Supplementary information：

Advancing 1.84 eV Wide-Bandgap Perovskite Solar Cells *via* Multidentate Molecular Engineering

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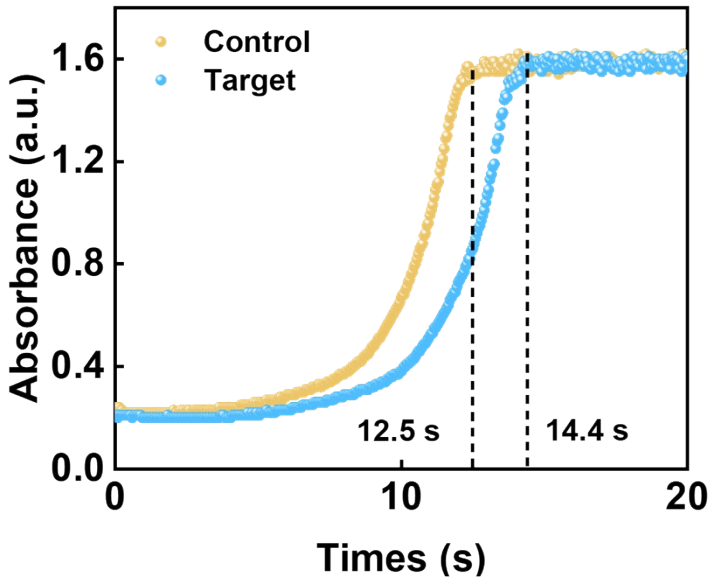


Fig. S1 Absorption spectral evolution curves of perovskite films during the annealing process.

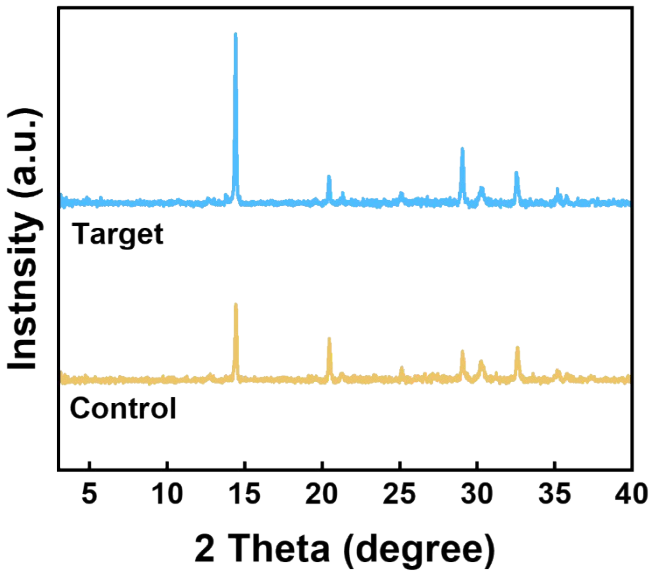


Fig. S2 XRD pattern of perovskite film deposited on ITO substrate.

