Supporting Information

Multifunctional Additive for Enhancing the Performance of Single-Junction Perovskite and Perovskite/Silicon Tandem Solar Cells

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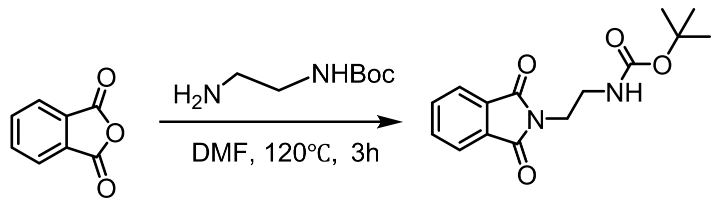
1. The synthetic method of BD-Boc

Benzene dicarboxylic anhydride (1.43 mmol), N-Boc-ethylenediamine (1.58 mmol) and 38 ml DMF were added into a 250 ml round-bottom flask. The mixture is stirred at 120 °C for 3 hours. Water was used to quench the reaction and ethyl acetate was used to extract the organic phase. By rotary evaporation, the solvent was removed, and the crude product was further purified using column chromatography (DCM and ethyl acetate). 342 mg light yellow powder (BD-Boc) is obtained after purification. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.85 (dd, J = 5.4, 3.1 Hz, 2H), 7.72 (dd, J = 5.4, 3.0 Hz, 2H), 4.89 (s, 1H), 3.94 – 3.73 (m, 2H), 3.53 – 3.34 (m, 2H), 1.34 (s, 9H).



BDA **BD-Boc**

Fig. S1. Synthesis mechanism of BD-Boc.

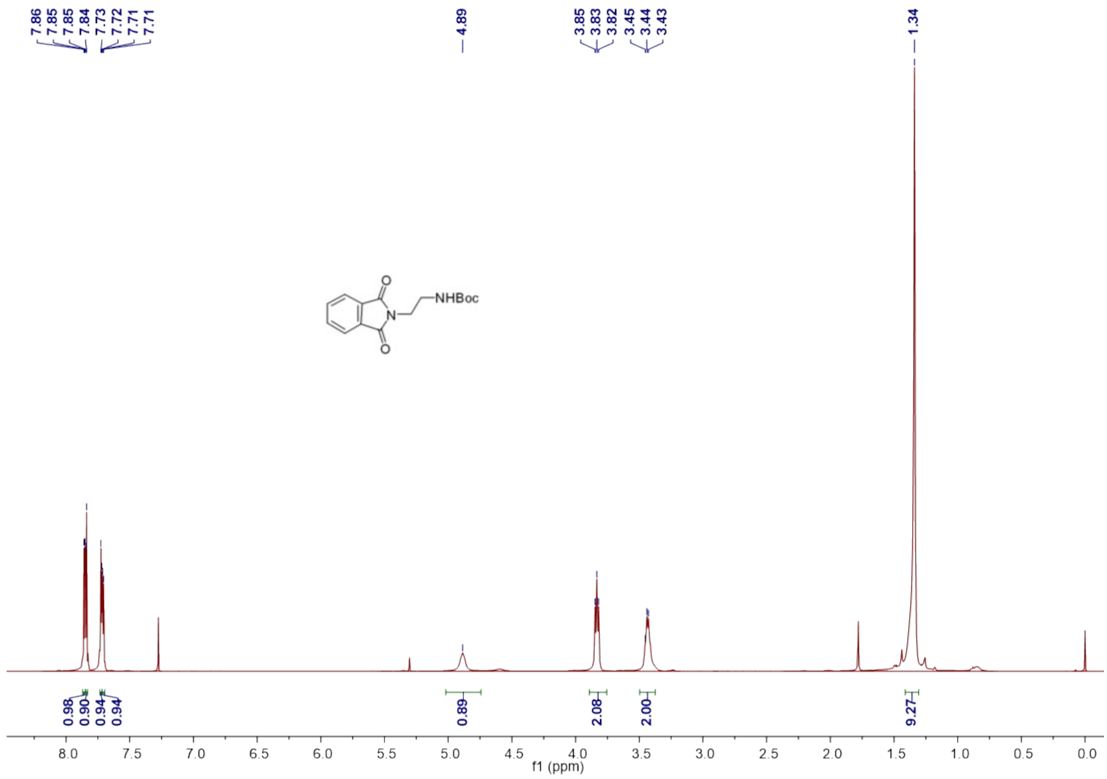


Fig. S2. NMR spectrum of BD-Boc.

