Supporting Information

Achieving near-unity photoluminescence quantum yield and high stability in CsPbI₃ nanoplatelets by hydroiodic acid-assisted ligand treatment

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Experimental section

Material

Lead iodide (PbI₂, Bidepharm, 99%), cesium carbonate (Cs₂CO₃, Aladdin, 99.99%), didodecylamine (DDDAm, TCI 97%), oleylamine (OLA, Alfa Aesar, 90%), hydrogen iodide (HI, Sigma-Aldrich), oleic acid (OA, Alfa Aesar,90%),1-octadecene (ODE, Alfa Aesar, 90%),hexane (Aladdin, anhydrous 99%), octane (Aladdin, anhydrous 99%), ethyl acetate (Aladdin, anhydrous 99.8%), toluene, poly (3,4-ethylenedioxythiophene):poly-(styrenesulfonate) (PEDOT:PSS), poly [bis(4-phenyl) (4-butylphenyl)amine] (Poly-TPD), 1,3,5-tris(2-N-phenylbenzimidazolyl)benzene (TPBi), LiF and Al were from Xi'an Yuri Solar Co., Ltd. All chemicals were used directly without further purification.

Synthesis of Cs-OA precursors

Typically, 0.4 g Cs_2CO_3 , 20 mL ODE, and 2 mL OA were loaded into a 50 mL 3-neck flask, and then the mixture was dried at 120 °C under vacuum. After 1 h, the mixture was heated to 150 °C under a flow of nitrogen to ensure more complete dissolution of Cs_2CO_3 . Subsequently, it was allowed to cool down naturally and stored in a 20 mL sample bottle for later synthesis use. The synthetic Cs-OA solution is preheated to 120 °C for the subsequent hot injection step.

Synthesis and purification of CsPbI₃ NPLs

We employed a modified hot-injection method for the synthesize of CsPbI₃ NPLs. Typically, 0.2 mmol PbI₂, 5.0 mL ODE, 0.5 mL OA, and 0.5 mL OLA were added into a 50 mL 3-neck flask, and then the mixture was dried at 120 °C under vacuum. After 30 min, it was heated to a temperature of 90 °C under nitrogen flow. After that, 0.5 mL of Cs-OA solution was quickly injected, and after about 10 s, the mixture solution was transferred into an ice bath.

For the synthesis of treated NPLs, during the synthesis, 70 µL HI was immediately injected into

the mixture solution after the injection of Cs-OA at 90 °C. After 10 s, the DDDAm in toluene was injected into the flask. After 30 s, the mixture solution was transferred into an ice bath.

To purify the NPLs, a TG16-WS tabletop high-speed centrifuge with a rotor size of 3 was used. The crude solution was centrifuged for 5 min at 8000 rpm. The supernatant was discarded carefully, and the precipitate was redispersed in toluene. Ethyl acetate was then added for washing (the volume ratio to NPLs solution is 2:1). The mixture solution was centrifuged for 5 min at 8000 rpm, and the obtained precipitate was redispersed into octane, stored in the sample for subsequent characterization and PeLED application.

Device fabrication

Indium tin oxide (ITO) substrates were successively cleaned with a glass water (2 vol.% Hellmanex III solution), deionized water, acetone, isopropanol, and ethanol at one time. After drying in a vacuum oven, UV-ozone irradiation was applied for 15 min. The hole injection layer materialand, PEDOT:PSS, was then spin-coated onto the ITO glass for 40 s at 5000 rpm, followed by annealing for 15 min at 150 °C in an air environment. Subsequently, the next operations were transferred to the glove box. The poly-TPD was dissolved in a chlorobenzene solution (4 mg/mL) and then spin-coated at 3000 rpm for 40 s as a hole transport layer, followed by annealing for 15 min at 120 °C. The purified NPLs (20 mg/mL) were coated for 30 s at 2000 rpm, and annealed for 10 min at 50 °C. Finally, TPBi, LiF, and Al layers were deposited in a thermal evaporation chamber with a vacuum pressure of $< 4 \times 10^{-4}$ Pa.

