Supporting Information

Regulated Crystallization with Minimized Degradation for Pure-Red Lead-Free Perovskite Light-Emitting Diodes

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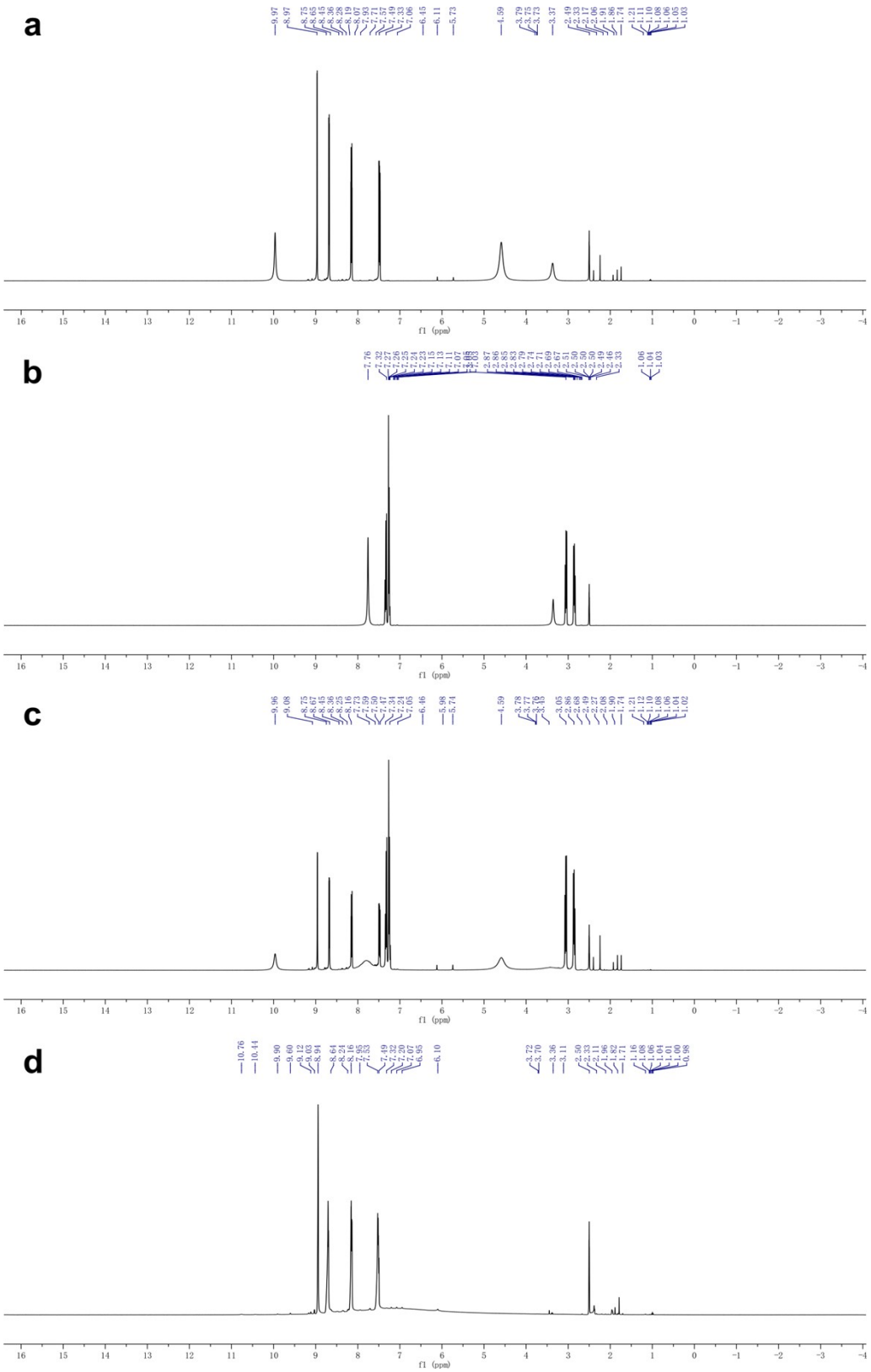

Figure S1. ^{1}H NMR spectra of (a) NHD, (b) PEAI, (c) PEAI/NHD mixture, and (d) SnI_2/NHD mixture.

