Supporting Information

Ligand-Engineered Aqueous Synthesis of Stable Perovskite Phosphors at Room Temperature

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Experimental section

Materials

Lead bromide ($PbBr_2$, 99% Macklin), Cesium bromide (CsBr, 99.5%, Aladdin), 2-Methylimidazole (2-MIM, 98% Macklin), Zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H_2O$, analytically pure (AR), KESHI), N,N-dimethylformamide (DMF, 99.5%, Aladdin), Sodium bromide(NaBr, AR, Sinopharm Chemical Reagent Co.,Ltd), Didodecyldimethylammonium bromide (DDAB, 98.%, Aladdin), Sodium dodecylbenzenesulfonate (SDBS, CP, Kermel), Tributylphosphine oxide (TBPO, 98%, Macklin). All reagents were used as received without further purification.

Synthesis of mesoporous ZIF-8: Hierarchical mesoporous ZIF-8 was synthesized by dissolving $Zn(NO_3)_2 \cdot 6H_2O$ (0.67 g, 2.25mM) and 2-methylimidazole (0.167g, 2.03mM) in 50 mL of N,N-dimethylformamide (DMF) under vigorous stirring, followed by the addition of sodium dodecylbenzenesulfonate (SDBS, 348.48 g/mol) as a mesoporous template. The homogeneous mixture was transferred into a Teflon-lined autoclave and hydrothermally treated at 140°C for 24 h. After cooling to room temperature, the white precipitate was collected via filtration, washed repeatedly with ethanol, and further immersed in ethanol for 48 h (ethanol refreshed every 12 h) to remove residual templates. The purified product was vacuum-dried at 150°C for 12 h and stored in a desiccator under inert conditions.

Synthesis of perovskite phosphors: 1 mM $PbBr_2$ (0.367 g) was dissolved in the NaBr aqueous solution (0.04 M, 10 mL) under magnetic stirring for 1 h and ultrasonication for 15 min to form a homogeneous $PbBr_2$/NaBr solution. Meanwhile,

mesoporous ZIF-8 (0.459 g) was ultrasonically dispersed in a 1.5 mM CsBr (0.32 g, 10 mL) aqueous solution for 20 min to obtain a ZIF-8/CsBr suspension. 2.5 mL of $PbBr_2$/NaBr solution was then incrementally added to the ZIF-8/CsBr suspension. Finally, rapid antisolvent crystallization was triggered by injecting 25 mL of ethanol into the above perovskite precursors, followed by 24 h of aging under stirring. The resulting yellow powders were collected via centrifugation and vacuum-dried. The pristine sample was named CPB.

For DDAB ligand and TBPO-modified perovskite phosphors (named CPB-DDAB and CPB-TBPO), 0.1 mL of DDAB (0.15 M) or 0.2 mL of TBPO ethanol solution (0.2 M) was introduced into the $PbBr_2$/NaBr solution. Then, 2.5 mL of $PbBr_2$/NaBr-DDAB or $PbBr_2$/NaBr-TBPO solution was incrementally added to the ZIF-8/CsBr suspension. Other synthesized processes were the same as the pristine CPB.

The light curing adhesive used is Norland-61. Simply put, 10 mg of phosphors are mixed with nitride system red phosphor and 200 mg UV-curable optical adhesive. To remove the bubbles in the optical adhesive, the resulting mixture is heated at 40 °C for 0.5 h. After that, the mixture was deposited on a 460 nm blue LED chip and then UV cured for 50 s (365 nm, 80 W cm^{-2}). White LED was synthesized successfully.

Characterization

The composition and phase structure of the catalyst were confirmed using X-ray diffraction (XRD, D8 Advance) in the 2θ range of 5–60º at a scan rate of 5º min^{-1}.The microstructure and elemental distribution of the samples were analyzed by scanning electron microscopy (SEM, Apreo S HiVac). Transmission electron microscopy (TEM,

FEI TalosF200S) was utilized to investigate the morphological structure, as well as analyze the content and distribution through methods such as element mapping, element linear scanning, and point scanning. The PL spectra and PLQYs were carried out on a Hamamatsu(C11347) absolute PLQY spectrometer by photoexcitation of octane suspension in a quartz colorimetric dish at a wavelength of 365 nm. Using Nicolet iS50 FT-IR with KBr pellet acquired the FTIR spectra. X-ray photoelectron specrtroscopy (XPS, ESCALAB XI+) was performed to examine the surface elemental composition and valence of the surface element and the C1s value was set at 284.8 eV for charge corrections.

