High-Efficiency and Stable Perovskite Solar Cells via DL-Methionine-Enhanced Crystallization and Defect Passivation

Xiaolan Li[a], Yanwei Xu[a], Fuxing Chen[a], Dongxing Lu[a], Yajun Zhu[a], Cong Li[b,*]

[a] Lanzhou Petrochemical University of Vocational Technology，Lanzhou 730060，China

[a] School of Advanced Materials and Nanotechnology, Xidian University, Xi'an 710126, China

***Corresponding author:**

Email: lc@xidian.edu.cn (C. Li)

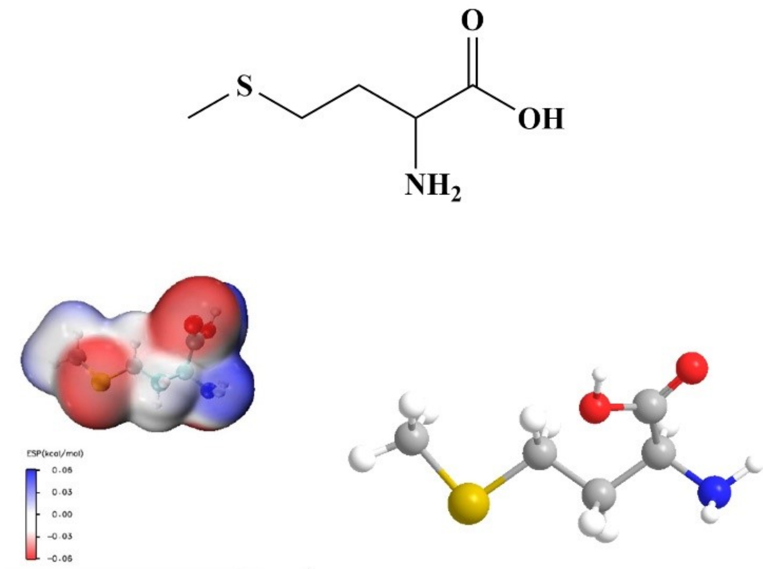


Figure S1. Molecular formula, electrostatic potential diagram, and ball and stick model of DL-methionine.

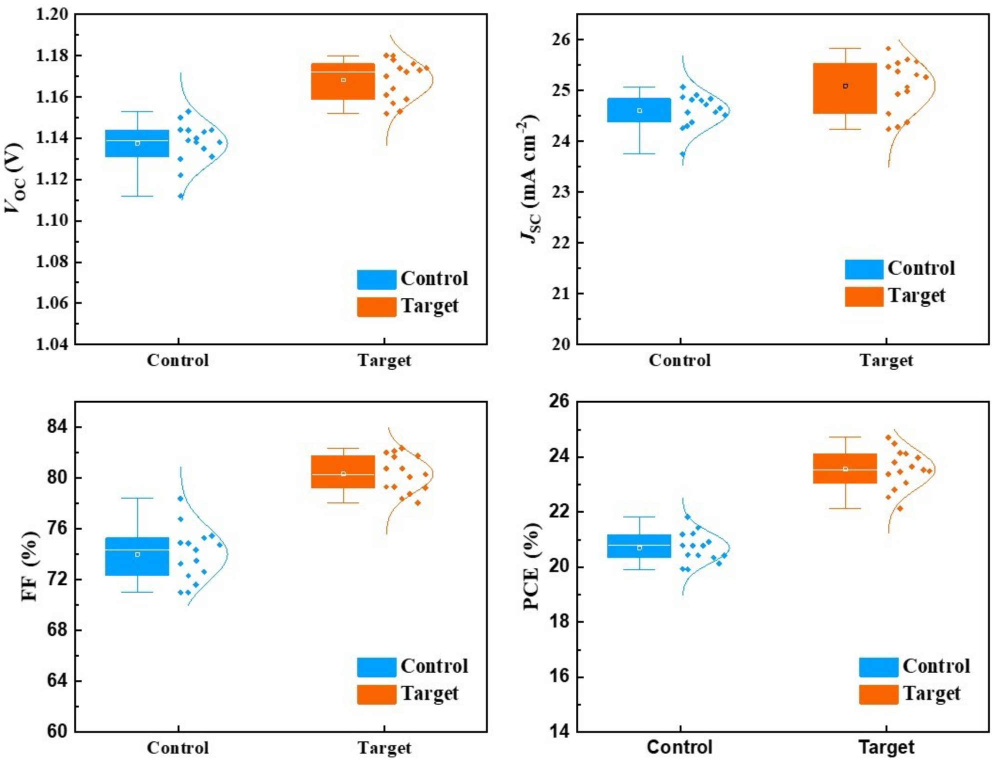
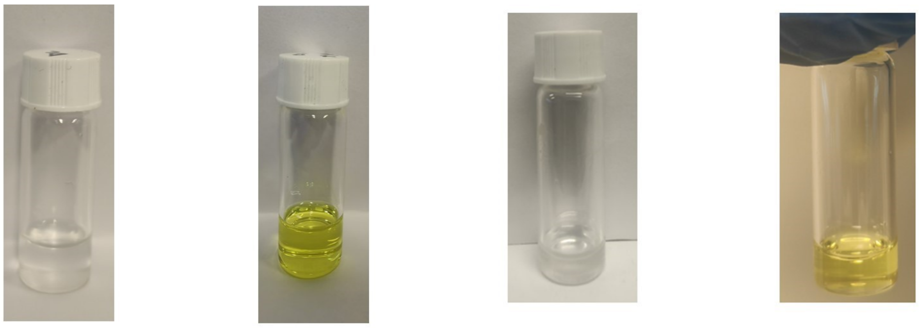


