

Supporting Information

Direct Integration of Conductive, Emissive Perovskite Nanocrystals Into Efficient Light-Emitting Diodes Without Post-Synthetic Ligand Exchange

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This supplementary material includes:

Figures S1–S26

Supplementary Note S1

Tables S1–S5

References

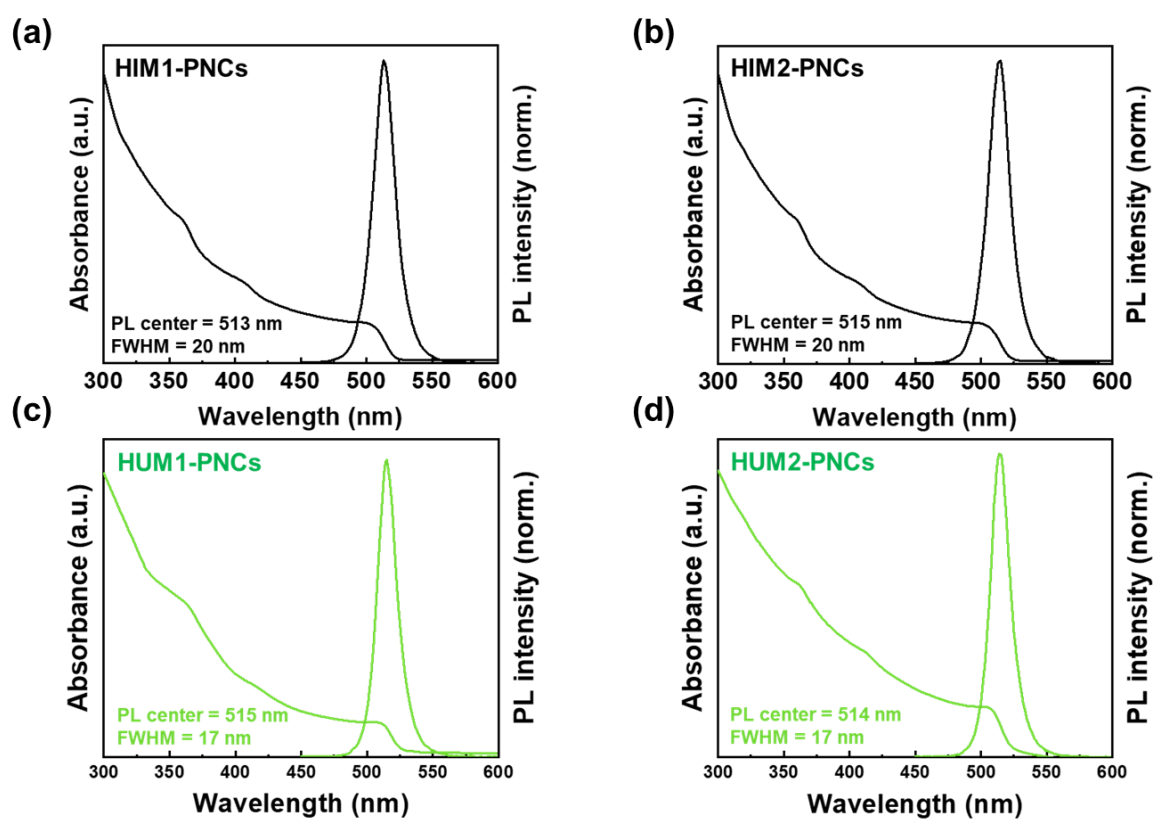


Figure S1. UV-Vis absorption and PL spectra of a) HIM1-, b) HIM2-, c) HUM1-, and d) HUM2-PNC solutions.

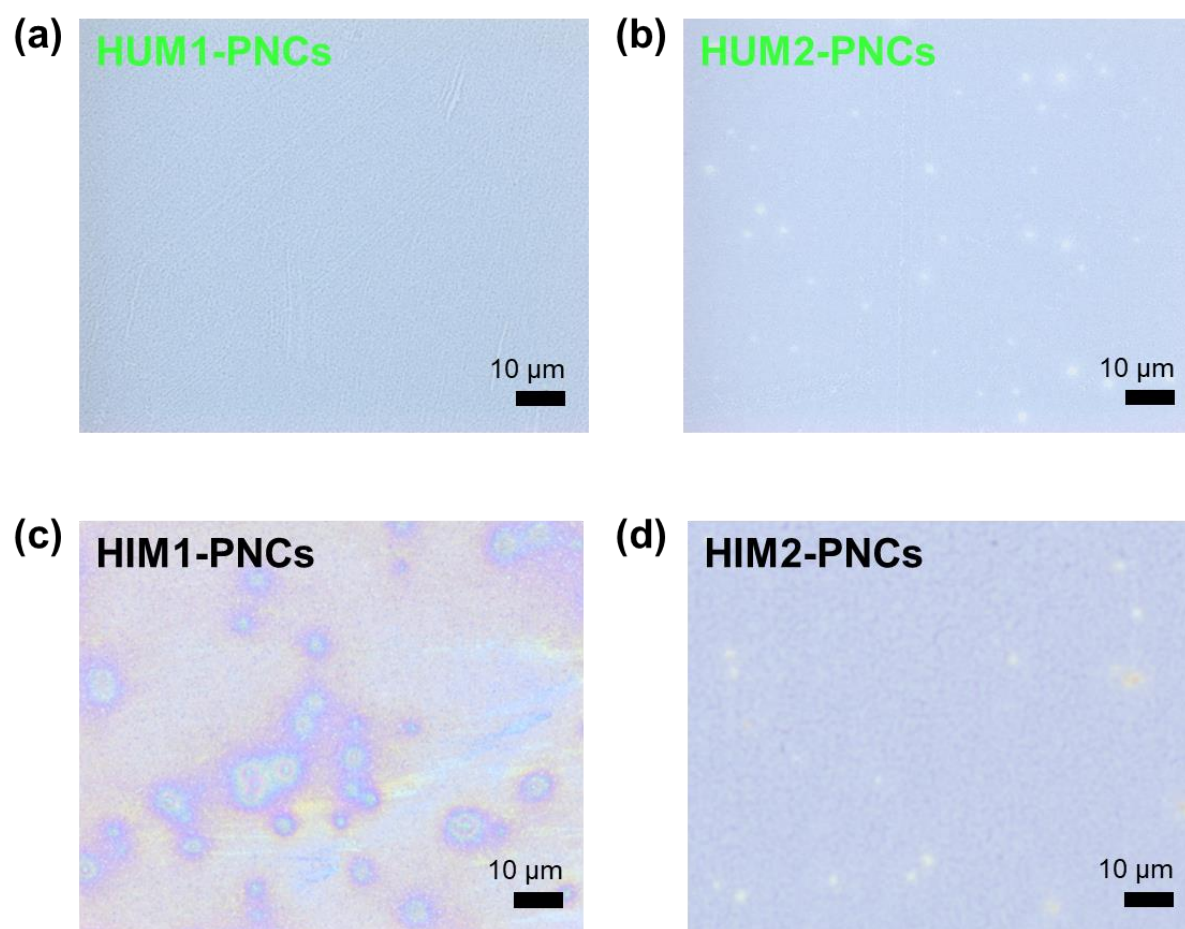


Figure S2. Optical microscopy images of **a)** HUM1-, **b)** HUM2-, **c)** HIM1-, and **d)** HIM2-PNC thin films.

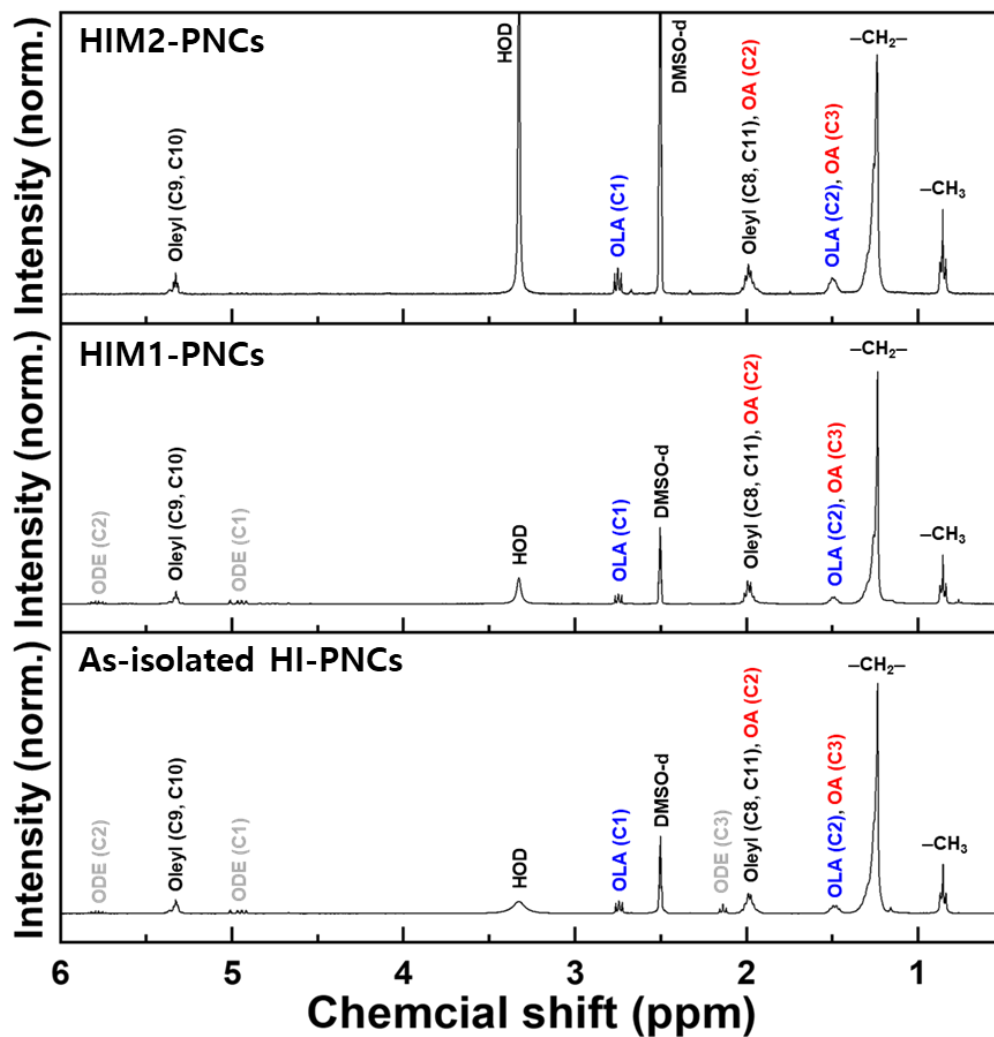
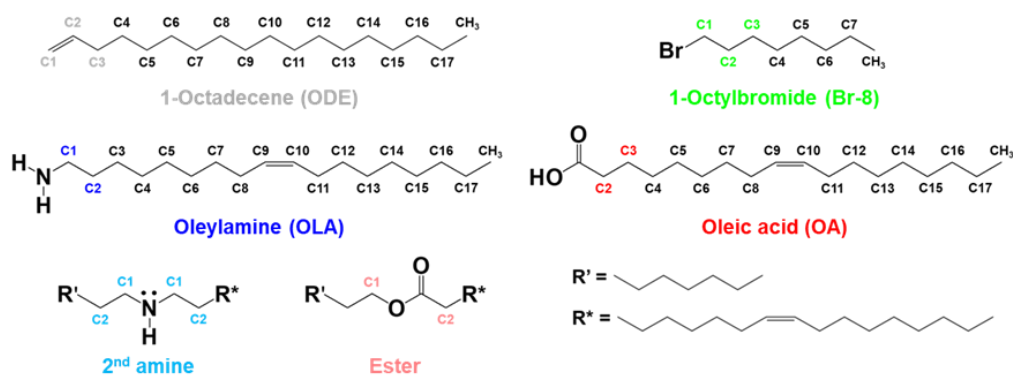


