Supporting information

Mixed Cation 2D Perovskite: A Novel Approach for Enhanced Perovskite Solar Cell Stability

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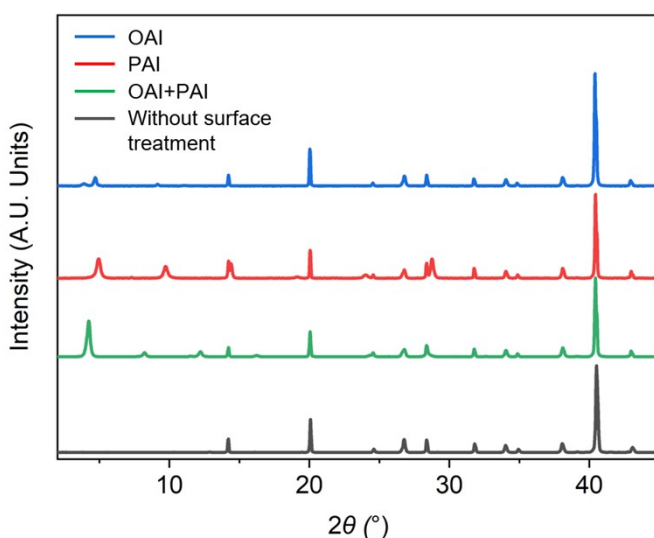

Figure S1. Full XRD spectra for OAI, PAI, OAI+PAI samples, and without surface treatment on the 3D perovskite.
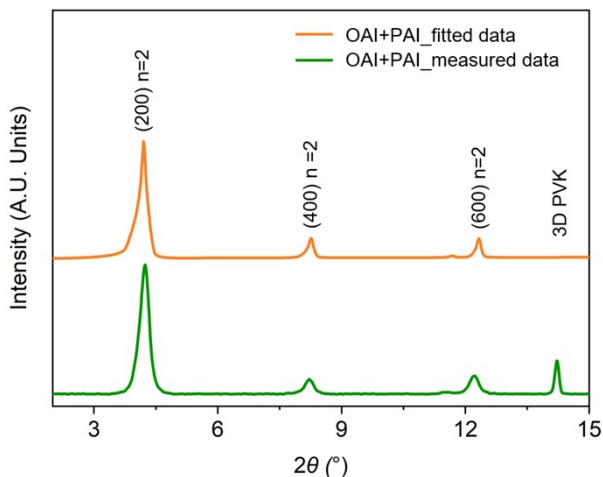
Figure S2. Sample measured and fitted XRD spectra for OAI+PAI.

Table S1. Fitted crystal data for 2D OAI+PAI, OAI, PAI

OAI+PAI
hkl Phase - 1 Lebail method

R-Bragg	99.961
Spacegroup	Pbca
Scale	1.60767e-007
Cell Mass	1.000
Cell Volume (Å^3)	3520.95291
Wt% - Rietveld	100.000

Double-Voigt|Approach

Cry size Lorentzian	194.9
k: 0.89 LVol-IB (nm)	110.417
k: 0.89 LVol-FWHM (nm)	73.443

Lattice parameters

a (Å)	43.6386405
b (Å)	9.0124875
c (Å)	8.9525000

h	k	l	m	d	Th2	I
2	0	0	2	21.81932	4.04632	1.17e+005
4	0	0	2	10.90966	8.09770	5.32e+004
1	1	0	4	8.82622	10.01358	3.88e-042
3	1	0	4	7.66119	11.54116	6.23e+003
6	0	0	2	7.27311	12.15925	1.03e+005
1	1	1	4	6.28525	14.07933	8.65e-041
1	-1	-1	4	6.28525	14.07933	5.09e-042
5	1	0	4	6.26969	14.11445	1.89e-041
3	1	1	4	5.82079	15.20917	2.03e-041
3	-1	-1	4	5.82079	15.20917	7.85e-043
8	0	0	2	5.45483	16.23619	1.3e-041
5	-1	-1	4	5.13554	17.25312	1.49e-041
5	1	1	4	5.13554	17.25312	7.32e-041
7	1	0	4	5.12704	17.28194	1.88e-041
0	2	0	2	4.50624	19.68497	4.25e-042
0	0	2	2	4.47625	19.81820	6.83e-042
7	-1	-1	4	4.44909	19.94042	2.29e-042
7	1	1	4	4.44909	19.94042	1.31e-042
2	2	0	4	4.41311	20.10467	3.71e-042
2	0	-2	2	4.38493	20.23524	1.06e-042
2	0	2	2	4.38493	20.23524	1.66e-042
10	0	0	2	4.36386	20.33395	9.81e-044
9	1	0	4	4.26999	20.78588	2.82e-043

