Supporting Information

Lead (Pb) Management in the Entire Life Cycle of Highly Efficient and Stable Perovskite Solar Cells

Kai Liu[a], Tianxiang Hu[a], Zenghua Cai[b], Fengcai Liu[a], Saqib Rafique[c], Xiaoguo Li[a], Liangliang Deng[a], Chongyuan Li[a], Yanyan Wang[a], Qiang Guo[a], Xiaofei Yue[a], Jiao Wang[a], Yingguo Yang[d], Chunxiao Cong[a], Shiyou Chen[e], Jia Zhang[f], Anran Yu[*a], Yiqiang Zhan[*a,f]

a. Center for Micro Nano Systems, School of Information Science and Technology (SIST), Fudan University, 200433 Shanghai, P. R. China

Email: aryu@fudan.edu.cn; yqzhan@fudan.edu.cn

b. Jiangsu Key Laboratory of Micro and Nano Heat Fluid Flow Technology and Energy Application, School of Physical Science and Technology, Suzhou University of Science and Technology, Suzhou, 215009, China

c. Oxide and chalcogenide MOCVD Centre Room 220, CSIM Building, Swansea University, Bay Campus, Fabian Way, Crymlyn Burrows, Skewen, Swansea, SA18EN

d. School of Microelectronics, Fudan University, Shanghai, 200433, P. R. China

e. Key Laboratory of Computational Physical Sciences (MOE), and State Key Laboratory of ASIC and System, School of Microelectronics, Fudan University, Shanghai 200433, China

f. State Key Laboratory of Photovoltaic Science and Technology, Institute of Optoelectronics, Fudan University, Shanghai 200438, China

* Corresponding author

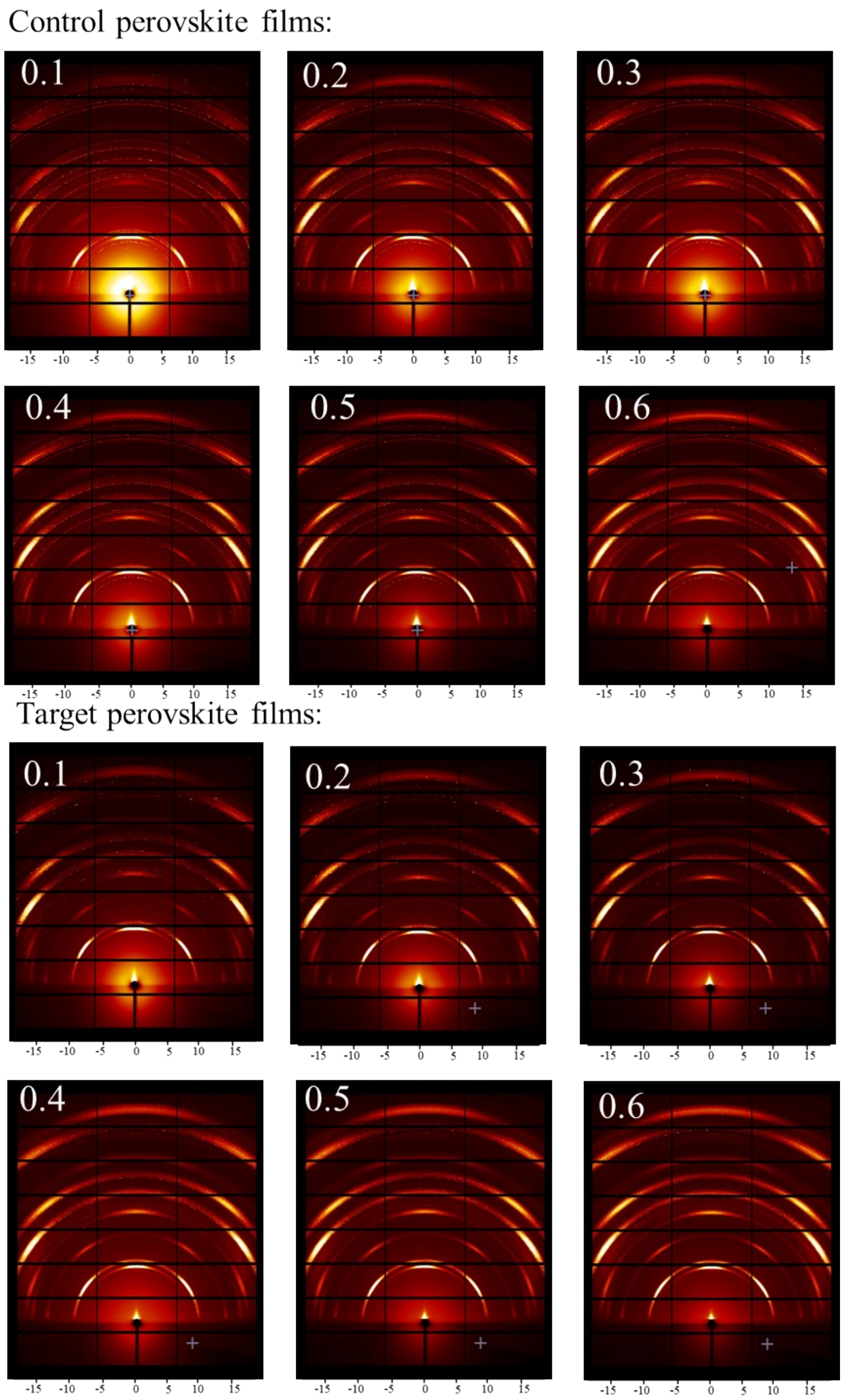


Figure S1. 2D-GIWAXS images of perovskite films at various incident angel ranging from 0.1° to 0.6°.