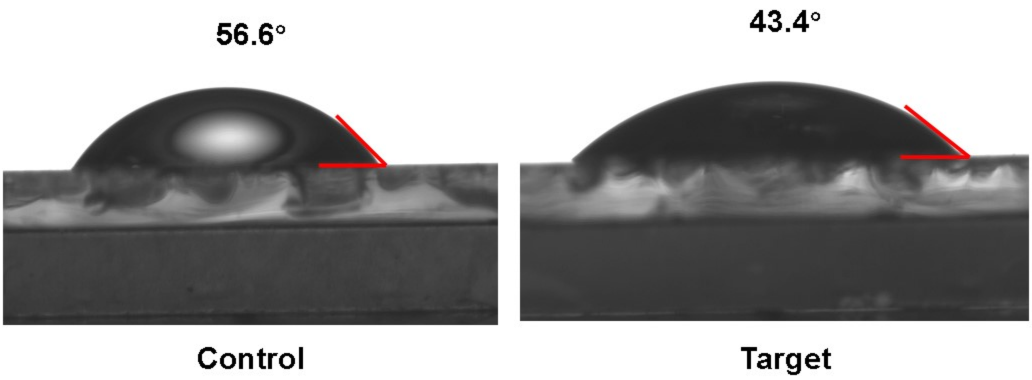


Fig. S3. Water droplet contact angles on surfaces of control and target perovskite films.

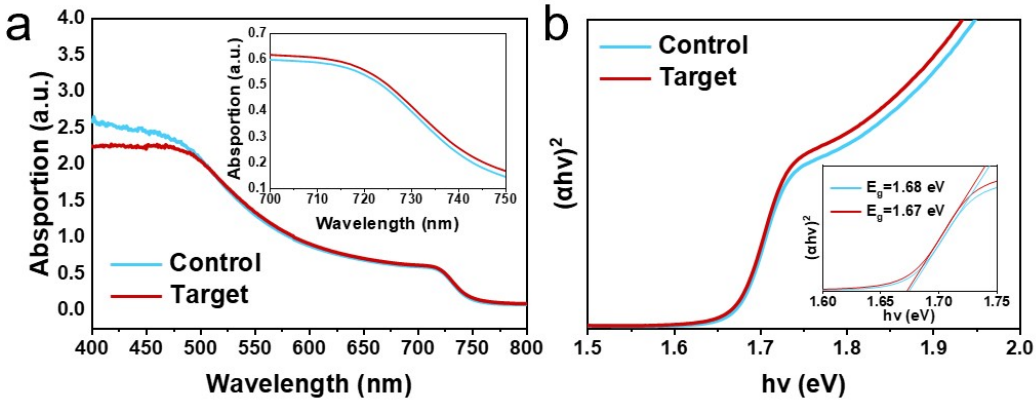


Fig. S4. (a) UV-vis absorption spectra of the control and target perovskite films. (b) Bandgap calculation plots for the control and target perovskite films.

