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Supporting Information

A Crystal Engineering Approach for Perovskite Solar Cells and Modules Fabrication out of the Glove Box

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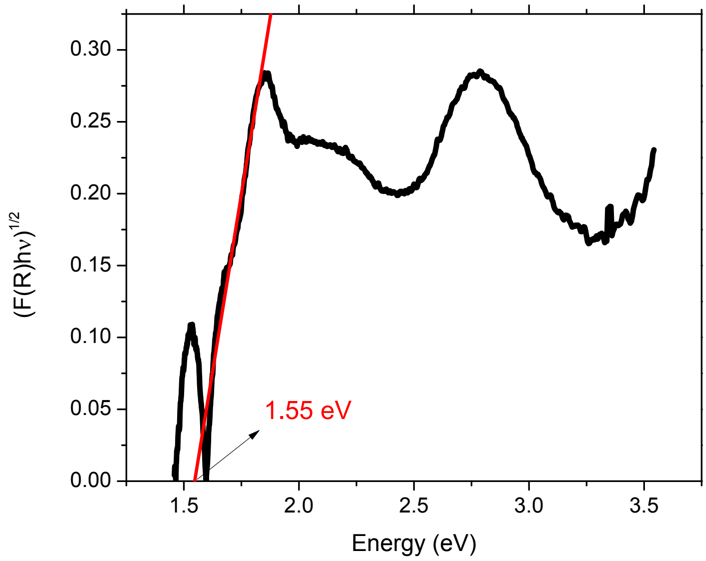

Fig. S1 Band gap energy of the CE fabricated perovskite layer evaluated by transformation of the diffuse-reflectance spectrum of the layer in Kubelka-Munk equation.

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Table S1 Average photovoltaic parameters of J-V measurement of the PSCs containing of spiro-OMeTAD and P3HT as HTM layer and HTM free devices.

HTL	Fabrication Method	Voc (V)	Jsc (mA/cm^2)	FF	PCE (%)
spiro-OMeTAD	CE	1.052 ± 0.028	22.375 ± 0.731	0.707 ± 0.020	16.639 ± 0.378
	OM	0.963 ± 0.033	18.450 ± 1.598	0.680 ± 0.064	12.002 ± 0.701
P3HT	CE	1.025 ± 0.011	21.605 ± 1.579	0.683 ± 0.050	15.110 ± 1.332
	OM	0.983 ± 0.011	15.037 ± 1.010	0.635 ± 0.020	9.377 ± 0.627
HTL free	CE	0.905 ± 0.026	11.162 ± 0.406	0.658 ± 0.019	6.652 ± 0.326
	OM	0.735 ± 0.128	9.500 ± 0.457	0.472 ± 0.086	3.323 ± 0.864

Table S2 Average photovoltaic parameters of J-V measurement of the PSCs containing of spiro-OMeTAD and P3HT as HTM layer and HTM free devices.

Reference	Scaffold layer	Perovskite structure	Perovskite deposition	HTM	Champion PCE (%)
[5]	mp-TiO2	(MA/FA)Pb(I/Br)3	2-step spin-spin coating (Iodide management)	PTAA	22.1
[18]	mp-TiO2	(MA/FA)Pb(I/Br)3	1-step spin coating	PTAA	20
[8]	mp-TiO2	(Rb/Cs/MA/FA)Pb(I/Br)3	1-step spin coating	Spiro	21.6
[9]	mp-TiO2	MAPbI3	2-step spin-dip coating	Spiro	15
[17]	mp-TiO2	MAPbI3	2-step spin-spin coating	Spiro	17.01
[22]	POM	MAPbI3	2-step spin-dip coating	Spiro	16.3
PA#	mp-TiO2	MAPbI3	2-step spin-dip coating (CE)	Spiro	17
[21]	mp-TiO2	MAPbI3	2-step spin-dip coating	P3HT	16.2
PA	mp-TiO2	MAPbI3	2-step spin-dip coating (CE)	P3HT	16.8
[24]	np-TiO2	MAPbI3	2-step in situ	No HTM	10.03
PA	mp-TiO2	MAPbI3	2-step spin-dip coating (CE)	No HTM	7