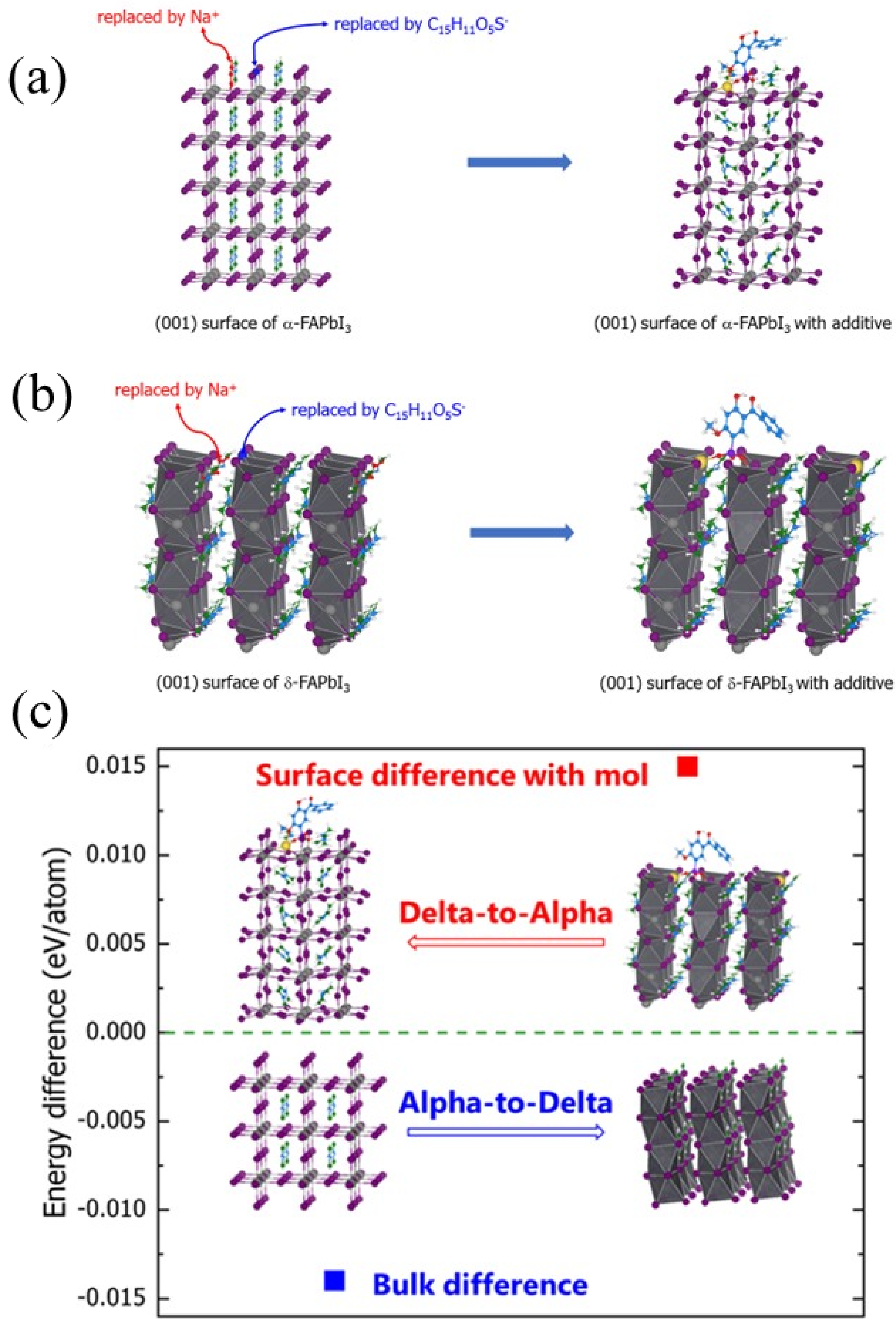


Figure S2. Theoretical simulation of FA perovskites with BP molecules. (a-b) The process of constructing the stable surface structure of $FAPbI_3$ with BP molecules. (c) DFT calculation results about the total energy difference w/wo BP-5 at the surface of $FAPbI_3$.

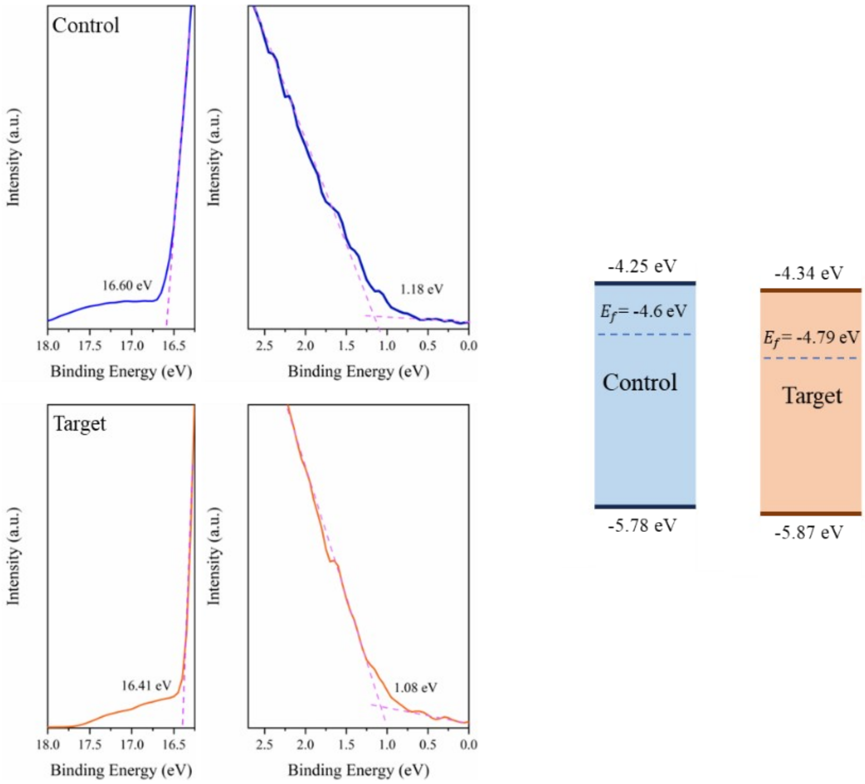


Figure S3. The UPS results and the energy band illustration of control and target perovskite films.