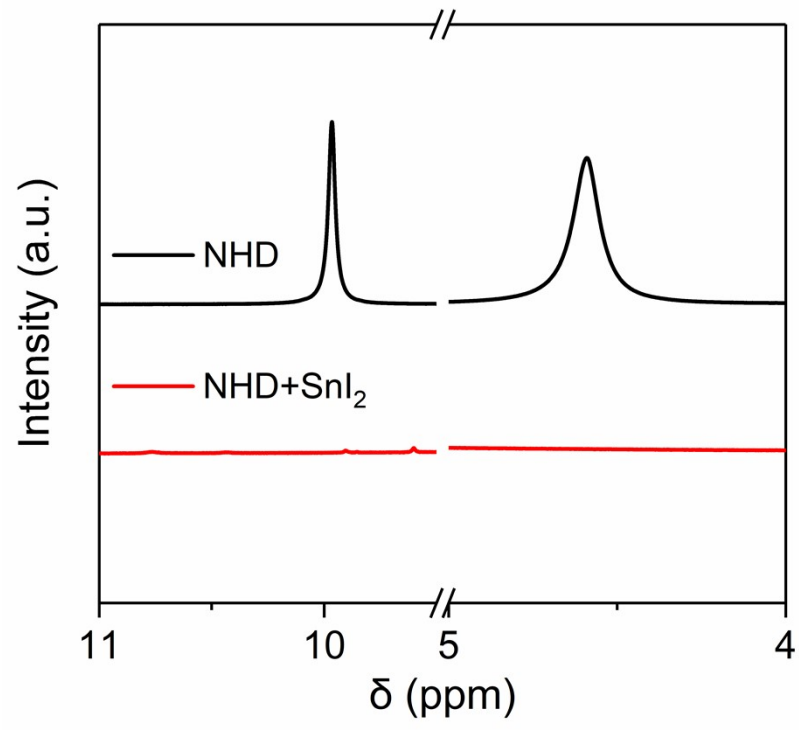

Figure S2. High-resolution ^{1}H NMR spectra of NHD and NHD/SnI_2 mixture.

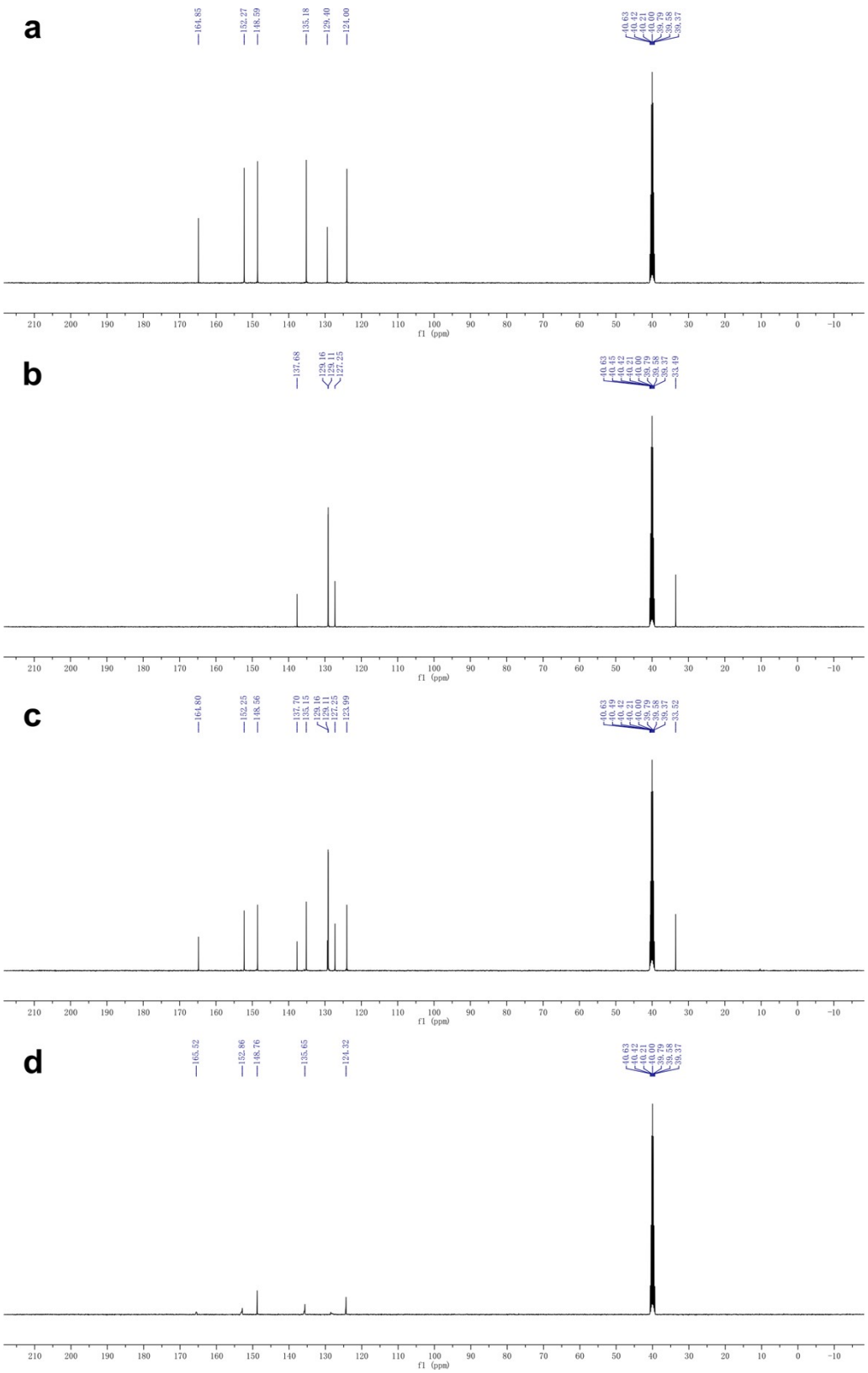

Figure S3. ^{13}C NMR spectra of (a) NHD, (b) PEAI, (c) PEAI/NHD mixture, and (d) SnI_2/NHD mixture.