PA: results of present article

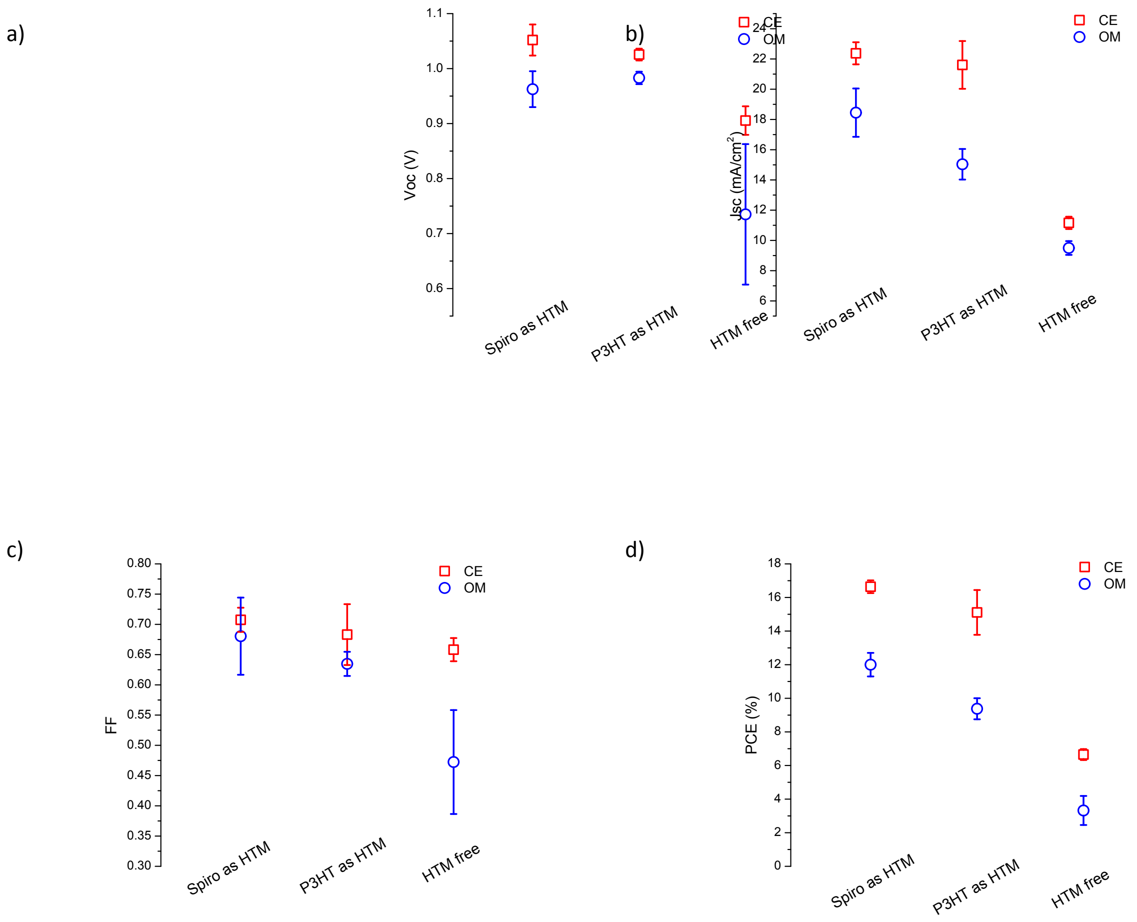

Fig. S2 Statistical results of photovoltaic parameters of V_{OC} (a), J_{SC} (b), fill factor (c) overall efficiency (d), under AM1.5G illumination, of the PSCs utilizing spiro-OMeTAD and P3HT as HTL layer and totally HTL-free devices, in comparison with ordinary sequential deposition of the perovskite layer (OM) and crystal engineering approach (CE).

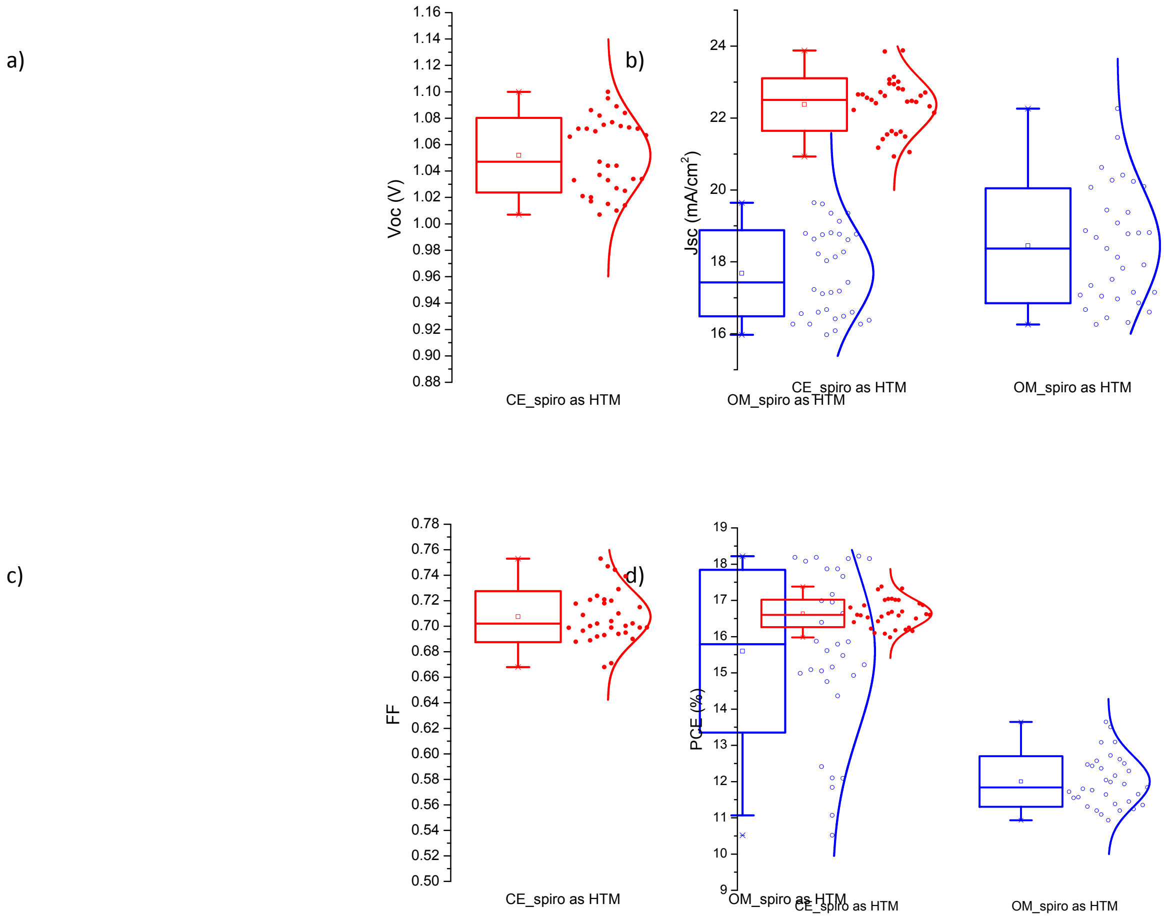

Fig. S3 Statistical results of photovoltaic J-V parameters of the PSCs containing spiro-OMeTAD as HTM layer under AM1.5G illumination with comparison between crystal engineering (CE) approach and ordinary sequential deposition method (OM).

