

Supporting Information

CsZnPbBr₃/ZnS Core/Shell Perovskite Nanocrystals for Stable and Efficient White Light-emitting Diodes

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Experimental section

Materials

PbBr₂ (98%), zinc acetate (99%), oleic acid (OA, 90%), oleylamine (OAm, 98%), and hydrobromic acid (HBr, 48% aqueous solution) were purchased from Aldrich. Cesium carbonate (Cs₂CO₃, >98%) was purchased from Aladdin. Sulfur (99.5) and 1-octadecene (ODE, 98%) were purchased from Alfa Aesar. Ethyl acetate, ethanol, hexane, and diethyl ether were purchased from Beijing Chemical Reagent Co., Ltd. All chemicals were used as received without further purification.

Synthesis of cesium oleate (CsOA)

The CsOA stock solution was formulated by adding 0.188 g Cs₂CO₃ and 1.5 mL OA in 10 mL ODE. The resultant mixture underwent initial degassing under vacuum at 120 °C to eliminate moisture, followed by additional heating to 150 °C. Subsequently, the solution was transferred and stored in a vial for subsequent utilization.

Synthesis of zinc oleate (Zn(OA)₂)

In a 25 mL three-neck flask, 1.2 mmol Zn(Ac)₂, 1 mL ODE, and 2.64 mL OA were mixed and heated to 120 °C under vacuum for 1 hour. Subsequently, the mixture was heated to 150 °C under a nitrogen atmosphere and maintained for 2 hours until a clear solution was obtained.

Synthesis of oleylammonium bromide (OAmBr)

100 mL ethanol and 12.5 mL OAm were mixed in a 250 mL flask. The mixture was stirred and cooled in an ice-water bath, and 8.56 mL HBr solution was added dropwise to generate OAmBr. The reaction mixture was stirred for 12 hours under a nitrogen atmosphere, and the solution was then subjected to rotary evaporation for drying. The resulting product was purified with diethyl ether through three cycles. The white OAmBr powder was obtained after vacuum drying at room temperature.

Synthesis of CsPbBr₃ PeNCs

In a 25 mL three-neck flask, 0.044 g PbBr₂, 0.1394 g OAmBr, and 10 mL ODE were loaded. The mixture was heated to 120 °C with stirring under a nitrogen flow and held for 10 minutes to remove water. Subsequently, 0.5 mL OA and 0.5 mL OAm were added. Once PbBr₂ was completely dissolved, the reaction temperature was raised to 180 °C. 1 mL CsOA was quickly injected, and the reaction was quenched immediately by using an ice-water bath. The product was purified twice with ethyl acetate/hexane.

Synthesis of CsZnPbBr₃/ZnS core/shell PeNCs

0.044 g PbBr₂, 0.1394 g OAmBr, 0.48 mL Zn(OA)₂ and 10 mL ODE were loaded into a 25 mL three-neck flask. The mixture was heated to 120 °C with stirring under a nitrogen flow and held for 10 minutes to remove water. Subsequently, 0.5 mL OA and 0.5 mL OAm were added. When PbBr₂ was completely dissolved, the reaction temperature was raised to 180 °C and then 1 mL CsOA was injected. After 10 s, a mixture of Zn(OA)₂ 0.67 mL OAm, 1 mL ODE and 0.6 mmol S powder was dropwise added. The core/shell PeNCs were purified twice with ethyl acetate/hexane.

Characterization

Transmission electron microscopy (TEM) was performed on JEM1011 (JEOL) electron microscopes at an operating voltage of 100 kV. High resolution TEM (HRTEM) and

energy dispersive spectrometer (EDS) elemental mapping were performed on a Tecnai G2 (FEI, USA) electron microscopes with an operating voltage of 200 kV. A DX-2700BH diffractometer (Dandong Haoyuan Instrument Co. Ltd.) with a Cu $K\alpha 1$ radiation ($K\alpha 1 = 1.5406 \text{ \AA}$) was utilized to measure the structure of PeNCs. Ultraviolet–visible (UV–vis) absorption spectroscopy was performed with a U-3100 spectrophotometer (Hitachi). The PL spectra of the samples were measured with a high-resolution spectrometer (wavelength resolution of 0.75 nm, Ocean Optics, USA) and F-2500 Fluorescent spectrophotometer (Hitachi). Photoluminescence quantum yield was measured by Edinburgh fluorescence spectrometer FS5. Time-resolved PL measurements were collected by fluorescence lifetime measurement system (QM/TM/NIR, PTI, America). The elemental analysis was studied by the XPS spectrum utilizing a Perkin-Elmer model PHI 5600.