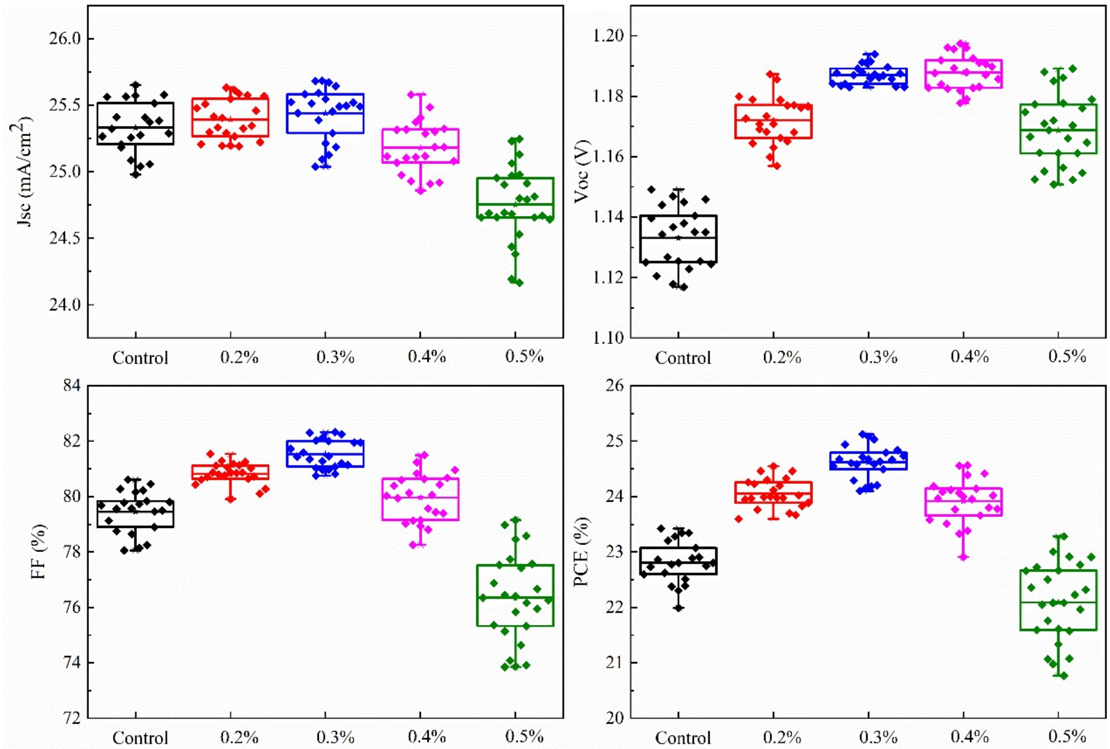
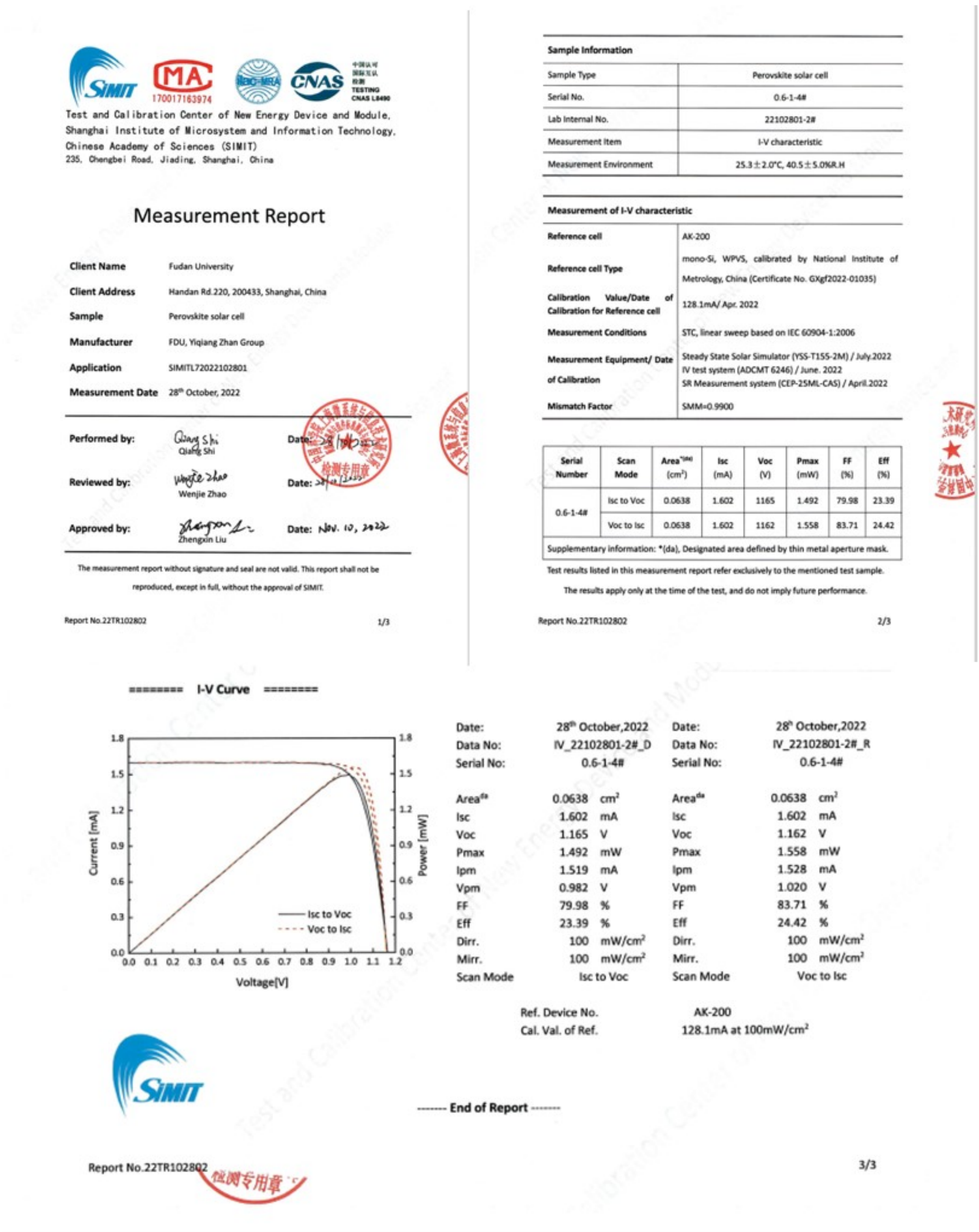


Figure S4. The photovoltaic parameters statistics of the control and target devices with different concentration of BP-5 molecules.



