Supporting information

Performance optimization of perovskite solar cells with automated spin-coating system and artificial intelligence technologies

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Table S1. List of experimental conditions for the initial data and the experimental conditions proposed by Bayesian optimization and each solar cell parameters.

Entry	Cycle	Spin coating speed (rpm)	Anneal temperature (℃)	MACl (mol%)	PbI_2 (mol%)	Jsc_forward (mA/cm2)	Jsc_reverse (mA/cm2)	Voc_forward (V)	Voc_reverse (V)	FF_forward	FF_reverse	PCE_forward (%)	PCE_reverse (%)
1	Initial	2000	70	0	80	3.4	3.5	0.55	0.23	0.47	0.43	0.9	0.4
2	Initial	4000	70	0	80	11.9	14.6	0.76	0.52	0.20	0.23	1.8	1.7
3	Initial	6000	70	40	80	0.9	1.0	0.43	0.26	0.48	0.38	0.2	0.1
4	Initial	2000	70	0	140	12.3	10.8	0.67	0.33	0.16	0.33	1.4	1.2
5	Initial	4000	70	0	140	4.0	4.5	0.63	0.26	0.24	0.33	0.6	0.4
6	Initial	2000	70	80	140	1.5	1.5	0.43	0.08	0.49	0.33	0.3	0.0
7	Initial	6000	70	80	140	14.9	15.7	0.74	0.74	0.26	0.17	2.9	1.9
8	Initial	2000	110	0	80	14.8	13.0	0.68	0.69	0.24	0.27	2.4	2.4
9	Initial	4000	110	0	80	4.0	4.1	0.62	0.25	0.42	0.47	1.0	0.5
10	Initial	6000	110	40	80	1.2	1.1	0.47	0.09	0.54	0.41	0.3	0.0
11	Initial	2000	110	0	140	19.6	19.4	0.76	0.80	0.26	0.40	3.9	6.3
12	Initial	4000	110	0	140	15.8	16.1	0.65	0.40	0.17	0.42	1.7	2.7
13	Initial	6000	110	80	140	4.5	5.1	0.50	0.19	0.35	0.32	0.8	0.3
14	Initial	2000	150	0	80	7.1	5.5	0.60	0.53	0.22	0.31	1.0	0.9
15	Initial	4000	150	0	80	15.6	14.6	0.60	0.60	0.27	0.23	2.5	2.0
16	Initial	6000	150	0	80	13.9	13.3	0.59	0.59	0.23	0.21	1.9	1.6
17	Initial	6000	150	40	80	5.9	5.3	0.55	0.44	0.29	0.34	1.0	0.8
18	Initial	2000	150	0	140	21.9	21.7	0.81	0.62	0.43	0.43	7.6	5.8
19	Initial	4000	150	0	140	22.6	22.4	0.76	0.78	0.34	0.39	5.8	6.8
20	Initial	6000	150	80	140	23.7	23.7	0.91	0.92	0.45	0.58	9.7	12.6
21	1st	5500	150	80	140	23.3	23.3	0.96	0.97	0.43	0.53	9.6	12.0
22	1st	6000	150	80	135	23.1	23.1	0.88	0.91	0.62	0.68	12.5	14.1
23	1st	6000	150	75	130	23.2	23.2	0.87	0.89	0.61	0.69	12.3	14.3
24	1st	5500	150	75	125	23.2	23.2	0.86	0.89	0.61	0.69	12.1	14.3
25	1st	6000	150	80	130	23.3	23.3	0.85	0.88	0.63	0.70	12.5	14.3
26	2nd	5000	150	50	130	23.2	23.2	1.06	1.09	0.65	0.76	15.8	19.0
27	2nd	5500	150	75	130	23.1	23.1	0.86	0.87	0.74	0.70	14.6	14.1
28	2nd	6000	150	70	130	23.1	23.1	0.83	0.83	0.73	0.74	14.0	14.2
29	2nd	5500	150	80	130	23.2	23.3	0.87	0.87	0.73	0.73	14.8	14.8
30	2nd	5500	150	80	120	23.0	22.9	0.75	0.74	0.67	0.65	11.4	11.1
31	3rd	4500	150	45	125	23.1	23.0	1.04	1.08	0.61	0.75	14.6	18.6
32	3rd	4500	150	50	135	23.0	23.0	1.04	1.08	0.58	0.72	14.0	18.1