OAI
hkl Phase - 1 Lebail method

R-Bragg	61.680
Spacegroup	Pbca
Scale	3.35345e-006
Cell Mass	1.000
Cell Volume (Å^3)	3492.44104
Wt% - Rietveld	100.000

Double-Voigt|Approach

Cry size Lorentzian   24.3
k: 1 LVol-IB (nm)   15.445
k: 0.89 LVol-FWHM (nm)   21.592
Lattice parameters
a (Å)   44.9283527
b (Å)   9.3781457
c (Å)   8.2887991

h	k	l	m	d	Th2	I
2	0	0	2	22.46418	3.93012	572
4	0	0	2	11.23209	7.86488	1.5e-010
1	1	0	4	9.18028	9.62645	3.88e-042
3	1	0	4	7.94834	11.12288	27
6	0	0	2	7.48806	11.80895	6.21e-022
5	1	0	4	6.48814	13.63693	1.89e-041
1	-1	-1	4	6.15216	14.38550	6.79e-018
1	1	1	4	6.15216	14.38550	1.18e-043
3	-1	-1	4	5.73691	15.43288	1.04e-029
3	1	1	4	5.73691	15.43288	4.53e-033
8	0	0	2	5.61604	15.76711	4.24e-030
7	1	0	4	5.29665	16.72450	1.49e-041
5	-1	-1	4	5.10907	17.34319	7.32e-041
5	1	1	4	5.10907	17.34319	1.88e-041
0	2	0	2	4.68907	18.91026	4.25e-042
2	2	0	4	4.59014	19.32169	6.83e-042
10	0	0	2	4.49284	19.74431	2.29e-042
7	1	1	4	4.46322	19.87667	2.29e-042
7	-1	-1	4	4.46322	19.87667	3.71e-042
9	1	0	4	4.40662	20.13459	1.06e-042
4	2	0	4	4.32714	20.50839	1.66e-042
0	0	2	2	4.14440	21.42311	9.81e-044

PAI
hkl Phase - 1 Lebail method
R-Bragg   99.968
Spacegroup   Pbca
Scale   1.37335e-009
Cell Mass   1.000
Cell Volume (Å^3)   3179.97927
Wt% - Rietveld   100.000
Double-Voigt|Approach
Cry size Lorentzian   67.3
k: 1 LVol-IB (nm)   42.845
k: 0.89 LVol-FWHM (nm)   59.898
Lattice parameters
a (Å)   37.7212996
b (Å)   9.1905125
c (Å)   9.1727152

h	k	l	m	d	Th2	I
2	0	0	2	18.86065	4.68140	1.42e+007
4	0	0	2	9.43032	9.37063	3.16e+007
1	1	0	4	8.92930	9.89769	8.48e-042
3	1	0	4	7.41978	11.91801	1.48e-041
1	1	1	4	6.39829	13.82935	5.16e-035
1	-1	-1	4	6.39829	13.82935	5.99e-035
6	0	0	2	6.28688	14.07566	7.03e+007

