SUPPLEMENTAL INFORMATION

Iodine-trapping Strategy for Light-heat Inverted Stable Perovskite Solar Cells under ISOS Protocols

Xiaodong Li*, Hui Yang, Acan Liu, Chunyan Lu, Haobo Yuan, Wenxiao Zhang and Junfeng Fang*

School of Physics and Electronic Science, Engineering Research Center of Nanophotonics & Advanced Instrument, Ministry of Education, East China Normal University, Shanghai 200062, China

E-mail: xdli@phy.ecnu.edu.cn; jffang@phy.ecnu.edu.cn

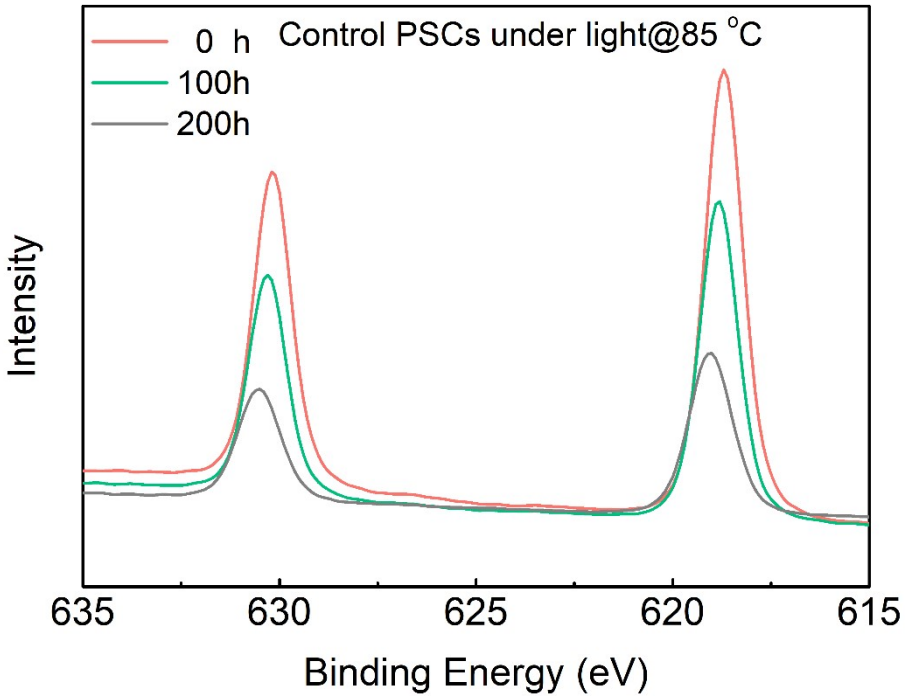

Fig. S1. XPS of I 3d in control PSCs under light and 85 °C aging.

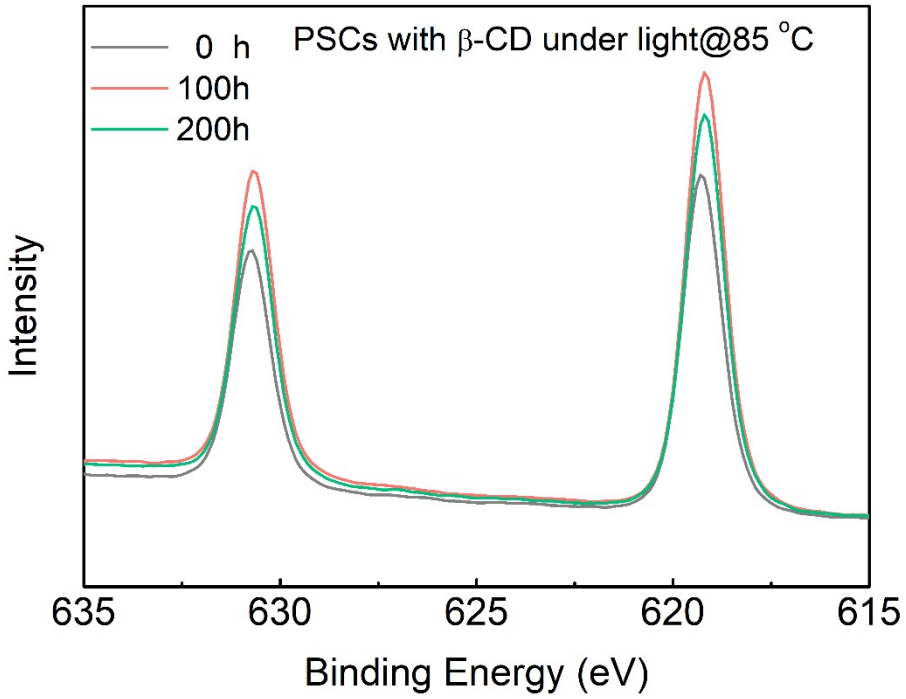

Fig. S2. XPS of I 3d in PSCs with β-CD under light and 85 °C aging.

