Supporting Information

Kinetically Regulated Growth of $CsPbBr_3$/CdSe Core/Shell Quantum Dots for Stable Light-Emitting Diodes

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1. Experimental Section

1.1 Materials

1-Octadecene (ODE, 90%) and Cs_2CO_3 (99.9%) were purchased from Alfa Aesar. Oleic acid (OA, 90%), oleylamine (OAm, 90%), and selenium (Se, 99.5%) were purchased from Sigma-Aldrich (Merck). Cadmium oxide (CdO, 99.99%) and lead acetate (Pb $(CH_3CO_2)_2 \cdot 3H_2O$, 99%) were purchased from Aladdin. Tri-n-octylphosphine (TOP, $C_{24}H_{51}P$, 90%), n-Hexane (C_6H_{14}, AR), and ethanol (C_2H_5OH, 99.8%) were purchased from Innochem.

1.2 Preparation of precursor solutions

Synthesis of the Cs-OA precursor, 0.4888 g (1.5 mmol) of Cs_2CO_3, 26 mL of ODE, and 4 mL of OA were added to a 50 mL three-neck flask. The mixture was degassed under vacuum with stirring and heated to 120 ^{o}C, maintained at this temperature for 10 minutes to remove residual water and oxygen. The temperature was then raised to 150 oC until the cesium carbonate powder was completely dissolved.

Synthesis of the Pb-OA precursor, 1.143 g (3 mmol) of lead acetate, 26 mL of ODE, and 4 mL of OA were added to a 50 mL three-neck flask. The mixture was heated to 120 ^{o}C under vacuum conditions for 30 minutes. The temperature was then raised to 150 ^{o}C until the lead acetate powder was completely dissolved.

Synthesis of the TOP-Se precursor, Selenium powder (4 mmol) was dissolved in 10 mL TOP to obtain a clear solution in a nitrogen-filled glovebox.

Synthesis of the $Cd(OA)_2$ precursor, 0.384 g cadmium oxide, 26 mL of ODE, and 4 mL OA were added to a 50 mL three-neck flask. The mixture was degassed under vacuum with stirring and heated to 120 ^{o}C, maintained at this temperature for 10 minutes to remove residual water and oxygen. Subsequently, under a nitrogen atmosphere, the temperature was raised to 240 ^{o}C and held until the cadmium oxide powder was completely dissolved.

1.3 Synthesis of $CsPbBr_3$/CdSe Pe-QDs

10 mL of ODE, 0.6 mmol of OAmBr, and 1 mL of Cs-OA were added to a three-neck flask. The mixture was then vacuumed at 120 ^{o}C for 1 h. At this temperature, 0.5

mL of OAm and 0.5 mL of OA were injected. The mixture was heated while being constantly stirred until all the reactants completely dissolved, producing a clear solution. Subsequently, the reaction mixture was heated to 180 ^oC. Then, 1 mL of Pb-OA and 0.25 mL of CdSe precursor were rapidly injected. After 5 min of reaction, the flask was immediately immersed in an ice-water bath to quench the reaction, allowing the product to cool down to room temperature.

Purification of $CsPbBr_3$/CdSe Pe-QDs, take 10 mL of the as-synthesized QDs solution and add 20 mL of ethyl acetate, then centrifuge at 7000 rpm for 3 min. Discard the supernatant and redisperse the precipitate in 5 mL of n-hexane. Repeat the process by adding 15 mL of ethyl acetate and centrifuging again. Finally, discard the supernatant and redisperse the final precipitate in 5 mL of n-hexane.

1.4 LED fabrication

Patterned indium tin oxide (ITO)-coated glasses were cleaned sequentially using soap, deionized water, ethanol, dichloromethane, isopropanol, and UV-ozone treatment for 15 min. PEDOT:PSS:KBr (2 mg/mL) solution was spin-coated onto the ITO-coated glass substrates at 4000 rpm for 40 s, followed by annealing in air at 150 ^oC for 15 min to form the hole-transporting layer. The substrates were then transferred into a N_2-filled glovebox, and PTAA (8 mg mL^{-1} chlorobenzene solution) was spin-coated on top of the PEDOT: PSS layer at 4000 rpm for 40 s and annealed at 120 ^oC for 20 min. Pe-QDs (8 mg/mL) were deposited by spin-coating at 2500 rpm for 30 s. An electron-transporting layer of TPBi (40 nm) and LiF/Al electrodes (1 nm/100 nm, respectively) were sequentially deposited by thermal evaporation in a vacuum deposition chamber (10^{-4} Pa pressure).