Figures and Tables

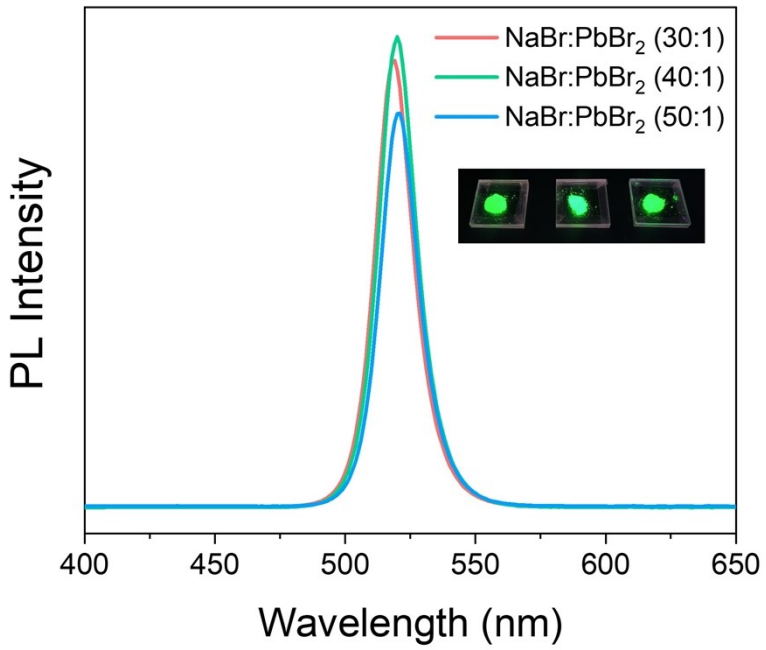

Figure S1. PL spectra of CPB-DDAB phosphors with various ratios of NaBr.

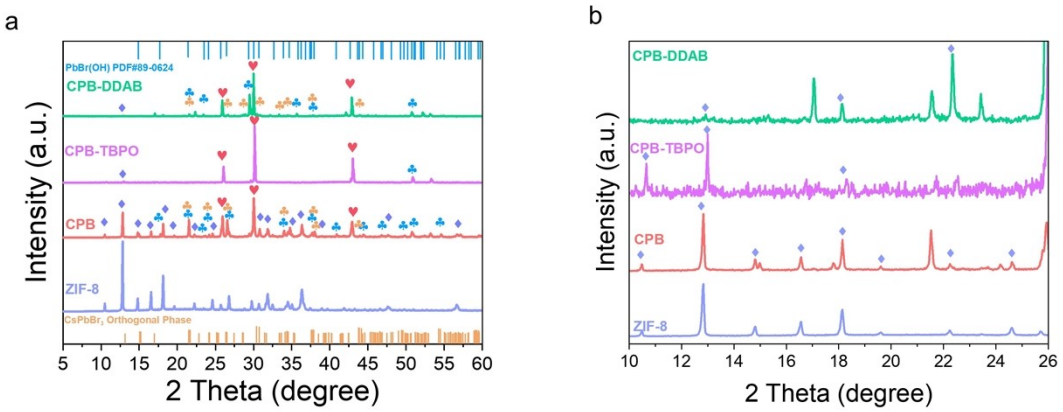

Figure S2. Powder XRD patterns of ZIF-8, CPB, CPB-TBPO and CPB-DDAB.

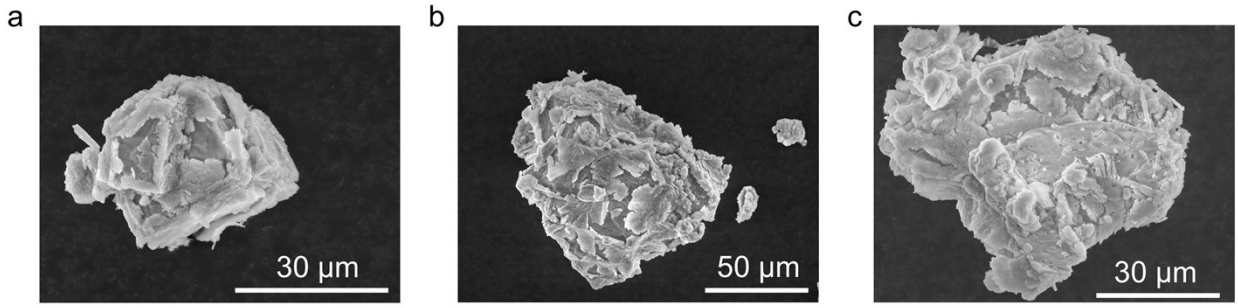

Figure S3. SEM image of (a) CPB, (b) CPB-TBPO, and CPB-DDAB phosphors.