Sample	Aging time	I	Pb	I/Pb	$Pb^0/(Pb^0+Pb^{2+})$
PSCs@β-CD	0 h	77.33%	22.67%	3.41	2.34%
	100 h	76.76%	23.24%	3.3	2.70%
	200 h	76.48%	23.52%	3.25	2.68%
Control PSCs	0 h	78.33%	21.67%	3.61	2.47%
	100 h	76.80%	23.20%	3.31	2.90%
	200 h	71.68%	28.32%	2.53	4.41%

Table S1. I, Pb atomic concentration, I/Pb atomic ratio and Pb0 content obtained from XPS.

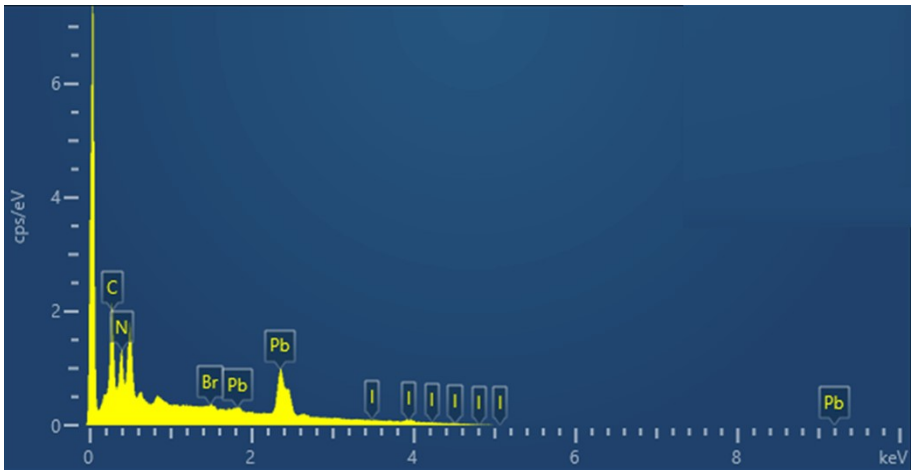

Fig. S3. I and Pb spectra during SEM-EDS mapping in control perovskite.

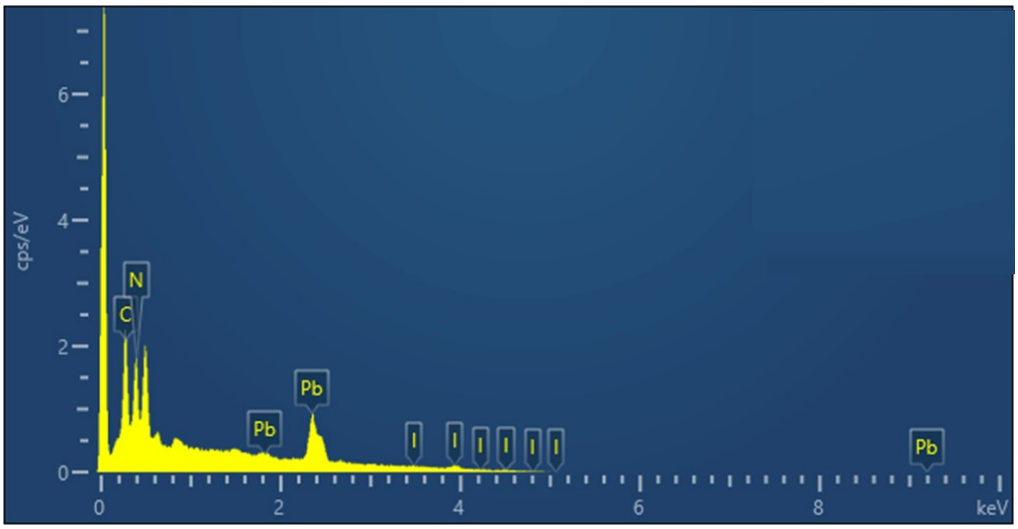

Fig. S4. I and Pb spectra during SEM-EDS mapping in perovskite with β-CD.