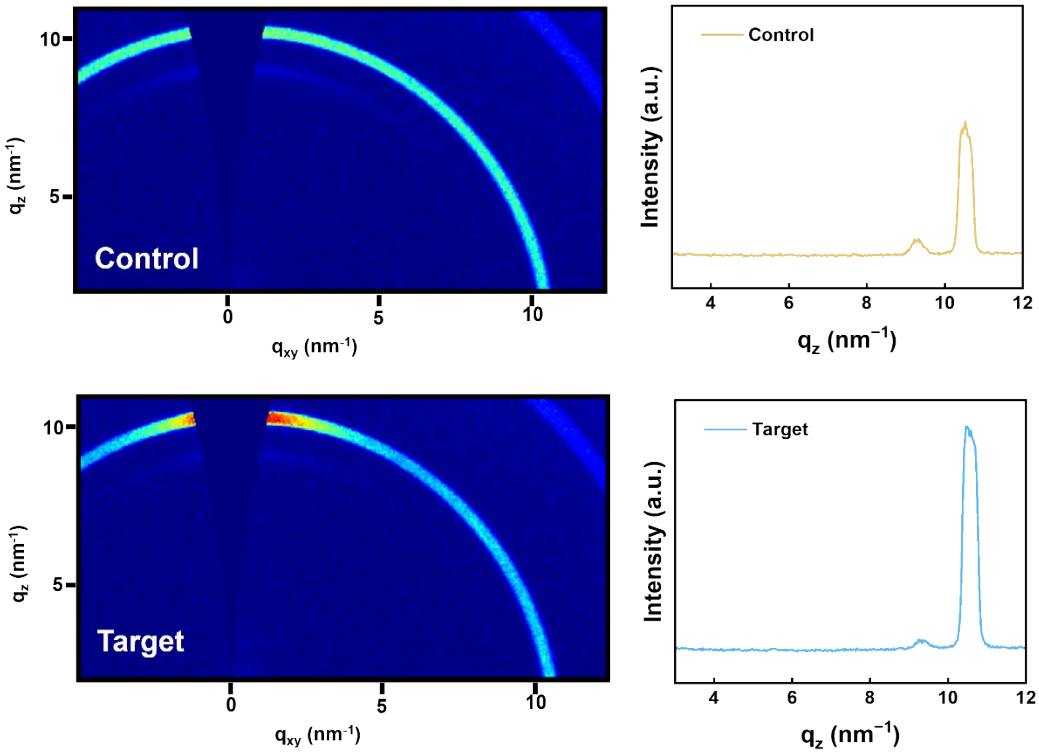


Fig. S3 2D GIWAXS diffraction patterns and their integrated intensity line profiles for perovskite thin films on glass substrates.

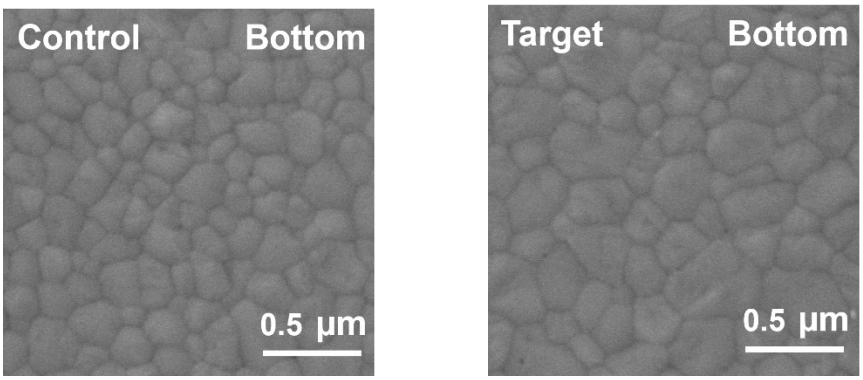


Fig. S4 Bottom SEM morphologies of control and H3TATB-doped samples.

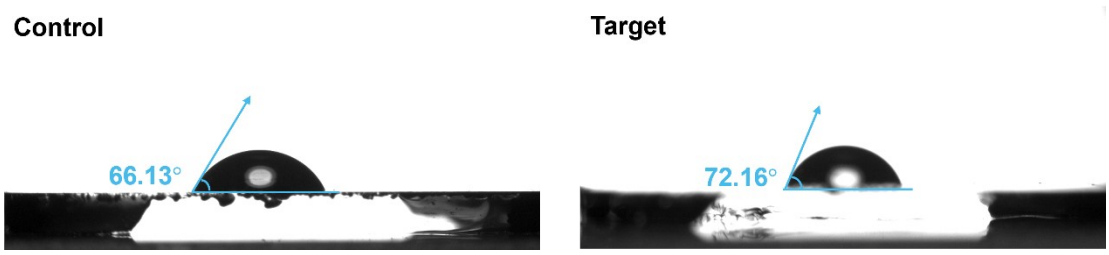


Fig. S5 Contact angle images of control and H3TATB-modified perovskite films.

