

Supporting Information

Multifunctional Ammonium Salts Synergizing Etching and Passivation Capability Enable Efficient Deep-Blue CsPbBr₃ Nanoplates

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Chemicals: The following chemicals were used without purification: cesium carbonate (Cs_2CO_3 , Energy Chemical, 99.9%), lead bromide (PbBr_2 , Youxuan Techonology, 99.9%), oleic acid (technical grade 90%), oleyl amine (technical grade 70%), toluene (for HPLC, $\geq 99.9\%$), acetone (for HPLC, $\geq 99.9\%$), hexane (for HPLC, $\geq 99.5\%$, GC) and ethyl acetate (ACS reagent, $\geq 99.5\%$) were purchased from Energy Chemical. Pyridine Hydrobromide (PyBr, Meryer, 98%), 2,6-Diaminopyridine (Aladdin, 98%), HBr (48 wt. % in H_2O), m-Phenylenediamine (MPD, 99%) was purchased from Meryer.

Synthesis of DaPyBr: 0.03 mol of 2,6-diaminopyridine was dissolved in 20 ml of anhydrous ethanol at room temperature with stirring until completely dissolved. Then, a solution containing 0.09 mol of HBr was added dropwise using a dropping funnel (over 40 minutes), with simultaneous stirring. After 3-4 hours of reaction, the resulting pale-yellow solution was transferred to a rotary evaporator and evaporated. The solid obtained after rotary evaporation was dissolved in a small amount of anhydrous ethanol, followed by recrystallization with diethyl ether. This procedure was repeated two to three times, yielding a final white solid, which was vacuum dried for 24 hours.

Synthesis of the untreated CsPbBr_3 NPLs: CsPbBr_3 nanoplates (NPLs) were synthesized with the method reported by Bohn et. al. 0.1 mmol Cs_2CO_3 powder was dissolved in 10 ml oleic acid at 100°C under continuous stirring to prepare the Cs-oleate precursor. 0.1 mmol PbBr_2 powder and 100 μl each of oleyl amine and oleic acid were

dissolved in 10 ml toluene at 100°C to form the PbBr₂-precursor solution. The Cs-oleate precursor (200ul) is added under vigorous stirring into a toluene solution containing the PbBr₂-oleylamine/oleic acid precursor (1ml) at room temperature. After 5 s, acetone is added to initiate the formation of NPLs. After 1 min of stirring, the solution is centrifuged at 4000 rpm for 3 min and the precipitate redispersed in 2 ml of hexane to obtain the untreated CsPbBr₃ NPLs.

Synthesis of the control CsPbBr₃ NPLs: The Cs-oleate precursor and PbBr₂-precursor solution are the same as mentioned above. Then, the Cs-oleate precursor (150 ul) is added under vigorous stirring into a toluene solution containing the PbBr₂-oleylamine/oleic acid precursor (1.5 ml) at room temperature. The remaining procedures are same as that of preparing the untreated NPLs.

Synthesis of DaPyBr-NPLs: First, different counts of DaPyBr (1.5 mg, 2.5mg, 3.5 mg, 4.5 mg, 5.5 mg) are dissolved in 2ml acetone to form different concentrations of DaPyBr acetone solution. The DaPyBr-CsPbBr₃ NPLs synthesis followed the same protocol as for CsPbBr₃ NPLs but including the addition of DaPyBr with the acetone step. Specifically, the Cs-oleate precursor (200 ul) is added under vigorous stirring into a toluene solution containing the PbBr₂-oleylamine/oleic acid precursor (1 ml) at room temperature. After 5 s, DaPyBr acetone solution is added to initiate the formation of NPLs. After 1 min of stirring, the solution is centrifuged at 4000 rpm for 3 min and the precipitate redispersed in 2 ml of hexane to obtain the untreated CsPbBr₃ NPLs.

Synthesis of PyBr-NPLs: Different counts of PyBr (1.5 mg, 2.5mg, 3.5 mg, 4.5 mg,

5.5 mg) are dissolved in 2 ml acetone to form different concentrations of PyBr acetone solution. The remaining procedures are same as that of preparing the DaPyBr-CsPbBr₃ NPLs.