Sample	I	Pb	C	I/Pb
Aged perovskite@β-CD	41.12%	13.29%	45.59%	3.09
Aged control perovskite	16.06%	37.92%	46.02%	2.36

Table S2. C, I and Pb atomic concentration and I/Pb atomic ration obtained in SEM-EDS mapping.

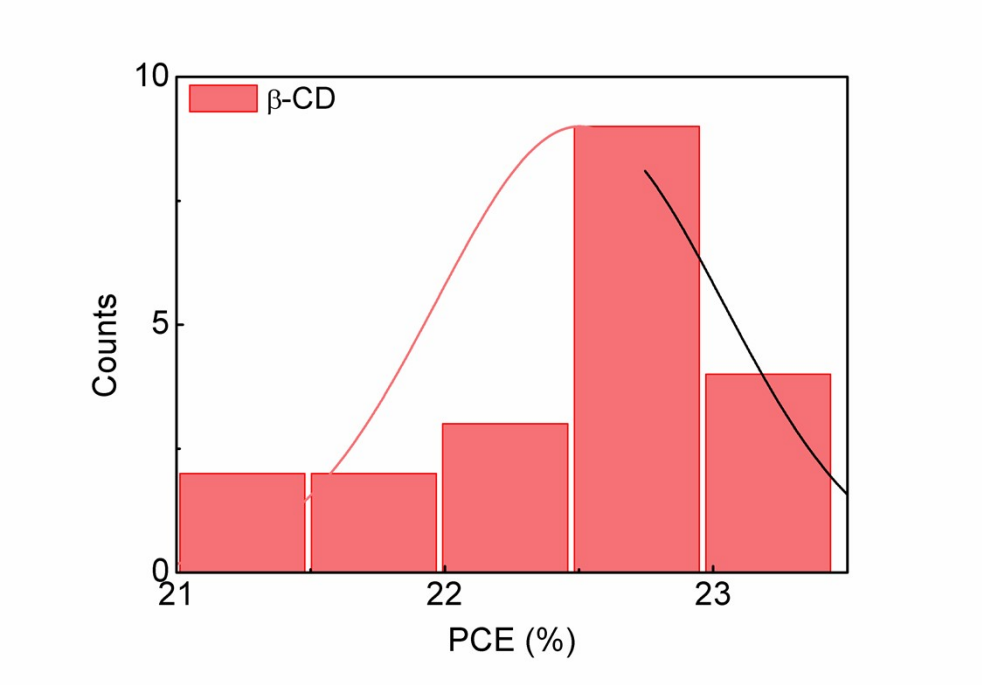

Fig. S5. Statistic distribution of device efficiency in PSCs with β-CD among 20 devices.

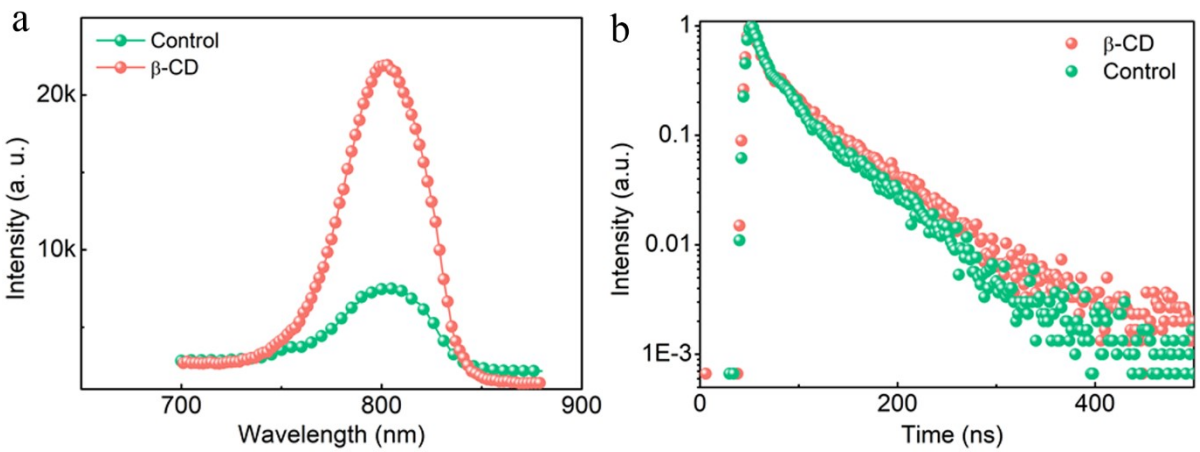

Fig. S6. Defects passivation in perovskite with β-CD. (a) PL and (b) TRPL of perovskite films.