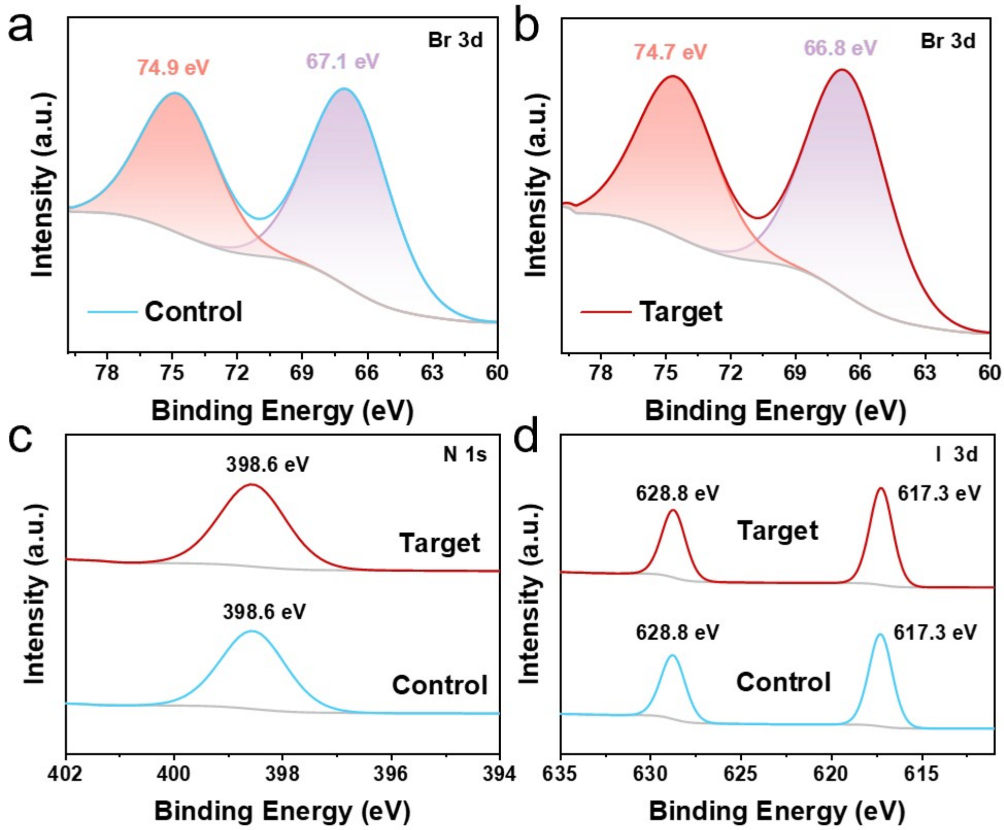


Fig. S5. XPS patterns: Br 3d core-level spectra of the (a) control and (b) target perovskite film. XPS patterns: (c) N 1s; (d) I 3d core-level spectra of the control and target perovskite films.

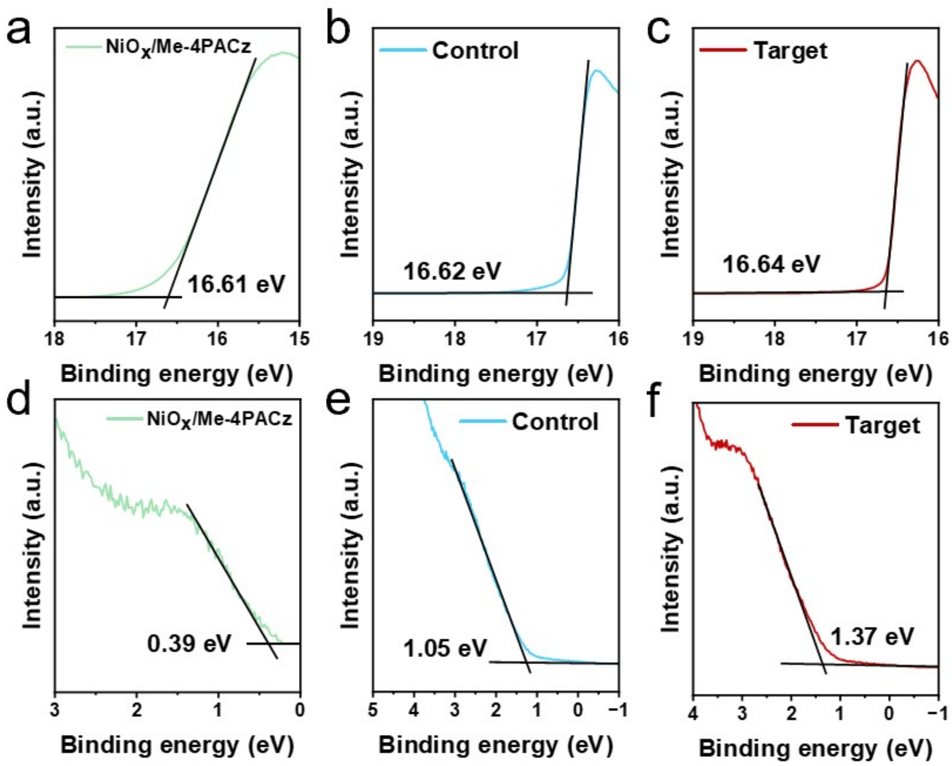


Fig. S6. UPS spectra of NiOx/Me-4PACz in the (a) onset and (d) cutoff energy region. UPS spectra of the (b) control and (c) target perovskite film in the onset energy region. UPS spectra of the (e) control and (f) target perovskite film in the cutoff energy region.