5	1	0	4	5.83123	15.18177	5.81e-042
3	-1	-1	4	5.76875	15.34718	1.89e-041
3	1	1	4	5.76875	15.34718	7.77e-042
5	-1	-1	4	4.92103	18.01130	1.14e-041
5	1	1	4	4.92103	18.01130	1.56e-040
8	0	0	2	4.71516	18.80467	4.53e-043
7	1	0	4	4.64860	19.07642	1.66e-042
0	2	0	2	4.59526	19.29998	2.29e-042
0	0	2	2	4.58636	19.33779	1.07e-042
2	2	0	4	4.46465	19.87020	6.69e-041
2	0	-2	2	4.45649	19.90697	1.31e-042
2	0	2	2	4.45649	19.90697	1.34e-042
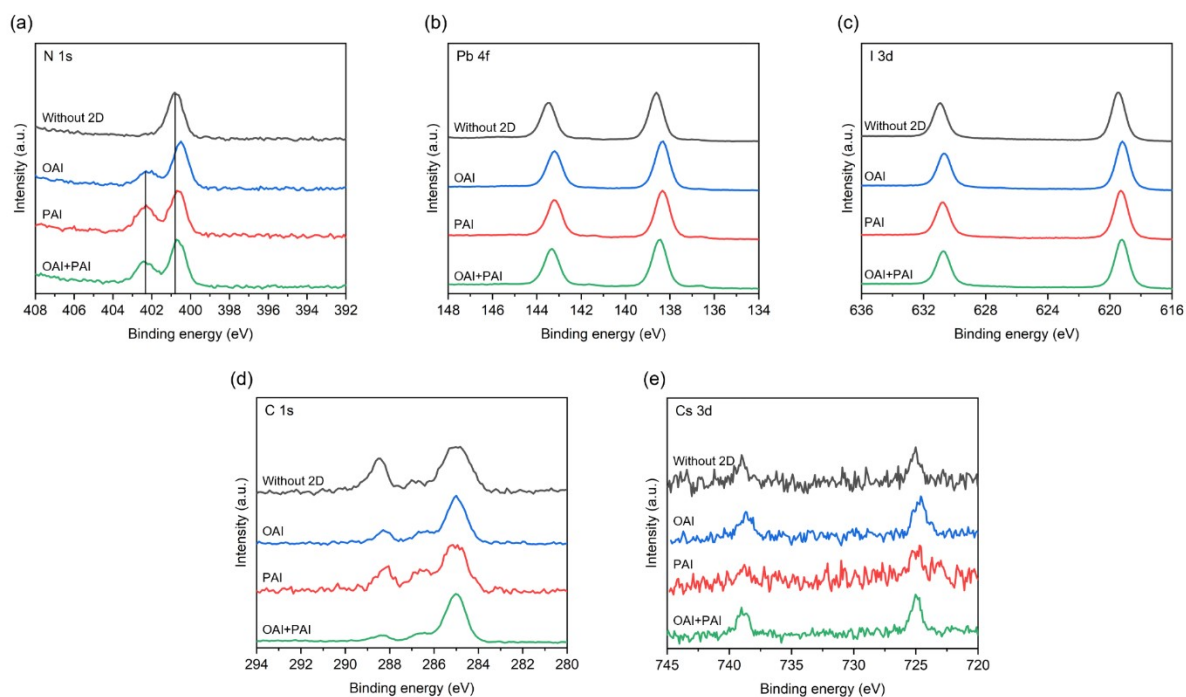