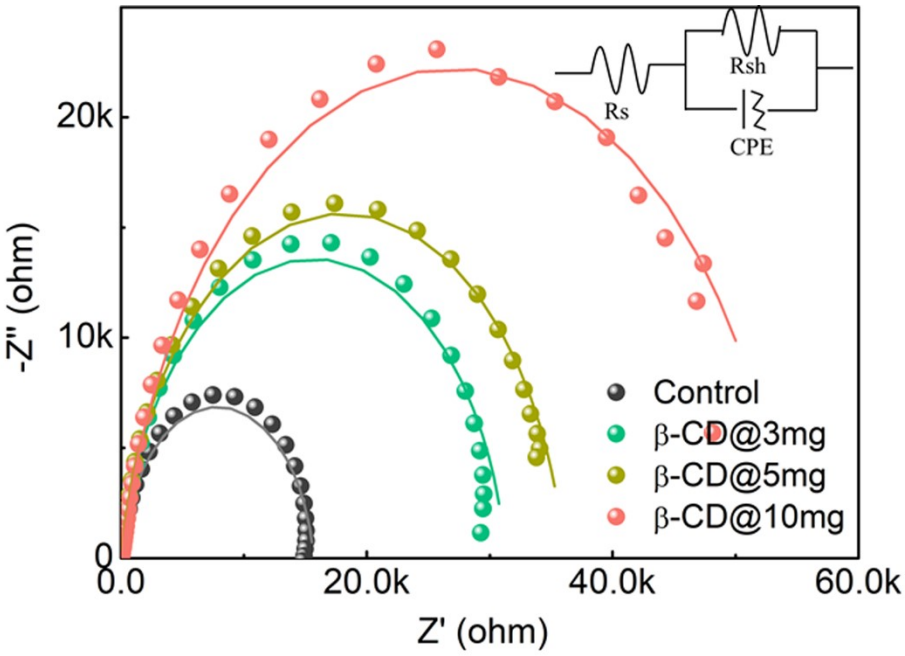

Fig. S7. Electrochemical impedance spectroscopy (EIS) of PSCs with different concentration of β-CD.

Concentration of β-CD (mg/mL)	Rs (Ω)	Rsh (kΩ)
0	27.2	15.6
3	32.2	31.3
5	31.1	36.0
10	50.6	54.0

Table S3. The series (Rs) resistance and shunt resistance (Rsh) of PSCs obtained from EIS fitting.

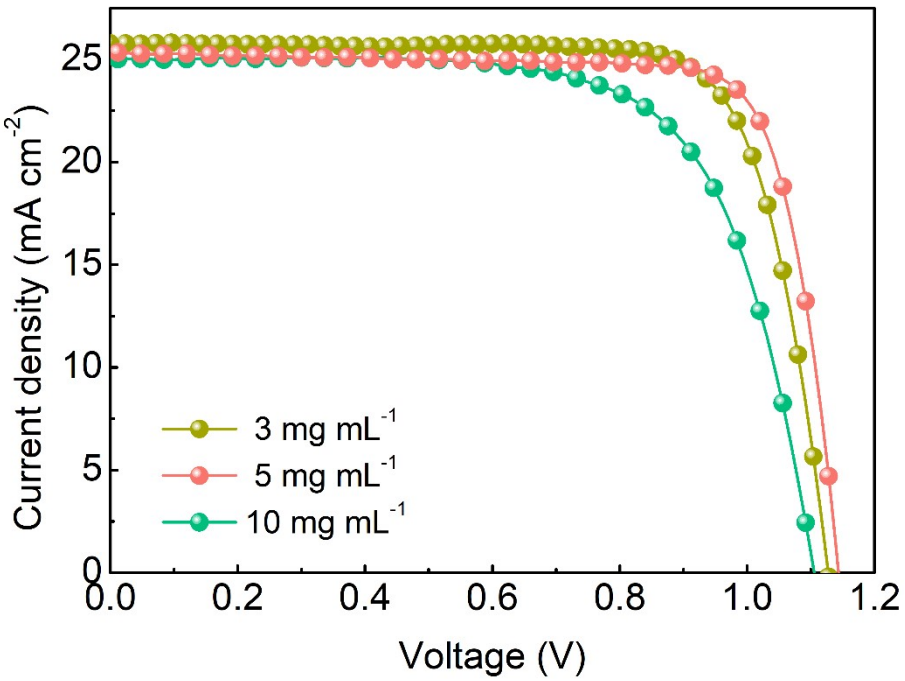

Fig. S8. The *J-V* curves of PSCs with different concentration of β-CD.