Characterization: Ultraviolet–visible (UV–vis) absorption spectra were measured by Thermo Evolution 300 UV–vis spectrometer. Steady-state photoluminescence (PL) spectra, time resolution photoluminescence (TRPL) spectra, and absolute photoluminescence quantum yield (PLQY) were carried out by a Horiba Fluorolog system with an excitation wavelength of 365 nm and a 395 nm nano-LED source. Atomic force microscopy (AFM) was performed by Bruker Dimension icon. X-ray diffraction (XRD) patterns were characterized by Rigaku Miniflex 600 with Cu K α radiation ($\lambda = 1.54178 \text{ \AA}$). Transmission electron microscope (TEM) was performed by JEM-2100F at 200 kV accelerating voltage. X-ray photoelectron spectroscopy (XPS) spectra were recorded by a Thermo ESCALAB-250 spectrometer.

Calculation:

The density functional theory (DFT) calculations were performed using a Castep module¹ to illustrate the phenomenon. The generalized gradient approximation (GGA) method with Perdew-Burke-Ernzerhof (PBE) function² was employed to describe the interactions between the valence electrons and the ionic core³. The energy cut-off for the plane-wave basis set was 400 eV was adopted. The threshold values of the convergence criteria were specified as follows: 0.002 \AA for maximum displacement, 0.05 eV/ \AA for the maximum force, 0.1 GPa for the maximum stress, 10^{-5} eV/atom for

energy, and 2.0×10^{-6} eV/atom for self-consistent field tolerance. The surface of CsPbBr₃ (100) was build. 15 Å vacuum space were implemented into the model to eliminate undesirable interactions between bottom side of the slab and the molecules in the vacuum space. The (2*2) unit cell of CsPbBr₃ (100) were used to prohibit lateral interactions between molecules on the surface.

The binding energy (ΔE) was calculated as

$$\Delta E = E_{\text{total}} - E_1 - E_2$$

where the total is the energy of the optimized system; E_1 is the energy of the CsPbBr₃ (100); E_2 is the energy of an optimized NH⁺ and NH₂ molecule within a 10 Å × 10 Å × 10 Å box.

It can be concluded that the binding energy (ΔE) between PyBr and the NPL surface is -3.38 eV, while ΔE between M-phenylenediamine and NPLs is -3.61 eV. The binding energy between DaPyBr and the NPL is $\Delta E = -4.48$ eV.