Figure S2. V_{OC}, J_{SC}, FF, and PCE distribution of 15 individual control and target PSCs.



DL-methionine in DMF   DL-methionine in PbI_2/DMF   DL-methionine in DMSO   DL-methionine in PbI_2/DMSO

Figure S3. Photos of 1 mg/ml methionine dissolved in DMF, PbI_2/DMF, DMSO, and PbI_2/DMSO solution.

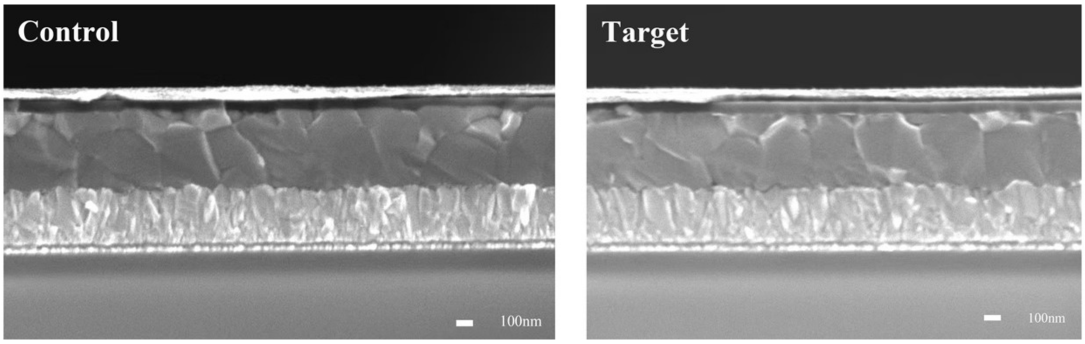


Figure S4. Cross-sectional SEM images of control and target PSCs.

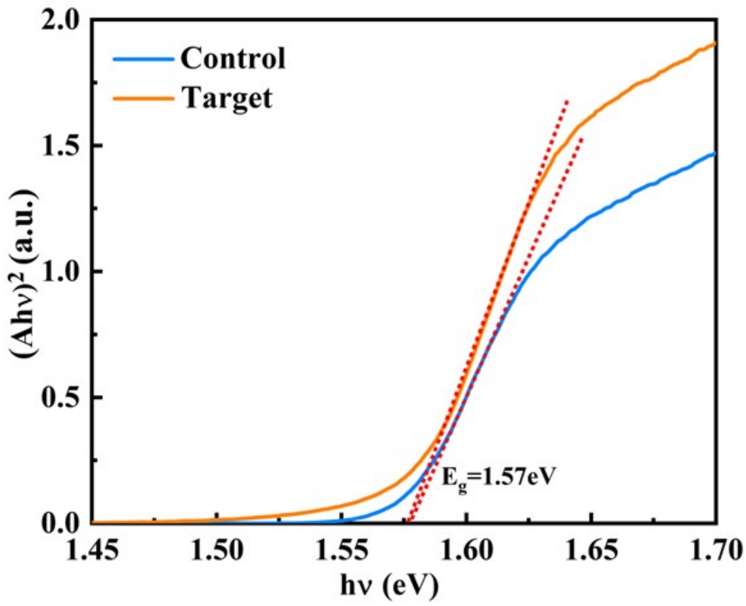


Figure S5. hv-$(ahv)^2$ curves of films.

