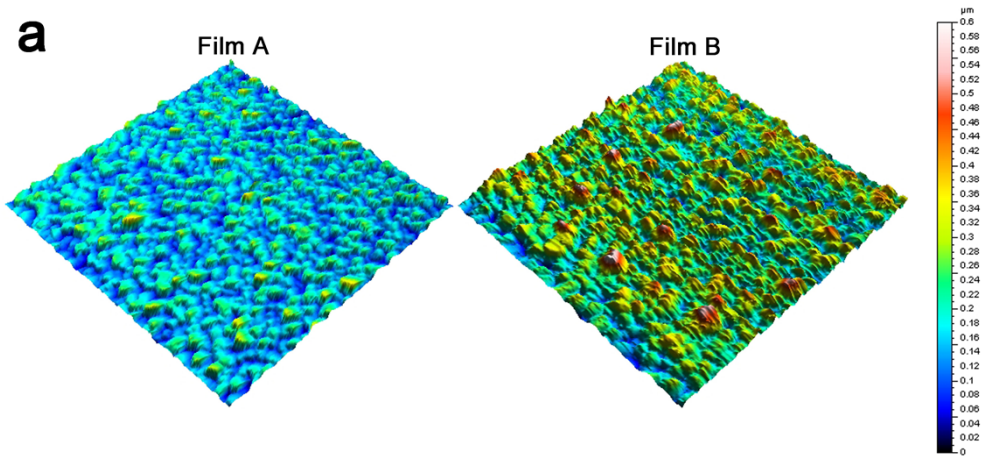


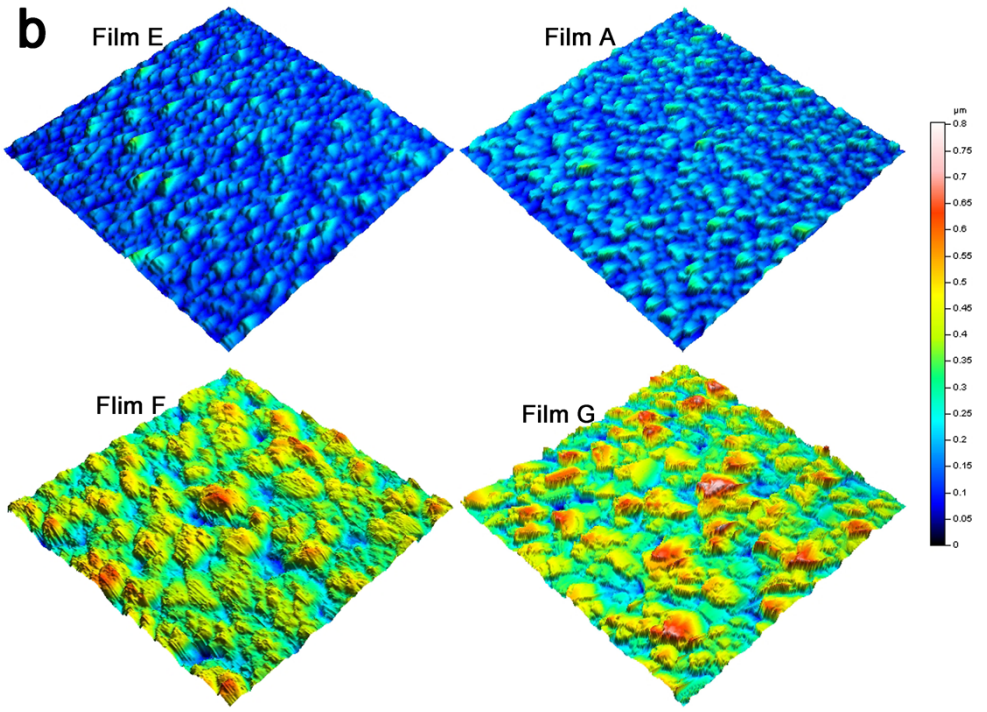
Figure S1. Top-view and cross-sectional SEM images of the $CH_3NH_3PbI_{3-x}Cl_x$ (x≈0.5) on top of the mesoporous TiO_2 by reacting a) and c) at 25 °C, b) and d) at 50 °C.



Average	J_{sc}	V_{oc}	FF	PCE
25ºC	13.1	0.91	0.31	3.7
50ºC	14.5	0.91	0.38	5.1

Figure S2. Top-view SEM images of $CH_3NH_3PbI_3$ on top of the compact TiO_2 by reacting a) at 25 °C, b) at 50 °C. c) Histogram plots of device performance parameters and the average performance for 20 individual cells by different reaction temperature based on the structure: FTO/compact TiO_2/$CH_3NH_3PbI_3$/spiro-MeOTAD/Ag. The thickness of each layer is in accordance with devices including mesoporous TiO_2, without further optimizing.





c

Device	A	B	E	F	G
Root Mean Square Roughness (nm)	44.5	78.2	44.1	87.8	118

Figure S3. 3D topographic images (10 × 10 µm) of a) Film A and B (scale bar=0.6 µm) and b) Film E, A, F and G (scale bar=0.8 µm) by tapping-mode atomic force microscopy. c) Surface roughness of the corresponding films.