Figure S3. ¹H-NMR spectra of HI-PNCs isolated via high-speed centrifugation before purification, and after subsequent purifications with MeOAc once and twice (*i.e.*, HIM1- and HIM2-PNCs). All samples were prepared in DMSO-*d*₆.

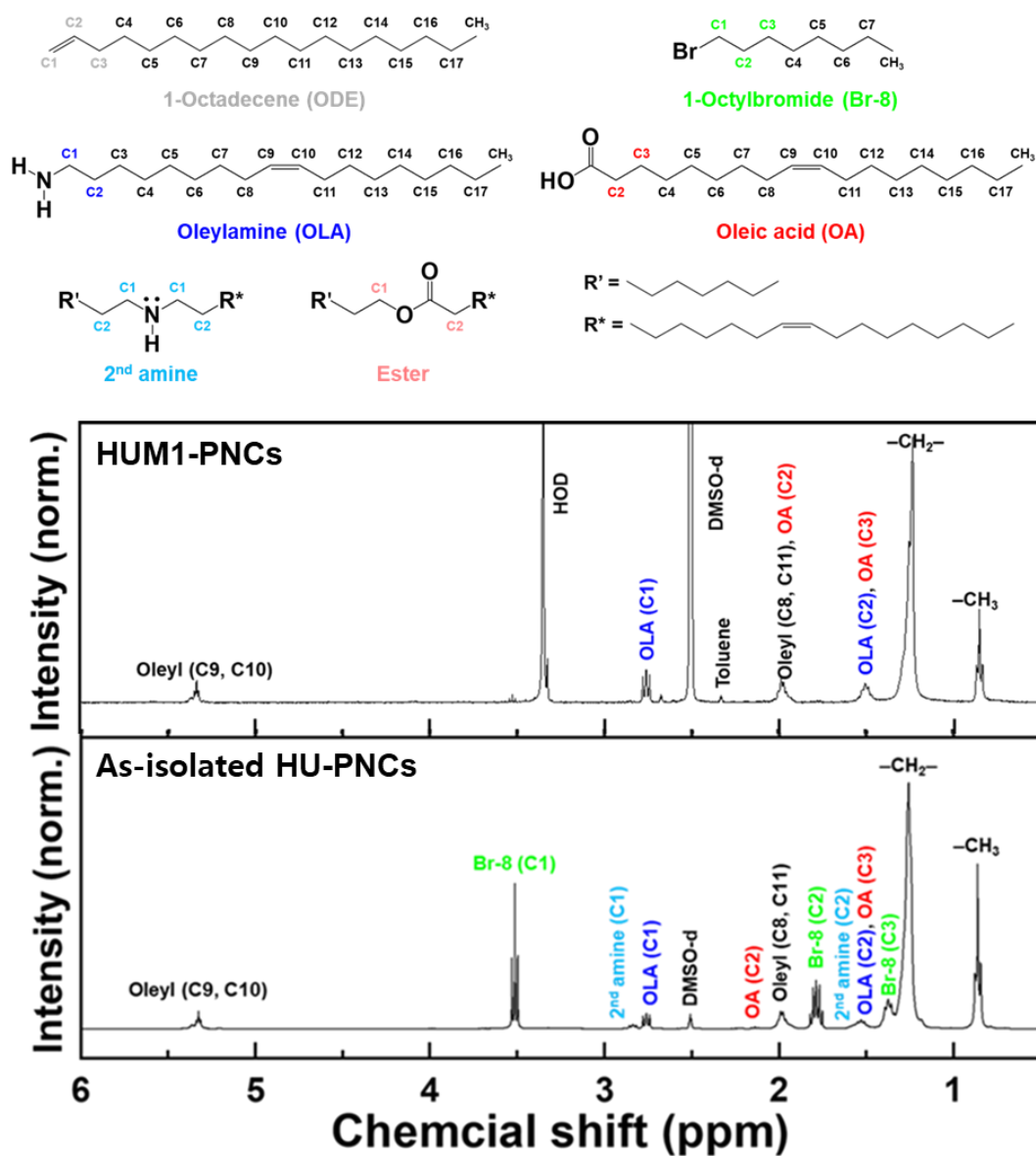


Figure S4. ^1H -NMR spectra of HU-PNCs isolated via high-speed centrifugation before purification, and after a subsequent purification with MeOAc (*i.e.*, HUM1-PNCs). All samples were prepared in DMSO- d_6 .

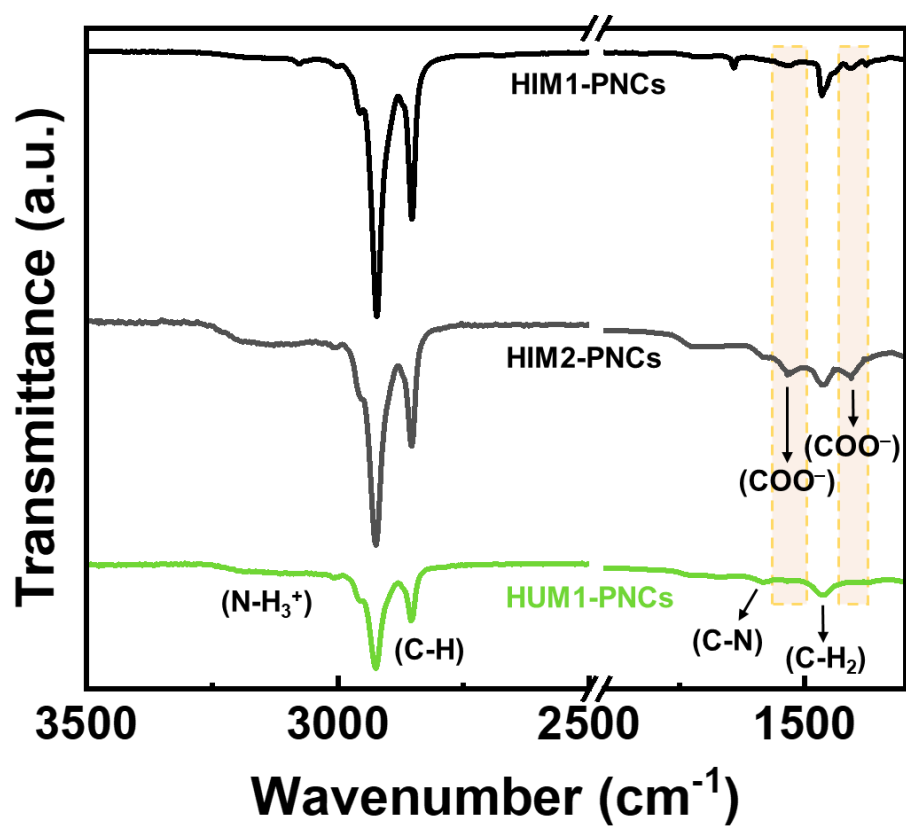


Figure S5. FT-IR spectra of HIM1-, HIM2-, and HUM1-PNCs.