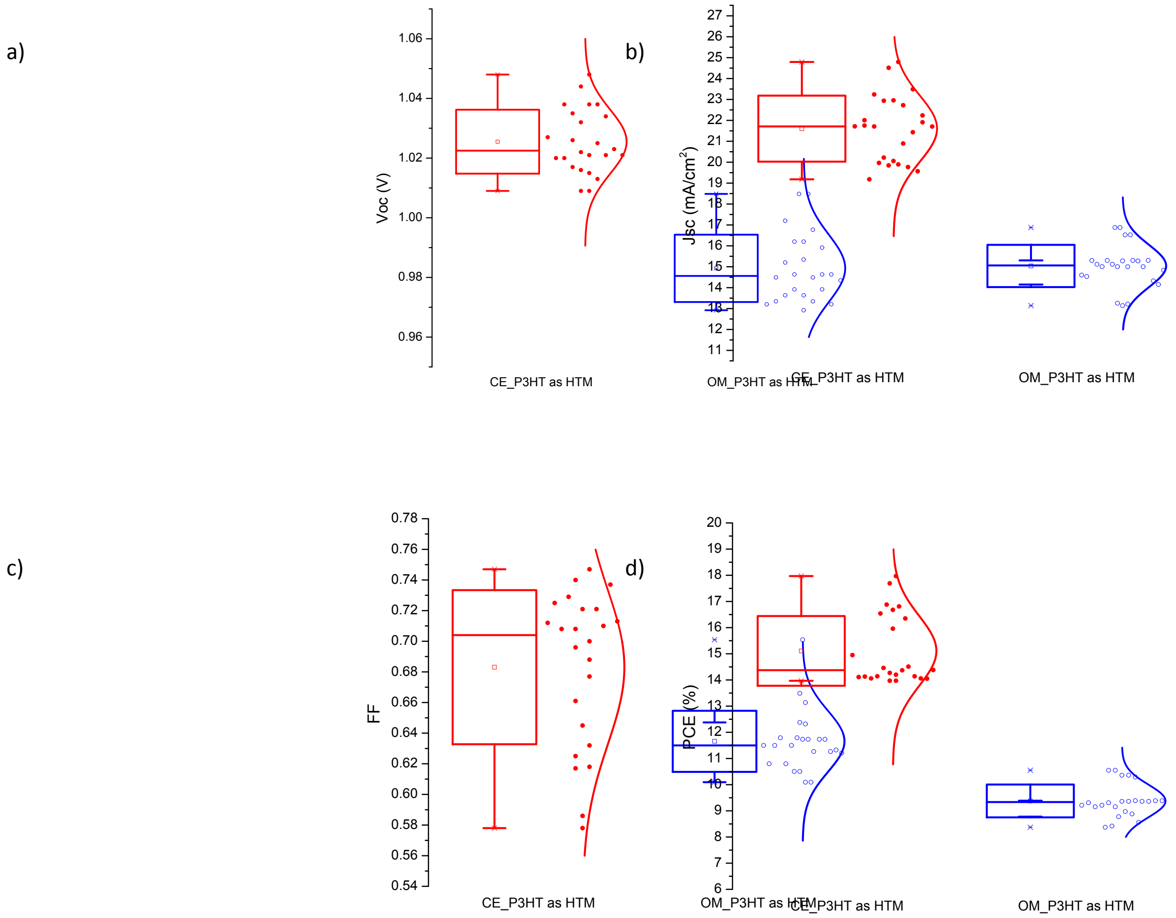

Fig. S4 Statistical results of photovoltaic J-V parameters of the PSCs containing P3HT as HTM layer under AM1.5G illumination with comparison between crystal engineering (CE) approach and ordinary sequential deposition method (OM).