33	3rd	5000	140	40	130	23.0	22.9	1.06	1.09	0.59	0.77	14.3	19.2
34	3rd	4500	150	50	130	23.4	23.4	1.06	1.09	0.63	0.72	15.6	18.4
35	3rd	5000	150	50	135	23.3	23.2	1.08	1.11	0.55	0.74	13.9	19.2
36	4th	5000	150	35	130	23.4	23.3	1.06	1.10	0.60	0.78	14.7	20.0
37	4th	5000	150	40	135	23.3	23.3	1.08	1.11	0.60	0.76	15.0	19.7
38	4th	2500	150	30	125	22.5	22.5	0.49	0.59	0.48	0.44	5.3	5.8
39	4th	5000	140	50	130	23.5	23.4	1.03	1.05	0.73	0.77	17.6	18.9
40	4th	5000	150	40	130	23.4	23.4	1.04	1.06	0.70	0.79	17.0	19.6
41	5th	5500	130	5	115	23.5	23.5	0.99	1.01	0.50	0.72	11.6	17.1
42	5th	5000	150	45	130	23.3	23.2	1.03	1.05	0.70	0.79	16.9	19.4
43	5th	5000	150	45	135	23.4	23.3	1.05	1.07	0.69	0.79	17.0	19.7
44	5th	3000	70	80	100	1.6	1.1	0.39	0.15	0.68	0.51	0.4	0.1
45	5th	5000	150	35	135	23.4	23.4	1.07	1.09	0.68	0.79	16.9	20.1
46	6th	6000	130	25	140	23.3	23.3	1.06	1.09	0.61	0.77	15.1	19.6
47	6th	4500	130	20	115	23.7	23.7	0.89	0.92	0.57	0.70	12.1	15.1
48	6th	6000	150	20	135	23.4	23.5	1.10	1.12	0.64	0.79	16.4	20.9
49	6th	5000	150	30	130	23.4	23.3	1.10	1.11	0.68	0.81	17.3	20.9
50	6th	6000	150	0	115	23.8	23.7	1.07	1.08	0.67	0.74	17.1	18.9
51	7th	6000	140	25	125	23.6	23.5	1.10	1.12	0.70	0.81	18.0	21.4
52	7th	6000	150	35	140	23.5	23.5	1.08	1.12	0.57	0.77	14.4	20.4
53	7th	6000	130	20	125	23.4	23.4	1.02	1.05	0.67	0.75	16.0	18.4
54	7th	6000	150	20	130	23.6	23.5	1.07	1.11	0.61	0.79	15.4	20.6
55	7th	6000	150	25	135	23.6	23.5	1.09	1.12	0.62	0.77	16.0	20.2
56	8th	6000	70	25	140	23.0	22.9	1.07	1.09	0.62	0.73	15.2	18.3
57	8th	6000	140	20	135	23.1	23.1	1.12	1.14	0.61	0.79	15.7	20.7
58	8th	6000	150	30	130	23.0	23.0	1.10	1.13	0.66	0.79	16.8	20.6
59	8th	5500	150	35	130	23.0	23.0	1.09	1.12	0.64	0.79	16.0	20.3
60	8th	6000	150	25	125	23.1	23.1	1.09	1.11	0.64	0.79	16.1	20.4
61	9th	5500	150	30	130	24.2	24.1	1.08	1.11	0.61	0.79	16.1	21.1
62	9th	3500	120	80	80	0.2	0.1	0.33	0.11	0.28	0.41	0.0	0.0
63	9th	6000	150	15	130	24.4	24.3	1.07	1.10	0.57	0.76	14.9	20.4
64	9th	5500	150	30	125	24.3	24.3	1.07	1.09	0.69	0.77	18.0	20.5
65	9th	6000	150	25	130	24.2	24.2	1.08	1.12	0.62	0.78	16.3	21.1