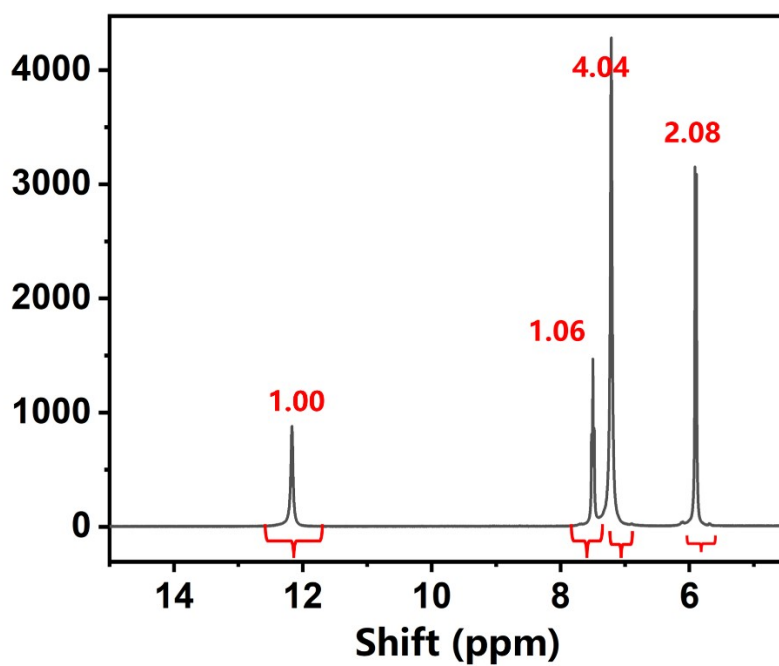


Figure S1. NMR (Nuclear Magnetic Resonance) hydrogen spectrum of DaPyBr

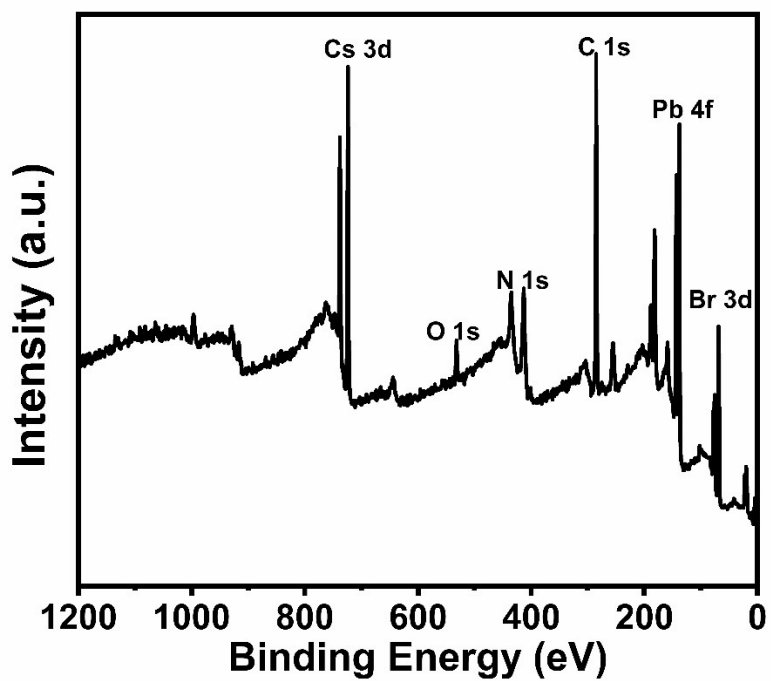


Figure S2. XPS spectrum of the untreated NPLs.

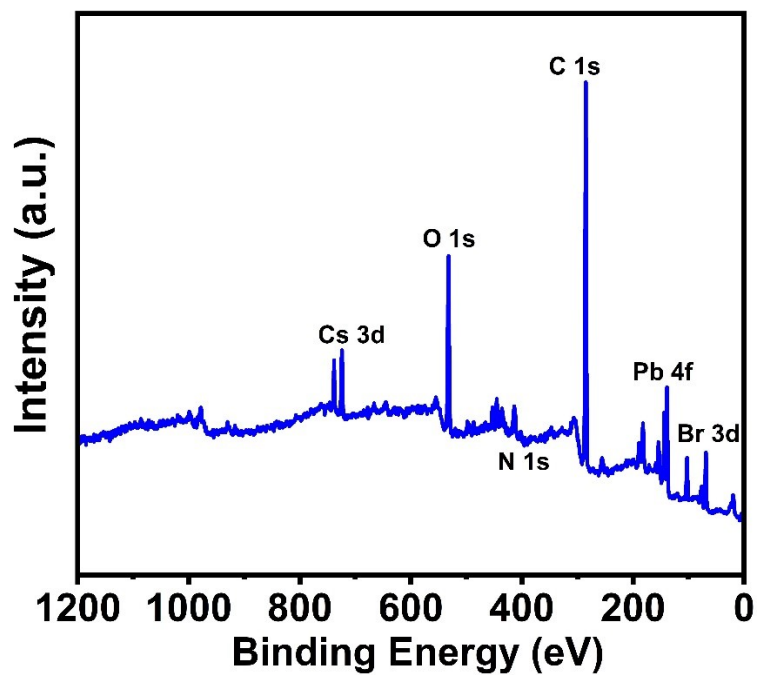


Figure S3. XPS spectrum of DaPyBr-NPLs

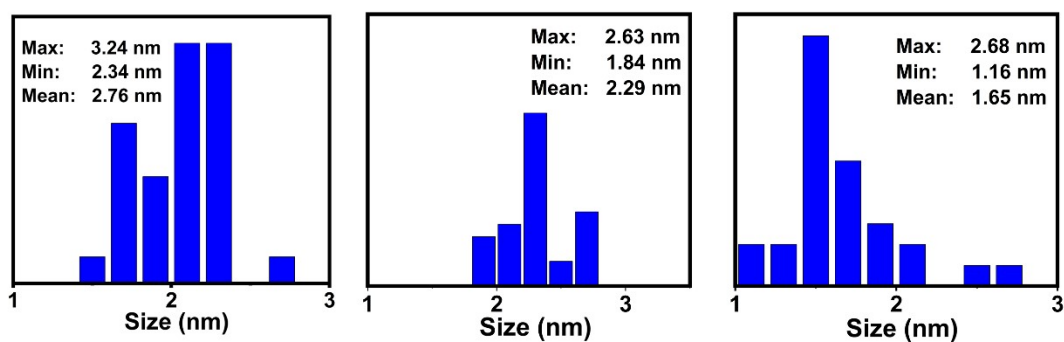


Figure S4. The particle size statistical analysis of the untreated NPLs and DaPyBr-NPLs with different concentrations.