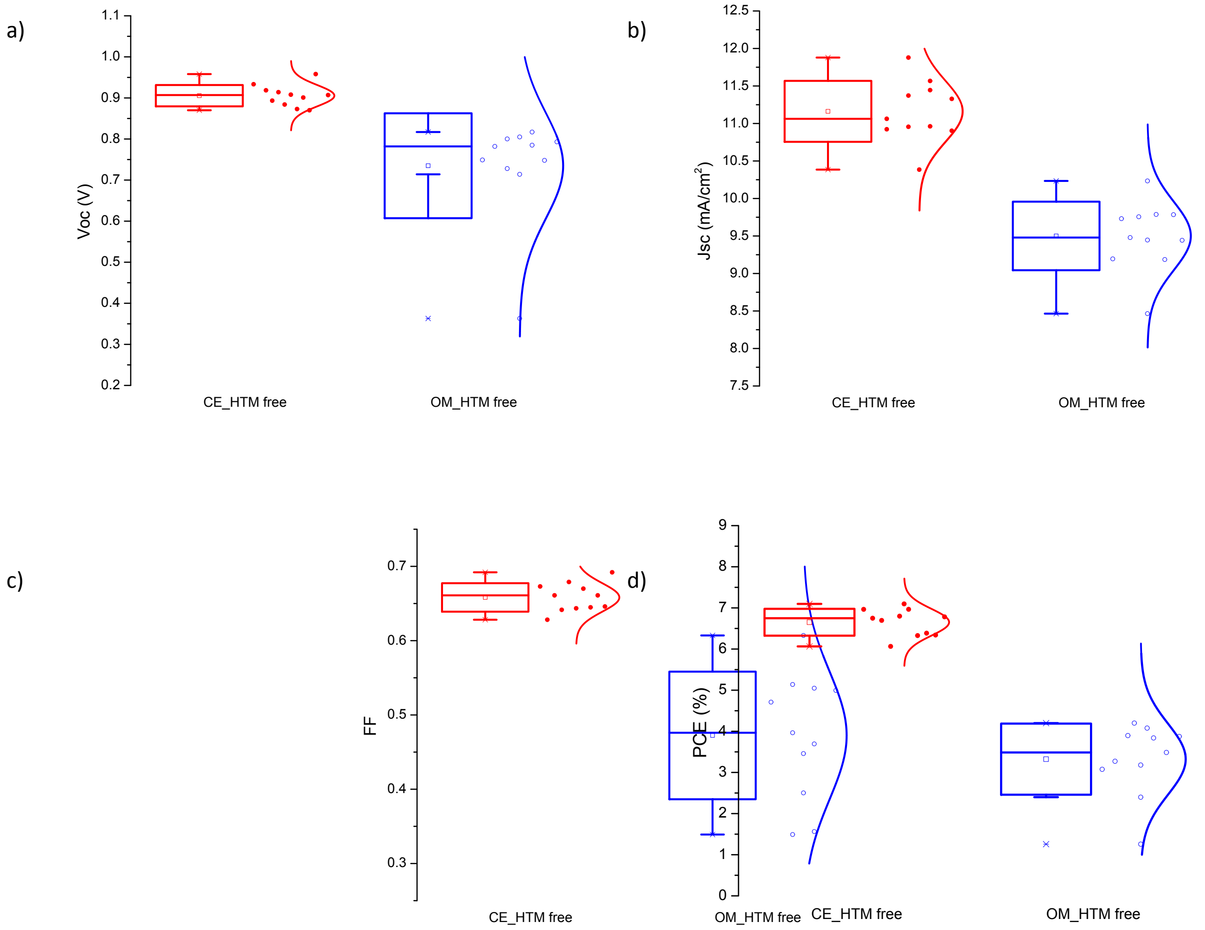

Fig. S5 Statistical results of photovoltaic J-V parameters of the PSCs without using any HTM layer under AM1.5G illumination with comparison between crystal engineering (CE) approach and ordinary sequential deposition method (OM).

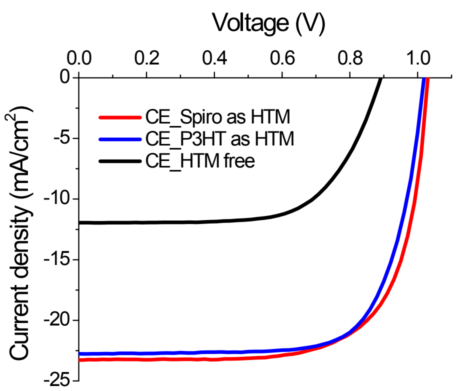

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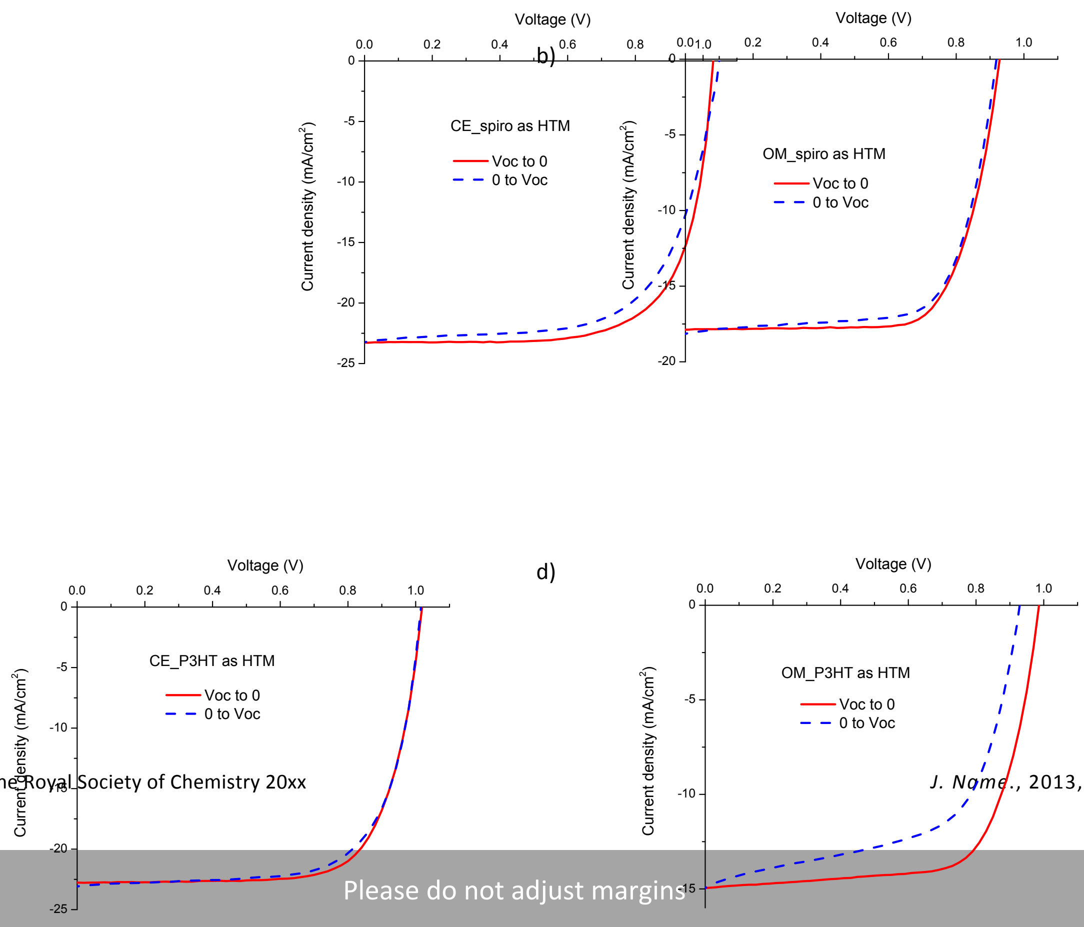
Fig. S6 J-V curves of the selected devices which fabricated by CE approach.

a) b) c) d)



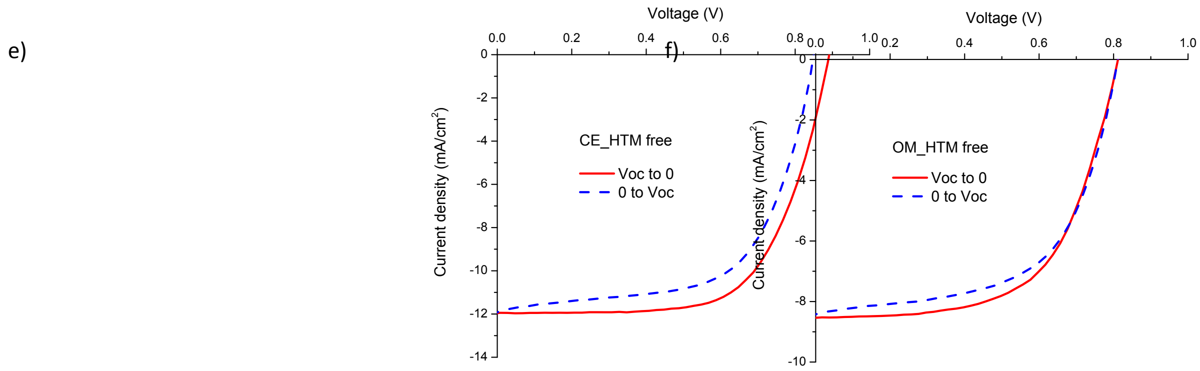


Fig. S7 J-V curves of selected PSCs containing spiro-OMeTAD (a,b) and P3HT (c,d) as HTM layer and totally HTM-free (e,f) devices in both forward and reverse voltage sweep. The curves are compared between crystal engineering (CE) approach (a,c,e) and ordinary sequential deposition (OM) method (b,d,f).