Test and Calibration Center of New Energy Device and Module, Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences (SIMIT)
235, Chengbei Road, Jiading, Shanghai, China

Measurement Report

Client Name	Fudan University
Client Address	Handan Rd.220, 200433, Shanghai, China
Sample	Perovskite solar cell
Manufacturer	FDU, Yiqiang Zhan Group
Application	SIMITL72022102801
Measurement Date	28th October, 2022

Performed by: Qiang Shi **Date:**
Reviewed by: Wenjie Zhao **Date:**
Approved by: Zhengxin Liu **Date:** Nov. 10, 2022

The measurement report without signature and seal are not valid. This report shall not be reproduced, except in full, without the approval of SIMIT.

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Sample Information

Sample Type	Perovskite solar cell
Serial No.	0.6-1-4#
Lab Internal No.	22102801-2#
Measurement Item	I-V characteristic
Measurement Environment	25.3±2.0°C, 40.5±5.0%R.H

Measurement of I-V characteristic

Reference cell	AK-200
Reference cell Type	mono-Si, WPVS, calibrated by National Institute of Metrology, China (Certificate No. GXgf2022-01035)
Calibration Value/Date of Calibration for Reference cell	128.1mA/ Apr. 2022
Measurement Conditions	STC, linear sweep based on IEC 60904-1:2006
Measurement Equipment/ Date of Calibration	Steady State Solar Simulator (YSS-T155-2M) / July.2022 IV test system (ADCMT 6246) / June. 2022 SR Measurement system (CEP-25ML-CAS) / April.2022
Mismatch Factor	SMM=0.9900

Serial Number	Scan Mode	Area*(da) (cm^2)	Isc (mA)	Voc (V)	Pmax (mW)	FF (%)	Eff (%)
0.6-1-4#	Isc to Voc	0.0638	1.602	1165	1.492	79.98	23.39
	Voc to Isc	0.0638	1.602	1162	1.558	83.71	24.42

Supplementary information: *(da), Designated area defined by thin metal aperture mask.

Test results listed in this measurement report refer exclusively to the mentioned test sample.
The results apply only at the time of the test, and do not imply future performance.

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======== **I-V Curve** ========

Date:	28th October,2022		Date:	28th October,2022	
Data No:	IV_22102801-2#_D		Data No:	IV_22102801-2#_R	
Serial No:	0.6-1-4#		Serial No:	0.6-1-4#	
Area(da)	0.0638	cm^2	Area(da)	0.0638	cm^2
Isc	1.602	mA	Isc	1.602	mA
Voc	1.165	V	Voc	1.162	V
Pmax	1.492	mW	Pmax	1.558	mW
Ipm	1.519	mA	Ipm	1.528	mA
Vpm	0.982	V	Vpm	1.020	V
FF	79.98	%	FF	83.71	%
Eff	23.39	%	Eff	24.42	%
Dirr.	100	mW/cm^2	Dirr.	100	mW/cm^2
Mirr.	100	mW/cm^2	Mirr.	100	mW/cm^2
Scan Mode	Isc to Voc		Scan Mode	Voc to Isc	

Ref. Device No. AK-200
Cal. Val. of Ref. 128.1mA at $100mW/cm^2$

------- **End of Report** -------

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Figure S5. The certified performance of the target device measured at SIMIT.

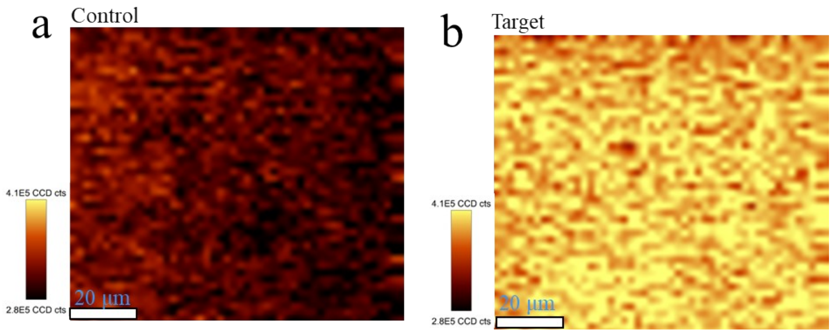


Figure S6. PL mapping images of control and target perovskite films.