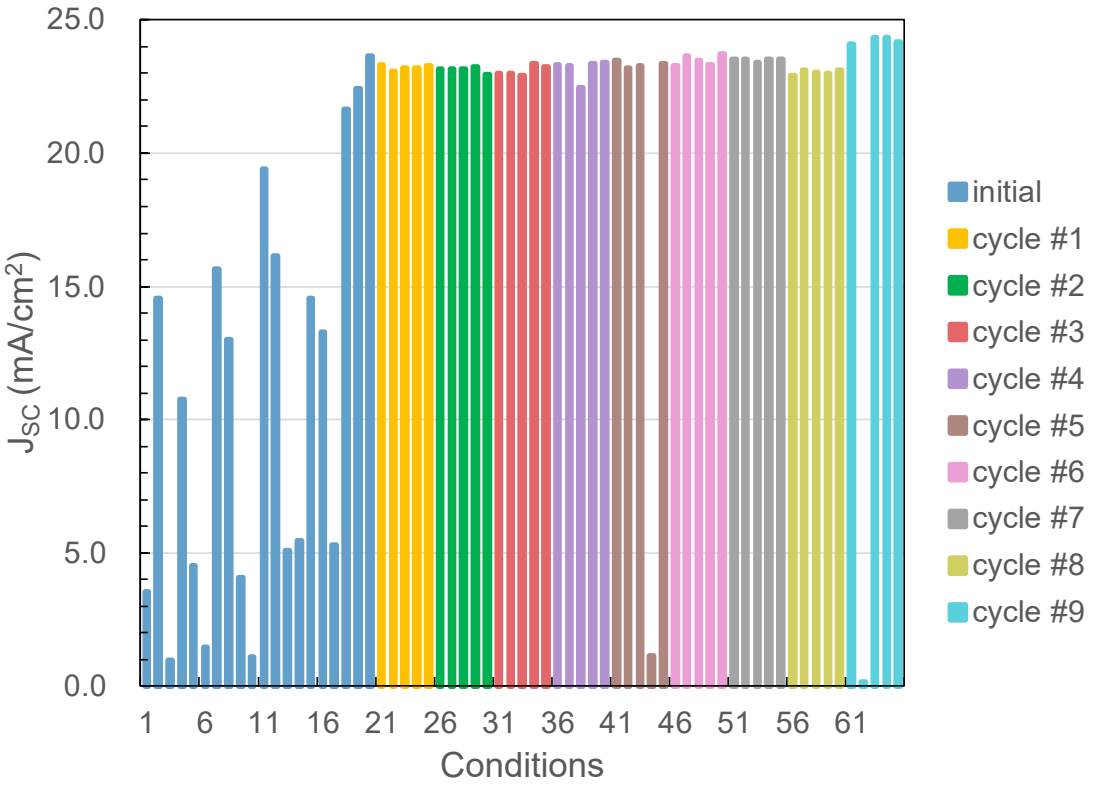

Figure S1. Progression of current density (J_{SC}) of reverse scan from initial data to the 9th cycle.