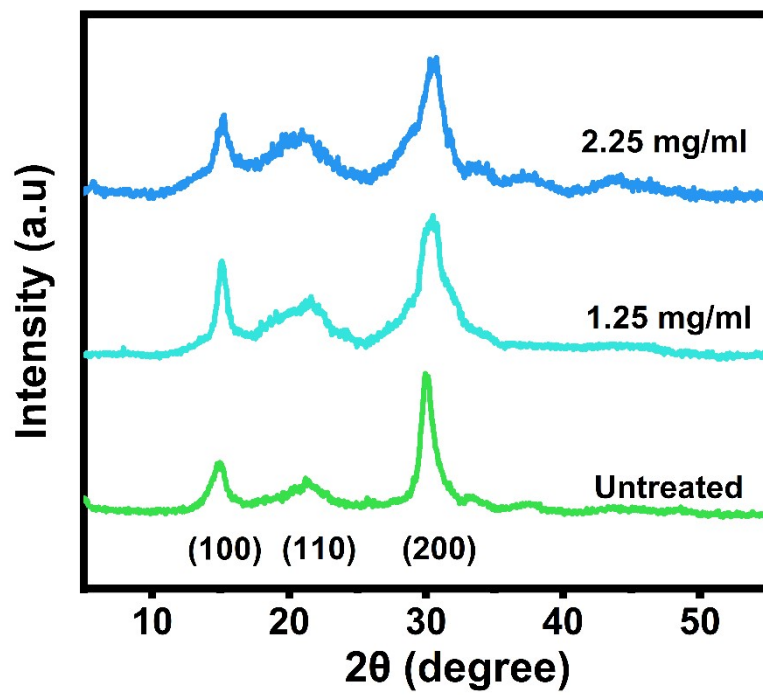


Figure S5. XRD patterns of the untreated CsPbBr₃ NPLs and different concentrations of DaPyBr.

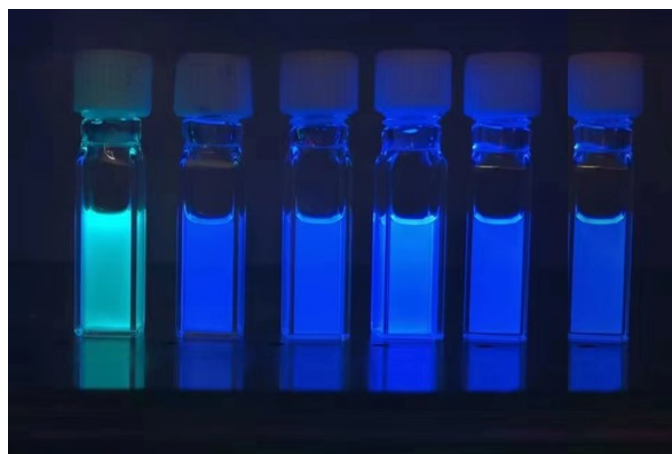
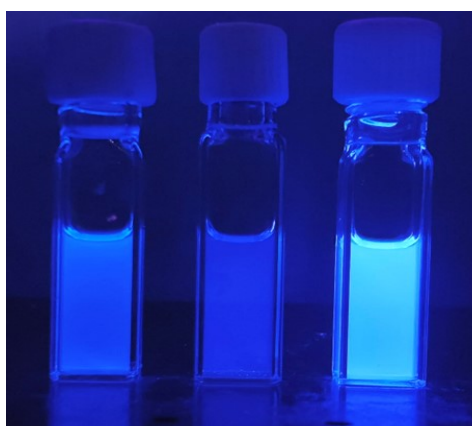


Figure S6. NPLs of different DaPyBr concentrations emission comparison under UV light



control-NPLs PyBr-NPLs DaPyBr-NPLs

Figure S7. The luminescence comparison after 45 days under UV light.

Concentration (mg/ml)	Max Emission Peak (nm)	FWHM (nm)	PLQY (%)	PL lifetime (ns)
0	474	24.97	15.28	3.68
0.75	461	19.79	17.99	3.62
1.25	461	18.86	30.96	3.70
1.75	459	20.35	79.06	4.70
2.25	456	22.17	99.16	7.62
2.75	455	19.70	62.13	4.90

Table S1. A summary of optical performance of DaPyBr-NPLs.

Concentration (mg/ml)	Max Emission Peak (nm)	FWHM (nm)	PLQY (%)
0	474	24.97	15.28
0.75	458	18.14	20.96
1.25	456	18.72	50.25
1.75	453	18.64	50.19
2.25	452	19.45	84.30
2.75	451	20.49	38.39

Table S2. A summary of optical performance of PyBr-NPLs

References

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2. Y. K. Zhang and W. T. Yang, *Phys. Rev. Lett.*, 1998, **80**, 890-890.
3. Vanderbilt, *Physical review. B, Condensed matter*, 1990, **41**, 7892-7895.