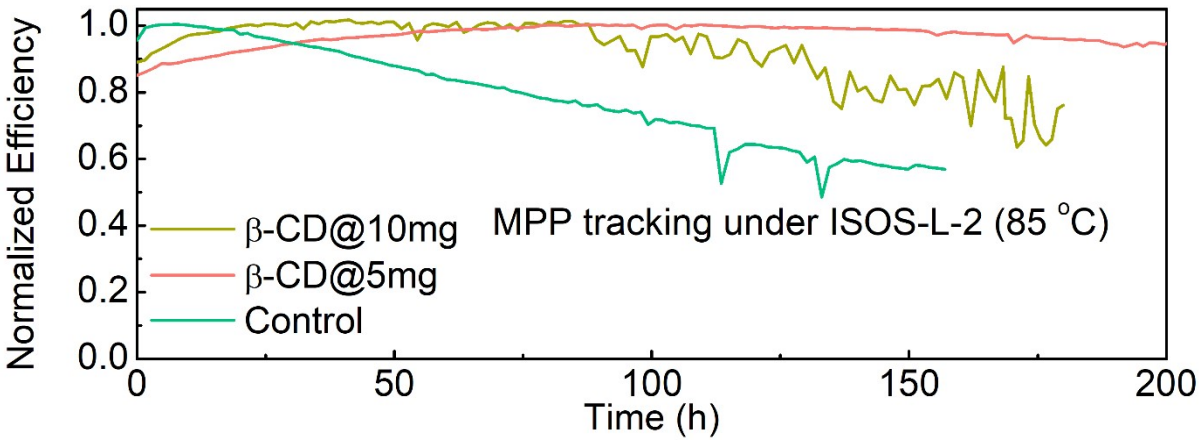

Fig. S9. The effect of β-CD concentration on device stability.

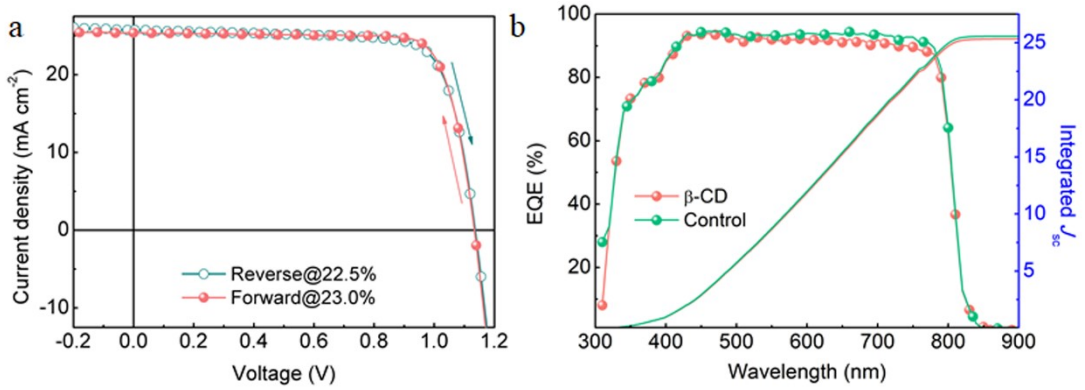

Fig. S10. (a) *J-V* hysteresis and (b) EQE and integrated current density in PSCs with β-CD.