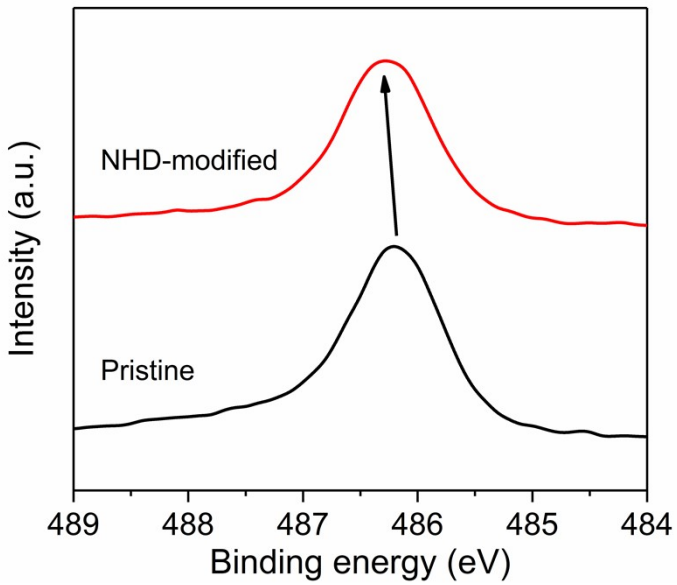

Figure S4. XPS spectra from the Sn *3d* core levels in PEA_2SnI_4 perovskite films with and without NHD.

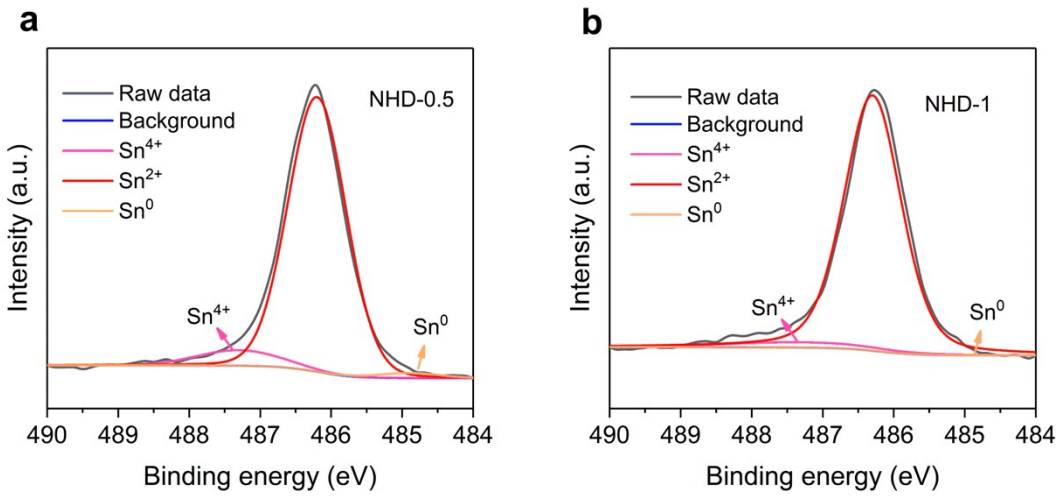

Figure S5. Fitting analysis of the XPS spectra from the Sn *3d* core levels PEA_2SnI_4 perovskite films with NHD at concentrations of (a) 0.5 and (b) 1 mg mL^{-1}.

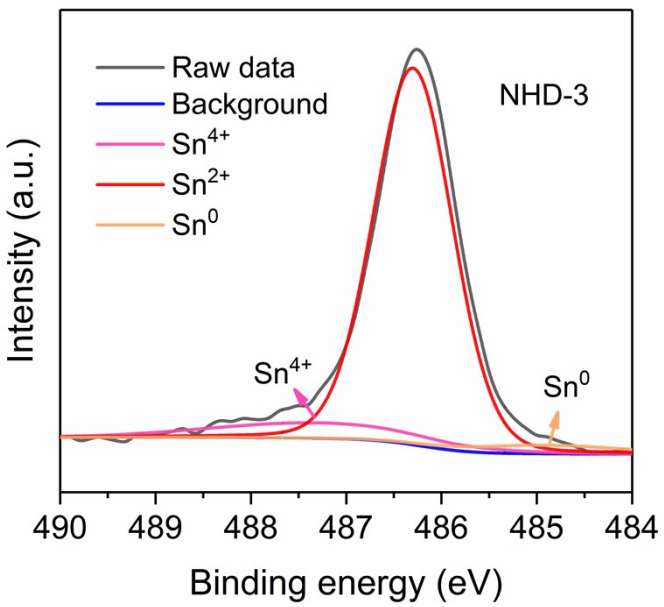

Figure S6. Fitting analysis of the XPS spectrum from the Sn *3d* core level PEA_2SnI_4 perovskite films with NHD at a concentration of 3 mg mL^{-1}.

