Improving the performance of pure-red 2D tin-based perovskite light-emitting diodes through N-methylthiourea ligand engineering-Supporting Information

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The section of calculation:

1. The bleach recovery kinetics were fitted by a double exponential decay equation:[1]

$$A(t) = A_1 e^{-\frac{t}{\tau_1}} + A_2 e^{-\frac{t}{\tau_2}} \tag{1}$$

Where A_1 and A_2 are the amplitudes of each component and τ_1 and τ_2 are the corresponding lifetimes. Then, the average lifetimes were calculated by the following equation:

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

The radiative (k_{rad}) and nonradiative recombination rates (k_{nonrad}) of the control and N-MTU-modified perovskite films were calculated by the following equations: [2]

$$\frac{1}{\tau_{avg}} = k_{rad} + k_{nonrad} \tag{3}$$

$$PLQY = \frac{k_{rad}}{k_{rad} + k_{nonrad}} \tag{4}$$

$$k_{\gamma ad} = \frac{PLQY}{\tau_{avg}} \tag{5}$$

$$k_{nonrad} = \frac{1}{\tau_{avg}} - k_{\gamma ad} \tag{6}$$

2. The characterization of space charge limited current (SCLC):

The charge carrier mobility was fitted by the current density-voltage (*J*-V) curves of only-electron/hole devices using SCLC measurements.

The Mott-Gurney law:[3,4]

$$J = \frac{9}{8}\mu\varepsilon_0\varepsilon_r\frac{V^2}{L^3} \tag{7}$$

Where J is the current density, μ is the charge carrier mobility, ε_0 is vacuum dielectric constant, ε_r is the relative dielectric constant ($\varepsilon_r = 3$), V is the applied voltage and L is the thickness between the cathode and anode of the perovskite film.

Poople-Frenkel law:[5]

$$\mu = \mu_0 e^{\gamma\sqrt{E}} \tag{8}$$

Where μ_0 is the zero electric filed mobility, γ is the electric filed dependence factor, and E is the electric field (E = V/L).

Combined the Mott-Gurney law and Poople-Frenkel law, we can deduce the relational expression as following:

$$\ln\left(\frac{J}{E^2}\right) = \ln\left(\frac{9\varepsilon_r\varepsilon_0\mu_0}{8\quad L}\right) + \gamma\sqrt{E} \tag{9}$$

There is a liner relationship between $\ln\left(\frac{J}{E^2}\right)$ and $\sqrt{E}$. The γ and μ_0 can be obtained by fitting the slope and intercept. Then, the field-dependent charge-carrier mobility under fixed electric field was got by substituting γ and μ_0 into Poople-Frenkel law.

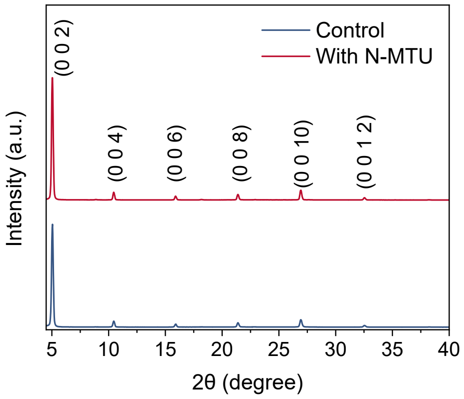

Figure S1. XRD patterns of the control and N-MTU-modified PEA_2SnI_4 film.

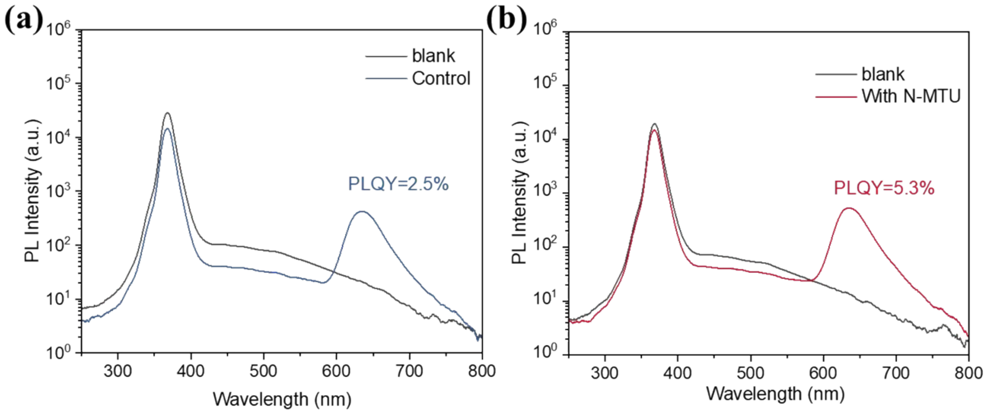

Figure S2. PL Spectra for the calculation of PLQYs of PEA_2SnI_4 films without (a) and with N-MTU additive (b).