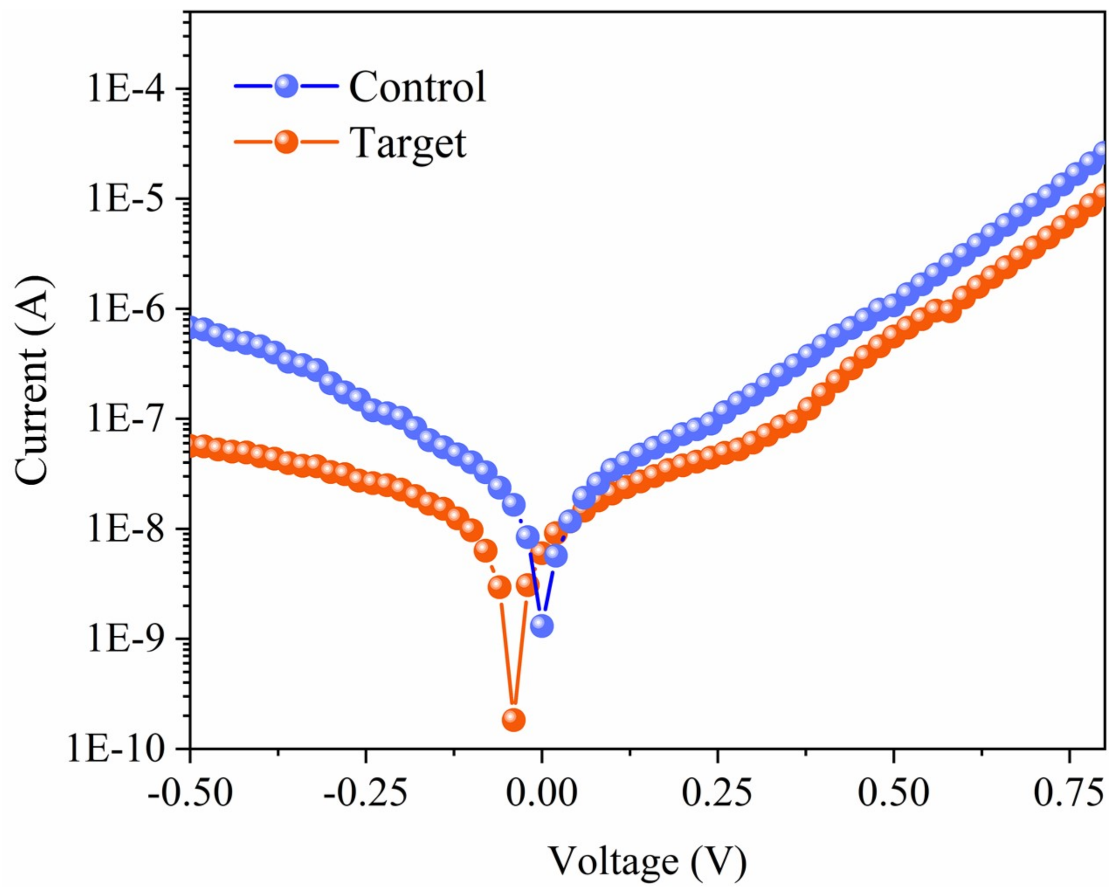
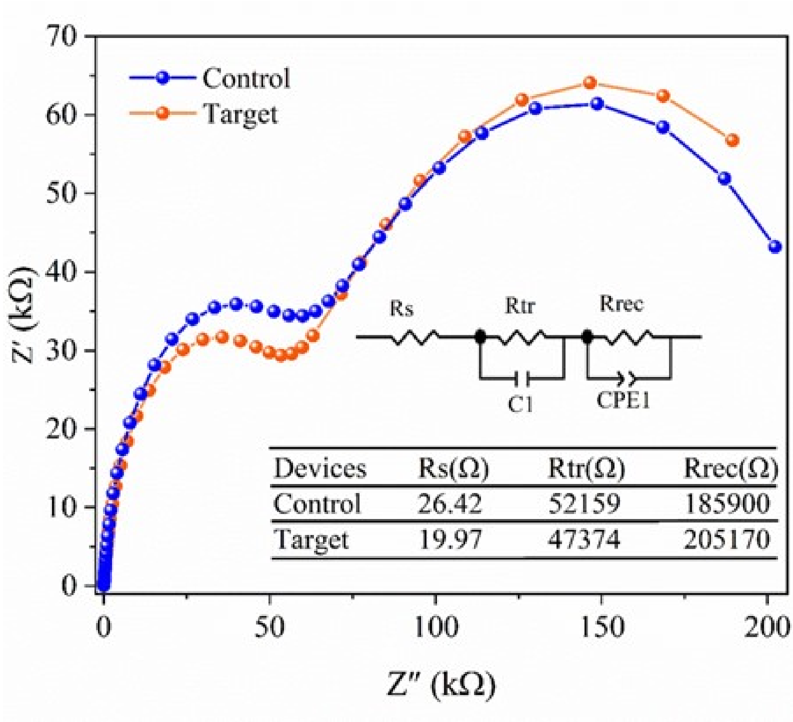


Figure S7. The dark current of the control and target device.



Devices	Rs(Ω)	Rtr(Ω)	Rrec(Ω)
Control	26.42	52159	185900
Target	19.97	47374	205170

Figure S8. The EIS spectra of control and target devices. The fitted results are presented in the inset-table.

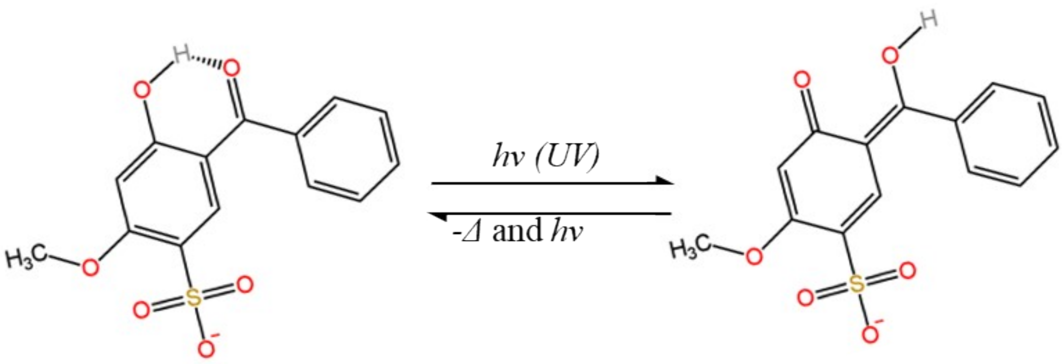


Figure S9. The mechanism of the UV absorption of BP-5 additives.