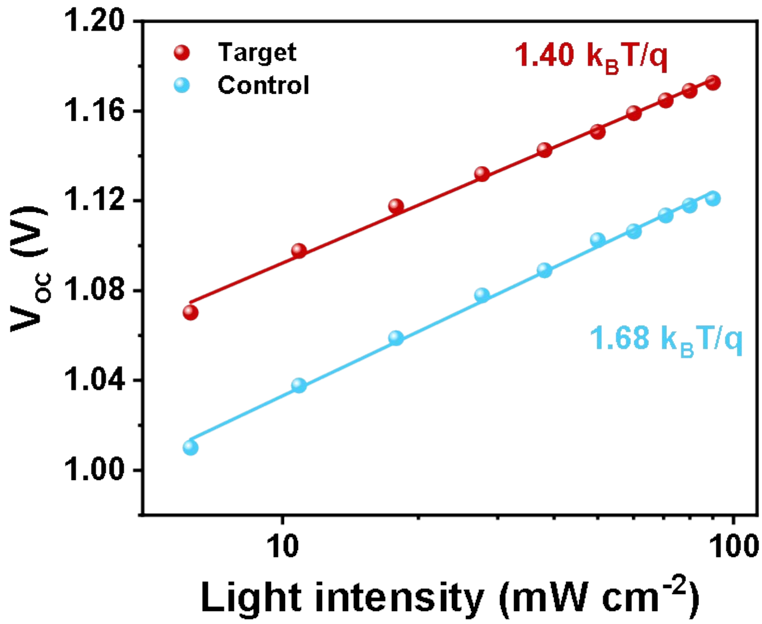


Fig. S7. V_{OC} versus light intensity for the control and target devices.

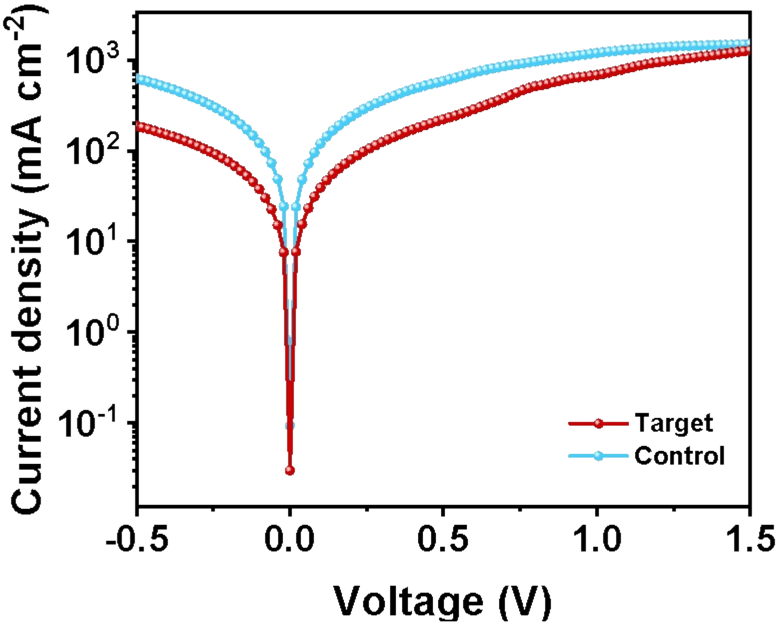


Fig. S8. Dark J−V curves of the control and target devices.