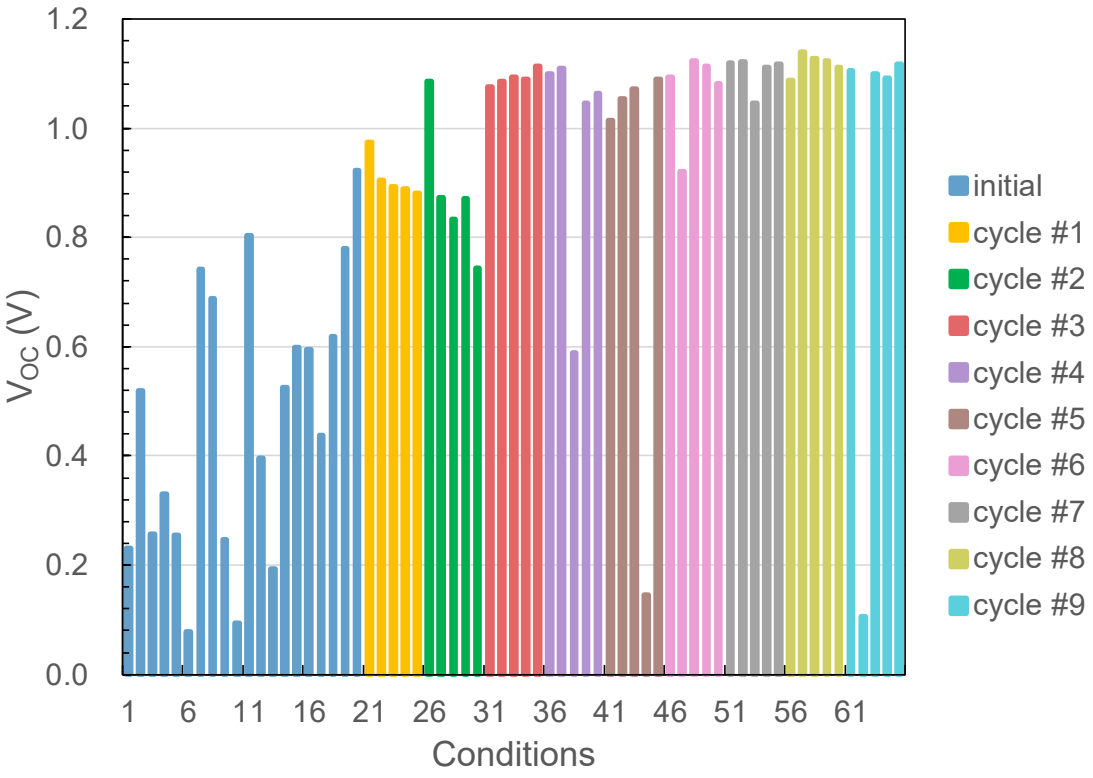

Figure S2. Progression of open circuit voltage (V_{OC}) of reverse scan from initial data to the 9th cycle.

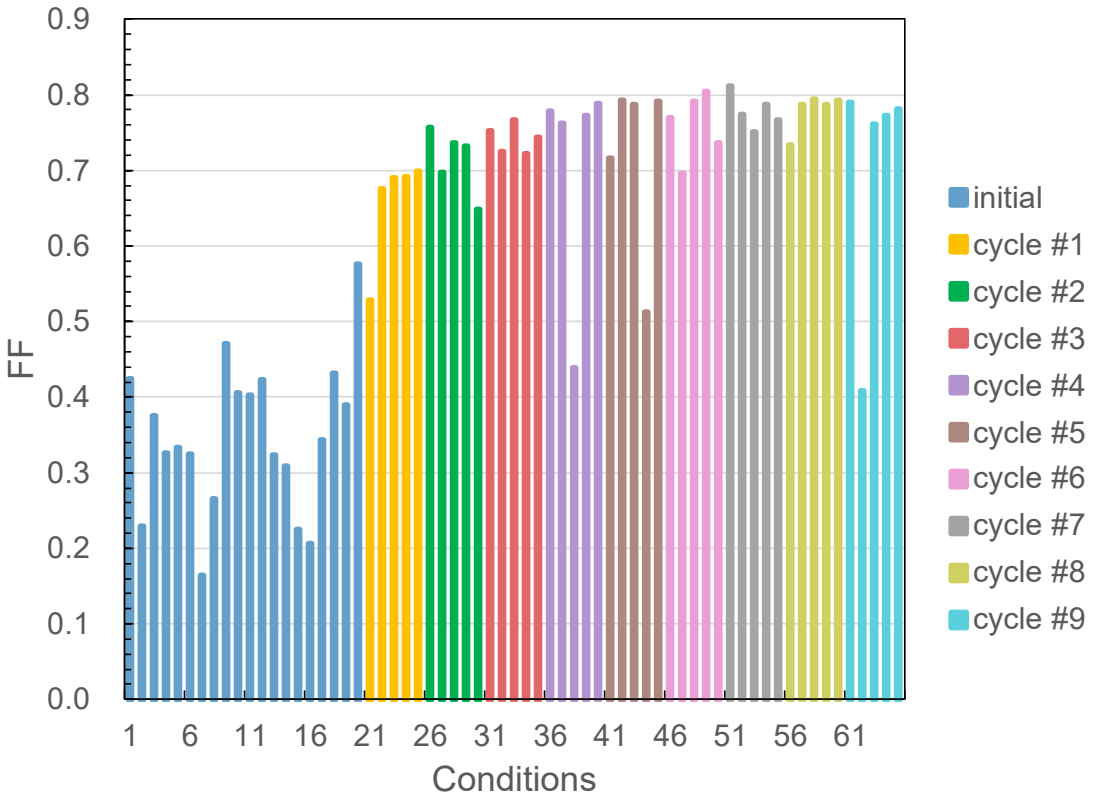

Figure S3. Progression of fill factor (FF) of reverse scan from initial data to the 9th cycle.