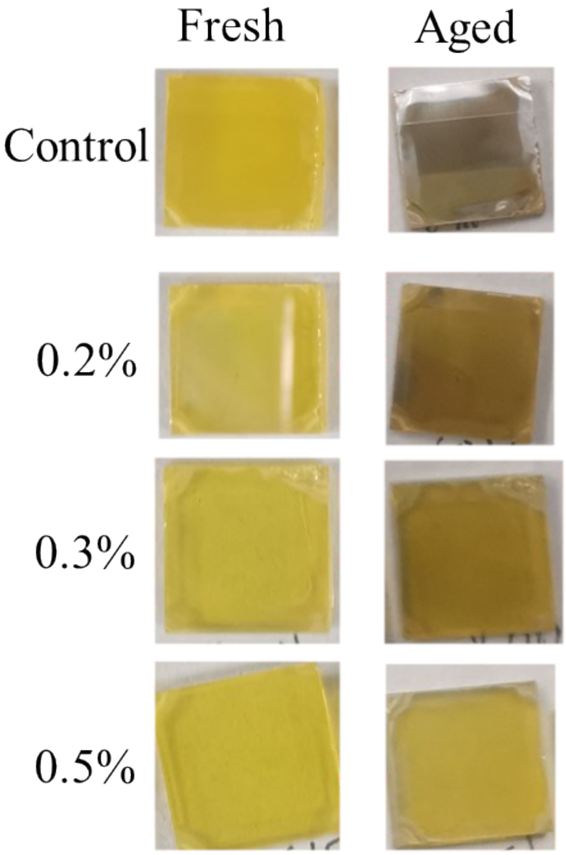


Figure S10. The digital photos of PbI_2 films with different concentration of BP-5 additives before and after UV aging test.

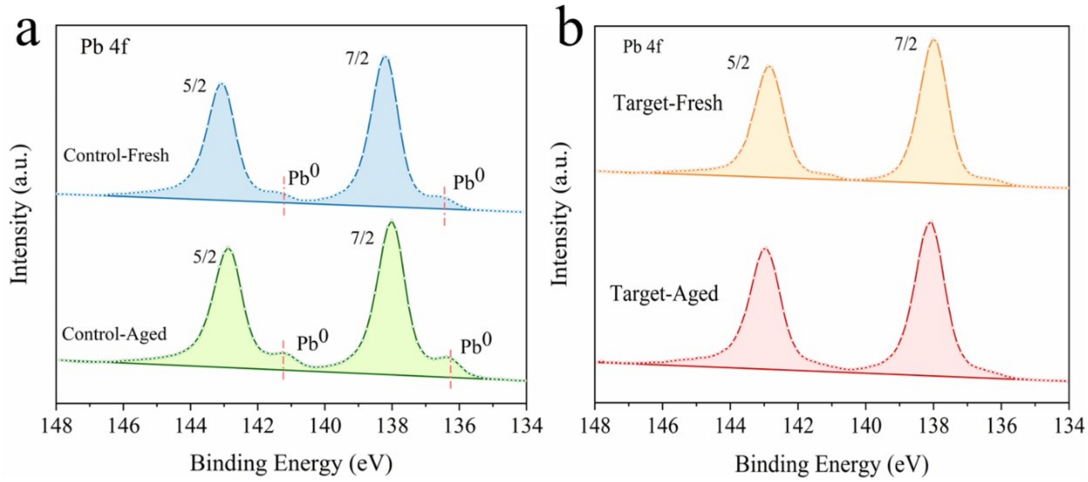


Figure S11. XPS analysis of control and target perovskite films. XPS spectra of control (a) and target films (b) before and after the UV aging period.

Figure S12. The pictures of different kinds of aqueous solutions. The clear black line indicates that there is no insoluble complex formed between BP-5 and lead iodide.

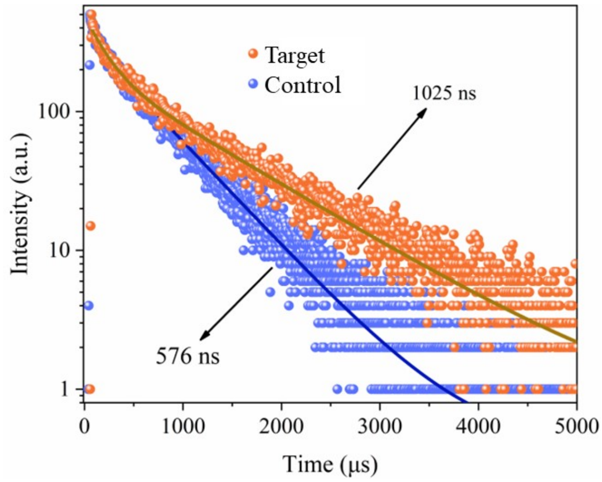


Figure S13. The TRPL results of control and target perovskite films (BP-9).

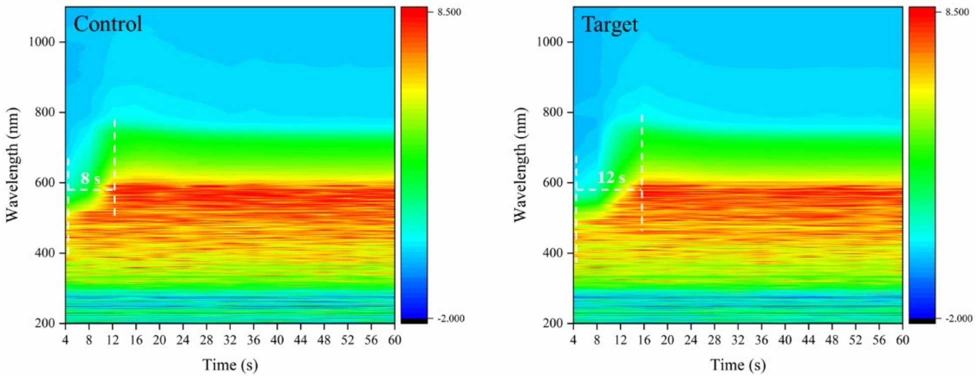


Figure S14. In-situ UV absorption spectra of control and target films during the initial annealing process at 150 °C

By using in-situ absorption spectra, we monitor the influence of BP-9 on crystallization process of perovskite. Figure S14 illustrates that the absorption edge of the control film shifts from 500 nm to 790 nm during the annealing process lasting from 4 s to 12 s, marking the transition from the intermediate phase to the perovskite phase. In contrast, the absorption edge of the target film extends to approximately 790 nm after 16 s of annealing, indicating a slower crystallization process in the target film. Moreover, the in-situ absorption spectra also revealed that after annealing for 16 s the control and target films both exhibit similar absorbance. This result suggests that the addition of BP additives cannot completely prevent the crystallization of perovskite, although it does slow down it through interaction.