Figure S3. XPS spectra of (a) N 1s, (b) Pb 4f, (c) I 3d, (d) C 1s, and (e) Cs 3d for the 2D/3D samples. The 3D perovskite sample without 2D layer was used as a reference.
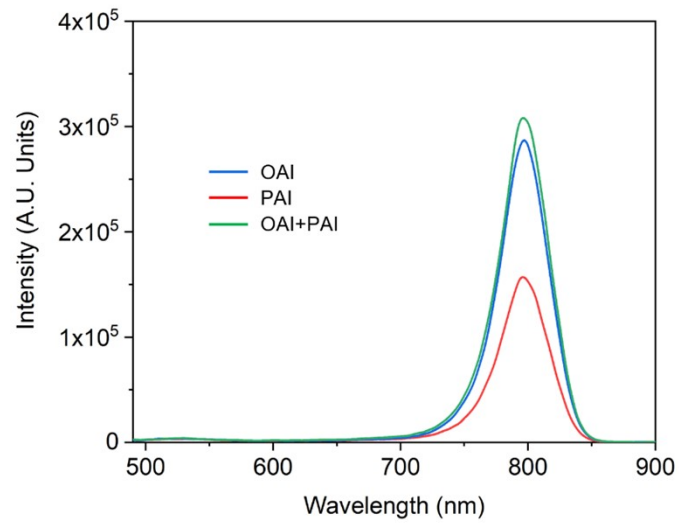
Figure S4. PL emission spectra for OAI, PAI, and OAI+PAI samples. The incident emission laser is from the glass side.
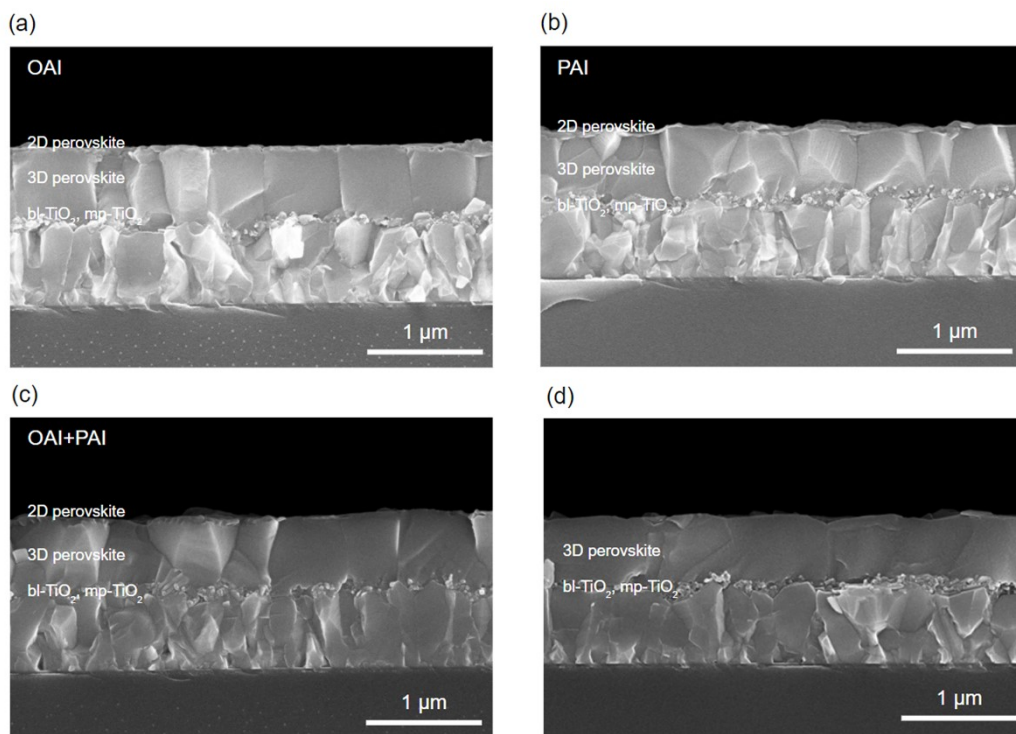
Figure S5: Cross-sectional scanning electron microscopy of the (a) OAI, (b) PAI, (c) OAI+PAI, and **(d) without surface treatment on the 3D perovskite.**