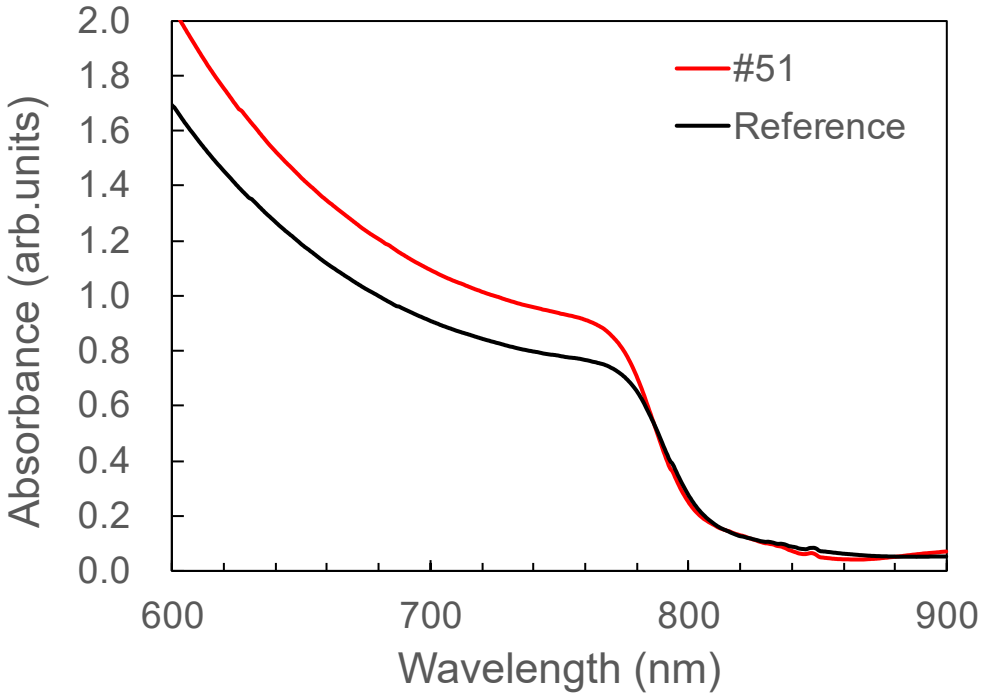

Figure S4. UV-vis absorption spectra of perovskite layer deposited on FTO/SnO_2 substrate. Red line and black line show the film fabricated under best condition optimized by Bayesian optimization and reference condition, respectively.