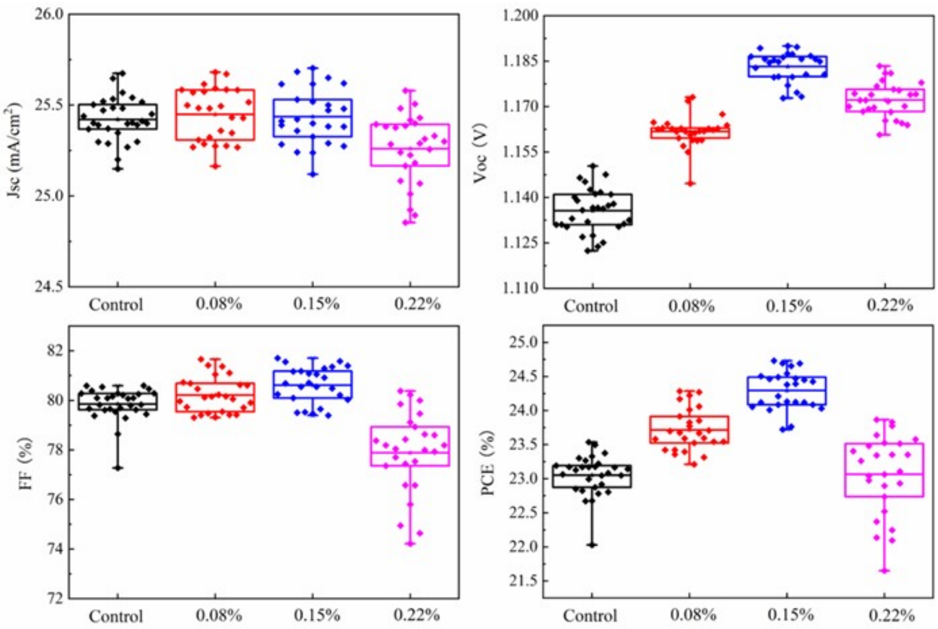


Figure S15. The photovoltaic parameters statistics of the control and target devices with different concentration of BP-9 molecules.

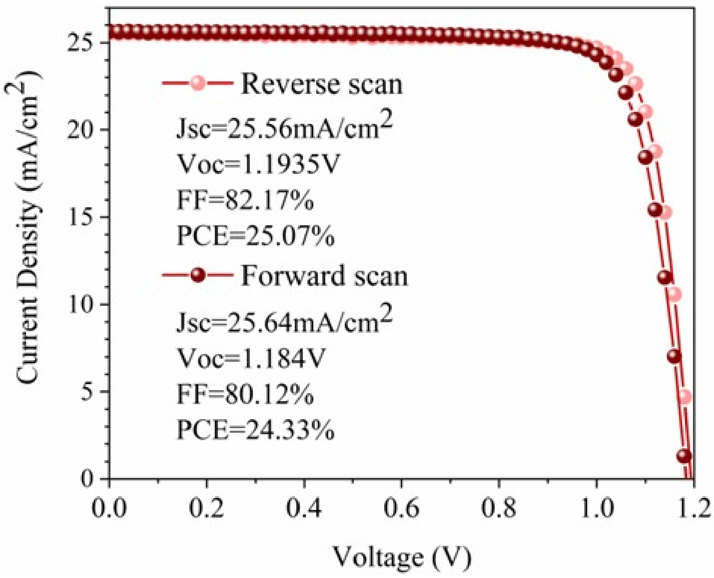


Figure S16. The efficiency of target devices (incorporated additives: BP-5 & BP-9). The device incorporated mixed additives (BP-5 &BP-9) shows similar efficiency in comparison to that of the champion device (incorporated additives: BP-5, Figure 3c)

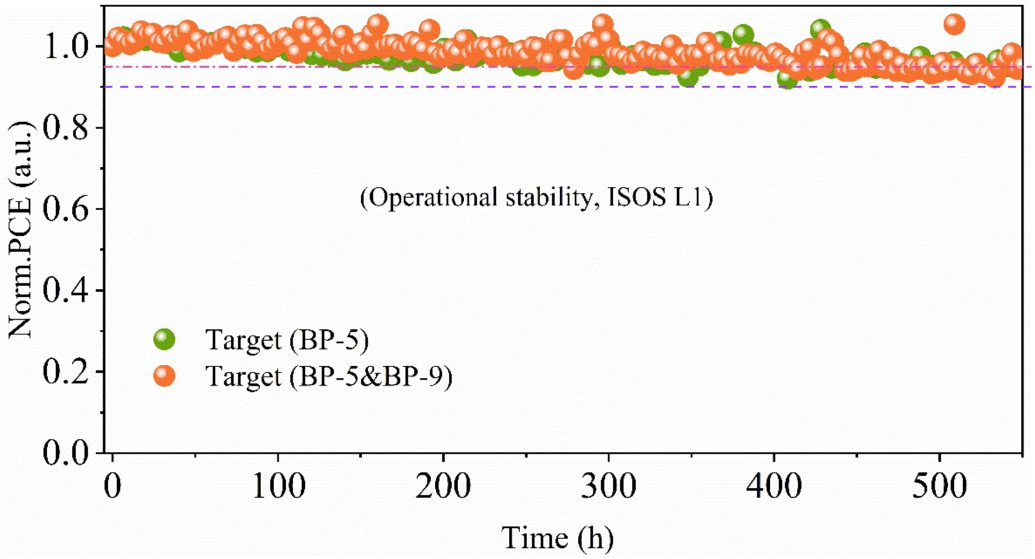


Figure S17. The operational stability of target devices. As shown in Figure S13, the target devices incorporated mixed additives (BP-5 and BP-9) also exhibits excellent operational stability.