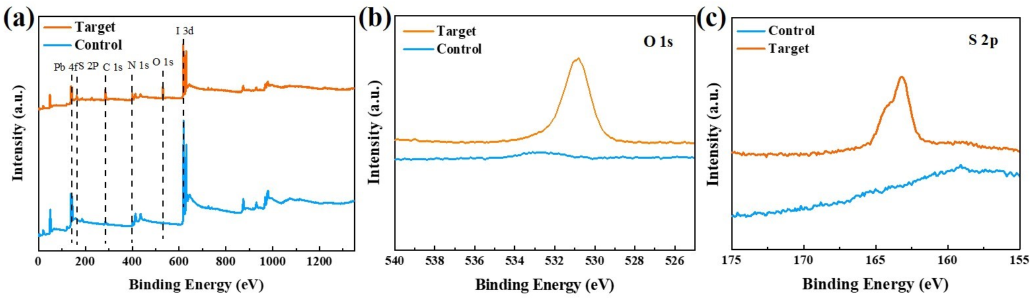


Figure S6. **(a)** XPS spectra of control and target perovskite films; XPS spectra of **(b)** O 1s and **(c)** S 2p in control and target perovskite films.

Table S1. Photovoltaic parameters of the champion control and target PSCs under forward and reverse scan.

	Scan	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
Control	Reverse	1.150	24.72	76.78	21.83
	Forward	1.142	24.56	74.77	20.97
Target	Reverse	1.180	25.54	82.01	24.72
	Forward	1.171	25.49	80.71	24.10

Table S2. The peak intensity of the perovskite films

		<100>	<110>	<111>	$I_{<100>}$ / $I_{<110>}$
Control	Intensity	557	204	194	2.73
Target	Intensity	1031	159	221	6.48

Table S3. TRPL fitting parameters of control and target perovskite films.

	A_1	τ_1 (ns)	A_2	τ_2 (ns)	τ_{avg} (ns)
Control	0.26	5.88	0.74	466.60	346.81
Target	0.21	8.59	0.79	536.86	425.92

Table S4. The fitted EIS results of control and target PSCs.

	R_S (Ω)	R_{rec} (Ω)	CPE-T	CPE-P
Control	62.15	961.3	5.3×10^{-9}	0.971
Target	35.66	1877	6.2×10^{-9}	0.973

Supplementary text: In electrochemical impedance spectroscopy (EIS) analysis, the constant-phase element (CPE) is used to describe non-ideal capacitive behavior. CPE-T (the admittance parameter of the constant-phase element) reflects the admittance magnitude of the CPE element, which is related to the conductivity of the electrode-electrolyte interface and indicates the ease or difficulty of charge transfer at the interface. CPE-P (the constant-phase element exponent, n) characterizes the non-ideal capacitive properties of the CPE. Its value range is typically between 0 and 1. When n=1, the CPE exhibits ideal capacitive behavior; when n deviates from 1, it means that factors such as roughness and inhomogeneity exist on the electrode surface, causing the capacitive behavior to deviate from the ideal state. The unit of CPE-T is $\Omega^{-1}\cdot s^n$, where n is the value of CPE-P; CPE-P is a dimensionless quantity and has no unit.

In the EIS fitting, the four parameters R_s, R_{rec}, CPE-T, and CPE-P are used to accurately describe the electrical characteristics of perovskite solar cells (PSCs). R_s represents the series resistance of the PSCs, reflecting the ease or difficulty of carrier transport; R_{rec} represents the recombination resistance of PSCs, reflecting the resistance to carrier recombination; In PSCs, CPE as a nonideal capacitor is commonly defined by two parameters: CPE-T and CPE-P. CPE-T is associated with the interfacial capacitance, while CPE-P is related to the characteristics of an ideal capacitor. [1, 2] In the equivalent circuit, CPE_{rec} is used to characterize the non-ideal capacitive characteristics presented in this recombination process. CPE_{rec} is determined from the CPE-T and CPE-P values by using the equation:[3] $CPE_{rec} = \frac{(CPE-T \times R_{rec})^{\frac{1}{CPE-P}}}{R_{rec}}$.

Reference:

1. Q. Wu, C. Xue, Y. Li, P. Zhou, W. Liu, J. Zhu, S. Dai, C. Zhu and S. Yang, *ACS Appl. Mater. Interfaces*, 2015, **7**, 28466-28473.
2. J. Han, D. Luo, W. Huang, F. Wang, C. Jia, X. Li and Y. Chen, *Dalton Trans.*, 2024, **53**, 8356-8368.
3. A. Shit and A. K. Nandi, *Phys. Chem. Chem. Phys.*, 2016, **18**, 10182-10190.