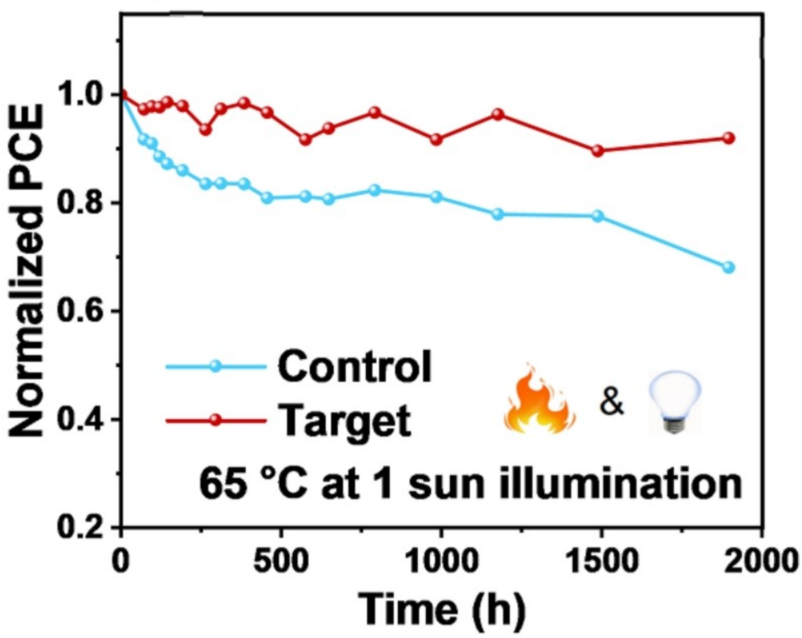


Fig. S9. PCE evolution of the unencapsulated PSCs stored in the glove box at 65 °C and 1 sun illumination.

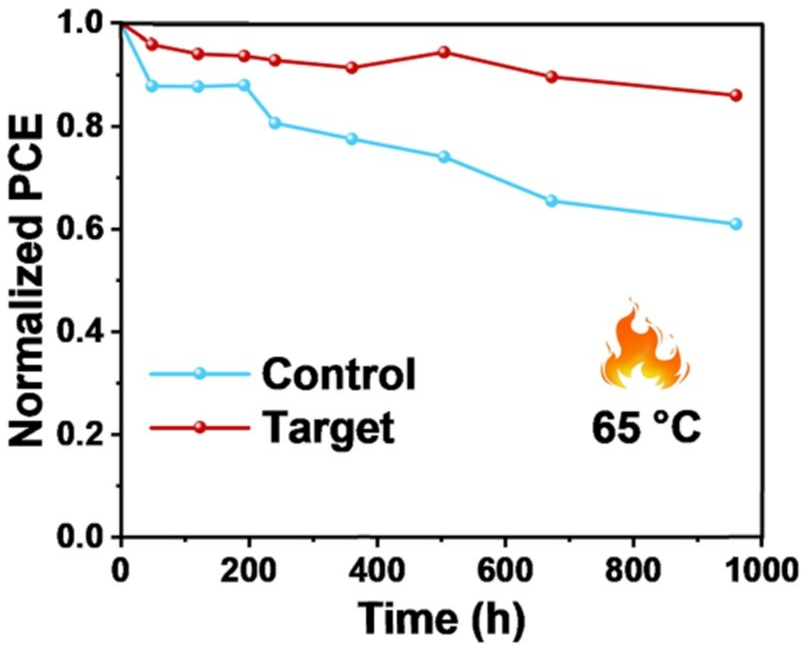


Fig. S10. PCE evolution of the unencapsulated PSCs stored in the glove box at 65 °C.