1.5 Characterization

The X-ray diffraction (XRD) patterns were obtained by using an 18-kW diffractometer with radiation. Transmission electron microscopy (TEM) and the high-resolution TEM (HRTEM) images were obtained using a JEOL JEM-2100 microscope operated at 200 kV. UV-vis absorption spectra were recorded using a Shimadzu UV-2600 spectrophotometer. X-ray photoelectron spectroscopy (XPS) was characterized by using a Thermo Scientific Escalab 250Xi. Photoluminescence (PL) and timeresolved PL (TRPL) spectra were collected with a Horiba spectrometer. A WITec alpha 300R confocal Raman microscopy was used to examine the Raman spectra of Pe-QDs. Electroluminescence (EL) spectroscopy was performed under forward bias conditions (0-10 V scanning, 0.5 V steps), current density-voltage (J-V), luminance-voltage (L-V), and EQE-J characteristics were conducted using a 50 mm-diameter EL-specialized integrating sphere (IS-E50) from Puyan Interconnect, coupled with a Keithley 2400 source meter. The device lifetime tests of PeLEDs were performed on a ZJZCL-1OLED aging lifespan test instrument without further packaging in a N_2 glovebox with O_2 and water below 0.1 ppm.

2. Supplementary Figures

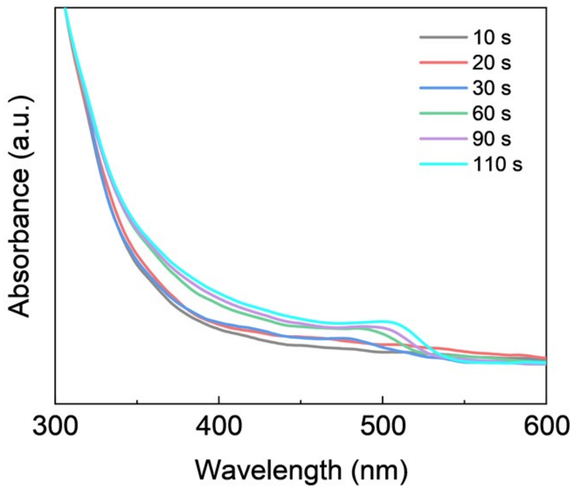

Fig. S1. Absorption of CdSe precursor at different stirring times at 180 °C.

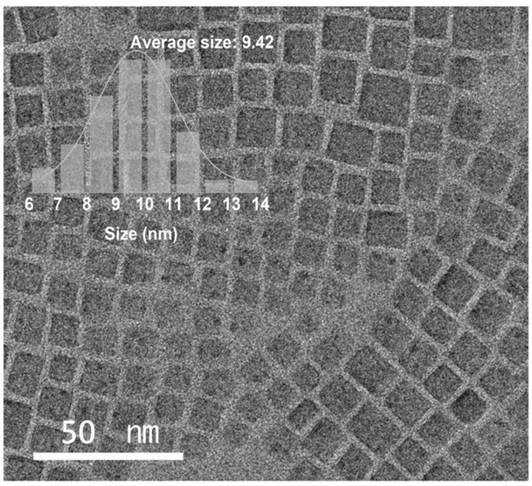

Fig. S2. TEM images of $CsPbBr_3$ Pe-QDs.