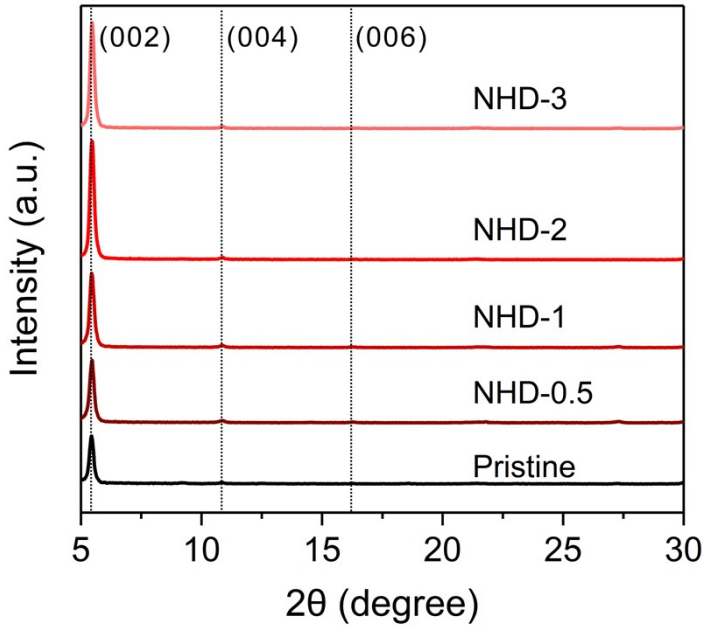

Figure S7. XRD patterns of perovskite films modified with different concentrations of NHD.

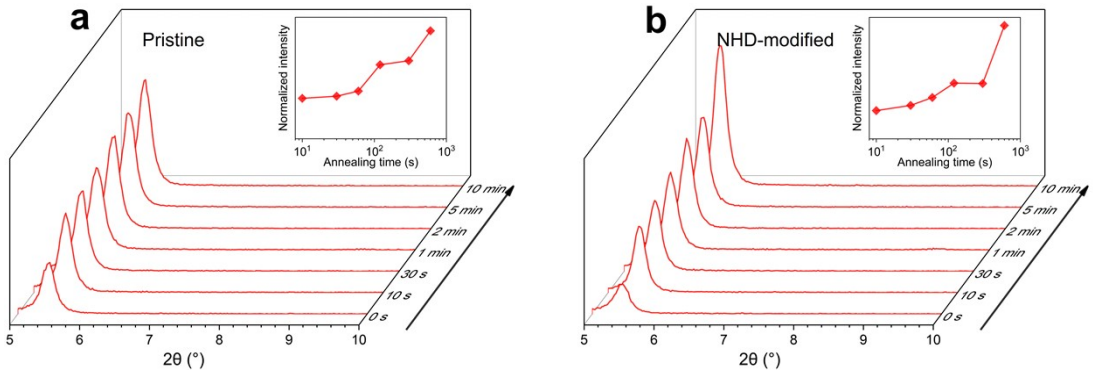

Figure S8. Crystallization kinetics of perovskite films. In situ XRD patterns with increasing annealing time of perovskite films (a) without and (b) with (2 mg mL^{-1}) NHD modification. The inserts are the normalized time-revolved diffraction intensities of the lattice plane (002) as obtained in (a) and (b).