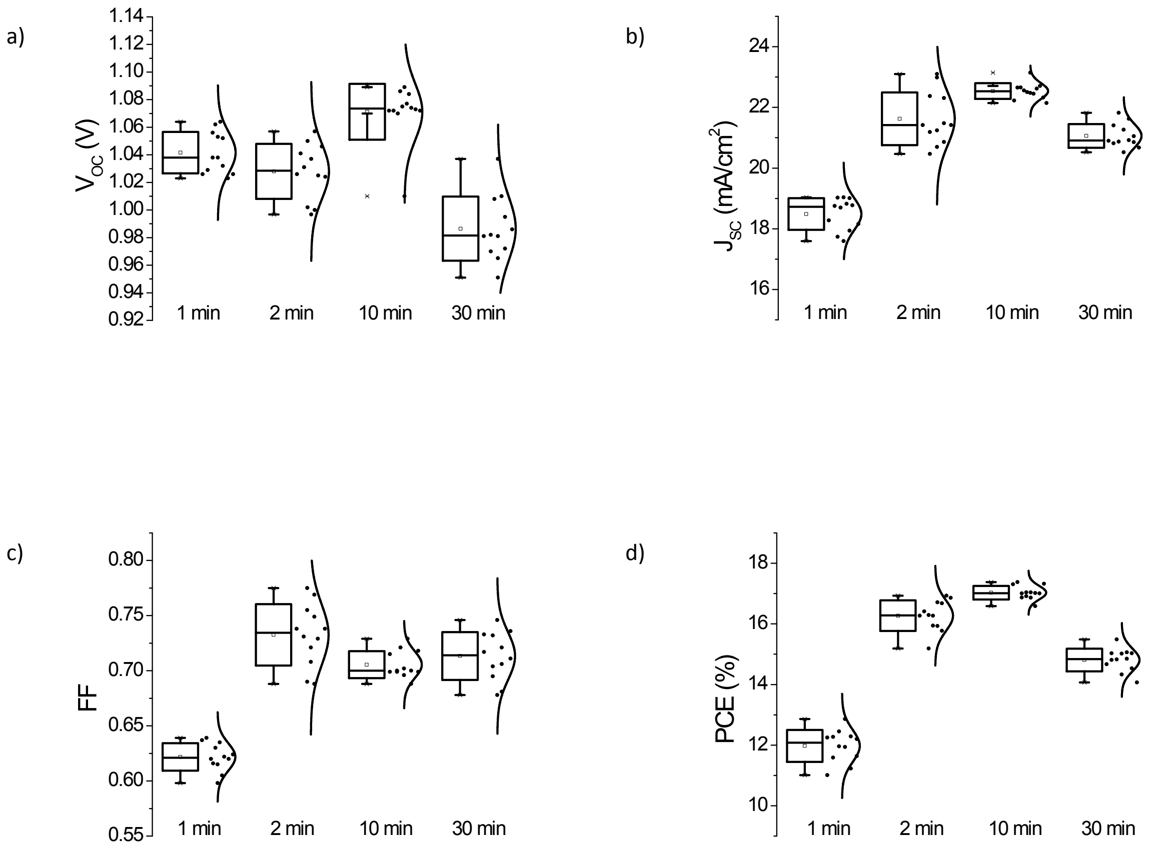


Fig. S8 Statistical results of J-V parameters of the PSCs that fabricated by CE approach. Effect of MAI dipping time (DT) is investigated on Voc (a), Jsc (b), FF (c) and overall efficiency (d).

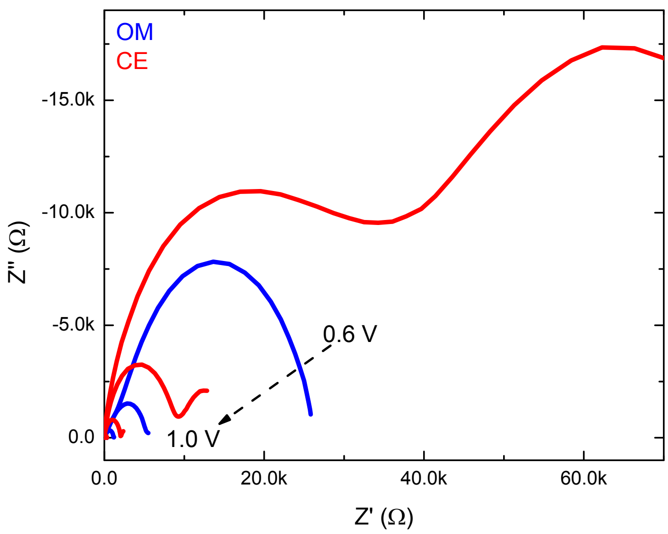


Fig. S9 Voltage dependent Nyquist plot of the Impedance for CE (red) and for OM (blue)