All sample bases used to calculate PLQY were ITO/PEDOT:PSS, and the subtraction of the bases was completed before the test.

Table S1. The calculated PL lifetimes data of perovskite films without and with N-MTU additive.

Calculated data	Control	With N-MTU
A_1	4535.68	4360.08
A_2	165.92	775.09

τ_1 (ns)	0.49	0.84
τ_2 (ns)	3.02	2.97
τ_{avg} (ns)	1.95	2.30
k_{rad} (μs^{-1})	12.82	23.04
k_{nonrad} (μs^{-1})	50.00	41.17

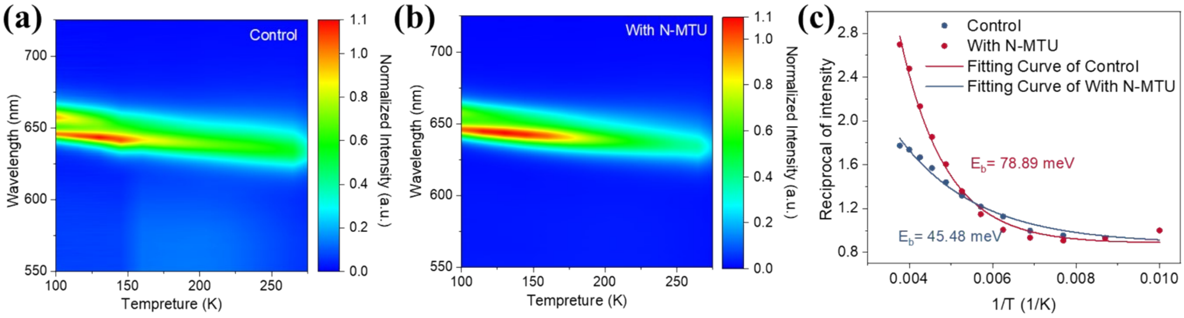

Figure S3. Pseudocolor maps of low-temperature-dependent PL spectra (a, b) and exciton binding energy fitting curves (c) of the control and N-MTU modified PEA_2SnI_4 films.

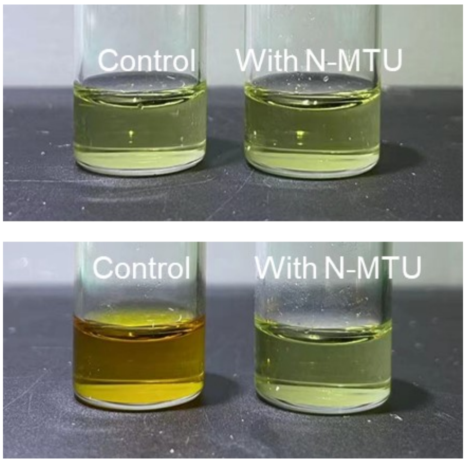

Figure S4. Photographs of the fresh PEA_2SnI_4 precursor solution (top) and aged PEA_2SnI_4 precursor solution in the air for 24h (bottom) without and with N-MTU ligands.

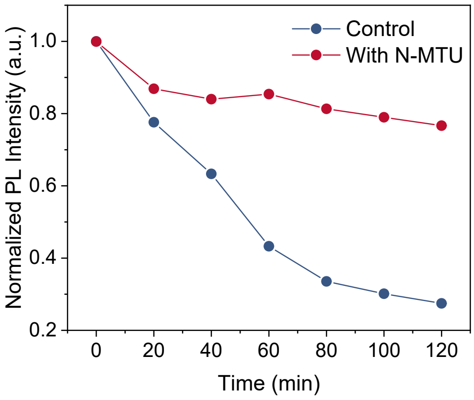

Figure S5. PL stability of PEA_2SnI_4 thin films with and without N-MTU ligands in air under room temperature.