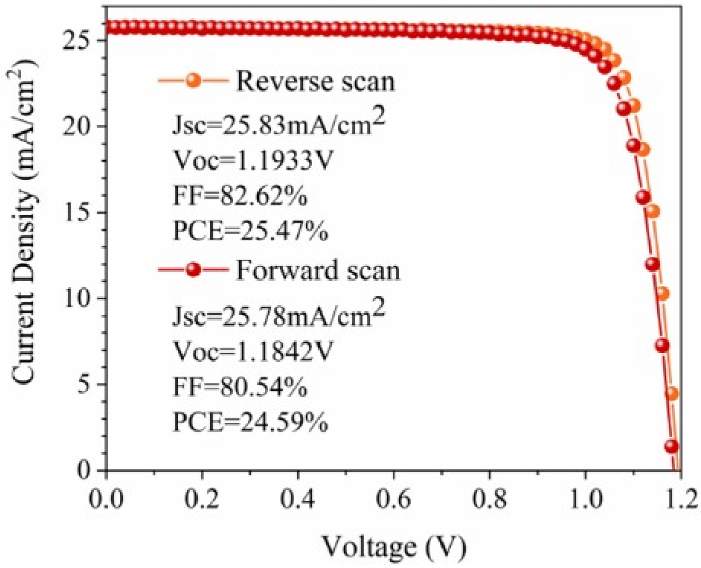


Figure S18. The efficiency of the latest n-i-p type PSCs fabricated by our strategy (BP-5 and BP-9)

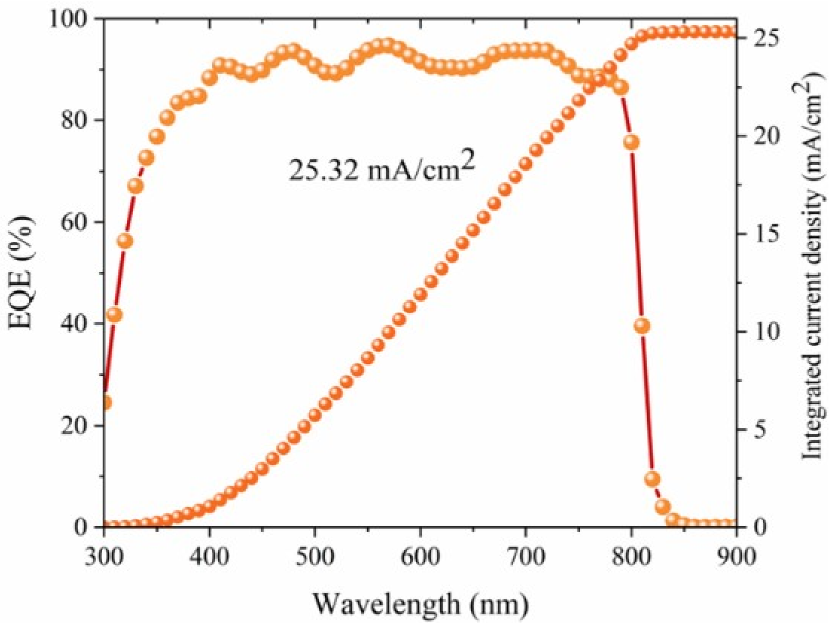


Figure S19. The EQE curve and the integrated J_{sc} of the target device.

Device	Scan Direction	Efficiency	Voc	FF	Jsc	Hysteresis Index
Control Device	Reverse scan	23.43%	1.1492V	79.70%	25.58 mA/cm^2	7.73%
	Forward scan	21.62%	1.1258V	75.62%	25.40 mA/cm^2	
Target Device	Reverse scan	25.13%	1.1913V	82.25%	25.64 mA/cm^2	2.51%
	Forward scan	24.50%	1.1832V	80.60%	25.69 mA/cm^2	

Table S1. The photovoltaic parameters of champion control and target devices.

Table S2. The efficiency of devices made by different additives (control, BP-5, BP-9, BP-5&BP-9).

Condition	PCE	FF	Voc	Jsc
Control	23.43%	79.70%	1.1492V	25.58 mA/cm2
BP-9	24.73%	81.36%	1.1865V	25.62 mA/cm2
BP-5	25.13%	82.25%	1.1913V	25.64 mA/cm2
BP-5&BP-9 (0.25%: 0.12%)	25.07%	82.17%	1.1935V	25.56 mA/cm2
BP-5&BP-9 (0.25%: 0.12%, Champion device)	25.47%	82.62%	1.1933V	25.83 mA/cm2
BP-5&BP-9 (0.25%: 0.15%)	24.82%	81.42%	1.1951V	25.51 mA/cm2
BP-5&BP-9 (0.25%: 0.18%)	24.05%	79.12%	1.1915 V	25.51 mA/cm2

Table S3. Photovoltaic performance (PCE and UV stability) of our devices compared with PSCs which were reported in the last two years.

Perovskite	PCE (%)	UV Stability (Time/retained initial efficiency)	Ref
$FA_xMA_{1-x}PbI_3$	**24.08%**	**300h/83% (UV: 365nm, 60 mW/cm^2)**	*Adv. Mater.*[1]
$FA_xMA_{1-x}PbI_3$	**24.26%**	**60h/100% (UV: 405nm, /)**	*Energy Environ. Sci.*[2]
$FA_xMA_{1-x}PbI_3$	**24.50%**	**1200h/87% (UV: 254nm, 50 mW/cm^2)**	*Joule* [3]
$FA_xMA_{1-x}PbI_3$	**24.58%**	**170h/86% (UV: 365nm, 60 mW/cm^2)**	*Adv. Energy Mater.*[4]
CsFAMA	**22.40%**	**70h/91% (UV: 365nm, 50 mW/cm^2)**	*Nano Energy*[5]
$FA_xMA_{1-x}PbI_3$	**24.50%**	**47h/90% (UV: 365nm, 119 mW/cm^2)**	*Adv. Funct. Mater.*[6]
FACsPb(I_xBr_{1-x})3	**21.54%**	**60h/85% (UV: 365nm, 60 mW/cm^2)**	*Small Methods*[7]
CsFAMA	**22.14%**	**72h/70% (UV: 275nm, 10 mW/cm^2)**	*J. Mater. Chem.*[8]
$FA_xMA_{1-x}PbI_3$	**25.47%**	**620h/96% (UV: 365nm, 22 mW/cm^2)**	**This work**

References

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