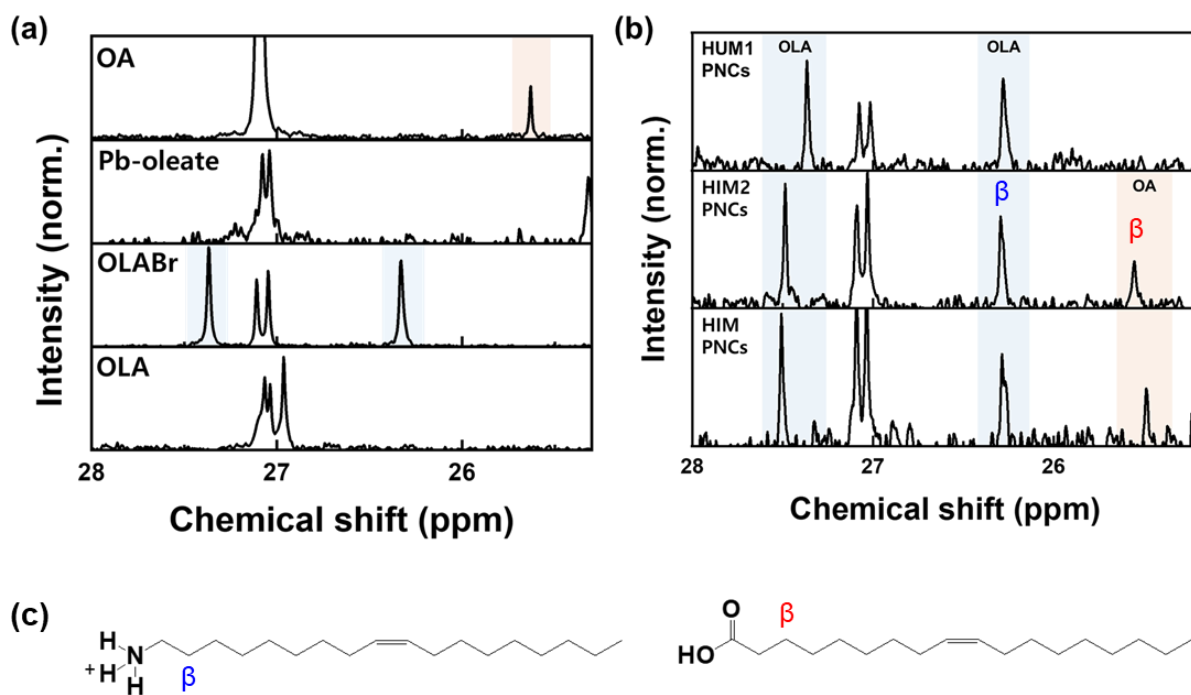


Figure S6. ^{13}C -NMR spectra of **a)** reference samples (i.e., OA, $\text{Pb}(\text{oleate})_2$, OAM-Br, and OLA) and **b)** HIM1-, HIM2-, and HUM1-PNC solution samples. All samples were prepared in $\text{DMSO-}d_6$. **c)** Molecular structures of OAM and OA indicating the assignments of their β -carbons. Based on the ^{13}C -NMR spectrum of the HIM2-PNCs, the relative integrated areas of the resonance peaks corresponding to the β -carbons of OLA and OA reveal that they constitute 65.16% and 34.84% of the surface ligands, respectively. These relative proportions were subsequently used to calculate the average molar mass of the surface ligands on the HIM2-PNCs (**Supplementary Note S1**).

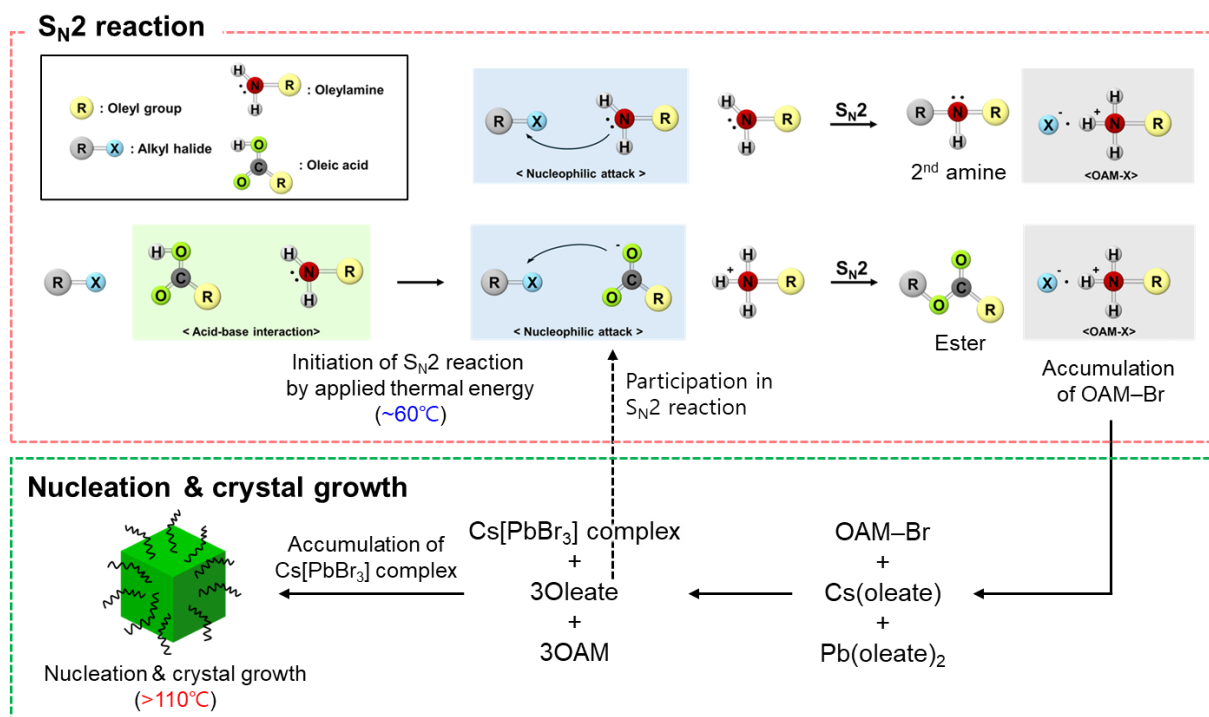


Figure S7. Schematic illustration depicting the S_N2 reaction pathway and the subsequent nucleation and crystal growth mechanism. In the S_N2-based heat-up synthesis, acid–base interactions between OA and OLA first generate oleate and ammonium species. Subsequently, the nucleophilic oleate and OLA gradually react with alkyl bromide via S_N2 pathways in the synthetic medium. As revealed in our previous study ^[26], the formation of the ester product begins at $\sim 60^\circ\text{C}$, whereas the nucleation and crystal growth of CsPbBr₃ PNCs begins at $\sim 110^\circ\text{C}$. This temporal separation indicates that the S_N2 reaction of oleate occurs predominantly in the solution, gradually releasing Br[−] prior to the formation of well-defined PNC surfaces. The accumulated Br[−] then promotes the formation of Cs[PbBr₃] monomers, which drive nucleation and crystal growth at elevated temperatures. Consequently, the substantial amount of OAM generated during this process plays a dominant role in determining the surface chemistry of the final HU-PNCs.

Supplementary Note S1. Calculation of ligand density based on TGA analysis

A protocol for calculating the ligand density of nanocrystals using the organic fraction extracted from TGA curves has been established in the literature [27]. The proposed formula is as follows:

$$\text{Ligand density} = \frac{\frac{m_{\text{particle}} \times \%_{\text{ligand}}}{\%_{\text{particle}}}}{\frac{M_{\text{ligand}}}{S}} \times N_A \quad (\text{Equation S1})$$

where M_{ligand} is the average molar mass of the ligands, m_{particle} is the mass of a single particle, $\%_{\text{particle}}$ and $\%_{\text{ligand}}$ are the weight percentages of the inorganic core and the organic shell, respectively, S is the surface area of one particle, and N_A is Avogadro's number ($6.02 \times 10^{23} \text{ mol}^{-1}$).

In this study, prior to calculating the ligand density, we obtained the average sizes of the HI- and HU-PNCs based on size distribution statistics derived from the HR-TEM images (Figure 1b and S8). Additionally, we determined the organic fractions from the TGA curves (Figure 2a) and the relative ligand ratio from the ^{13}C -NMR spectrum of the HIM2-PNCs (Figure S6). The OLA-to-OA molar ratio in the HIM2-PNCs was 65.16:34.84. Using this ratio, the average molar mass of the ligands on the HI-PNCs was calculated as follows:

$$M_{\text{OLA}} = 268.49 \text{ g mol}^{-1}$$

$$M_{\text{OA}} = 281.46 \text{ g mol}^{-1}$$

$$M_{\text{ligand, HI}} = (0.6516 \times 268.49) + (0.3484 \times 281.46) = 273.01 \text{ g mol}^{-1}$$

The values of the estimated parameters are summarized in Table S1. Assuming a cubic morphology based on the average sizes of the PNCs, we calculated the surface areas of the HI- and HU-PNCs. Furthermore, using the density of bulk CsPbBr_3 (4.42 g cm^{-3}), we determined the mass of the inorganic core for the HI- and HU-PNCs as follows:

$$S_{\text{HI}} = 6 \times (11.01 \text{ nm})^2 = 727.32 \text{ nm}^2$$

$$S_{\text{HU}} = 6 \times (13.32 \text{ nm})^2 = 1064.53 \text{ nm}^2$$

$$m_{\text{particle, HI}} = 4.42 \text{ g cm}^{-3} \times (11.01 \times 10^{-7} \text{ cm})^3 = 5.899 \times 10^{-18} \text{ g}$$

$$m_{\text{particle, HU}} = 4.42 \text{ g cm}^{-3} \times (13.32 \times 10^{-7} \text{ cm})^3 = 1.045 \times 10^{-17} \text{ g}$$

By substituting these calculated terms into Equation S1, the ligand densities of the HI- and HU-PNCs were obtained as follows:

$$\text{Ligand density, HI} = \frac{\frac{(5.899 \times 10^{-18}) \times 0.204}{0.796}}{\frac{273.01}{727.32}} \times N_A = 4.59 \text{ nm}^{-2}$$

$$\text{Ligand density, HU} = \frac{\frac{(1.045 \times 10^{-17}) \times 0.120}{0.880}}{\frac{268.49}{1064.53}} \times N_A = 3.00 \text{ nm}^{-2}$$

The calculated ligand density of the HU-PNCs was 3.00 nm^{-2} , which is significantly lower than the 4.59 nm^{-2} observed for the HI-PNCs, representing an approximately 34.6% reduction.