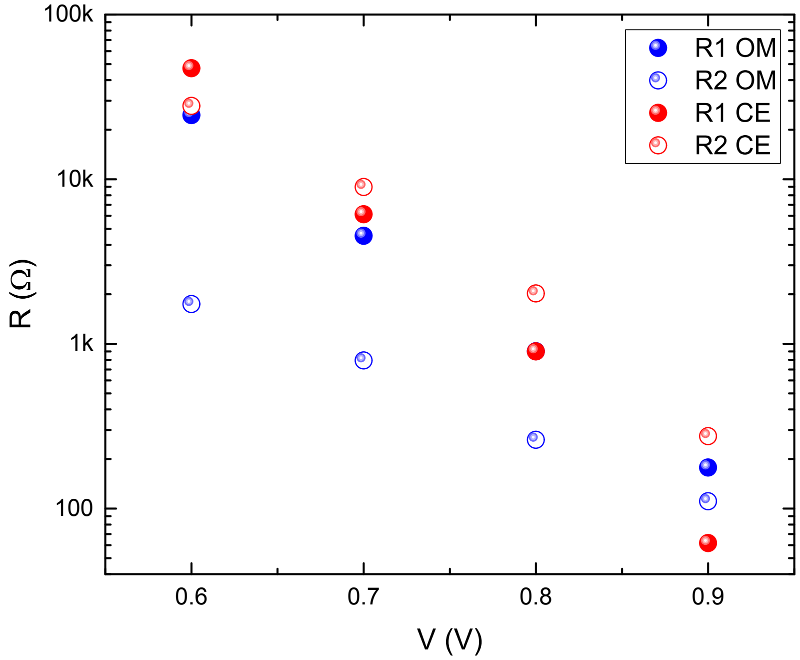


Fig. S10 Resistive element (R1 and R2) extracted from EIS analysis of the model reported in Figure 6c.

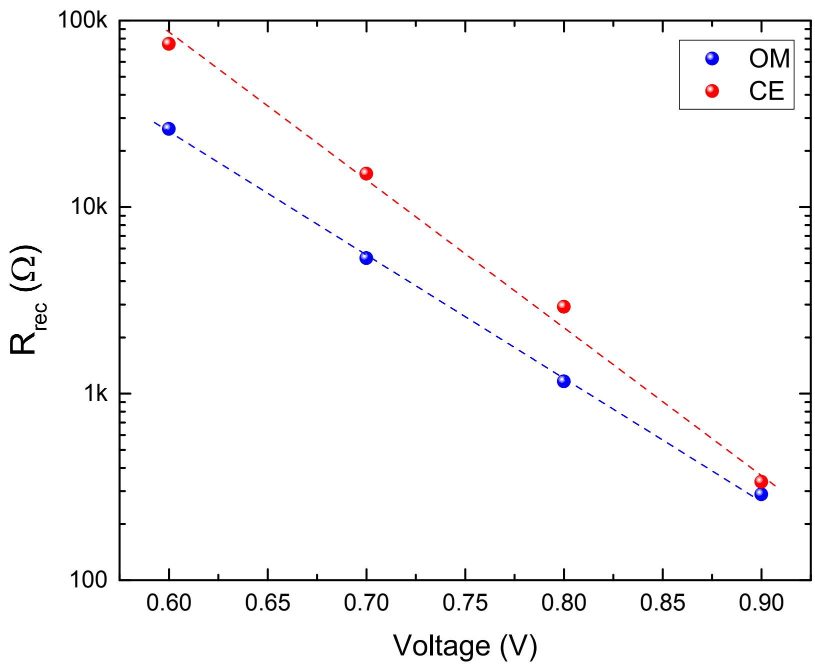


Fig. S11 Recombination resistance estimated from EIS analysis for the CE and the OM.

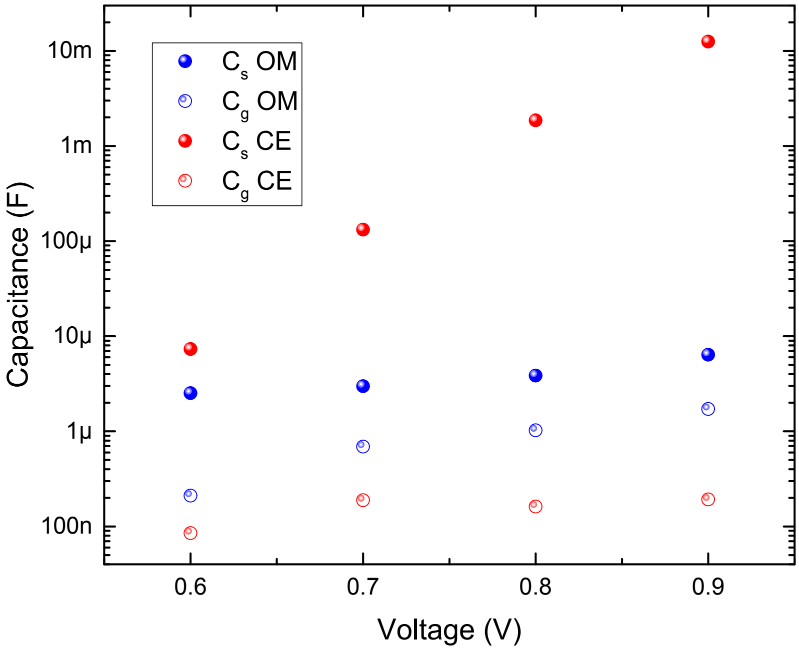
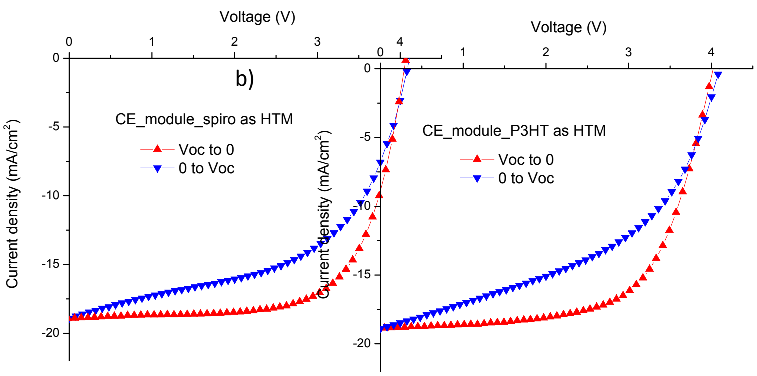