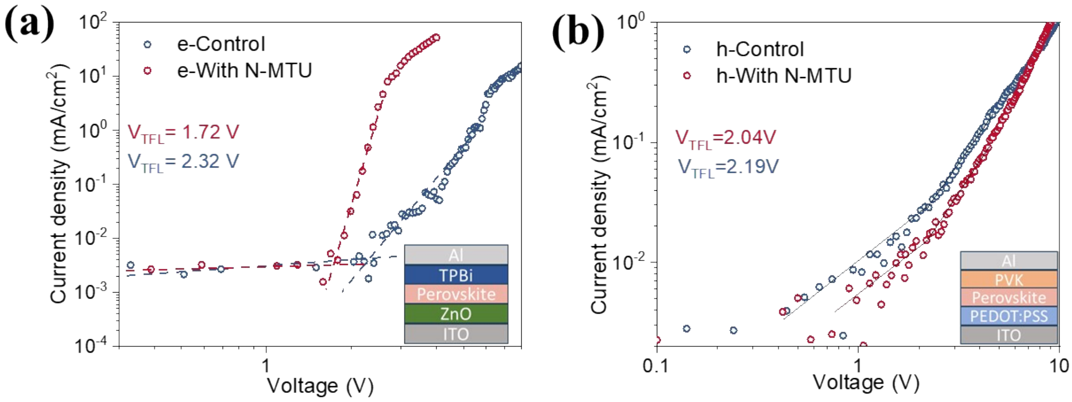

Figure S6. SCLC characteristics of the electron-only (a) and hole-only (b) devices consisting of pristine and N-MTU-modified PEA_2SnI_4 emission layer.

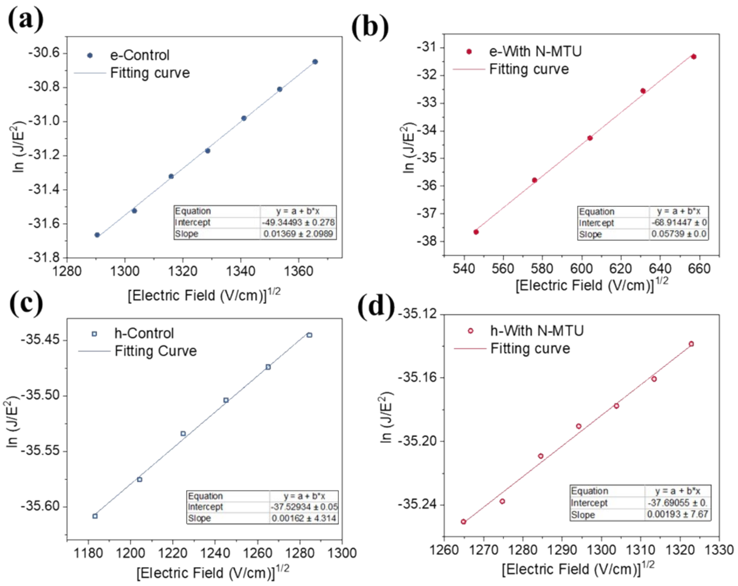

Figure S7. $\ln\left(\frac{J}{E^2}\right)$-$\sqrt{E}$ **curves of electron-only devices (a, b) and hole-only devices (c, d).**

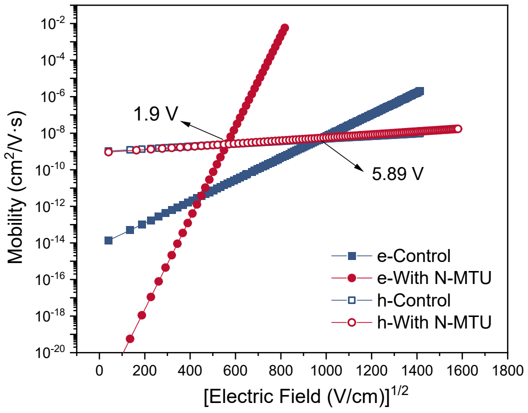

Figure S8. Electric filed-dependent charge-carrier mobility of the electron-only and hole-only devices.

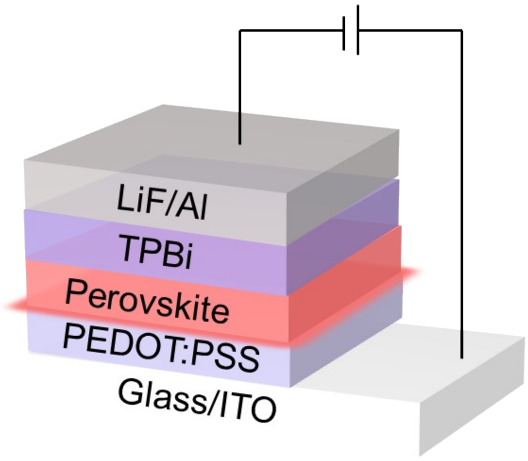

Figure S9. Device structure of the PeLEDs.