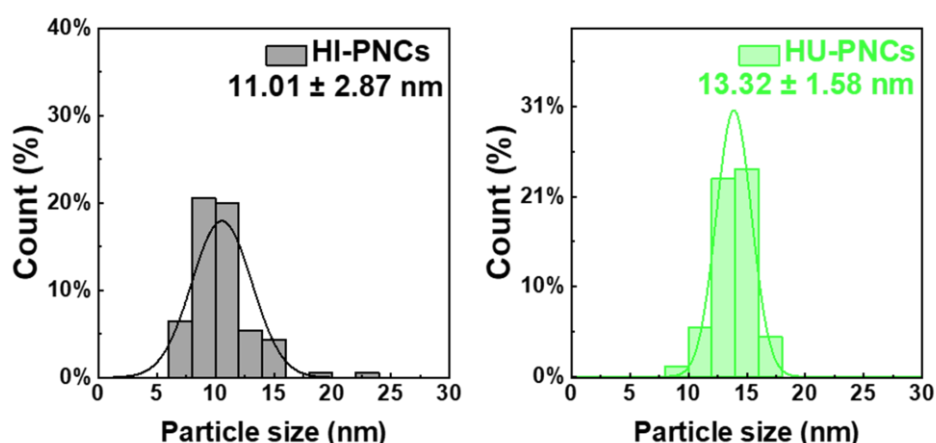


Figure S8. Size distribution histograms of HI- and HU-PNCs derived from the HR-TEM images shown in **Figure 1b**. The average particle sizes, determined via Gaussian fitting, are $11.01 \pm 2.87 \text{ nm}$ and $13.32 \pm 1.58 \text{ nm}$ for the HI- and HU-PNCs, respectively.

Table S1. Parameters used to calculate the ligand density.

	Average size (nm) ¹⁾	Organic fraction from TGA curves (wt%) ²⁾	Ratio of ligand ³⁾	Average molar mass of ligand (g mol ⁻¹)	Ligand density (nm ⁻²)
HI-PNCs	11.01 ± 2.87	20.4	OLA:OA = 65.16:34.84	273.01	4.59
HU-PNCs	13.32 ± 1.58	12.0	OLA 100%	268.49	3.00

¹⁾Derived from the HR-TEM images (**Figure 1b** and **Figure S8**); ²⁾derived from the TGA curves (**Figure 2a**); ³⁾derived from the ¹³C-NMR spectrum (**Figure S6**)

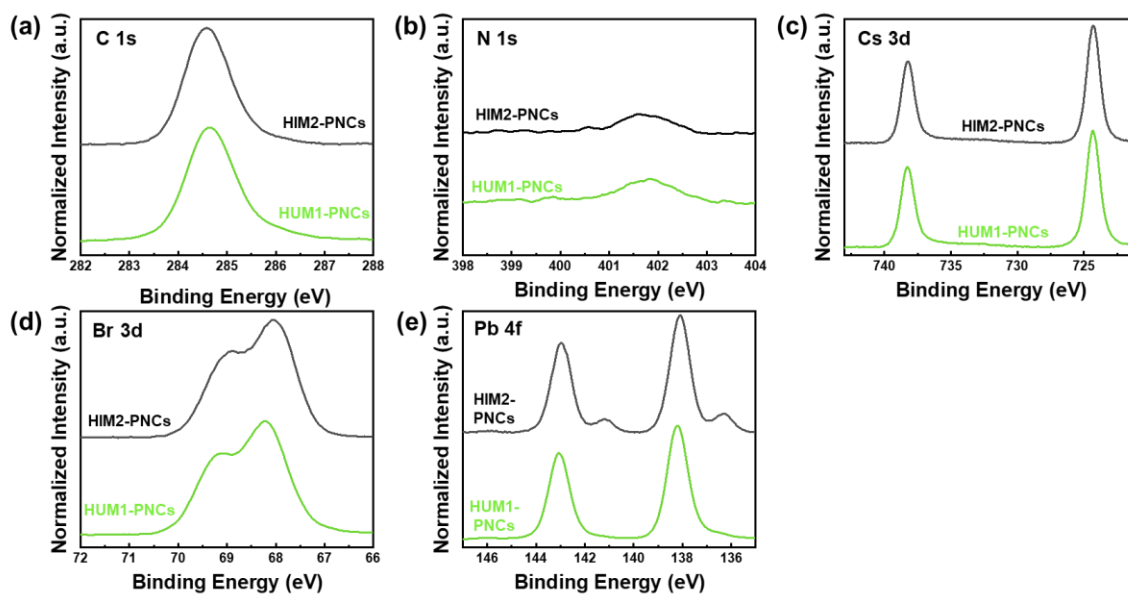


Figure S9. XPS **a)** C 1s, **b)** N 1s, **c)** Cs 3d, **d)** Br 3d, and **e)** Pb 4f spectra of HIM2- and HUM1-PNCs. The binding energies were calibrated using C 1s peak at 284.5 eV.

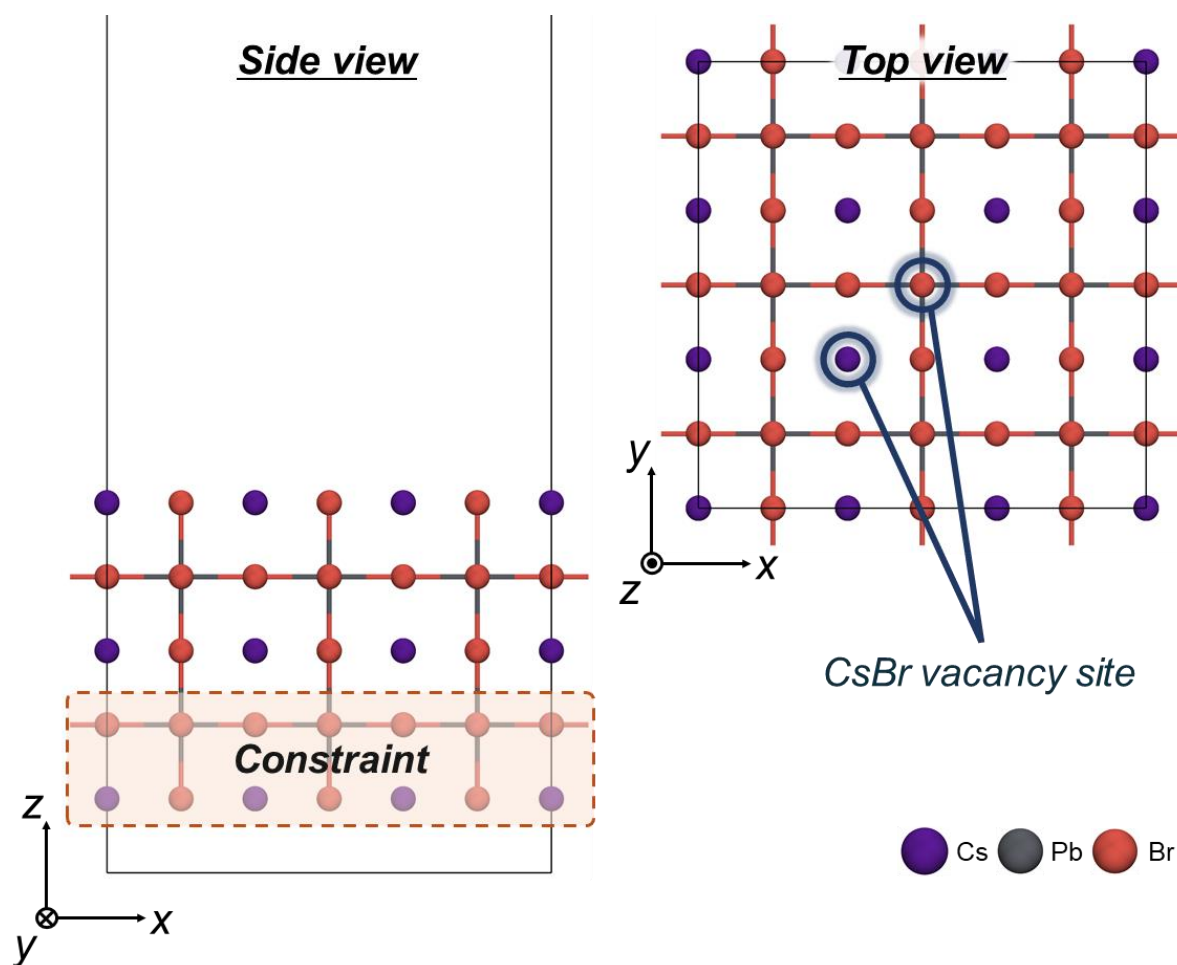


Figure S10. Model system for the (100) surface of CsPbBr_3 perovskite with CsBr vacancy. Note that the surface consists of five layers with 3×3 supercell of the CsPbBr_3 unit cell along x- and y- axes. From the bottom side, two layers are constrained and the vacuum spacing is over 30 Å.