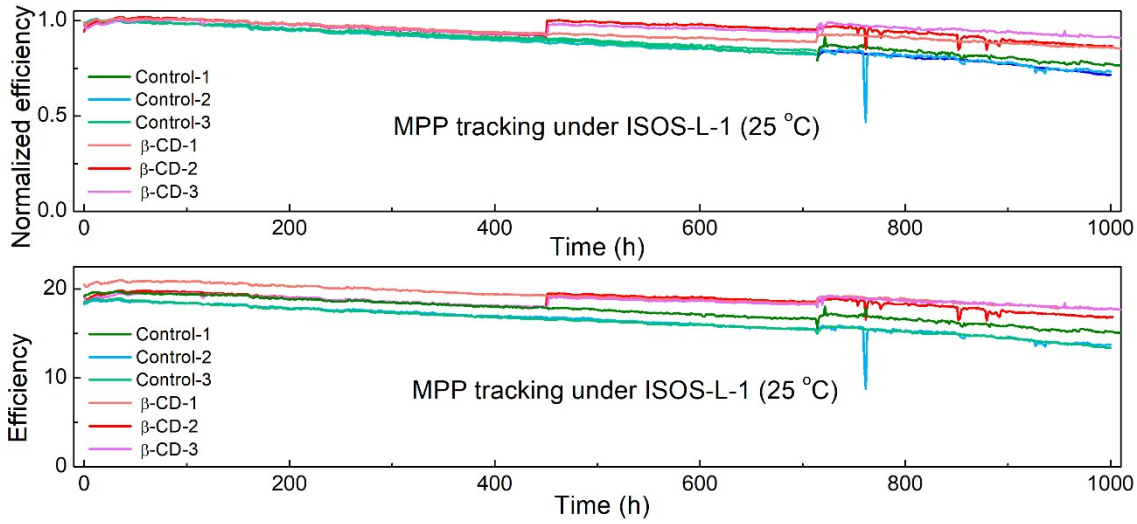

Fig. S11. MPP tracking and non-normalized data of 3 separated PSCs under ISOS-L-1 conditions at 25 °C.

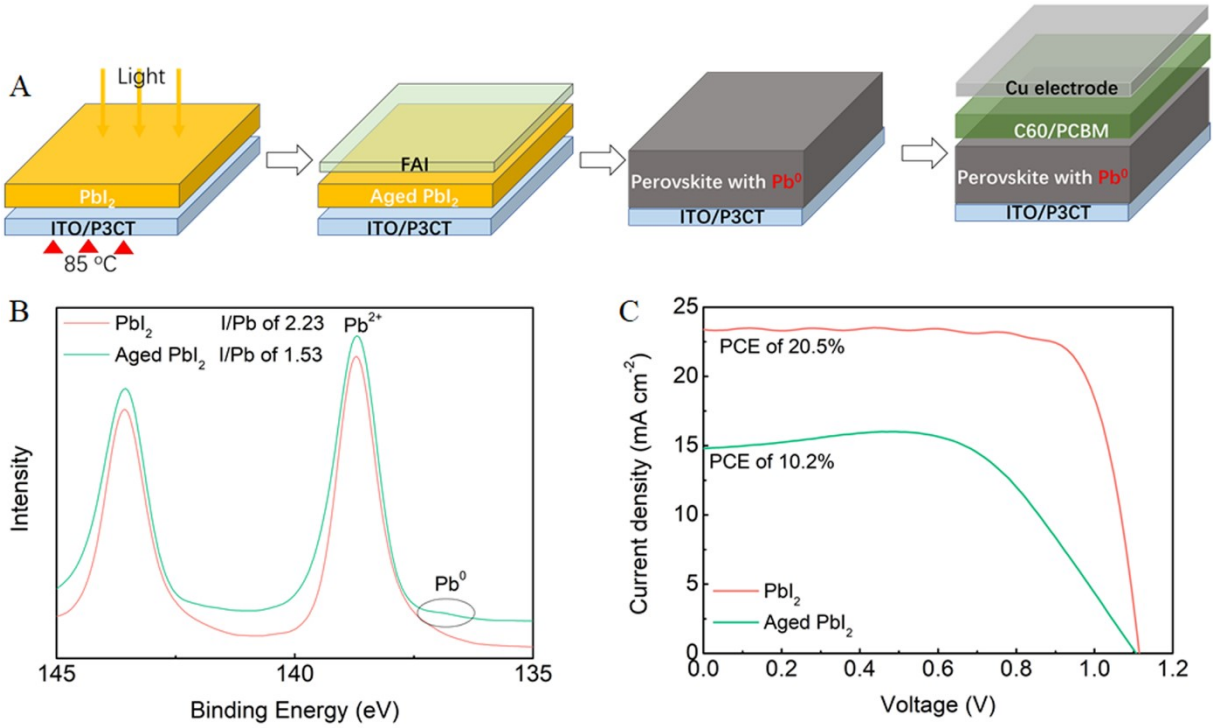

Fig. S12. Extra experiment to verify the harmful effect of Pb^0 on devices performance. (a) Schematic diagram of experiment, (b) XPS of Pb 4f in fresh and aged PbI_2 films, (c) *J-V* curves of PSCs fabricated with fresh or aged PbI2 films with typical two-step method.