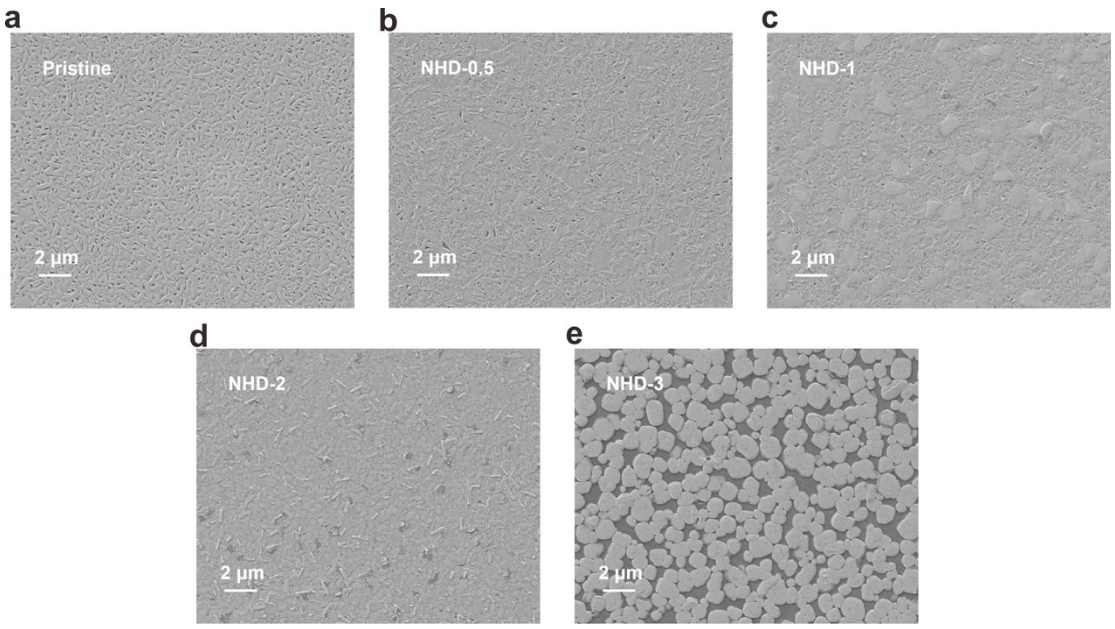

Figure S9. Top-view SEM images of PEA_2SnI_4 films (a) without and with NHD at concentrations of (b) 0.5, (c) 1, (d) 2, and (e) 3 mg mL^{-1}.

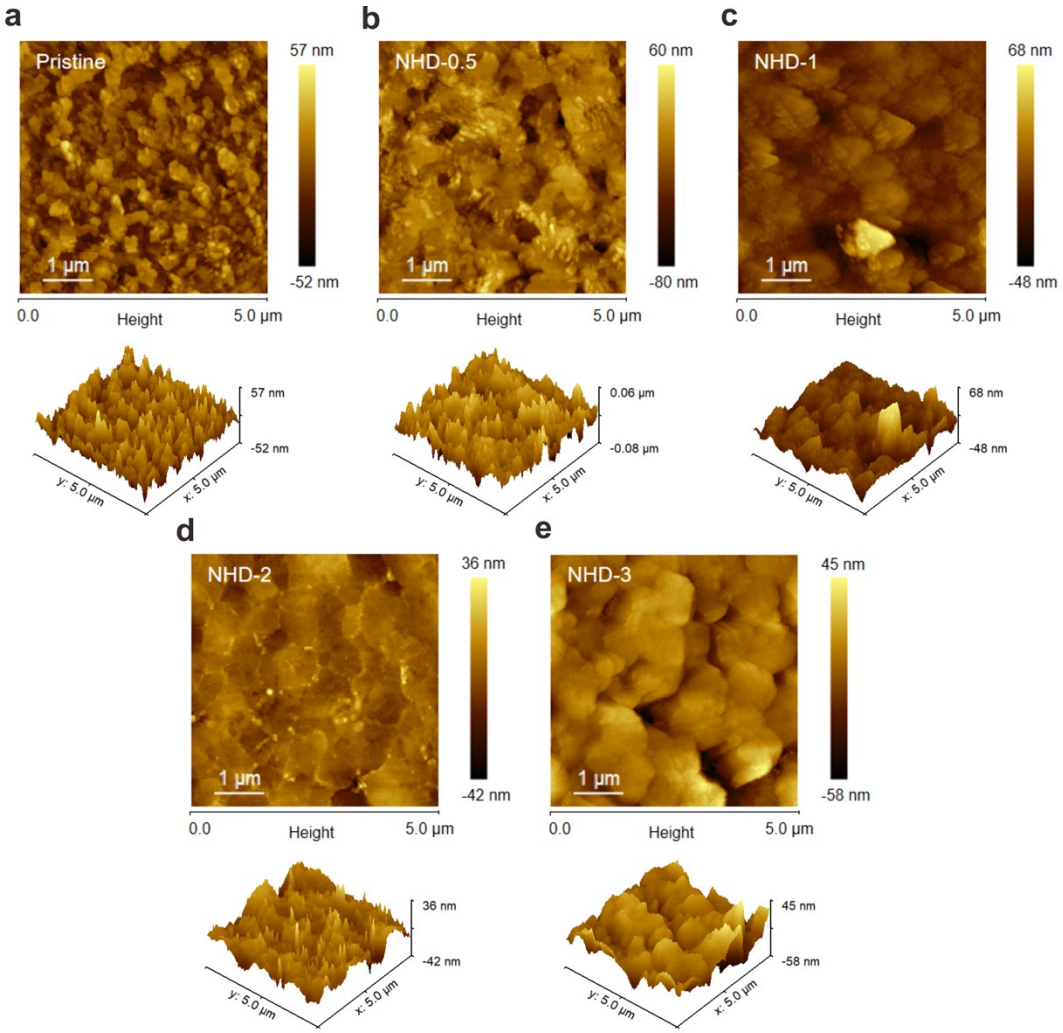

Figure S10. Height-mode AFM images of PEA_2SnI_4 films (a) without and with NHD at concentrations of (b) 0.5, (c) 1, (d) 2, and (e) 3 mg mL^{-1}, the RMS values of which are 10.7, 12.6, 8.0, 4.86, and 9.83 nm, respectively.