Fig. S12 Geometric capacitance (C_g) for the dielectric response of perovskite layer and surface charge accumulation capacitance (C_s) for the interfaces extracted from impedance model of figure 6c.

a)



c)

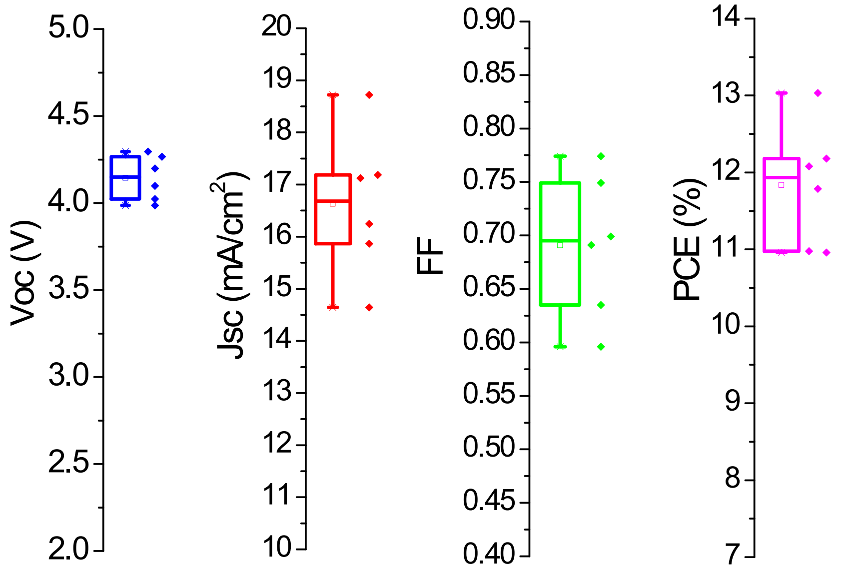


Fig. S13 J-V curves of selected perovskite solar modules containing spiro-OMeTAD (a) and P3HT (b) as HTLlayers in both forward and reverse voltage sweep. Statistical results of J-V parameters of the perovskite solar modules containing spiro-OMeTAD as HTM which are fabricated by CE approach (c).

Table S3 Photovoltaic parameters of J-V measurement in both forward and reverse voltage sweep of the perovskite solar modules containing of spiro-OMeTAD and P3HT as HTLs and fabricated by CE approach.

HTM	Fabrication Method	Potential Scan Direction	Voc (V)	Jsc (mA/cm^2)	FF	PCE (%)
spiro-OMeTAD	CE	Voc to 0	3.986	18.723	0.699	13.03
		0 to Voc	4.136	19.388	0.511	10.24
P3HT	CE	Voc to 0	3.972	18.652	0.656	12.14
		0 to Voc	4.130	19.224	0.457	9.08

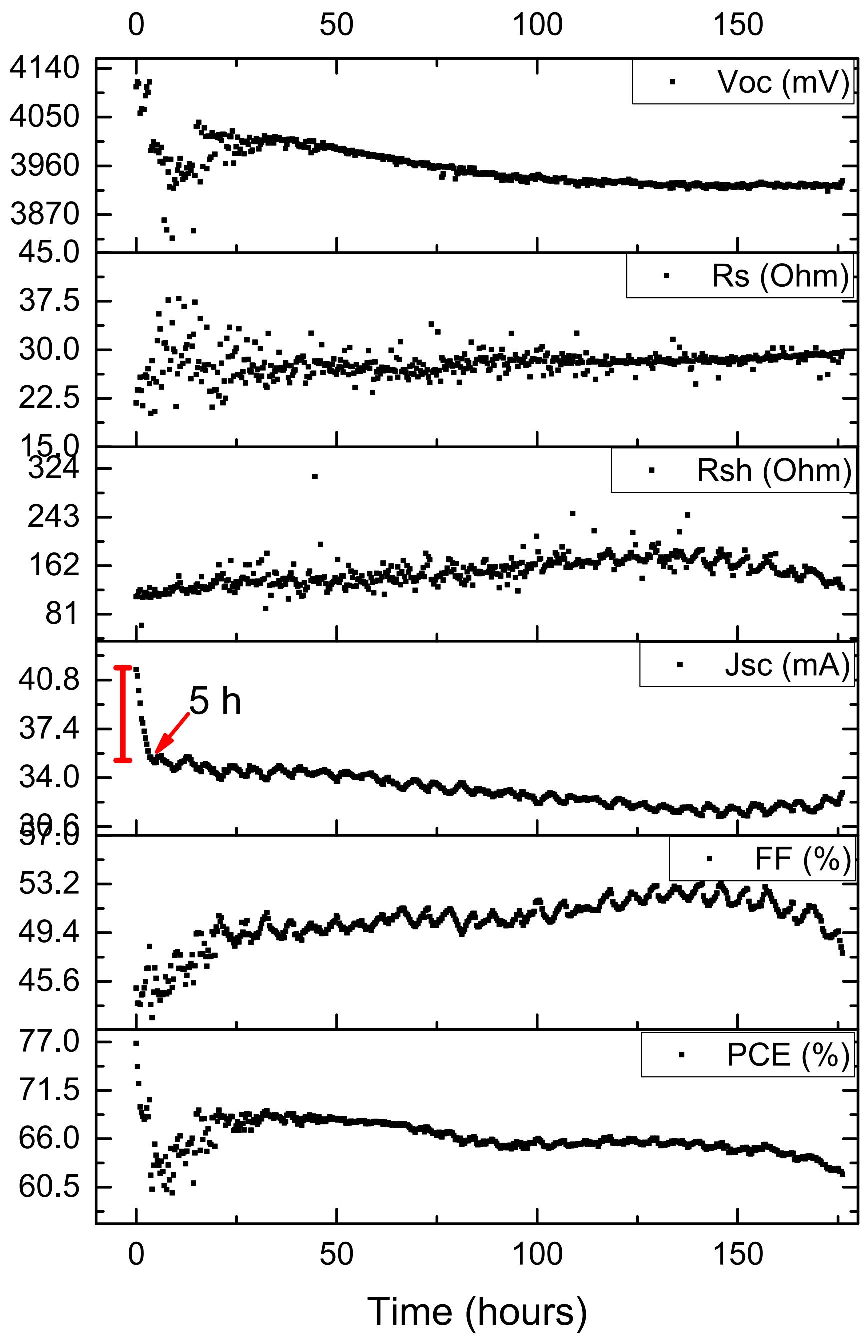


Fig. S14 PV parameters extracted from an IV scan every 20 minutes during the light soaking test of the perovskite module.