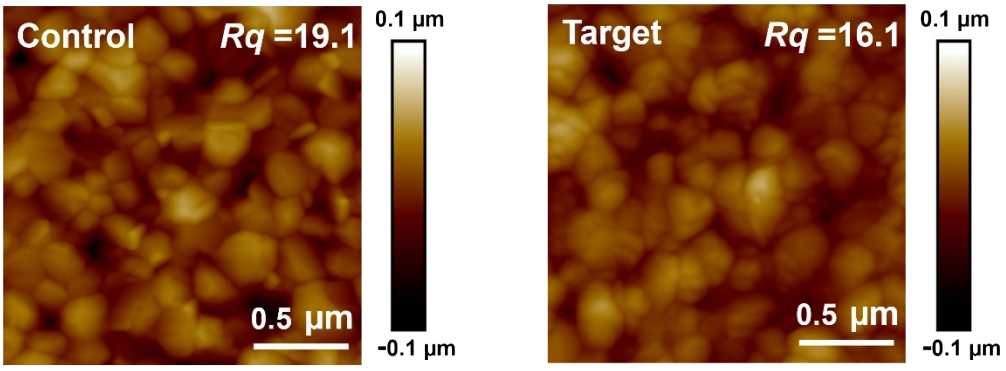


Fig. S6 AFM morphologies of the perovskite surfaces in control and H3TATB-modified samples.

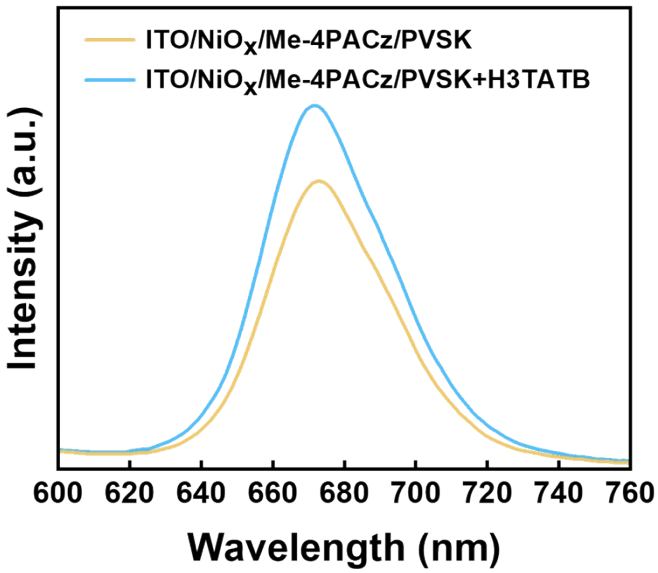


Fig. S7 PL spectra of perovskite thin films spin-coated onto ITO/NiOx/Me-4PACz.

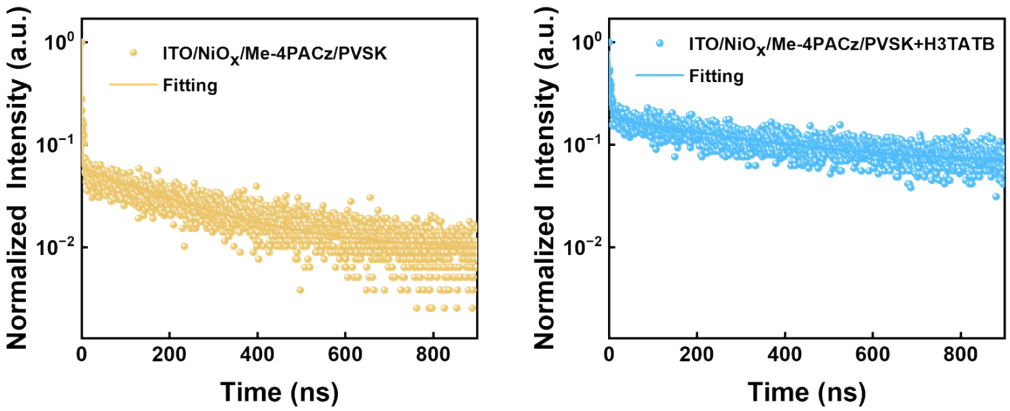
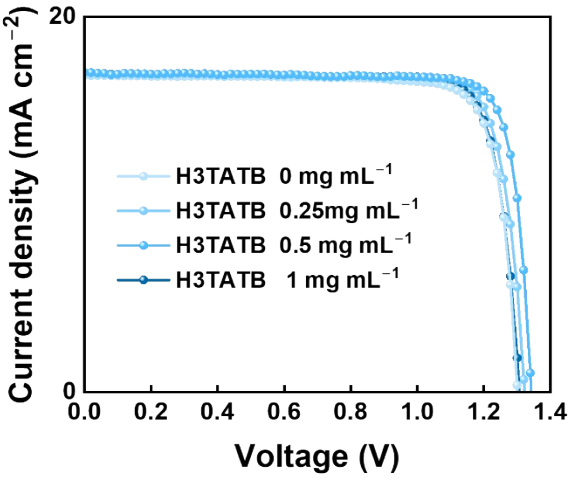