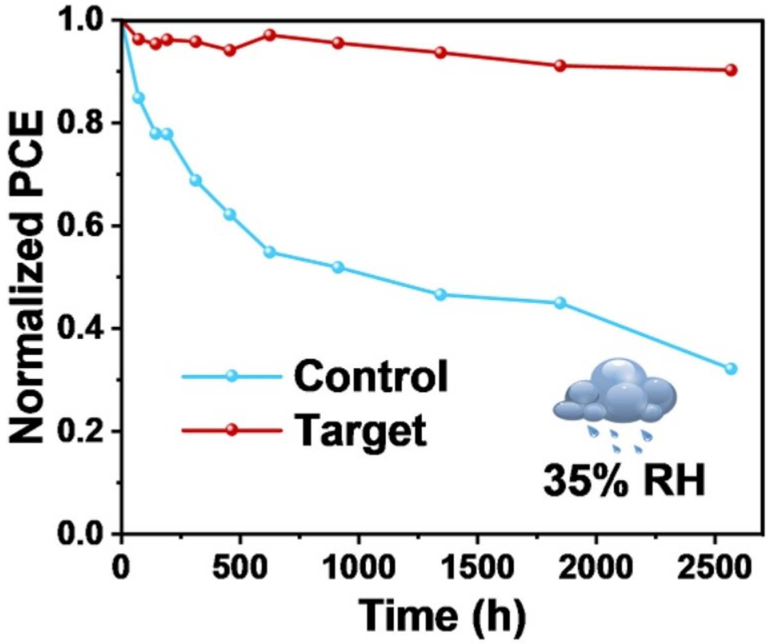


Fig. S11. PCE evolution of the unencapsulated PSCs stored in air at room temperature (RT) with a relative humidity (RH) of 35± 5%.

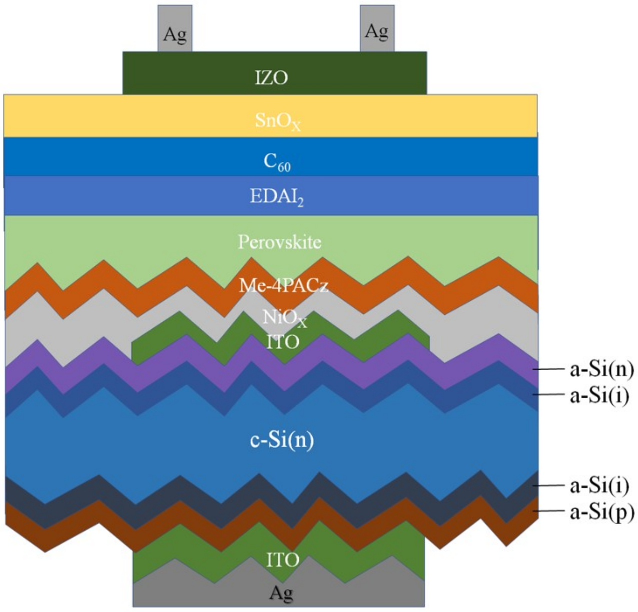


Fig. S12. Device schematics of textured, 2-terminal perovskite/silicon TSCs studied in this work.

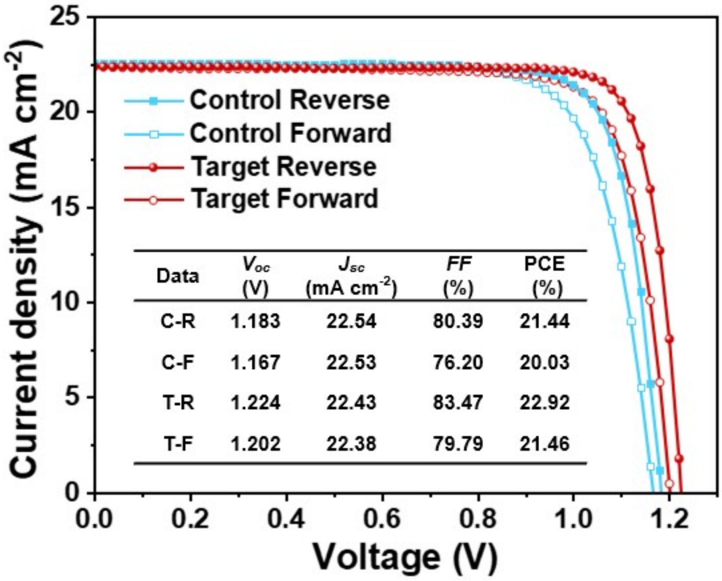


Fig. S13. J–V curves for control and target PSCs.