Table S2: TRLP parameters for the 2D/3D samples **and without treatment**.

| | A_1 | t_1 | A_2 | t_2 |
	[-]	[ns]	[-]	[ns]
OAI	0.273	167.9	0.630	939.6
PAI	0.461	128.1	0.497	574.8
OAI+PAI	0.240	160.6	0.632	979.5
Without surface treatment	0.161	338.6	0.654	1463.5

† The equation for the fitting is $y = y_0 + A_1 \times \exp(-(x-x_0)/\tau a) + A_2 \times \exp(-(x-x_0)/\tau b)$.
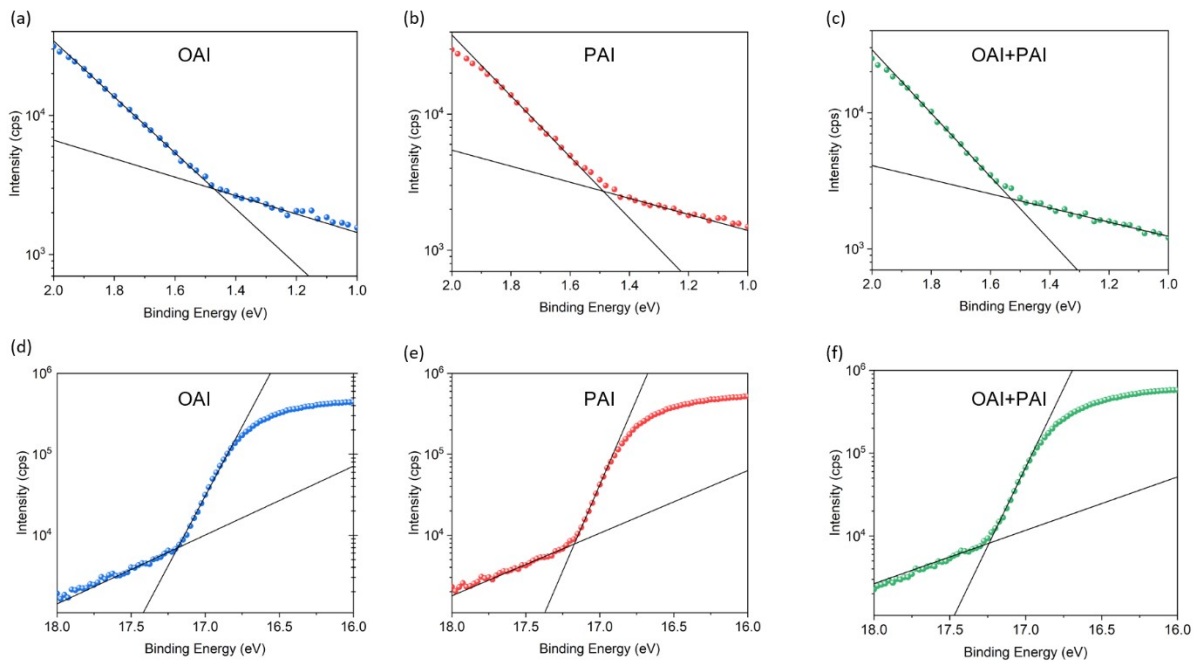