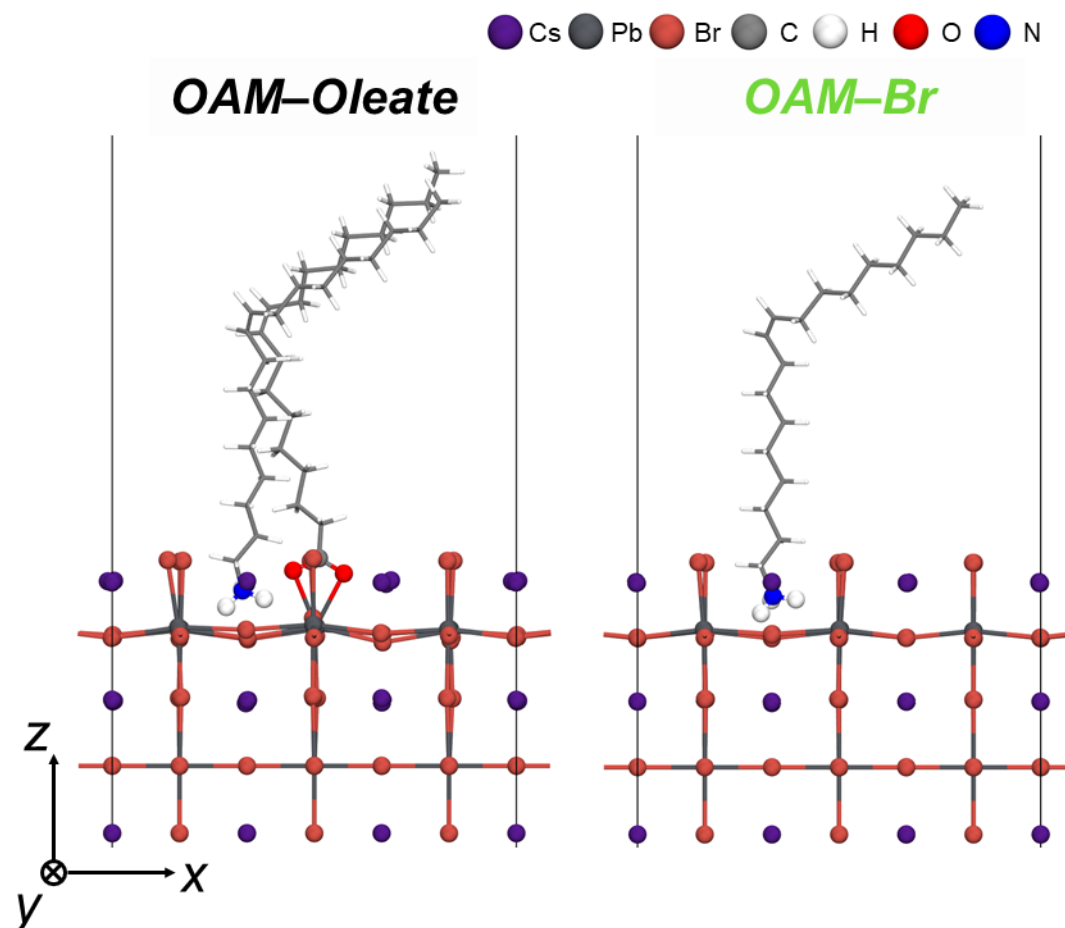


Figure S11. Geometrically optimized structures of OAM–oleate and OAM–Br passivated to the (100) surface of CsPbBr₃ perovskite. Note that the vacuum spacing is over 15 Å.

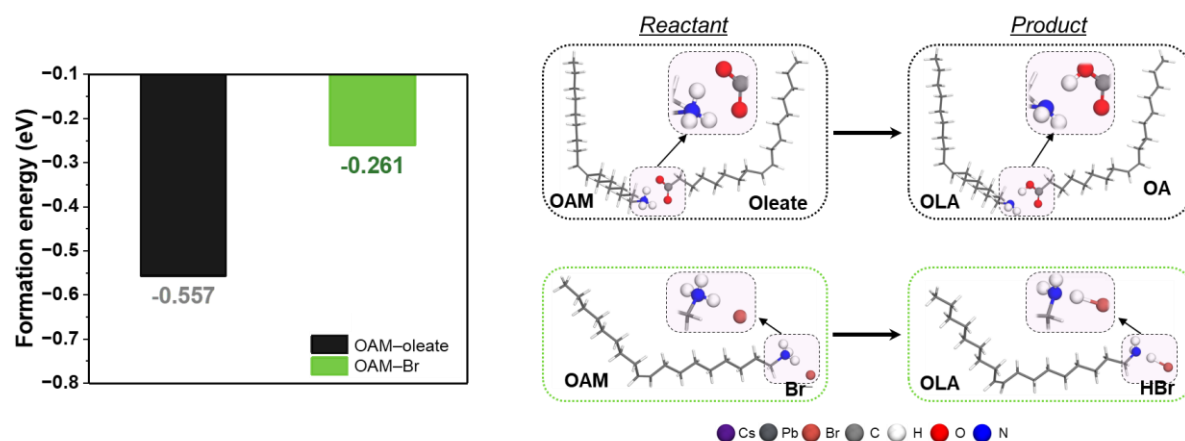


Figure S12. Formation energies of the deprotonation reaction of OAM in the presence of oleate or Br⁻ and corresponding optimized structures. Note that the NH³⁺ group in the reactant is constrained.

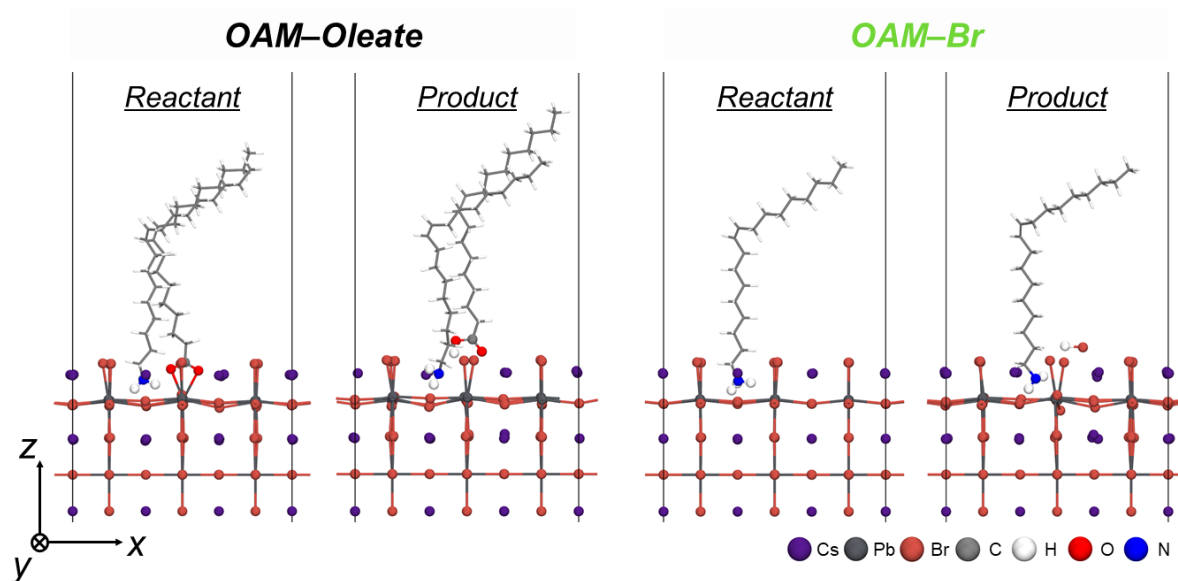


Figure S13. Geometrically optimized structures of deprotonation reaction of OAM in the presence of oleate or Br⁻ on the (100) surface of CsPbBr₃ perovskite.

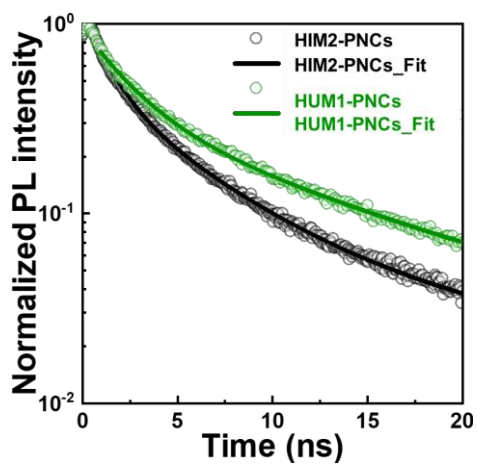


Figure S14. TR-PL plots and fitted curves of HIM2- and HUM1-PNC solutions.

Table S2. Calculated parameters of TR-PL triexponential fitting of HIM2- and HUM1-PNC solutions.

	A_1 (%)	τ_1 (ns)	A_2 (%)	τ_2 (ns)	A_3 (%)	τ_3 (ns)	τ_{avg} (ns)
HIM2-PNCs	32.984	0.61864	50.075	2.81369	17.373	10.77618	6.96244
HUM1-PNCs	15.91	2.224989	44.19	9.059621	39.90	26.58882	7.39213

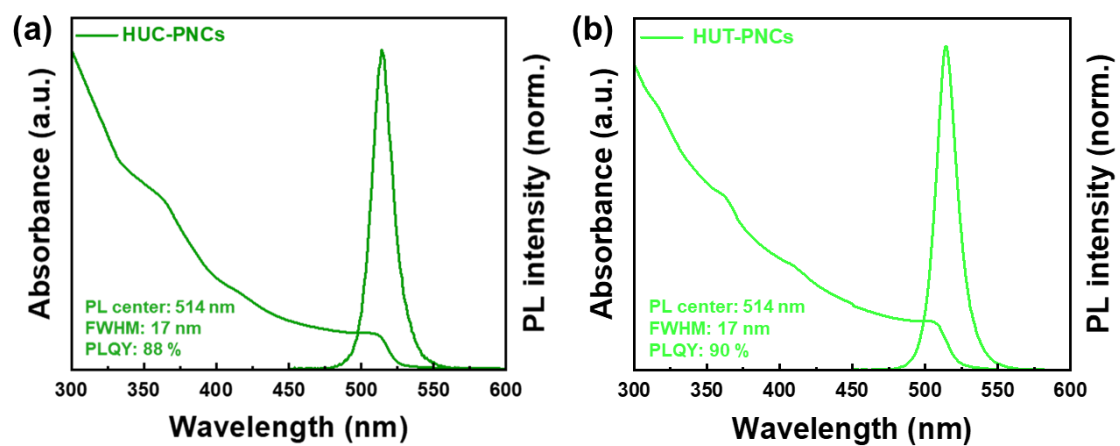


Figure S15. UV–Vis absorption and PL spectra of (a) HUC- and (b) HUT-PNC solutions.