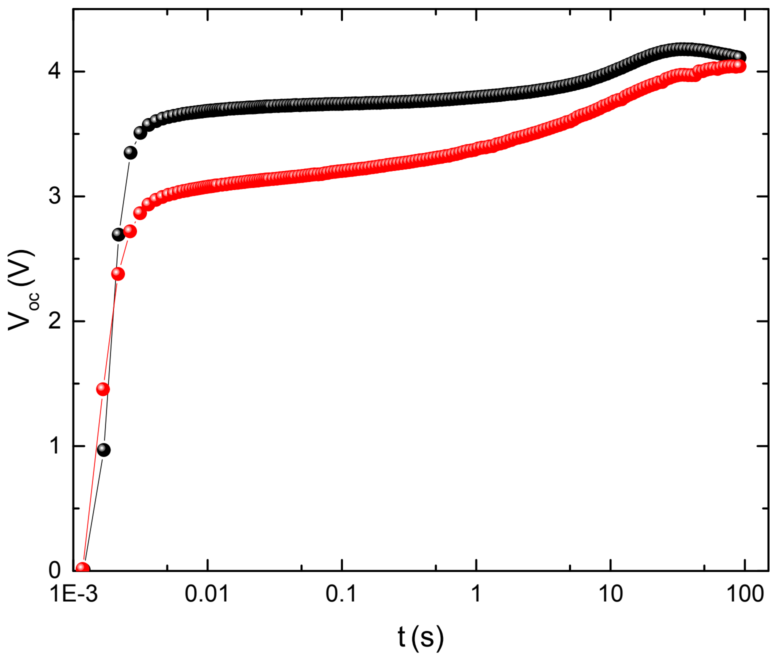


Fig. S15 Open-voltage voltage rise test of Perovskite module before after switching the light condition from dark to 1 SUN before (black) and after (red) the light soaking stress test.

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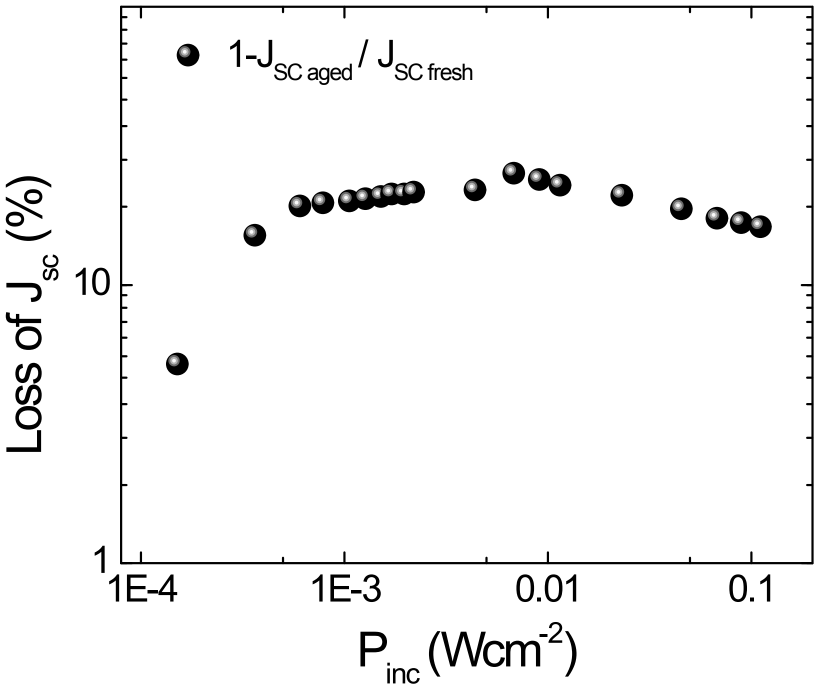

Fig. S16 percentage loss of J_{sc} after light soaking test evaluated for different light intensity levels.

a) b)

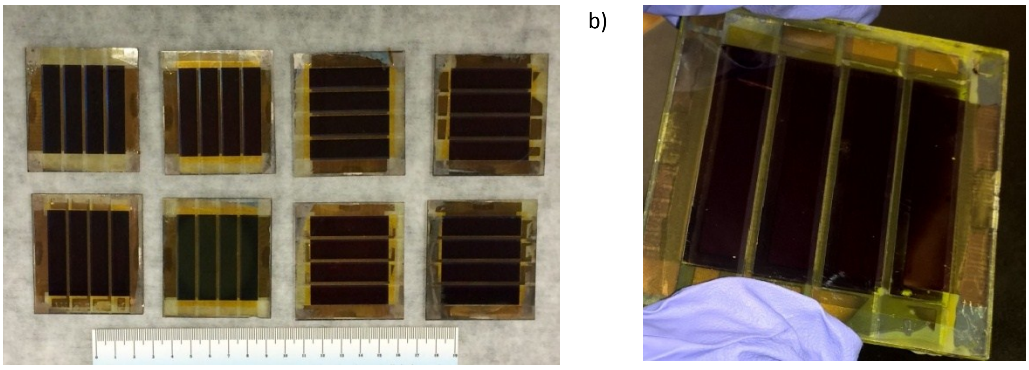

Fig. S17 a) CE Fresh modulus. b) CE module 7 months after light soaking test.

Table S4 photovoltaic parameters of the CE module 7 months after light soaking test.

HTM	Fabrication Method	Potential Scan Direction	Voc (V)	Jsc (mA/cm^2)	FF	PCE (%)
spiro-OMeTAD	CE	Voc to 0	4.064	13.315	0.642	8.69
		0 to Voc	3.994	17.876	0.339	6.05