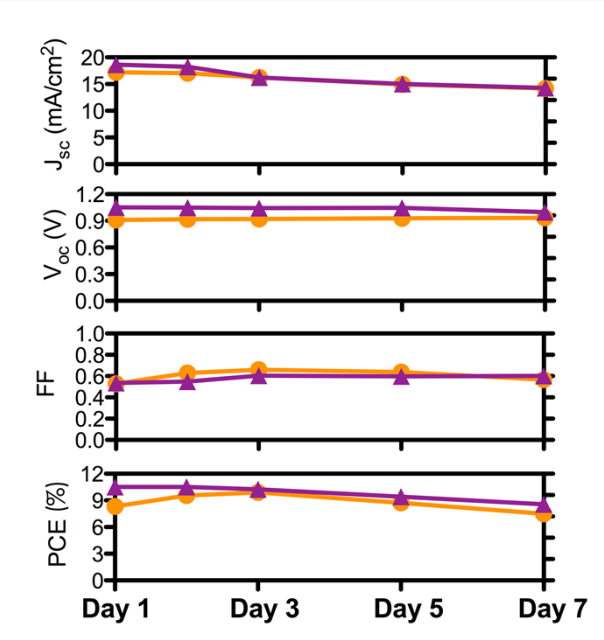


Figure S4. Evolution of photovoltaic parameters under 100 mW cm^{-2} simulated AM1.5G irradiation. Device B (orange) and Device D (purple) were stored in air at room temperature in the dark (humidity: 10%~40%) without encapsulation.

Table S1. Photovoltaic parameters of a batch of twenty Device A-F measured under 100 mW cm^{-2} simulated AM1.5G irradiation.

Device A	V_{oc} (V)	J_{sc} (mA cm^{-2})	*FF*	PCE (%)	Device B	V_{oc} (V)	J_{sc} (mA cm^{-2})	*FF*	PCE (%)
1	0.94	14.0	0.54	7.2	1	0.93	16.0	0.52	7.7
2	0.91	15.1	0.53	7.2	2	0.93	16.1	0.52	7.7
3	0.91	15.1	0.57	7.8	3	0.92	16.0	0.53	7.8
4	0.91	15.9	0.57	8.3	4	0.93	16.1	0.52	7.7
5	0.91	15.3	0.55	7.6	5	0.90	16.5	0.52	7.7
6	0.92	14.6	0.55	7.4	6	0.92	16.1	0.53	7.8
7	0.90	16.0	0.54	7.8	7	0.92	16.4	0.52	7.9
8	0.91	14.4	0.54	7.2	8	0.95	16.6	0.55	8.7
9	0.92	15.8	0.51	7.4	9	0.92	16.0	0.53	7.9
10	0.91	15.4	0.52	7.3	10	0.91	16.7	0.52	7.9
11	0.91	16.0	0.56	8.2	11	0.91	16.5	0.52	7.8
12	0.90	15.7	0.55	7.8	12	0.91	16.8	0.52	8.0
13	0.91	15.9	0.54	7.7	13	0.90	16.7	0.52	7.8
14	0.91	15.9	0.55	8.0	14	0.92	16.4	0.53	8.0
15	0.91	15.2	0.59	8.1	15	0.94	15.7	0.52	7.7
16	0.91	15.6	0.60	8.5	16	0.92	16.6	0.53	8.2
17	0.91	15.3	0.52	7.3	17	0.91	16.6	0.53	8.0
18	0.91	14.7	0.54	7.3	18	0.94	16.2	0.56	8.4
19	0.93	15.1	0.56	7.8	19	0.93	15.9	0.53	7.8
20	0.93	15.2	0.50	7.1	20	0.93	15.8	0.53	7.8

Average	0.91	15.3	0.55	7.6

21	0.91	17.7	0.63	10.2
22	0.91	16.3	0.69	10.2
23	0.94	18.1	0.59	10.0
24	0.92	16.2	0.66	9.9
25	0.92	17.1	0.61	9.6
26	0.94	16.0	0.64	9.6
27	0.89	16.8	0.63	9.4
28	0.93	16.5	0.59	9.1
29	0.91	17.0	0.57	8.8
30	0.94	17.5	0.52	8.5
31	0.94	17.3	0.52	8.4
32	0.94	17.5	0.51	8.4
33	0.91	17.2	0.53	8.3
34	0.93	15.8	0.61	9.0
35	0.93	15.7	0.63	9.1
36	0.92	15.6	0.60	8.5
37	0.91	15.9	0.57	8.3
38	0.90	16.3	0.56	8.3
39	0.91	16.1	0.56	8.1
40	0.91	16.0	0.57	8.2
Average	0.92	16.5	0.56	8.4