Fig. S8 Time-resolved photoluminescence (TRPL) decay curves of perovskite films spin-coated on ITO/NiO_x/Me-4PACz substrates. The fitted carrier lifetimes are tabulated.



Concentration	V_{OC} (V)	J_{SC} (mA cm^{-2})	*FF* (%)	*PCE* (%)
H3TATB 0 mg	1.30	16.98	82.31	18.17
H3TATB 0.25 mg	1.32	17.06	83.35	18.77
H3TATB 0.5 mg	1.34	17.07	84.19	19.26
H3TATB 1 mg	1.30	17.02	82.84	18.33

Fig. S9 J-V curves and parameters of PSCs with H3TATB modification at different concentration

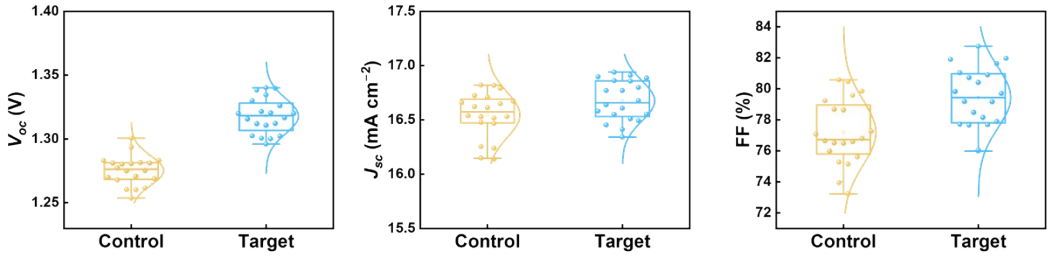


Fig. S10 Distribution statistics of J_{SC}, V_{OC}, and FF with and without H3TATB modification.