To exclude other components (FAI or CsI species) effect and provide direct evidence of Pb^0 on devices performance, we spin-coat PbI_2 on ITO/P3CT substrates and then put these samples under light-heat (85 °C) conditions for 50 h to introduce some Pb^0 sites. Then we further spin-coat fresh FAI solution onto these PbI_2 films to fabricate PSCs using typical two-step methods.

XPS results on aged PbI_2 show that I/Pb atom ratio decreases from initial 2.23 to 1.53 after aging, together with obvious Pb^0 signals (~136.8 eV, Fig. S12b), further confirming the existence of iodine escaping and Pb^0 generation even in PbI_2 films. For PSCs with fresh PbI_2, good efficiency of over 20% can be obtained (Fig. S12c). While in PSCs with aged PbI_2 containing Pb^0 sites, much lower efficiency of ~10% is obtained. Since the FAI component in PSCs is fresh without any degradation, such low efficiency can only be caused by Pb^0, confirming its harmful effect on PSCs performance.

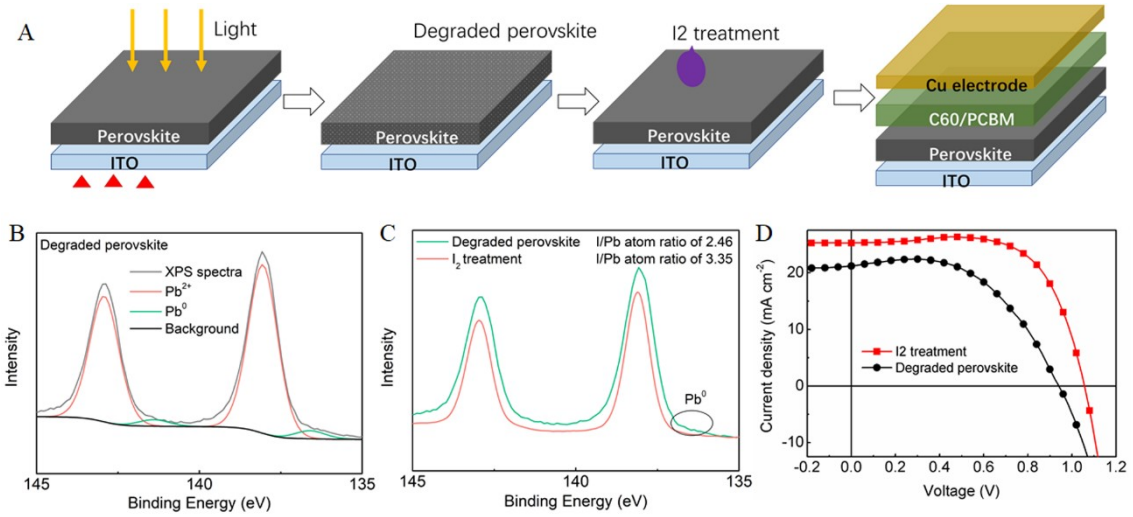

Fig. S13. Extra experiment to verify the beneficial effect of effect of I_2 trapping on device. (a) Schematic diagram of experiment, (b) XPS of Pb 4f in aged perovskite films, (c) XPS of Pb 4f in aged perovskite films after I_2 treatment, (d) *J-V* curves of PSCs fabricated with aged perovskite films with or without I_2 treatment.

To confirm that I_2 can indeed react with the Pb^0 generated in perovskite films, we design an experiment as shown in Fig. S13a. Perovskite films are aged under light and 85 °C conditions for 200 h. Then the degraded perovskite films are treated with I_2 solution in CB (1-3 mg/mL). XPS results demonstrate that Pb^0 sites appear in aged perovskite films (Fig. S13b). However, after I_2 treatment, the Pb^0 peaks disappear, indicating its reaction with I_2. I/Pb atom ratio is also increased from 2.46 of degraded perovskite to 3.35 after I_2 treatment (Fig. S13c), further demonstrating the reaction between I_2 and Pb^0 even in real perovskite conditions.