Temperature cycling test:

Both CsPbBr_3 and $\text{CsZnPbBr}_3/\text{ZnS}$ PeNCs films were placed on a heating platform initially set to 300 K and left for 3 minutes. Subsequently, they were heated in increments of 30 K and maintained at each temperature for 3 minutes, during which their PL intensity were tested. Upon reaching 450 K, the heating platform was cooled in steps of 30 K, and their PL intensity were simultaneously tested. This process of heating and cooling was repeated three times.

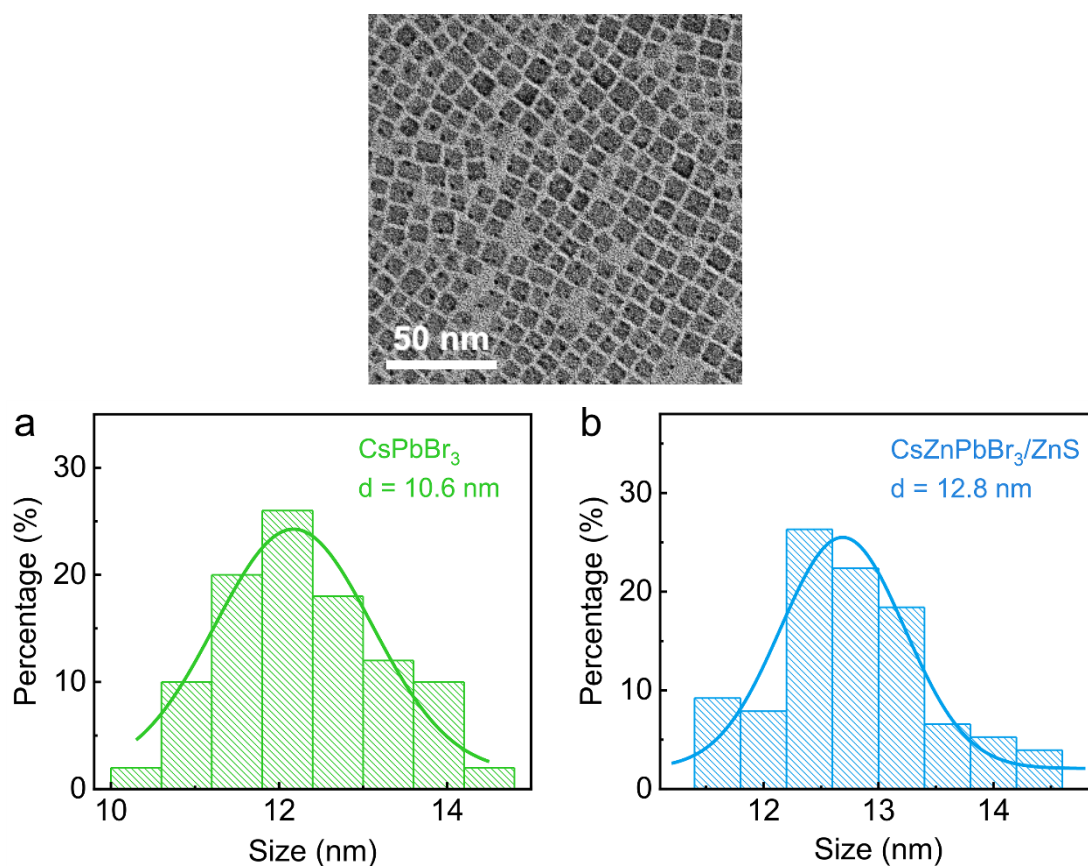


Figure S1. Size distribution histograms of (a) pristine CsPbBr_3 and (b) $\text{CsZnPbBr}_3/\text{ZnS}$ PeNCs.