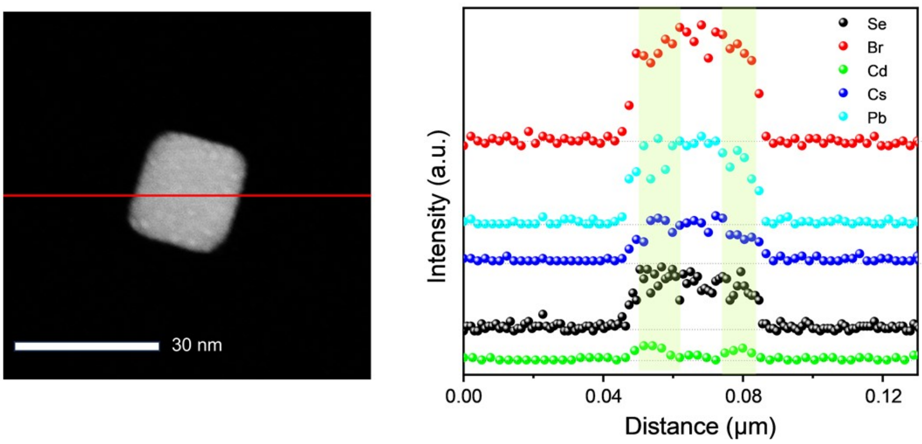

Fig. S3. EDS line scan of $CsPbBr_3$/CdSe Pe-QDs.

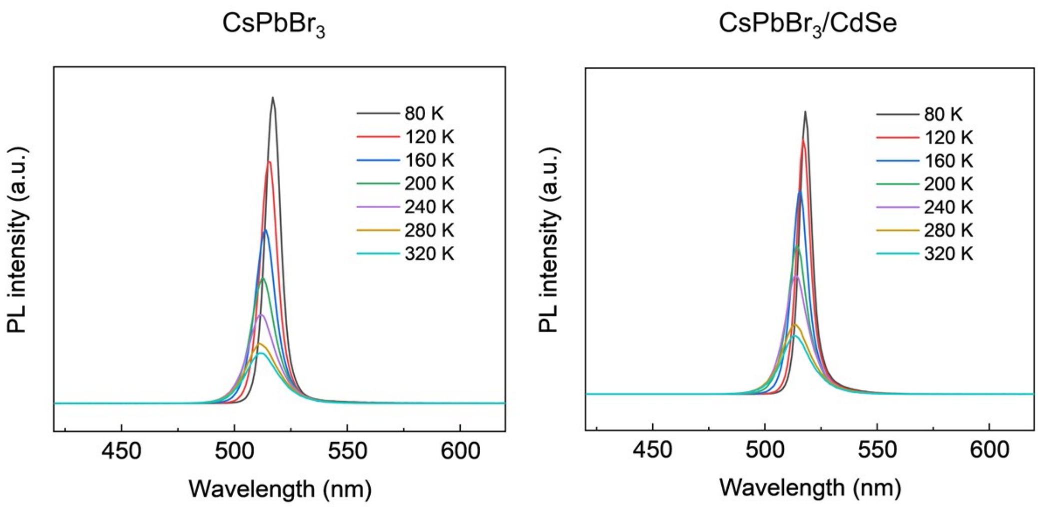

Fig. S4. Temperature-dependent fluorescence PL spectra of $CsPbBr_3$ Pe-QDs and $CsPbBr_3$/CdSe C/S Pe-QDs.

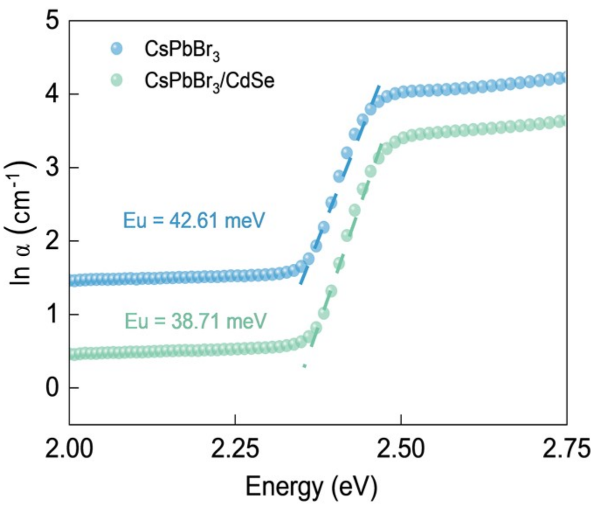

Fig. S5. Urbach energies.