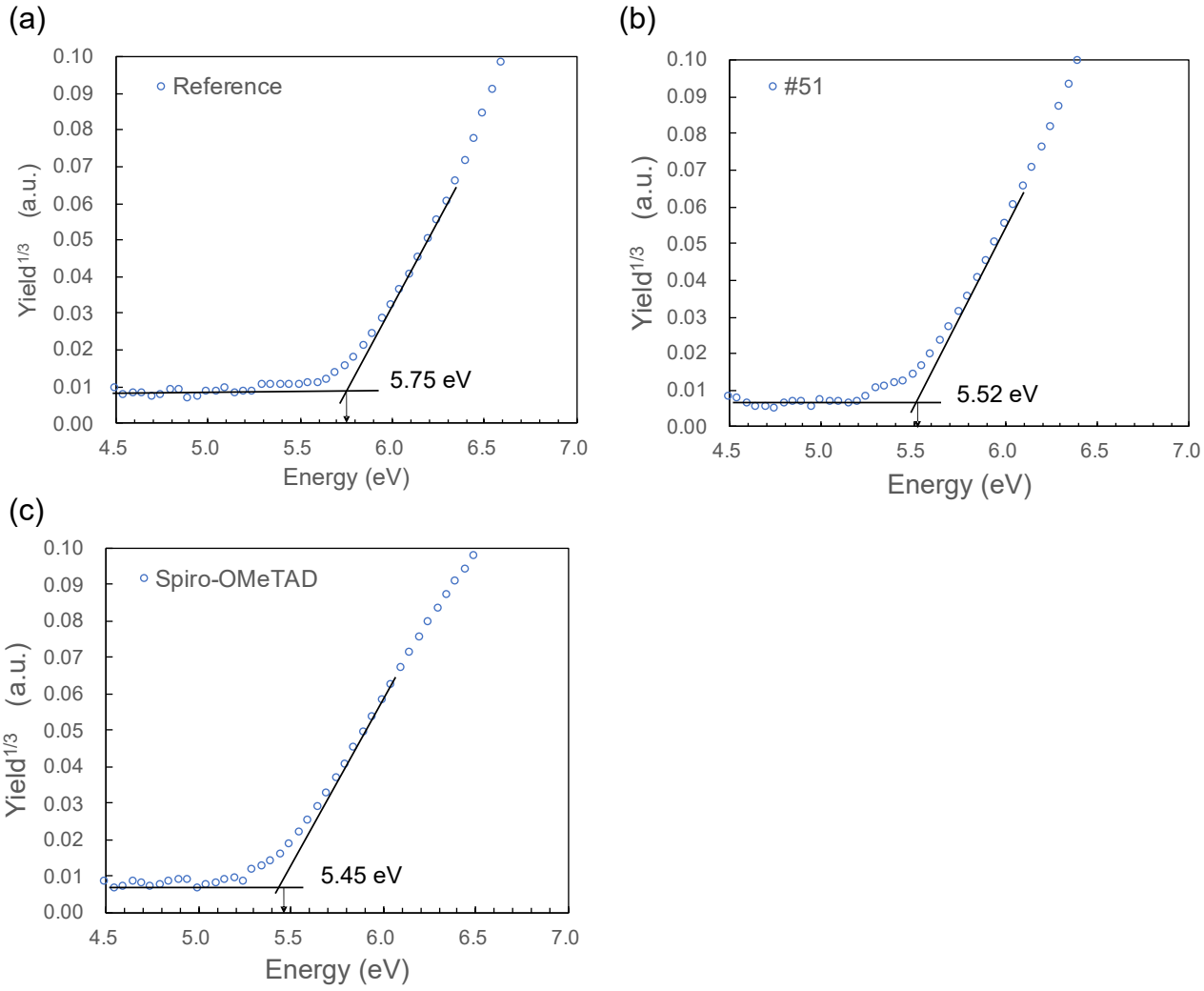

Figure S5. Photoelectron yield spectroscopy spectra of perovskite layer deposited on FTO/SnO_2 substrate. (a) Perovskite layer prepared under reference condition. (b) Perovskite layer prepared under #51 (best) condition. (c) Hole transport layer (Spiro-OMeTAD).