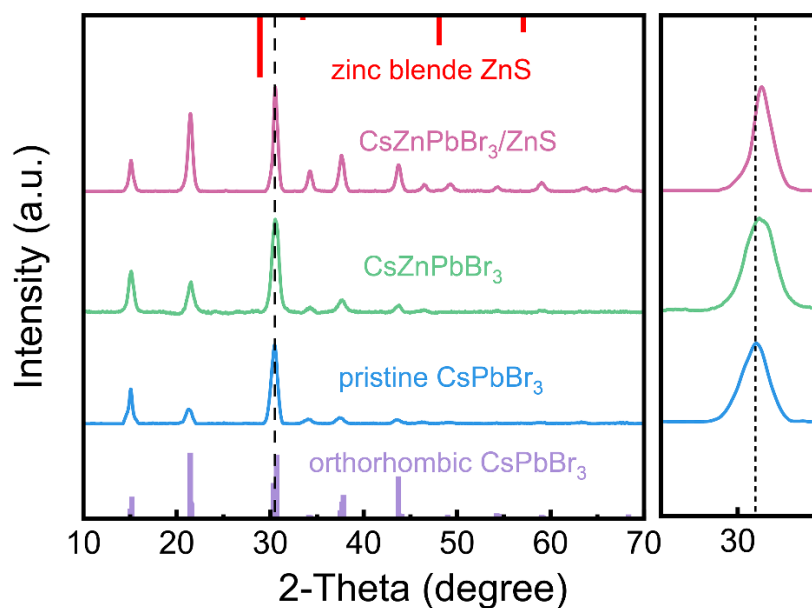


Figure S2. XRD patterns of pristine CsPbBr₃, CsZnPbBr₃ and CsZnPbBr₃/ZnS PeNCs.

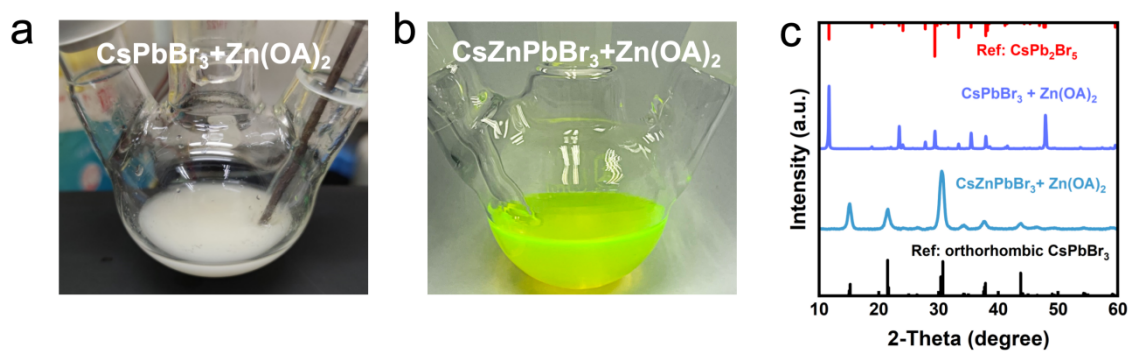


Figure S3. Pictures of (a) pristine CsPbBr₃ PeNCs and (b) CsZnPbBr₃ PeNCs solutions after adding Zn precursor at 180 °C, and (c) the corresponding XRD patterns.