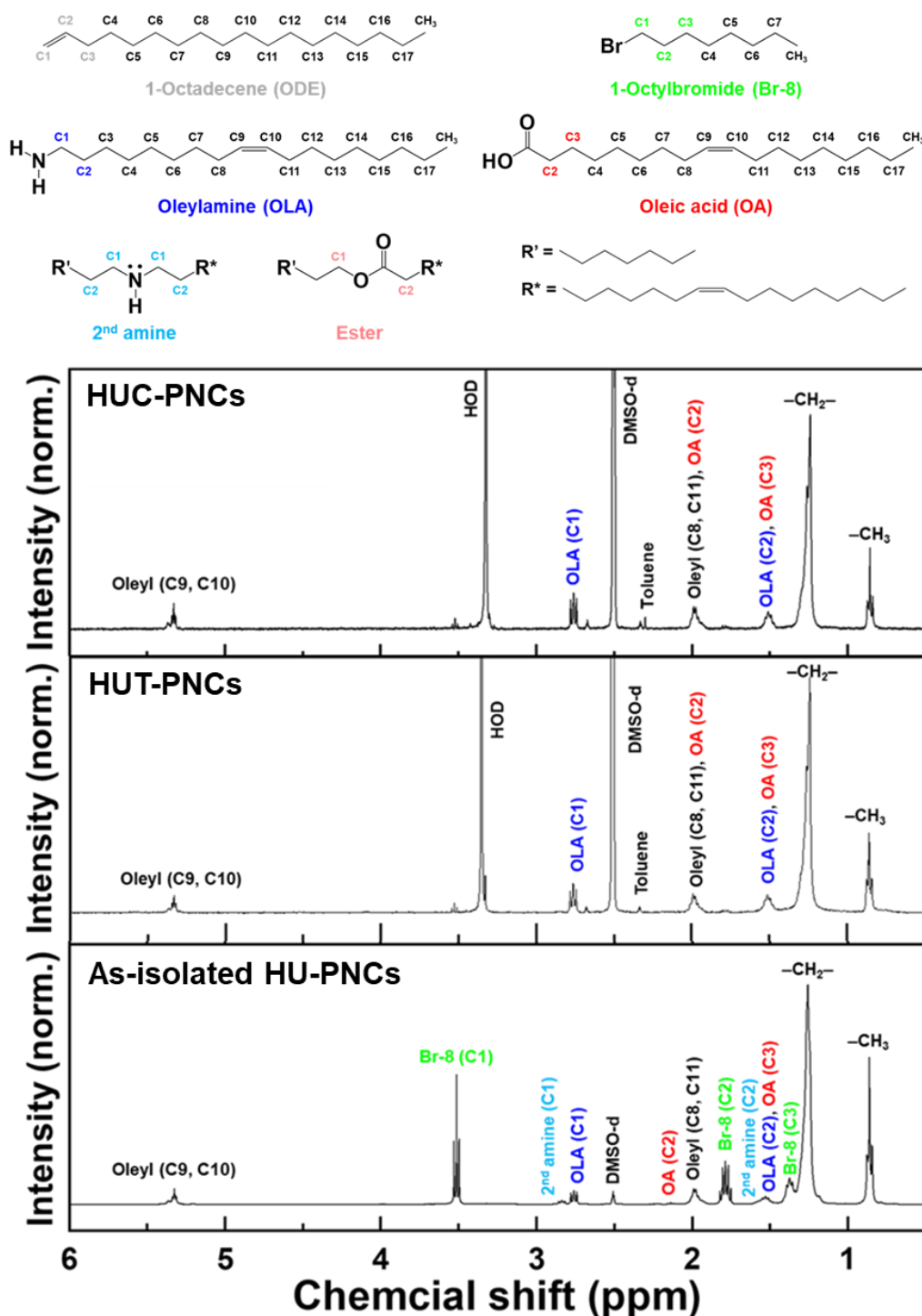


Figure S16. ^1H -NMR spectra of HU-PNCs isolated via high-speed centrifugation before purification, and after a subsequent purification with each anti-solvent (CB and toluene) (*i.e.*, HUC- and HUT-PNCs). All samples were prepared in $\text{DMSO}-d_6$. The ^1H -NMR spectrum of as-isolated HU-PNCs is identical to that shown in **Figure S4**. Resonances associated with unreacted precursors (*e.g.*, 1-octylbromide) were eliminated in HUC- and HUT-PNCs, similar to HUM1-PNCs (**Figure S4**). This finding indicates that HU-PNCs with low ligand density can be purified from unreacted chemicals even in nonpolar solvents (CB and toluene). This, in turn, suggests that the weight loss below 430°C identified by TGA originate from surface ligands.

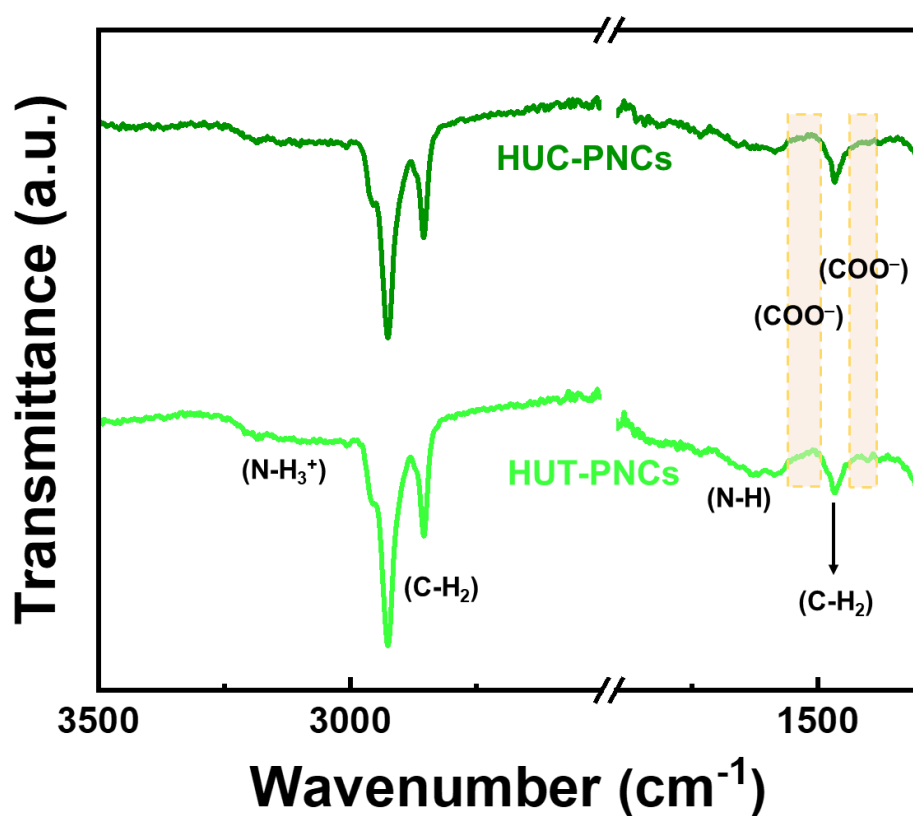


Figure S17. FT-IR spectra of HUC- and HUT-PNCs. Carboxylate-related peaks (*i.e.*, symmetric and asymmetric COO⁻) were not shown in the FT-IR spectra of HUC- and HUT-PNCs.

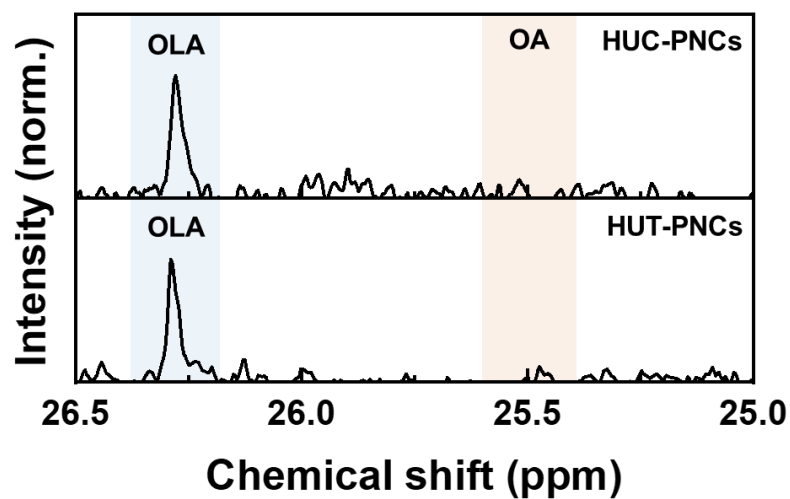


Figure S18. ^{13}C -NMR spectra of HUC-, HUT-PNC solution samples prepared in $\text{DMSO-}d_6$.