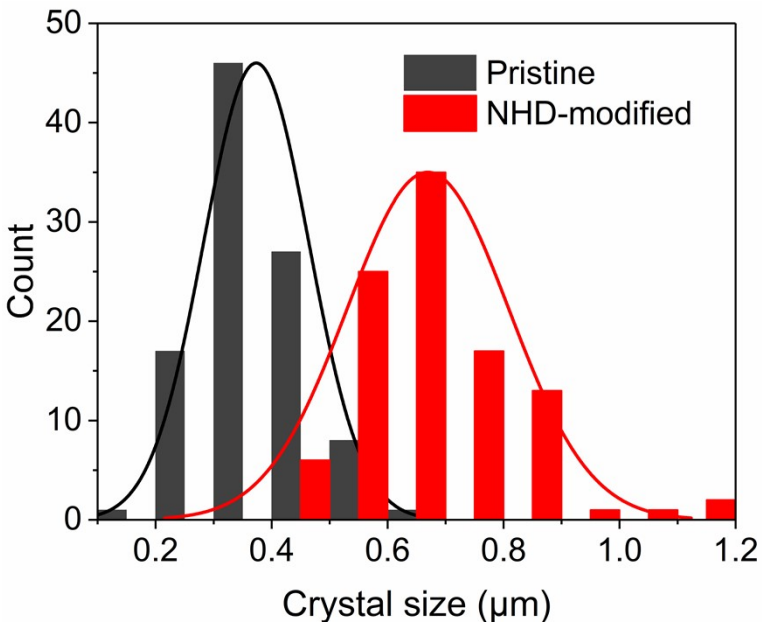

Figure S11. Histogram of grain size distribution with and without NHD modification.

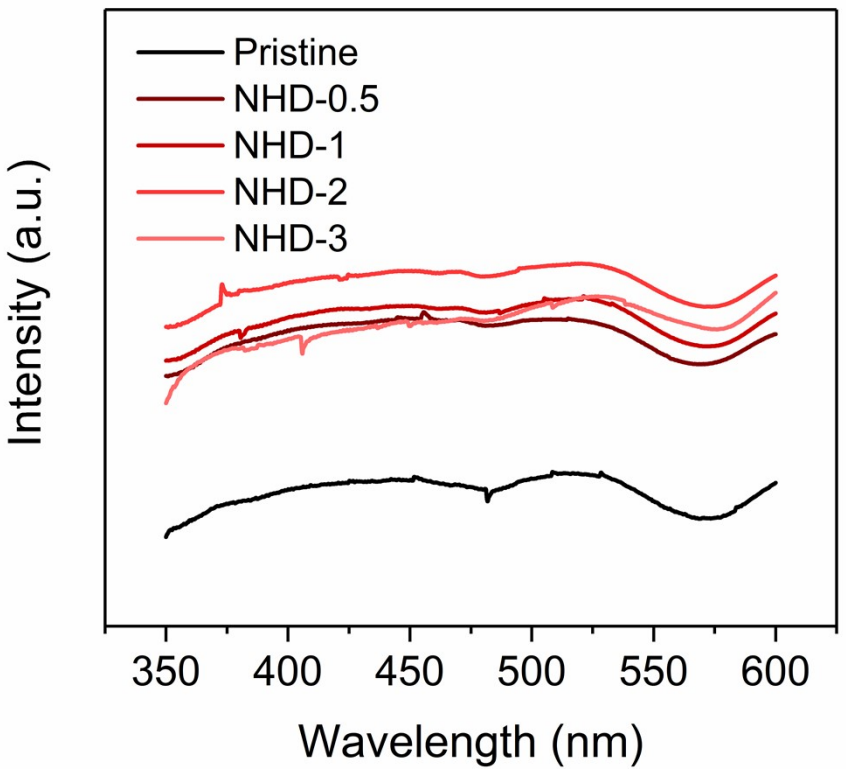

Figure S12. PLE characteristics of perovskite films with different doping concentration of NHD.