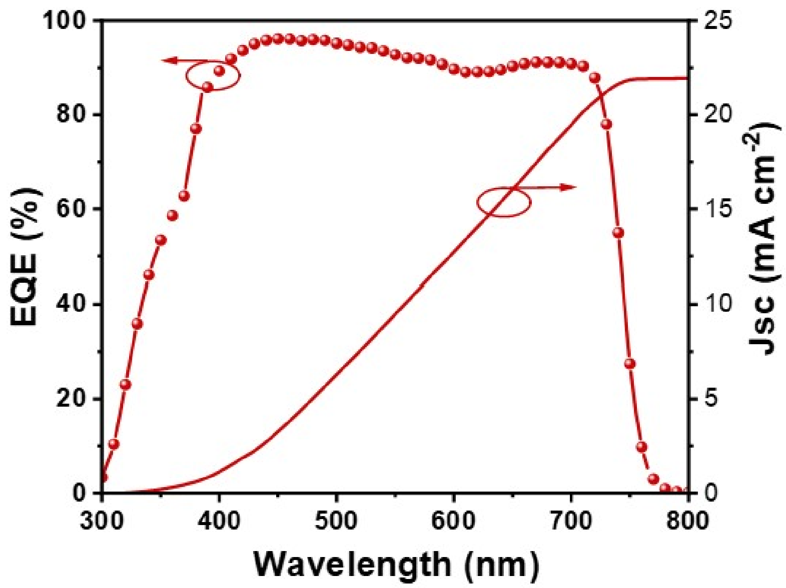


Fig. S14. The EQE spectra with the integrated J_{SC} of the target PSC.

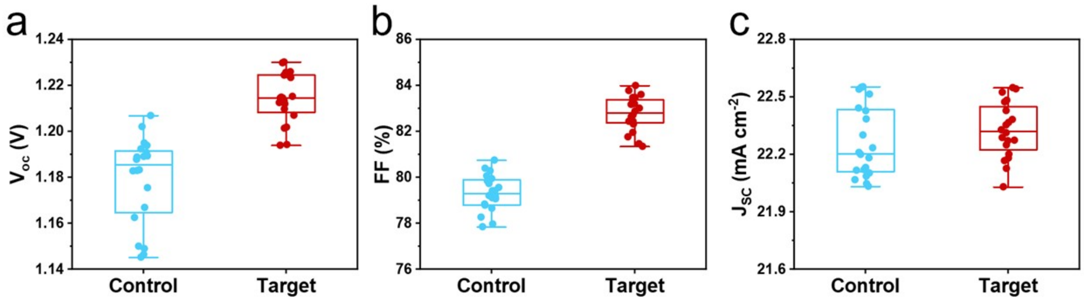


Fig. S15. Photovoltaic parameters including (a) V_{oc}, (b) FF, and (c) J_{SC} of devices with and without additive BD-Boc.

Table S1. Parameters for TRPL of the control and target wide-bandgap perovskite films.

Samples	A_1	T_1(ns)	A_2	τ_2 (ns)	τ_{ave}(ns)
Control	0.652	82.2	0.338	173.3	129.8
Target	0.544	158.9	0.455	369.4	297.9

The average carrier lifetime (τ_{ave}) was calculated according to the equation:

$$\tau_{ave} = \frac{A_1\tau_1^2 + A_2\tau_2^2}{A_1\tau_1 + A_2\tau_2}$$

Where A_1 and A_2 are constants, τ_1 is the fast transient component and τ_2 is the slow component.

Table S2. Photovoltaic performance parameters of the best control and target wide-bandgap perovskite solar cells under different scan directions.

Samples	Scan direction	V_{OC} (V)	J_{SC} (mA cm^{-2})	*FF* (%)	PCE (%)
Control	Reverse	1.183	22.54	80.39	21.44
	Forward	1.167	22.53	76.20	20.03
Target	Reverse	1.224	22.43	83.47	22.92
	Forward	1.202	22.38	79.79	21.46

Table S3. Photovoltaic performance parameters of the wide-bandgap perovskite solar cells prepared by adding different concentrations of BD-Boc to the perovskite precursor solution.

Samples	Scan direction	V_{OC} (V)	J_{SC} (mA cm^{-2})	*FF* (%)	PCE (%)
0.1mg	Reverse	1.222	21.75	84.24	22.49
0.3mg	Reverse	1.224	22.43	83.47	22.92
0.5mg	Reverse	1.214	21.61	82.66	22.18