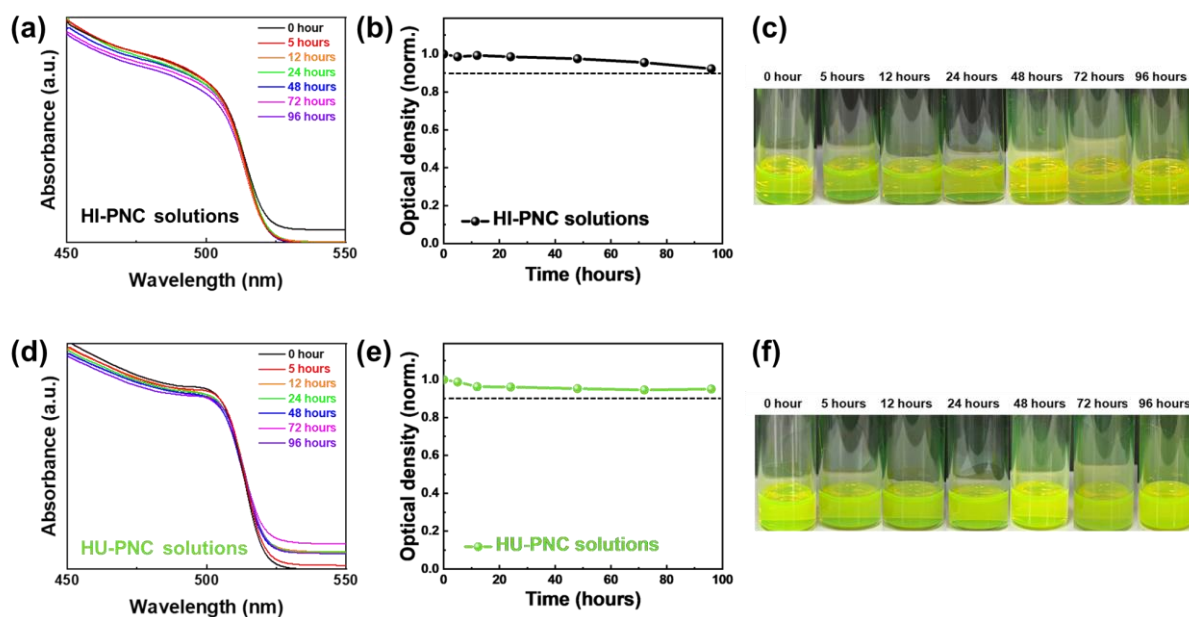


Figure S19. Data for the HIM2-PNC solutions showing **a)** solution-phase UV–Vis absorption spectra, **b)** changes in optical density at 500 nm, and **c)** photographic images. Corresponding data for the HUT-PNC solutions showing **d)** solution-phase UV–Vis absorption spectra, **e)** changes in optical density at 500 nm, and **f)** photographic images.

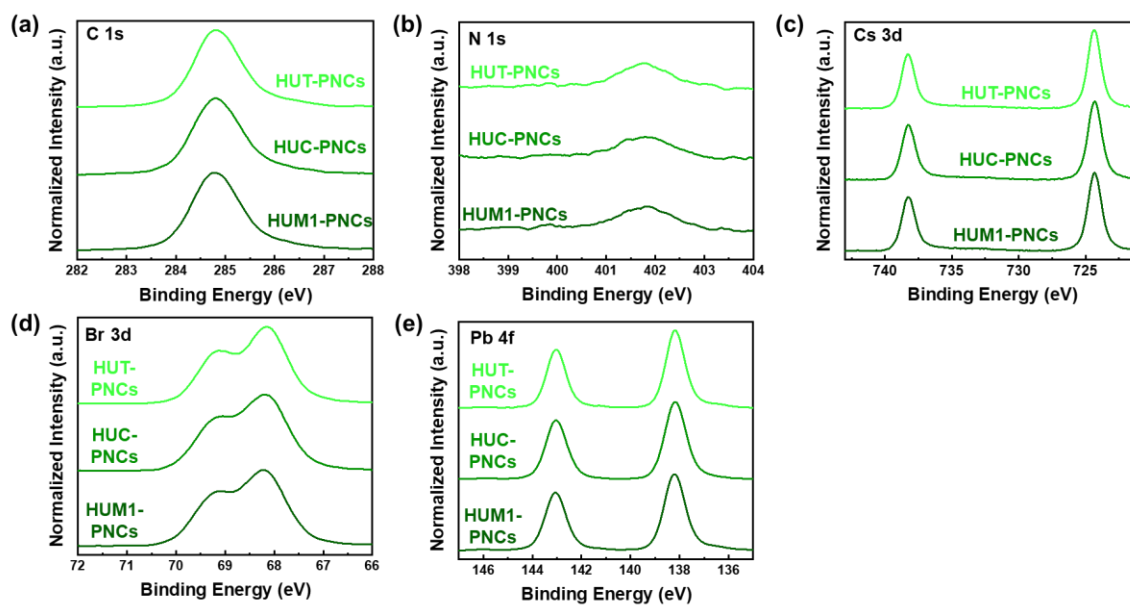


Figure S20. XPS **a)** C 1s, **b)** N 1s, **c)** Cs 3d, **d)** Br 3d, and **e)** Pb 4f spectra of HUM1-, HUC-, and HUT-PNCs. The binding energies were calibrated using C 1s peak at 284.5 eV. The XPS spectra of HUM1-PNCs are identical to those shown in **Figure S9**.

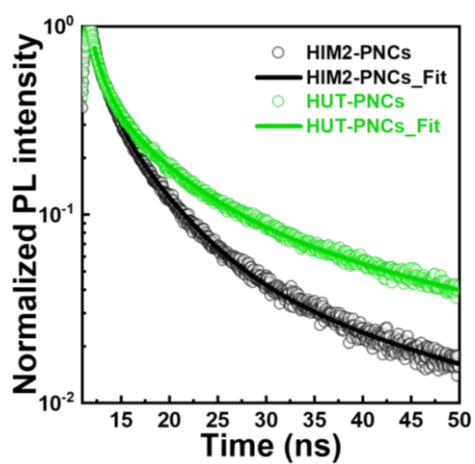


Figure S21. TR-PL plots and fitted curves of HIM2- and HUT-PNC solutions. The data for HIM2-PNCs are identical to those shown in **Figure S14**.

Table S3. Calculated parameters of TR-PL triexponential fitting of HIM2- and HUT-PNC solutions. The data for HIM2-PNCs are identical those shown in **Table S2**.

	A_1 (%)	τ_1 (ns)	A_2 (%)	τ_2 (ns)	A_3 (%)	τ_3 (ns)	τ_{avg} (ns)
HIM2-PNCs	32.984	0.61864	50.075	2.81369	17.373	10.77618	6.96244
HUT-PNCs	43.409	0.75606	37.813	3.6384	19.551	16.36867	11.75261

Table S4. Calculated parameters of TR-PL triexponential fitting of HIM2- and HUT-PNC thin films, measured at various emission photon energies including PL emission maximum.

	A_1 (%)	τ_1 (ns)	A_2 (%)	τ_2 (ns)	A_3 (%)	τ_3 (ns)	τ_{avg} (ns)
HIM2-PNCs at 2.44 eV	0.47466	0.72749	0.45978	3.18346	0.11555	11.63307	6.51654
HIM2-PNCs at 2.39 eV	0.27948	0.93258	0.55609	3.42566	0.17795	11.50893	7.20028
HIM2-PNCs at 2.34 eV	0.30681	1.04322	0.55575	3.77189	0.15092	13.29539	7.89501
HUT-PNCs at 2.44 eV	0.19538	0.75055	0.34226	4.26358	0.45092	15.91325	13.72418
HUT-PNCs at 2.39 eV	0.07836	0.47441	0.37788	4.82582	0.53598	16.78607	14.72147
HUT-PNCs at 2.34 eV	0.0645	0.61797	0.37278	4.91015	0.53564	16.92869	14.85801

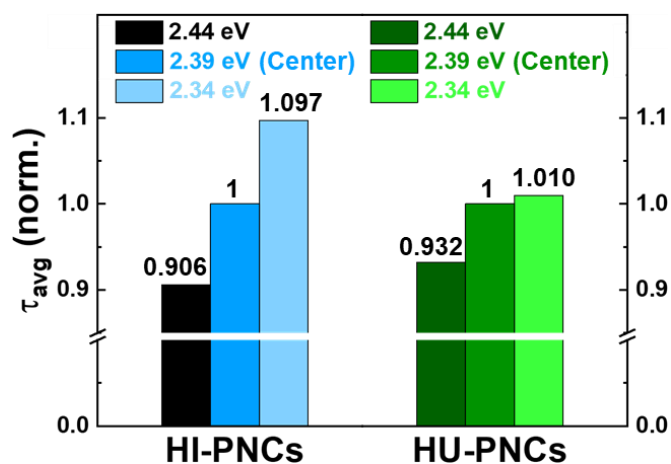


Figure S22. Average PL decay lifetimes of HIM2- and HUT-PNC thin films at various emission photon energies including PL emission maximum (**Figure 4c** and **Table S4**), normalized to the respective average lifetimes corresponding to PL emission maximum.

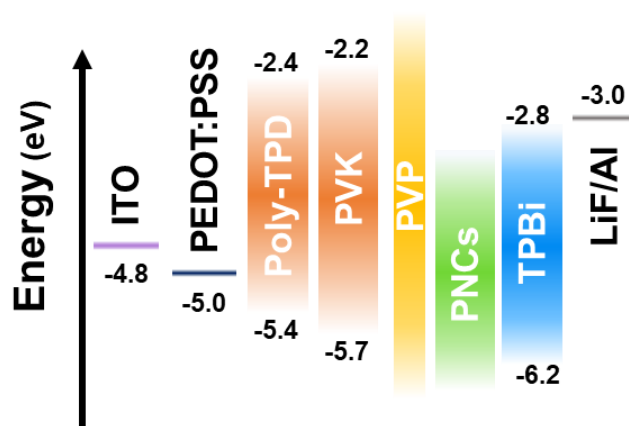


Figure S23. Energy band diagram of HI- and HU-PNC LEDs.

Table S5. Summary of LED device performance reported to date for colloidal PNCs integrated without post-synthetic surface engineering (*e.g.*, ligand exchange, additives).