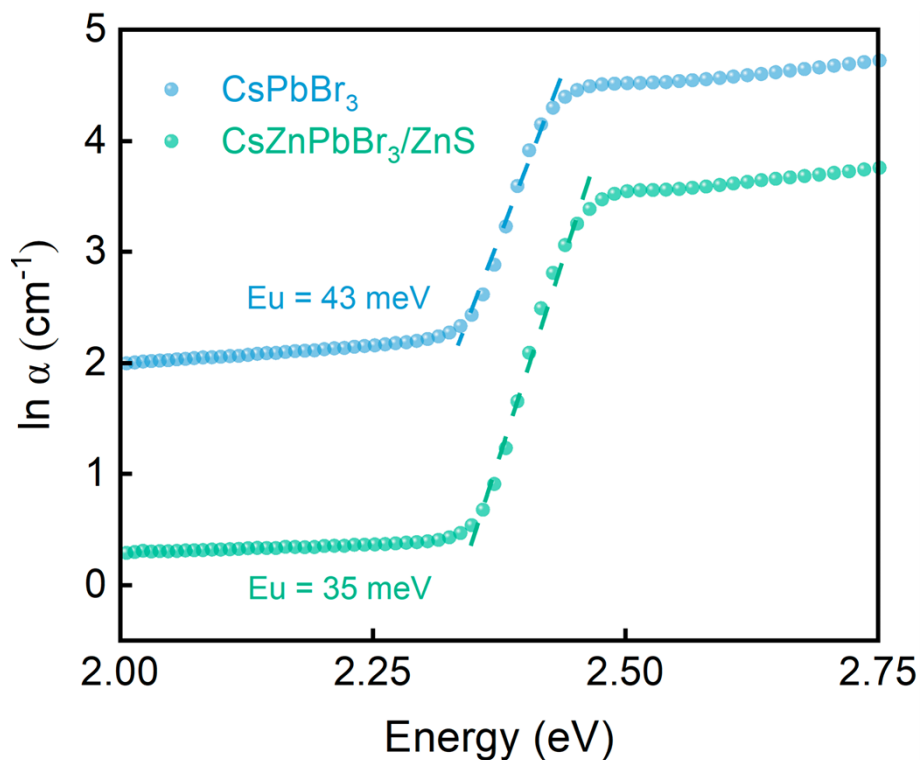


Figure S4. Urbach Energy of pristine CsPbBr_3 , CsZnPbBr_3 and $\text{CsZnPbBr}_3/\text{ZnS}$ PeNCs.

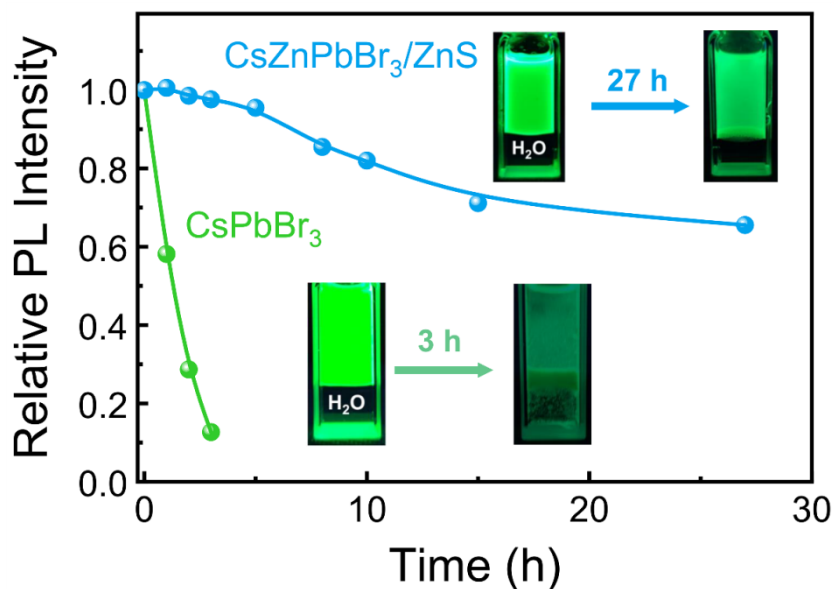


Figure S5. Relative PL intensity values of CsPbBr_3 and $\text{CsZnPbBr}_3/\text{ZnS}$ PeNCs as a function of time in a hexane/water mixture. Inset shows digital photographs of the samples under UV light (365 nm).