In addition, if we use such degraded perovskite films to fabricate PSCs, much lower efficiency of ~11% is obtained (Fig. S13d). However, if we treat such perovskite film with I_2 to eliminate Pb^0 sites and then fabricate PSCs, the devices efficiency can recover to over 18% (Fig. S13d). This result directly demonstrates that iodine escaping and Pb^0 generation in perovskite films indeed strongly affect devices performance and that I_2 can react with and eliminate Pb^0 sites to inhibit its harmful influence on devices performance.

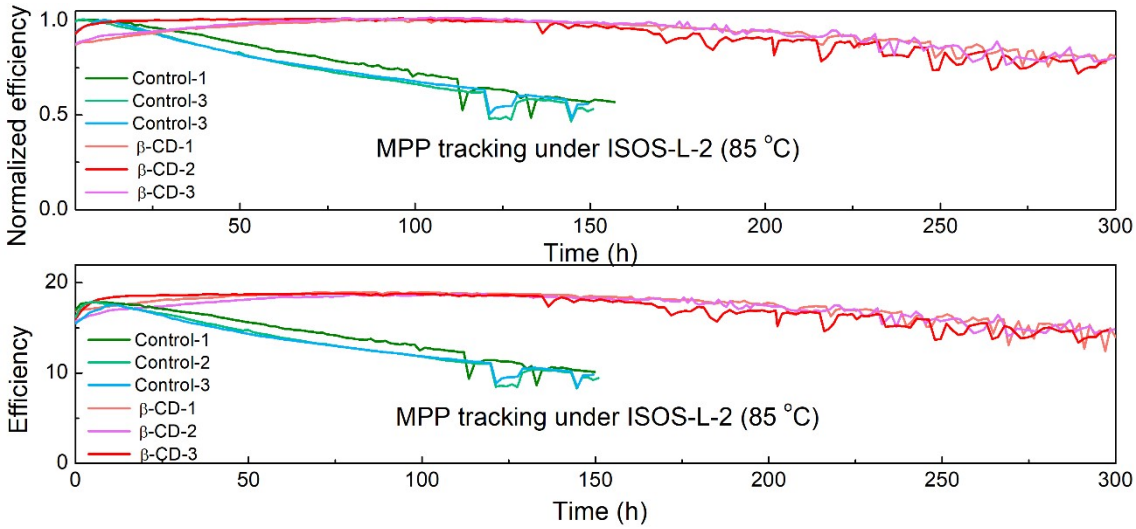

Fig. S14. MPP tracking and non-normalized data of 3 separated PSCs under ISOS-L-2 conditions at 85 °C.

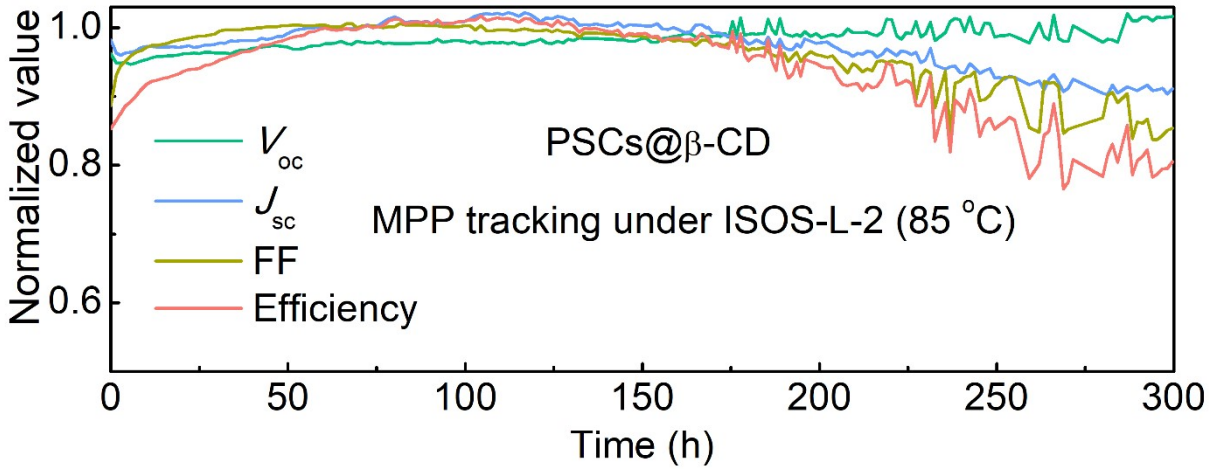

Fig. S15. Specific parameter variation during MPP tracking under ISOS-L-2 (take device of β-CD-2 in Fig. S14 for example).