Year	PNC composition	Surface ligand	EQE (% @ bias)	Turn-on voltage (V @ 0.1 cd m ⁻²)	EL peak (nm)	EL FWHM (nm)	Ref.
2026	CsPbBr ₃	OAM, Br	11.4 @3.0	2.7	514	17	This work
2025	CsPbBr ₃	OAM, Oleate	0.49	~2.3	513	17	[1]
2024	CsPbBr ₃	DOPA ¹⁾ , TOPO ²⁾ , TOAB ³⁾ , Lecithin	7.3	2.3	517	16.2	[2]
2024	CsPbBr ₃	OcAm ⁴⁾ , OTA ⁵⁾	5.46	2.4	464	23	[3]
2024	CsPbBr ₃	OAM, Oleate	4.06	-	-	-	[4]
2024	CsPbBr ₃	OAM, Oleate	~4	3.2	520	-	[5]
2024	CsPbBr ₃	OAM, Oleate	1.79	3.2	510	-	[6]
2024	CsPbBr ₃	OAM, Oleate	0.3	3.3	512	21	[7]
2024	CsPbBr ₃	OAM, Oleate	0.23				[8]
2023	CsPbBr ₃	OTAc ⁵⁾ , TOAB ⁶⁾ , DDAB ⁷⁾	7.95 @3.4	-	519	-	[9]
2023	CsPbBr ₃	OAM, Oleate	3.32		~517	~17	[10]
2023	CsPbBr ₃	OAM, Oleate	2.17	-	-	-	[11]
2023	CsPbBr ₃	OAM, Oleate	0.2	3.8	~517		[12]
2022	CsPbI ₃	OAM, Oleate, Lecithin	7.1	2.99	634	39	[13]
2022	CsPbBr ₃	OAM, Oleate	1.3	3.6	516	18	[14]
2022	CsPbBr ₃	OTAc ⁵⁾ , TOAB ⁶⁾ , DDAB ⁷⁾	0.053	7.0	518.9	24.8	[15]
2021	MAPbBr ₃	OAM, Oleate	3.5 @5.0	3	524	22.4	[16]
2021	CsPbBr ₃	OAM, Oleate	0.87	4.7	517	17	[17]
2021	CsPbBr ₃	OAM, Oleate	0.4	5.2	-	-	[18]
2021	CsPbBr ₃	OAM, Oleate	0.09	2.6	512	-	[19]
2020	FAPbBr ₃	OAM, Oleate	8.1	2.6	512	20	[20]
2020	CsPbBr ₃	PEG ⁸⁾	0.29	-	520	21	[21]
2019	CsPbBr ₃	OAM, Oleate, HPA ⁹⁾	9.8	3.0	515	-	[22]
2019	CsPbBr ₃	OAM, Oleate	0.3	4.6	516	20	[23]
2018	CsPbBr ₃	Oleate, OPA ¹⁰⁾ , TOPO ¹¹⁾	6.5@3.4	2.8	516	-	[24]
2018	CsPbBr ₃	Sulfobetaine ¹²⁾	2.5@3.5	-	518	-	[25]

¹⁾Diisooctylphosphinic acid; ²⁾Trioctylphosphine oxide; ³⁾Trioctyl (propyl-3-sulfonate) ammonium betaine; ⁴⁾n-Octylamine; ⁵⁾n-Octanoic acid; ⁶⁾Tetraoctylammonium bromide ⁷⁾Didodecyldimethylammonium bromide; ⁸⁾Polyethylene glycol; ⁹⁾Hexylphosphonic acid; ¹⁰⁾Octylphosphonic acid; ¹¹⁾Trioctylphosphine oxide; ¹²⁾3-(N,N-dimethylotadecylammonio)propanesulfonate (C18-sulfobetaine)

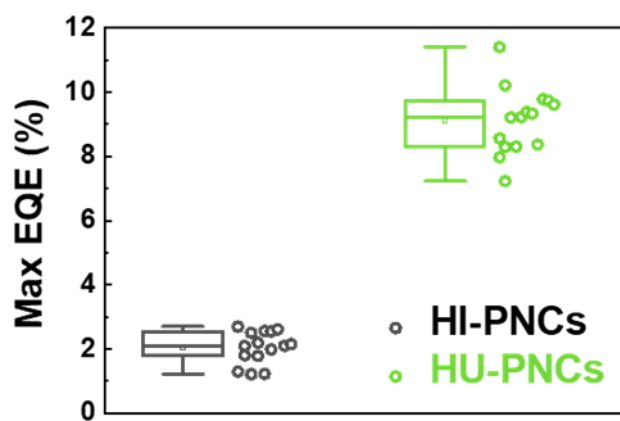


Figure S24. Statistical distribution of maximum EQE values from 15 devices each for HI- and HU-PNC LEDs.

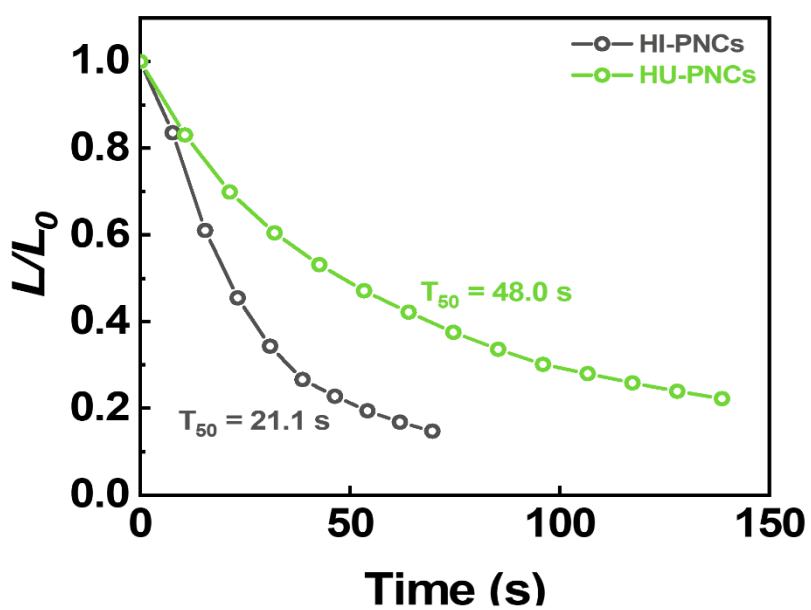


Figure S25. Operational stability of HI- and HU-PNC LEDs under a constant initial luminance of $\sim 50 \text{ cd m}^{-2}$.

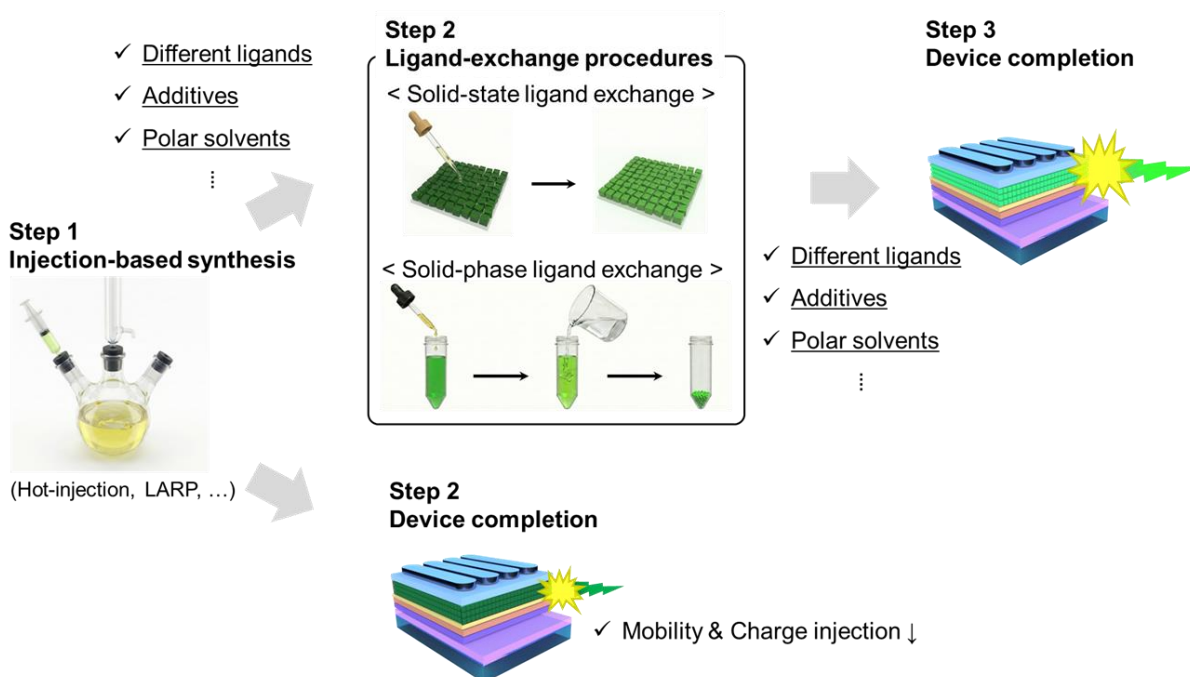
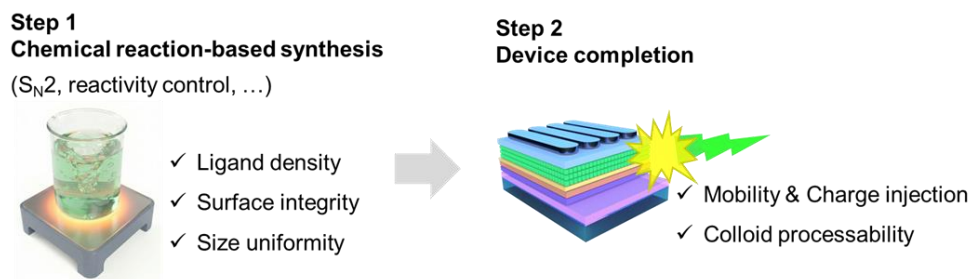
(a) Conventional PNC-based LED fabrication process**(b) Direct integration of colloidal PNCs to LED devices (This work)**

Figure S26. a) Schematic of conventional device fabrication processes for PNCs prepared by conventional injection-based methods. These require post-synthetic procedures such as ligand exchange, additives, and polar solvents, and are subject to a trade-off between ligand density and mobility. b) Direct device integration of charge-conductive colloidal PNCs obtained via S_N2 reaction-based synthesis.

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