Figure S6. Measurement of the energy band diagram parameters. UPS spectrum edge of work function for (a) OAI, (b) PAI, and (c) OAI+PAI samples. (b) Valence band edge of perovskite layer for € PAI, (f) OAI, and (g) OAI+PAI samples.
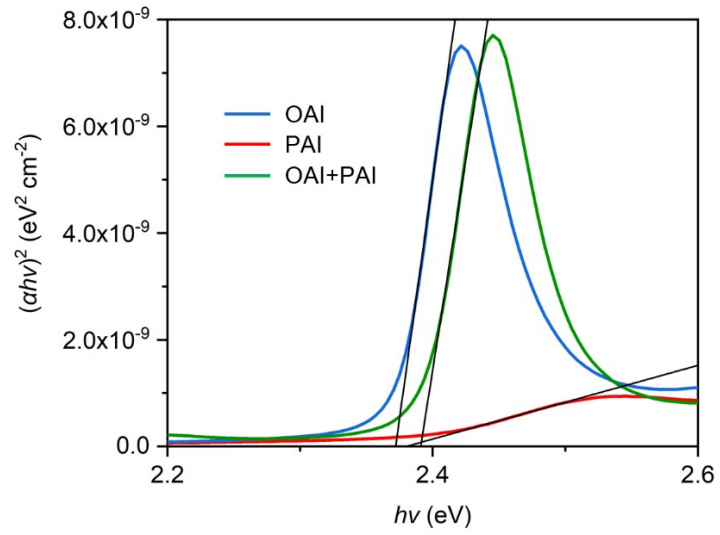
Figure S7. Tauc plot as obtained from UV-VIS to determine the bandgap of (a) OAI, (b) PAI, and (c) OAI+PAI samples.
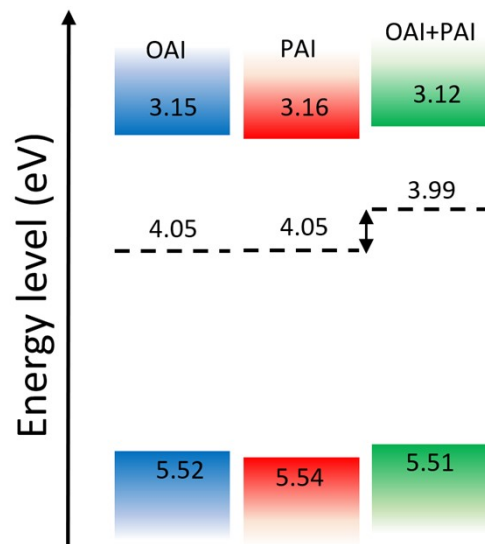
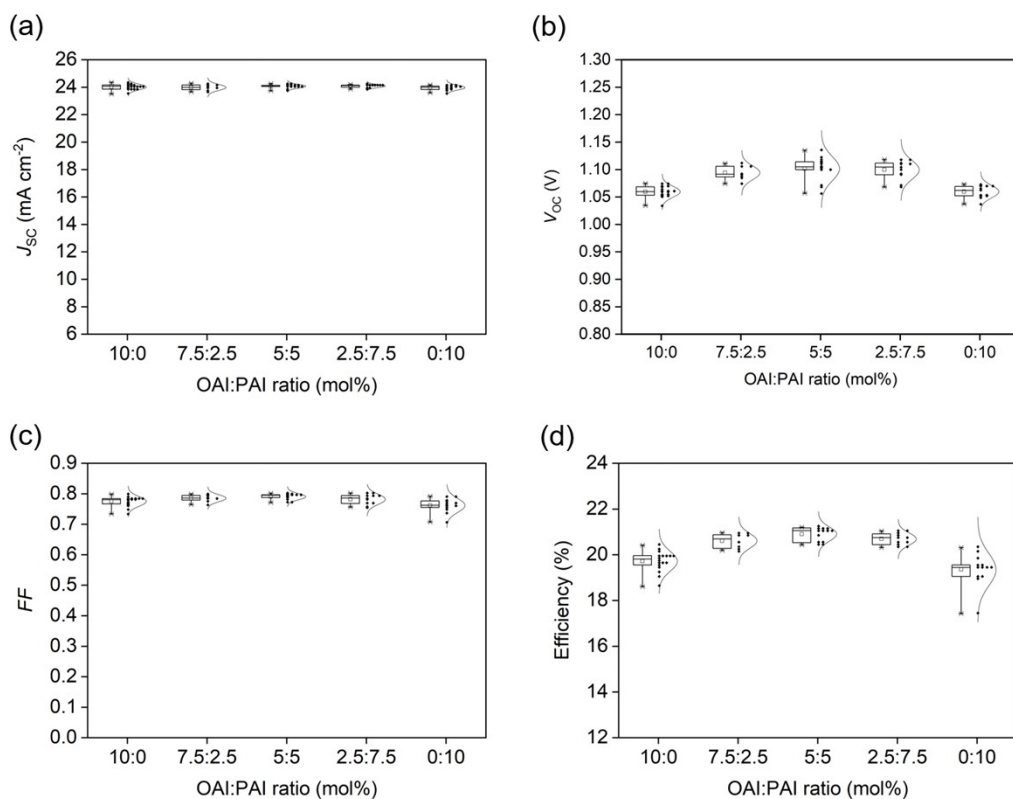Figure S8. Energy band diagram for OAI, PAI and OAI+PAI samples.