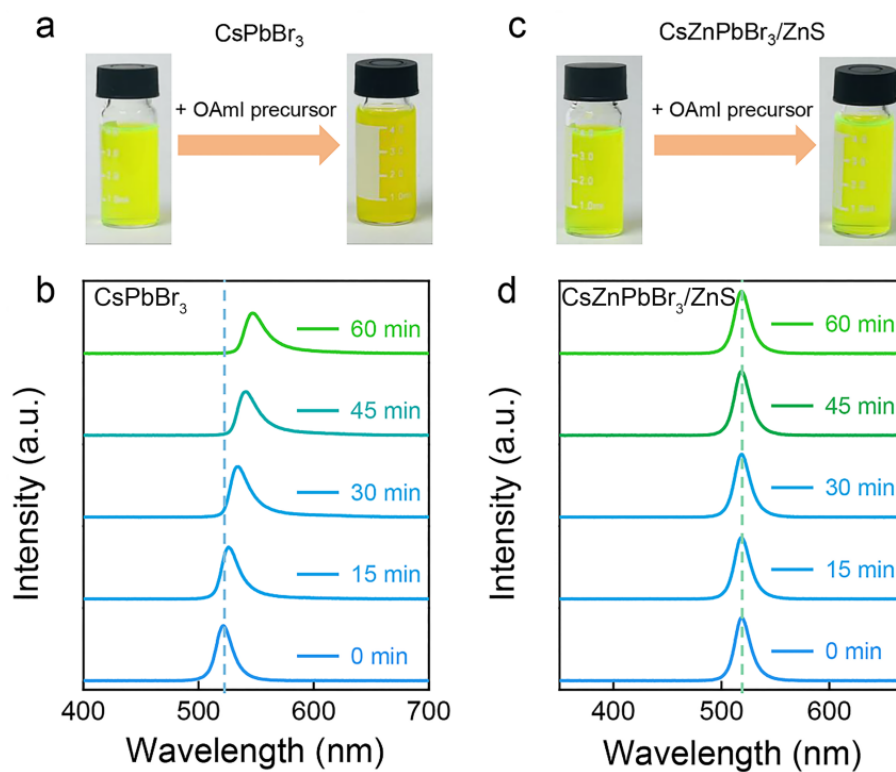


Figure S6. Physical images of (a) pristine CsPbBr₃ and (c) CsZnPbBr₃/ZnS core/shell PeNCs solution after adding OAmI. (b, d) the corresponding evolution of PL spectra, respectively.

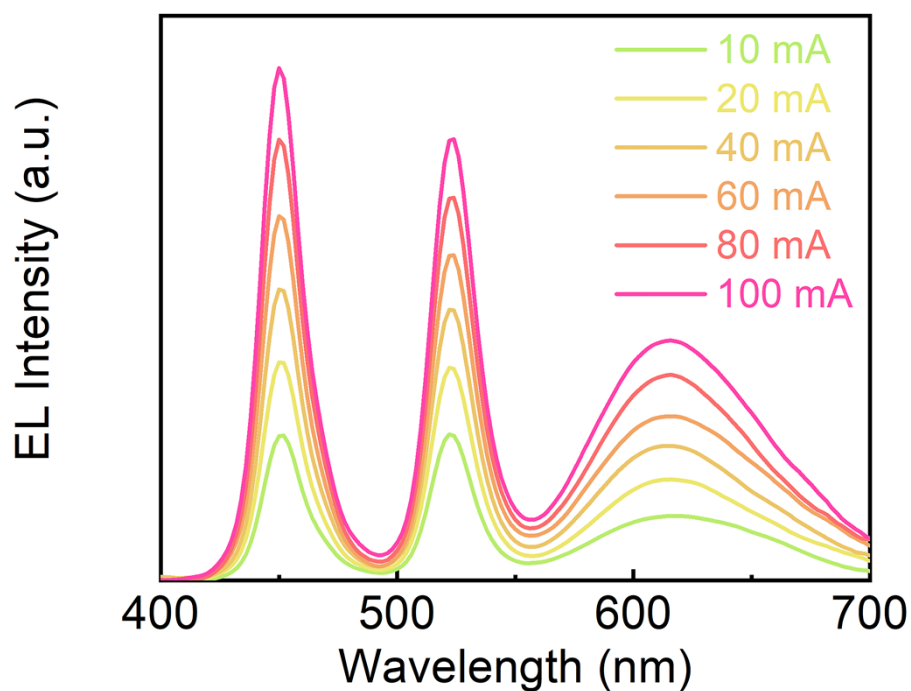


Figure S7. EL spectra of the WLED device under different driving currents ranging from 10 to 100 mA.

Table S1. Detailed information of time resolved PL decay of pristine CsPbBr₃, CsZnPbBr₃ and CsZnPbBr₃/ZnS PeNCs.

Sample	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)	τ_{ave} (ns)	R^2
CsPbBr ₃	14.97	51.64	52.07	48.36	32.91	0.9996
CsZnPbBr ₃	20.12	30.42	74.58	69.58	58.01	0.9995
CsZnPbBr ₃ /ZnS	24.52	23.11	92.62	76.89	76.88	0.9995