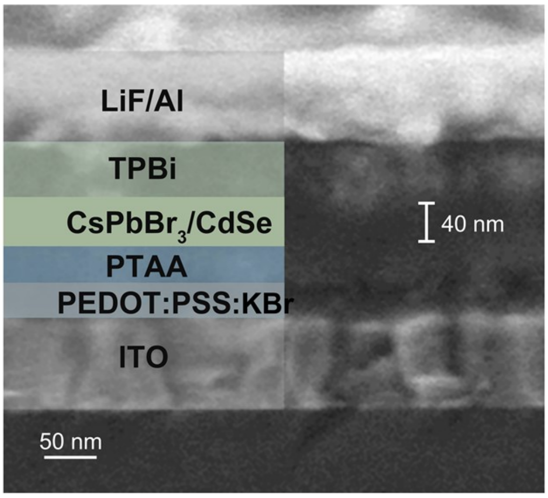

Fig. S6. SEM image of the cross-section of a $CsPbBr_3$/CdSe PeLED.

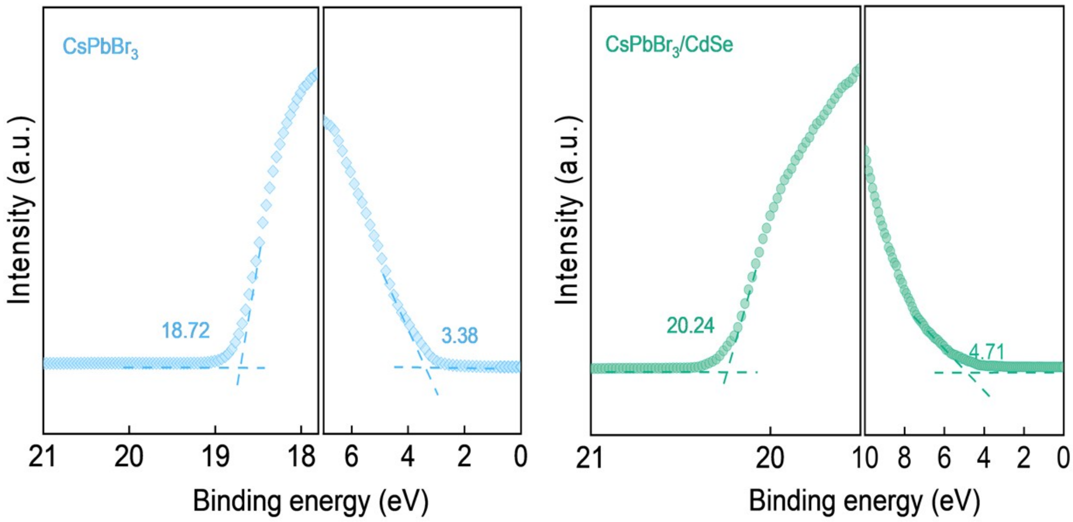

Fig. S7. UPS data of pristine and $CsPbBr_3$/CdSe Pe-QDs.

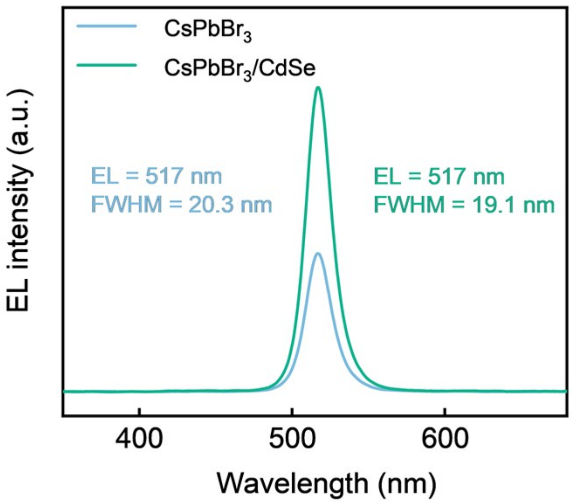

Fig. S8. EL spectra of $CsPbBr_3$ and $CsPbBr_3$/CdSe PeLEDs.

Table S1. PL lifetimes of the $CsPbBr_3$ and $CsPbBr_3/CdSe$, according to tri-exponential fits.

	A_1	τ_1	A_2	τ_2	A_3	τ_3	τ_{avg} (ns)
$CsPbBr_3$	0.26	10.87	0.5	3.7	0.02	58.4	18.1
$CsPbBr_3/CdSe$	0.3	15.7	0.5	4.6	0.04	88.5	37.7