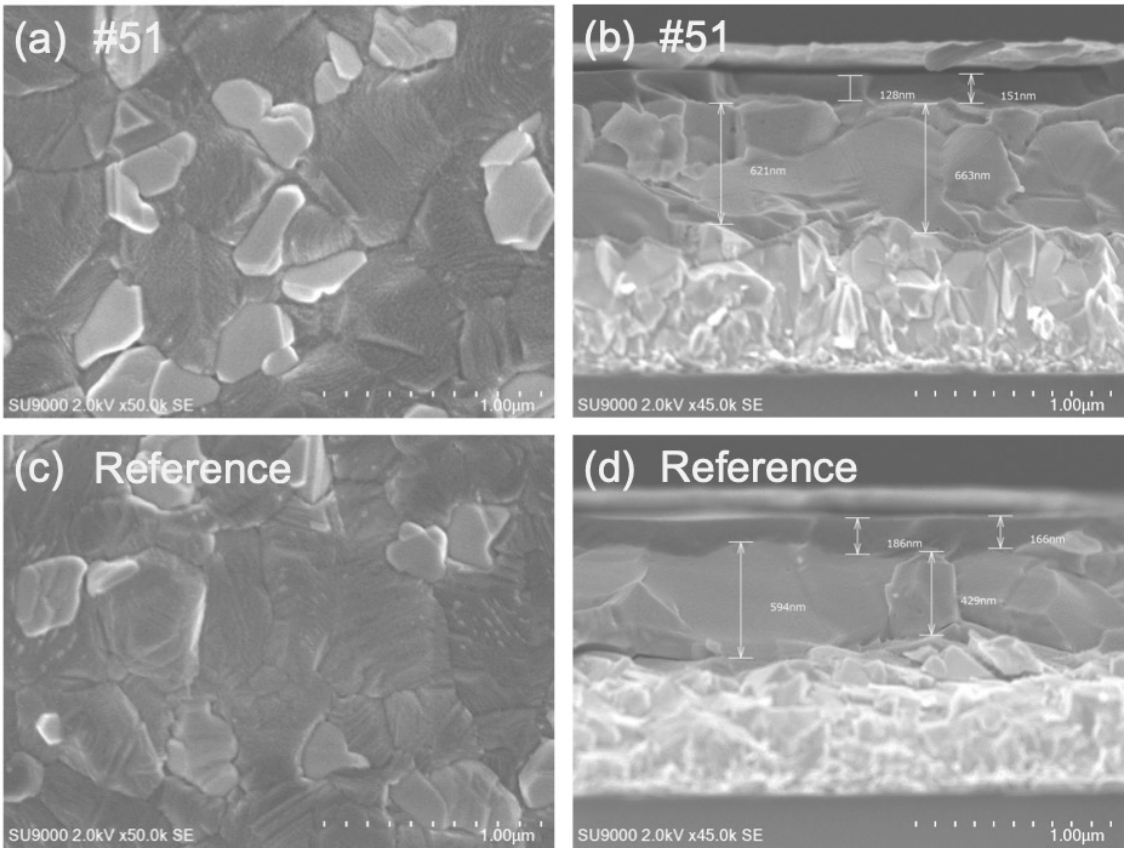

Figure S6. (a) Top view of the perovskite layer fabricated under the best conditions optimized by Bayesian optimization. (b) Cross-sectional view of the solar cell fabricated under the best conditions optimized by Bayesian optimization. (c) Top view of the perovskite layer fabricated under the reference condition. (d) Cross-sectional view of the solar cell fabricated under the reference condition.

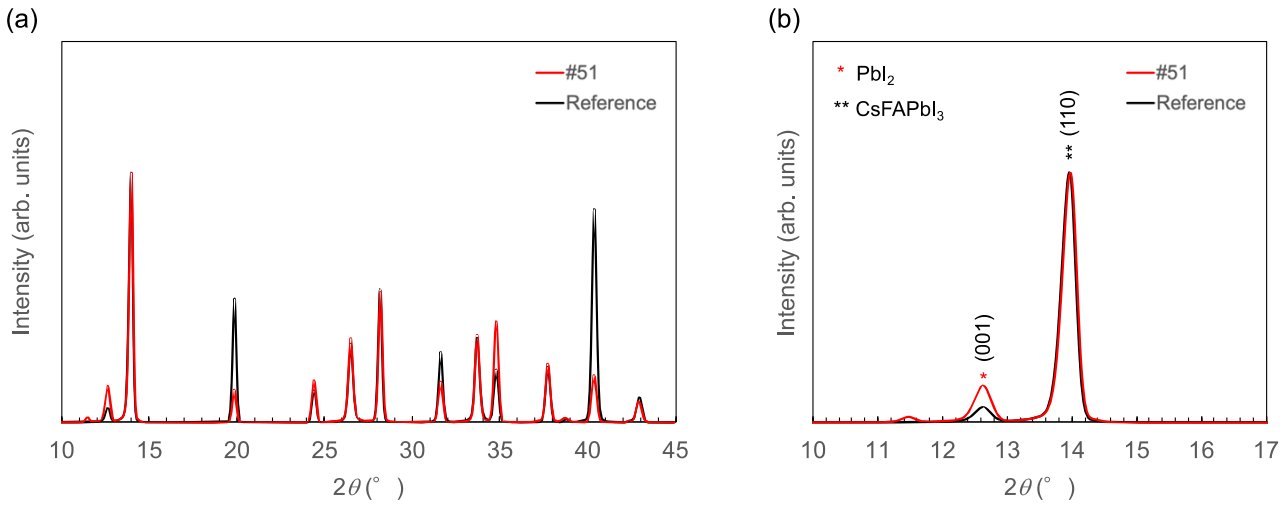

Figure S7. (a) XRD spectra of perovskite films prepared under condition of #51 and reference conditions. Red line and black line show XRD spectra of #51 and reference, respectively. (b) XRD spectra with expanded range of 10-17°.

Table S2. Peak information for the (110) plane of $CsFAPbI_3$ prepared with #51 condition and reference condition.

	2θ (°)	Intensity (cps)	FWHM (°)	Integrated Intensity (cps°)	Integral breadth (°)
#51	13.969	26534	0.256	7760	0.2924
Reference	13.954	26699	0.252	7827	0.2932