Characterization

The UV-visible absorption spectra were measured by a LAMBDA 750 UV spectrophotometer. The PL spectra were measured by a Horiba Jobin-Yvon Fluromax-4 spectrofluorometer. The PL decay curves were collected using a time-correlated single-photon counting (TCSPC) spectrofluorometer equipped with a 368 nm laser. The PL QYs were measured under the excitation at 345 nm on an absolute PL QY system (Hamamatsu Photonics Co., Ltd., C11347) equipped with an integrating sphere. Transmission electron microscope (TEM) was performed on a FEI TECNAI G2 F30 operating at an accelerating voltage of 300 kV. The X-ray diffraction (XRD) patterns were performed on a SMARTLAB3KW X-ray diffractometer equipped with Cu-K α radiation (λ = 1.540 Å). The X-ray photoelectron spectroscopy (XPS) measurements were performed on an ESCALAB 250Xi+ X-ray photoelectron spectrometer. The electroluminescent properties were measured using a computer-controlled programmable power supply (Keithley model 2400) and a commercial system (XPQY-EQE, Guangzhou Xi Pu Optoelectronics Technology Co., Ltd.) in a glove box filled with nitrogen.



Fig. S1.PL spectra of CsPbI₃ NPLs synthesized at different reaction temperatures.



Fig. S2. The relationship between the PL QY of $CsPbI_3$ NPLs and their synthesis temperature.



Fig. S3. PL and absorption spectra of DDDAm ligand-assisted CsPbI₃ NPLs.



Fig.S4. PL spectra of CsPbI₃ NPLs after addition of different amounts of HI.



Fig. S5. The PL QYs and FWHMs of CsPbI₃ NPLs after addition of different amounts of HI.



Fig. S6. PL QYs of treated CsPbI₃ NPLs synthesized in different batches.



Fig. S7. XRD patterns of treated CsPbI₃ NPLs in the 2° - 50° range.

Sample	Diffraction Peaks	d ₀₀₂ [Å]	20[°] (004)	20[°] (006)	20[°] (008)	2θ[°] (0010)	20[°] (0012)	2θ[°] (0014)
Etched CsPbI ₃	n=3	5.13	3.48	5.22	6.96	8.7	10.46	12.2
Treated CsPbI ₃	n=3	5.13	3.48	5.22	6.96	8.7	10.46	12.2

Table S1. XRD diffraction angles for etched and treated $CsPbI_3 NPLs$.

Sample	$A_1[\%]$	$\tau_1 [ns]$	A ₂ [%]	$\tau_2 [ns]$	$\tau_{avg} \left[ns \right]$	k _{nr} [ns ⁻¹]
Pristine CsPbI ₃	52	7.82	48	26.8	22.22	0.0387
Etched CsPbI ₃	43	6.57	57	16.4	14.08	0.037
Treated CsPbI ₃	48	6.4	52	20.26	17.08	0.029

Table S2. PL lifetimes fitting parameters of pristine, etched, and treated CsPbI₃ NPLs.



Fig. S8. Temperature-dependent PL spectra of pristine CsPbI₃ NPLs (a) and treated CsPbI₃ NPLs (b) in the temperature range of 80 to 300 K, excited at a wavelength 345 nm.



Fig. S9. PL QYs variation of treated CsPbI₃ NPLs over a period of 87 days.



Fig. S10. The PL spectra variation of pristine CsPbI₃ NPLs film (a) and treated CsPbI₃ NPLs film (b) after a week of storage in ambient conditions.



Fig S11. Evolution of 2D pseudo color PL spectra of pristine and treated NPLs under UV irradiation at 100 mW cm⁻² power (a, b) and 500 mW cm⁻² power (c, d).

Sample	PL [nm]	PLQY [%]	FWHM [nm]	Stability	Ref.
CsPbI3	625	90	34	PL QY retains 80% on the 8 th day	1
CsPbI3	596	84.6	28	not mention	2
CsPbI3	626	99.4	28	not mention	2
CsPbBr ₃	460	96	12	PL maintained for 400 min under UV illumination	3
CsPbBr ₃	463	92	16	PL maintained for 30 h under UV irradiation	4
CsPbI3	600	95	22	PL QY retains 61.8% on the 87 th day XRD remains unchanged over a 21- day period	This work

Table S3. Optical characterization of CsPbX₃ NPLs with quantum confinement effect published to date.



Fig. S12. CIE coordinate of a PeLED prepared based on treated $CsPbI_3 NPLs_1$



Fig. S13. EQE of treated CsPbI₃ NPL PeLED as a function of luminance.



Fig. S14. The luminous efficiency of treated CsPbI₃ NPL PeLED as a function of current density.

Material	EL peak [nm]	Luminance [cd m ⁻²]	EQE [%]	Ref.
CsPbI ₃	636	100	5.3	1
CsPbBr ₃	464	40	0.3	5
CsPbBr ₃	460	158	3.18	6
CsPbBr ₃	463	141	2	7
CsPbI ₃	600	472.85	2.98	This work

Table S4. Summary of the EL performance of $CsPbX_3$ NPLs PeLEDs with strong quantum confinement effect released to date.

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