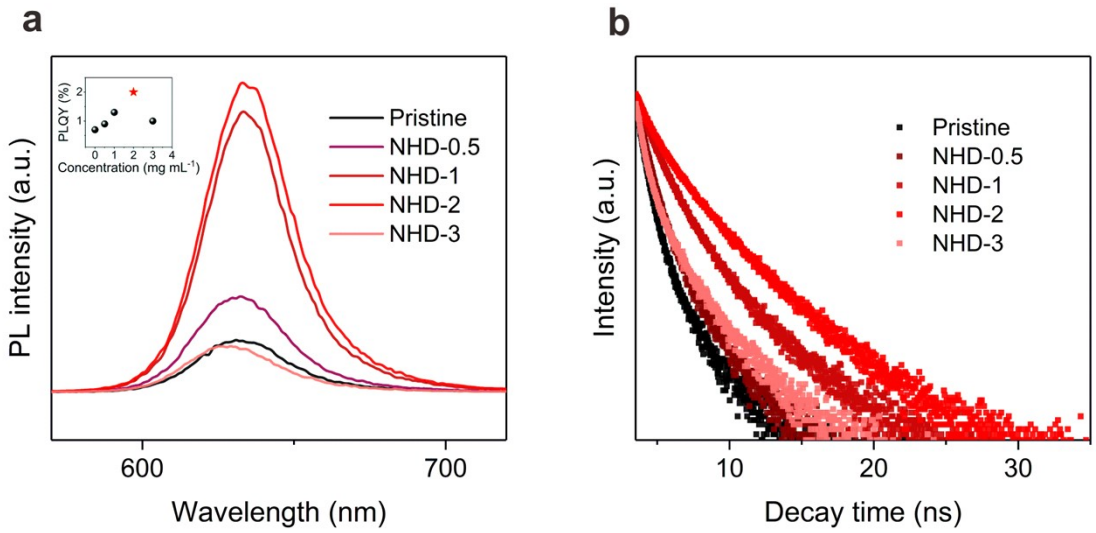

Figure S13. PL characteristics of perovskite films. (a) PL and (b) TRPL spectra of PEA_2SnI_4 perovskite films with different doping concentrations of NHD. The inset is PLQY of films without and with different concentrations of NHD.

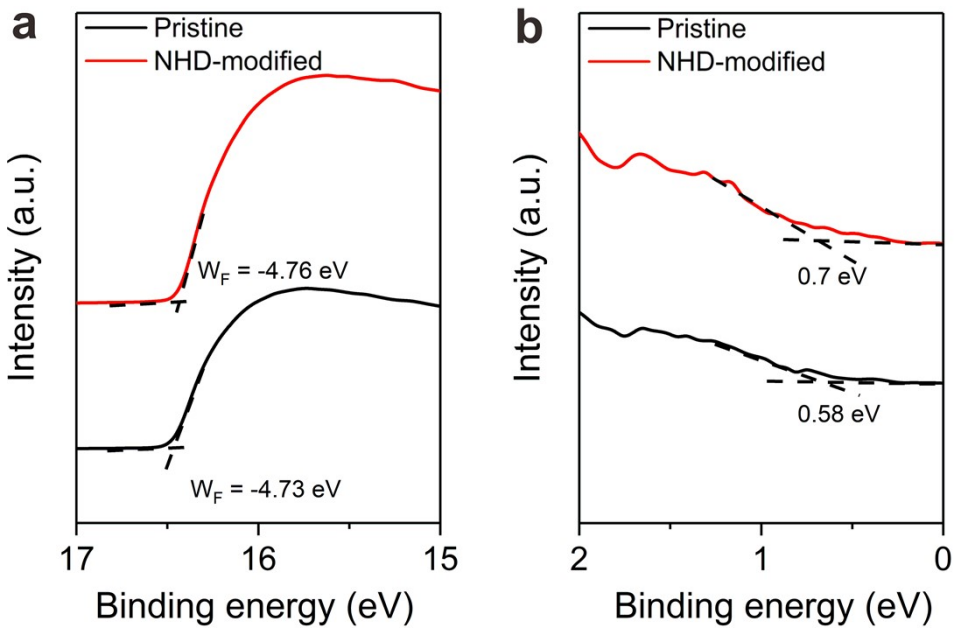

Figure S14. Energy-level measurement of PEA_2SnI_4 films. (a) Secondary-electron cutoff and (b) onset regions of the UPS spectra of pristine and NHD-modified perovskite films.

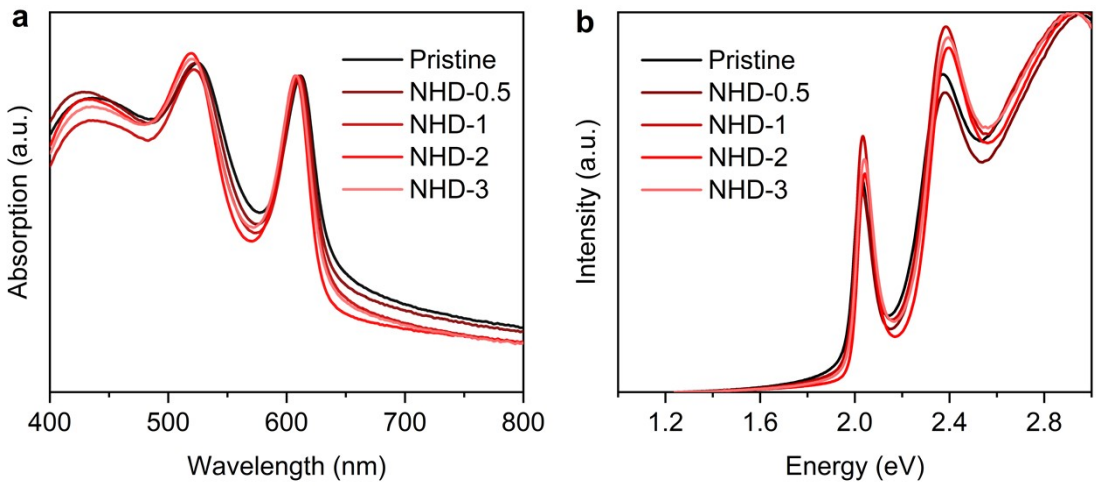

Figure S15. Absorption spectra and bandgap measurement of perovskite films. (a) Absorption spectra of PEA_2SnI_4 films with different doping concentrations of NHD. (b) Tauc plot analysis of the absorption measurement.