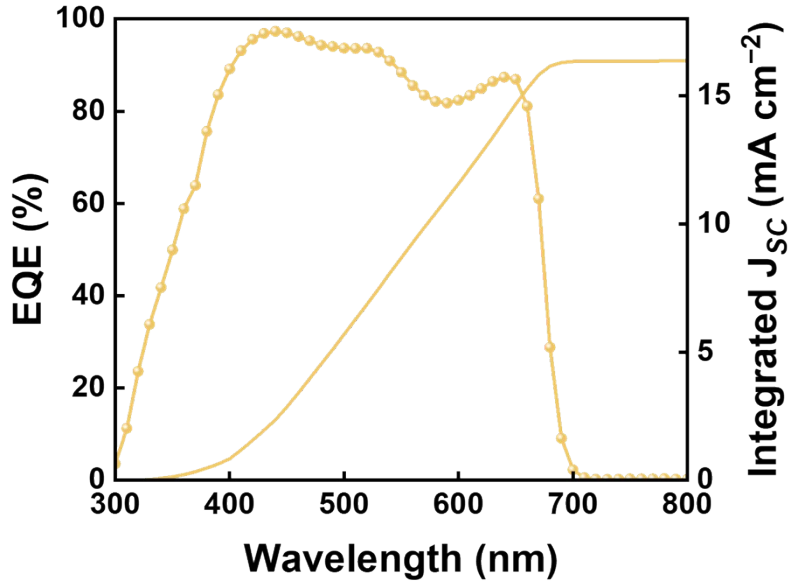


Fig. S11 The external quantum efficiency (EQE) spectra of the control group

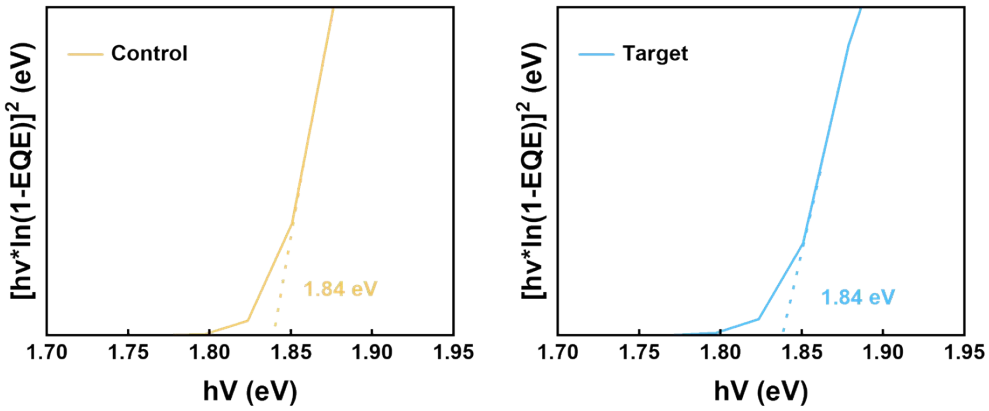


Fig. S12 EQE integral plots and band gap calculation plots.

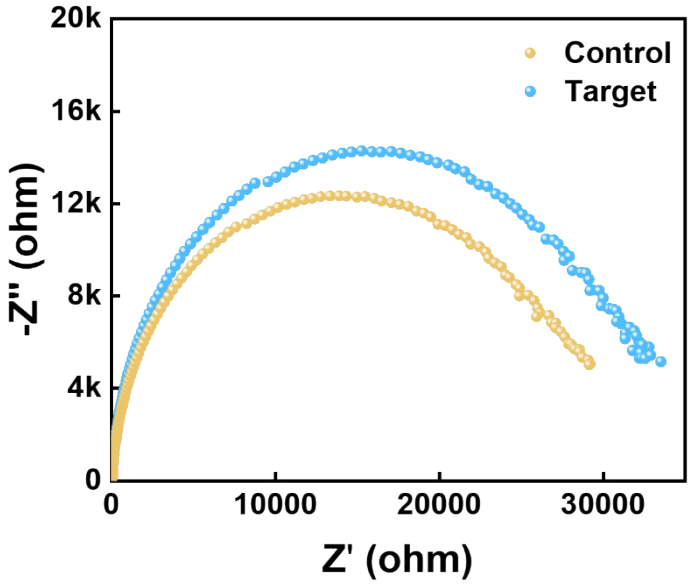


Fig. S13 Electrochemical impedance spectroscopy (EIS) measurement under 1 V bias voltage.

Table S1 the carrier lifetimes of perovskite films deposited on ITO.

Sample	A1 (%)	τ_1 (ns)	A2 (%)	τ_2 (ns)	τ_{ave} (ns)
Control	73.8	3.1	26.2	164.1	45.3
H3TATB	53.7	3.2	46.3	530.8	247.5

Table S2 the carrier lifetimes of perovskite films deposited on ITO/NiOx/Me-4PACz

Sample	A1 (%)	τ_1 (ns)	A2 (%)	τ_2 (ns)	τ_{ave} (ns)
Control	93.2	43.3	5.1	220.2	12.0
H3TATB	62.2	2.1	12.4	255.5	60.9