Figure S9. Statics of the device performance of each condition. The total number of the device was over 50.
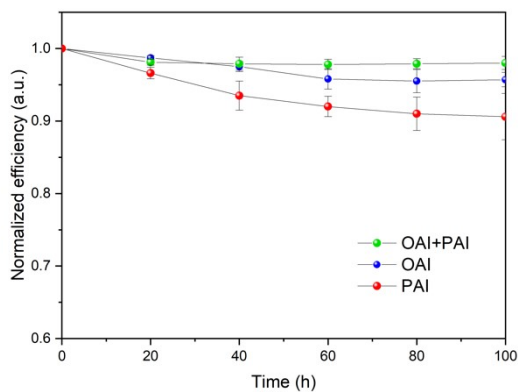

Figure S10. Stability result for each condition. The stability test was performed by maximum-power point tracking (MPPT) with the condition under AM1.5G with N$_2$ ambient with 0%RH at 25°C. The total number of devices was two for each condition.
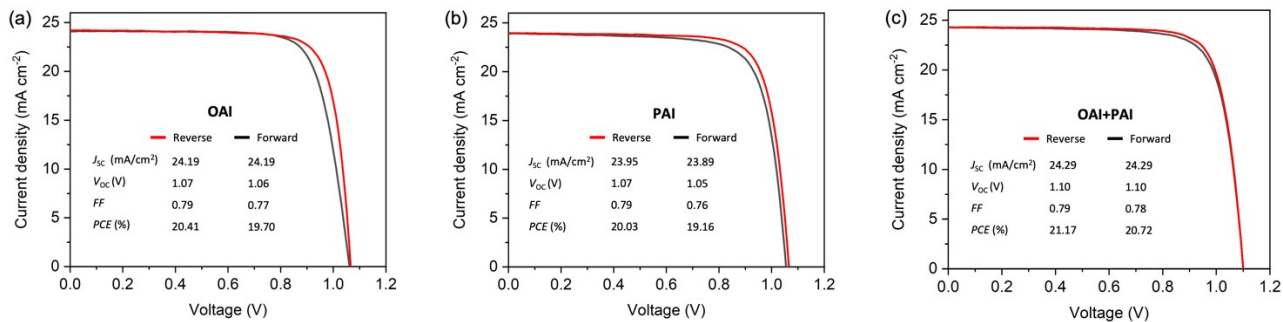
Figure S11. Hysteresis behavior for (a) OAI, (b) PAI and (c) OAI+PAI samples.
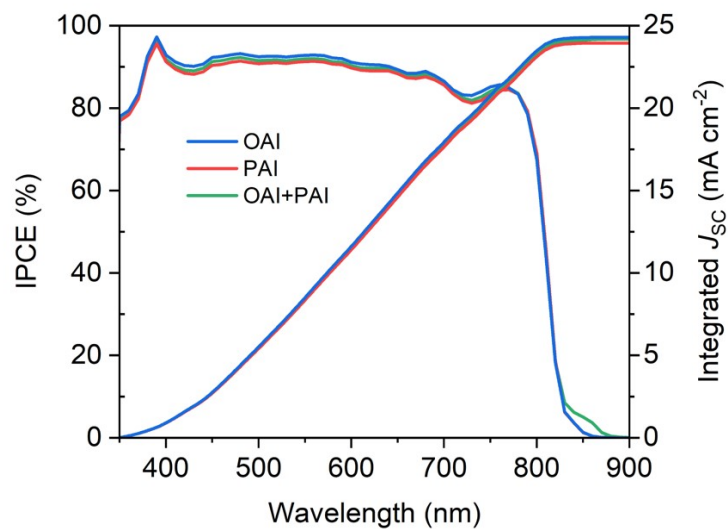

Figure S12. Incident photon to current efficiency (IPCE) for OAI, PAI and OAI+PAI samples.
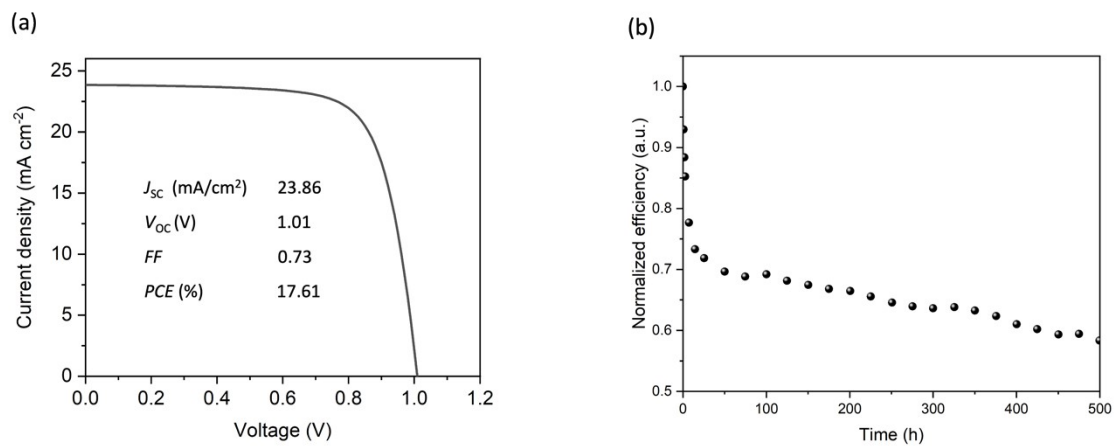
Figure S13. (a) JV characteristic and (b) MPPT stability measurement for PSC devices without 2D perovskite layers at the 3D PVK/HTL interface.