Device C	V_{oc} (V)	J_{sc} (mA cm^{-2})	*FF*	PCE (%)
1	0.97	15.4	0.58	8.6
2	0.97	15.3	0.55	8.2
3	1.00	15.5	0.57	8.8
4	0.96	15.7	0.55	8.3
5	0.98	15.3	0.56	8.3
6	0.97	15.2	0.54	7.9
7	0.99	15.1	0.57	8.5
8	1.01	15.1	0.60	9.2
9	0.99	15.6	0.57	8.9
10	0.97	16.2	0.57	8.9
11	0.96	16.0	0.56	8.6
12	0.96	15.2	0.56	8.2
13	0.97	15.5	0.58	8.7
14	0.97	15.3	0.57	8.5
15	0.96	15.5	0.57	8.5
16	0.98	15.1	0.58	8.5
17	1.02	15.3	0.57	8.9
18	0.96	15.5	0.56	8.3
19	0.95	15.8	0.57	8.5
20	0.95	15.9	0.55	8.3
Average	0.97	15.5	0.57	8.5
22	1.04	17.6	0.58	10.7
23	1.04	16.3	0.61	10.2
24	1.04	16.2	0.56	9.5
25	0.97	15.9	0.60	9.2
Average	0.99	17.2	0.56	9.5

Device D	V_{oc} (V)	J_{sc} (mA cm^{-2})	*FF*	PCE (%)
1	0.96	16.8	0.53	8.6
2	1.05	18.7	0.54	10.5
3	0.94	17.6	0.52	8.6
4	0.97	17.7	0.55	9.5
5	1.05	18.2	0.55	10.5
6	1.00	18.2	0.59	10.8
7	0.99	19.0	0.56	10.5
8	1.00	15.6	0.57	9.2
9	1.00	15.5	0.57	8.8
10	1.04	15.4	0.58	9.3
11	0.96	18.1	0.55	9.5
12	0.96	17.7	0.56	9.5
13	0.95	17.2	0.55	9.0
14	0.97	16.9	0.55	9.0
15	0.96	16.7	0.53	8.4
16	0.96	17.1	0.54	8.9
17	0.96	16.0	0.56	8.7
18	0.96	16.7	0.56	9.0
19	0.93	18.2	0.55	9.3
20	0.98	18.0	0.56	9.8
21	0.99	17.8	0.52	9.1

Device E	V_{oc} (V)	J_{sc} (mA cm^{-2})	*FF*	PCE (%)
1	0.91	15.5	0.43	6.1
2	0.95	15.0	0.46	6.5
3	0.95	14.4	0.46	6.3
4	0.95	13.8	0.48	6.3
5	0.94	14.2	0.46	6.1
6	0.94	14.6	0.47	6.4
7	0.93	14.9	0.45	6.3
8	0.94	14.9	0.46	6.4
9	0.93	15.1	0.46	6.4
10	0.95	14.2	0.46	6.2
11	0.93	14.4	0.46	6.1
12	0.92	15.2	0.43	6.0

Device F	V_{oc} (V)	J_{sc} (mA cm^{-2})	*FF*	PCE (%)
1	0.80	18.5	0.52	7.6
2	0.90	18.9	0.46	7.8
3	0.89	18.3	0.45	7.4
4	0.90	18.6	0.45	7.6
5	0.90	17.4	0.45	7.1
6	0.90	18.8	0.44	7.5
7	0.90	17.7	0.46	7.3
8	0.89	17.6	0.46	7.2
9	0.89	17.4	0.47	7.2
10	0.89	18.0	0.47	7.5
11	0.89	17.6	0.47	7.4
12	0.89	18.5	0.47	7.7

13	0.92	14.6	0.44	6.0	13	0.89	16.7	0.50	7.4
14	0.91	14.9	0.44	6.0	14	0.89	16.3	0.49	7.1
15	0.91	15.1	0.45	6.2	15	0.89	16.0	0.49	7.0
16	0.95	14.4	0.46	6.4	16	0.88	16.5	0.50	7.3
17	0.94	15.3	0.46	6.6	17	0.90	16.3	0.49	7.1
18	0.93	15.3	0.45	6.4	18	0.86	17.3	0.49	7.3
19	0.90	15.8	0.44	6.2	19	0.85	17.7	0.49	7.4
20	0.96	14.4	0.43	5.9	20	0.87	16.8	0.49	7.1
Average	0.93	14.8	0.45	6.2	**Average**	0.88	17.5	0.48	7.4

Device G	V_{oc} **(V)**	J_{sc} **(mA cm^{-2})**	***FF***	**PCE (%)**
1	0.82	16.2	0.44	5.8
2	0.77	17.2	0.51	6.8
3	0.75	15.8	0.53	6.3
4	0.73	16.2	0.52	6.2
5	0.73	15.6	0.51	5.8
6	0.72	14.7	0.48	5.0
7	0.71	15.4	0.49	5.3
8	0.72	16.4	0.49	5.8
9	0.71	17.0	0.49	6.0
10	0.70	16.5	0.48	5.6
11	0.82	16.2	0.43	5.8
12	0.72	15.6	0.51	5.7
13	0.80	17.1	0.45	6.2
14	0.77	15.5	0.43	5.1
15	0.70	15.5	0.48	5.3
16	0.69	15.3	0.48	5.1
17	0.75	14.5	0.50	5.4
18	0.74	15.6	0.50	5.8
19	0.74	15.6	0.48	5.5
20	0.73	15.1	0.52	5.8
Average	0.74	15.8	0.49	5.7