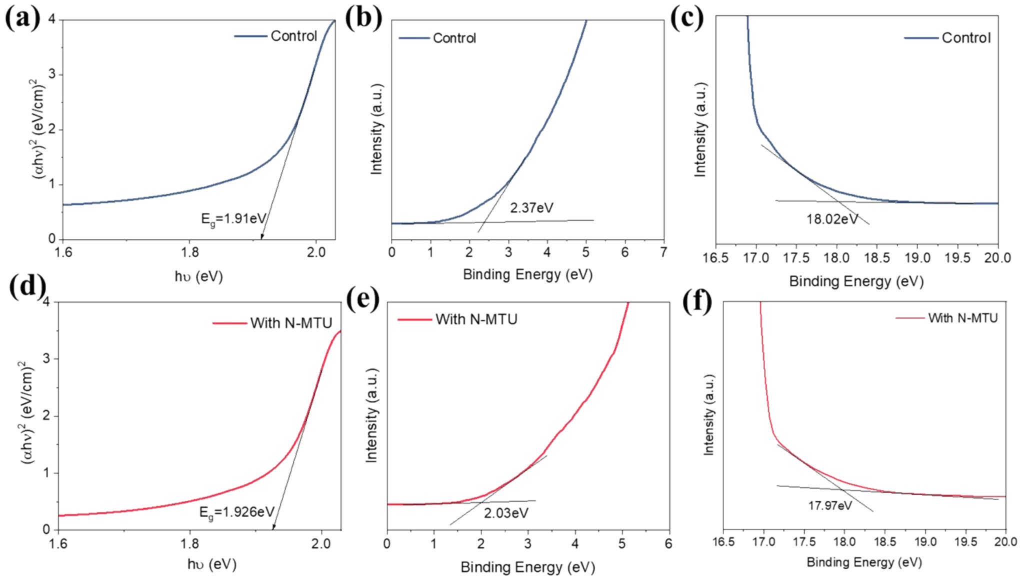

Figure S10. Tauc-plots from the UV–vis absorbance characterizations of control (a) and N-MTU modified PEA_2SnI_4 films (d). The UPS spectra of control (b, c) and N-MTU-modified PEA_2SnI_4 films (e, f).

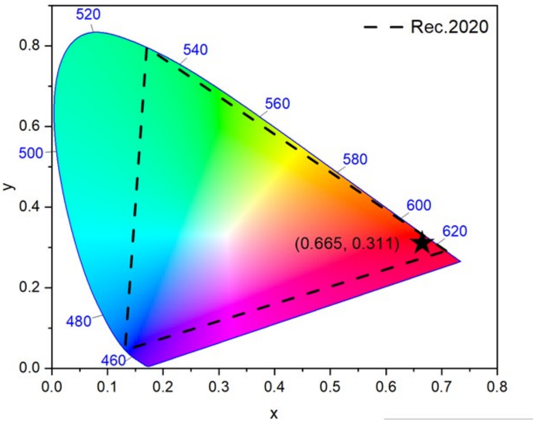

Figure S11. CIE chromaticity coordinates of the N-MTU-based PEA_2SnI_4 PeLED and ITU-R Recommendation BT.2020 (Rec.2020) standards.

Table S2. Summary of the device performances of the reported pure-red PEA_2SnI_4-based PeLEDs.

References	EQE_{MAX} (%)	L_{MAX} (cd/m^2)	EL Peak (nm)	FWHM (nm)
ACS Photonics 2020, 7, 1915-1922	0.72	132	632	/
Adv. Sci. 2020, 7, 1903213	0.3	70	633	24
J. Phys. D: Appl. Phys. 2020, 53, 414005	0.52	355	630	29
Sci. Adv. 2020, 6, eabb0253	5	170	632	21
Adv. Funct. Mater. 2021, 31, 2106974.	0.361	68.84	/	/
J. Mater. Chem. C 2021, 9, 12079-12085	1.48	221	633	/
ACS Appl. Mater. Interfaces 2022, 14, 22941–22949	0.4	43.3	625	28
ACS Energy Lett. 2022, 7, 3653-3655	1	30	630	25
Adv. Funct. Mater. 2023, 2301304	3.51	451	630	23
J. Mater. Chem. C 2023, 11, 9916-9924	0.86	50	630	23
Angew.Chem. Int.Ed. 2023, e202312728	9.32	328.2	625	33
This Work	**2.35**	**509.4**	**626**	**33**

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