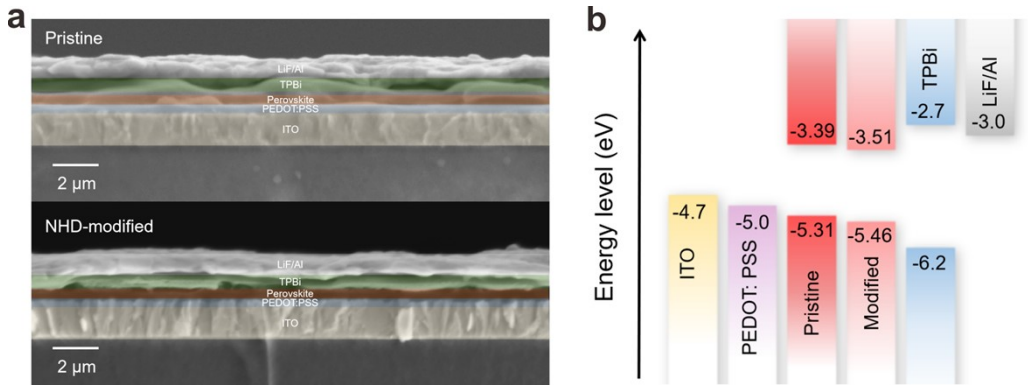

Figure S16. (a) Cross-section SEM image of devices with and without NHD modified.

(b) Schematic energy-level alignment of pristine and NHD-modified devices.

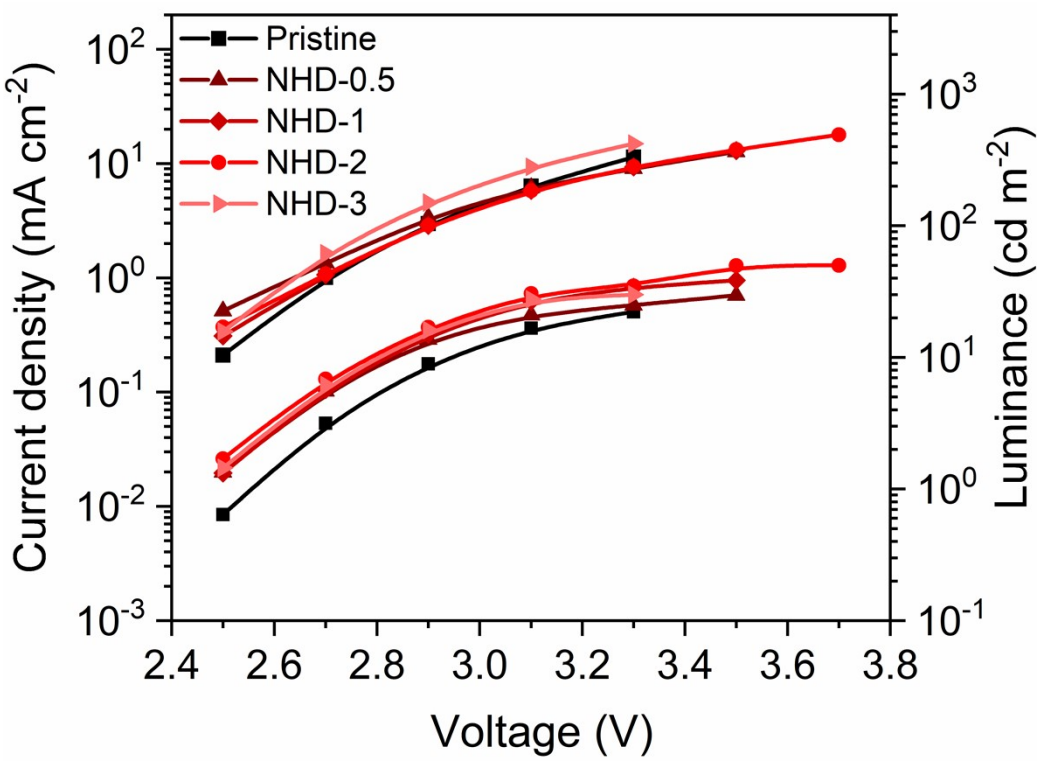

Figure S17. *J-V-L* curves of devices with different doping concentration of NHD.

Table S1. The fitted TRPL lifetimes of perovskite films.

perovskites	τ_{ave} (ns)	τ_1 (ns)	τ_2 (ns)
pristine	0.74	0.14	0.68
NHD-0.5	0.91	0.55	1.25
NHD-1	2.32	0.83	2.71
NHD-2	3.66	0.93	4.77
NHD-3	0.88	0.50	1.31

Table S2. Performance Parameters of pristine and NHD-modified devices.

Devices	Peak (nm)	CIE coordinates	FWHM (nm)	L_{max} (cd m^{-2})	EQE_{max} (%)	CE (cd A^{-1})
Pristine	628	(0.69, 0.30)	30	22	0.30	0.31
NHD-modified	628	(0.69, 0.30)	30	50	0.86	1.1