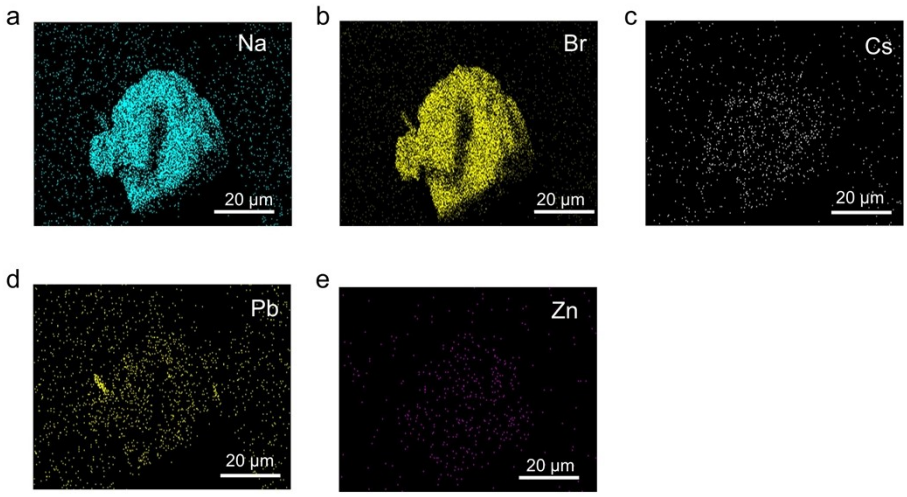

Figure S4. Elemental mapping of Na, Br, Cs, Pb and Zn of CPB.

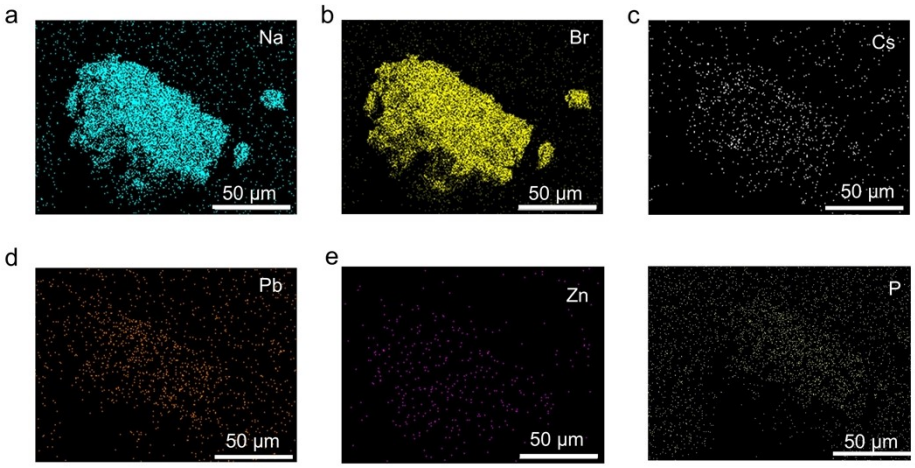

Figure S5. Elemental mapping of Na, Br, Cs, Pb, Zn, and P of CPB-TBPO.

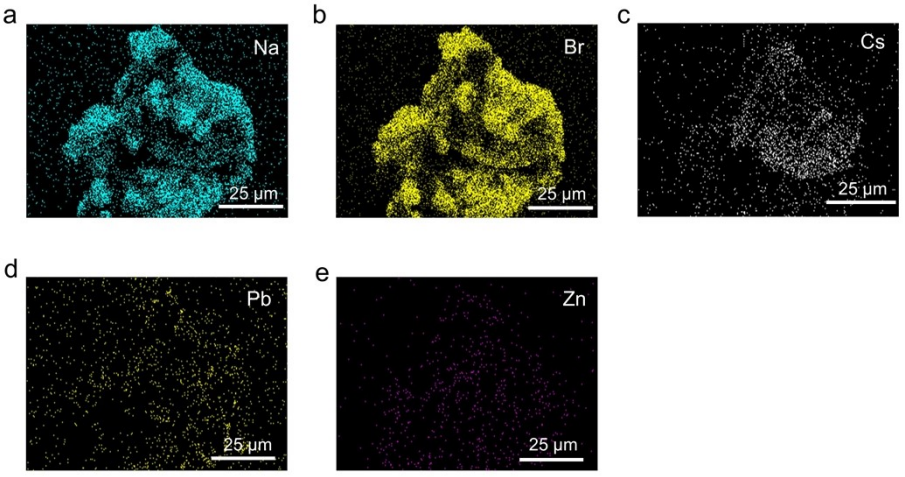

Figure S6. Elemental mapping of Na, Br, Cs, Pb, and Zn of CPB-DDAB.

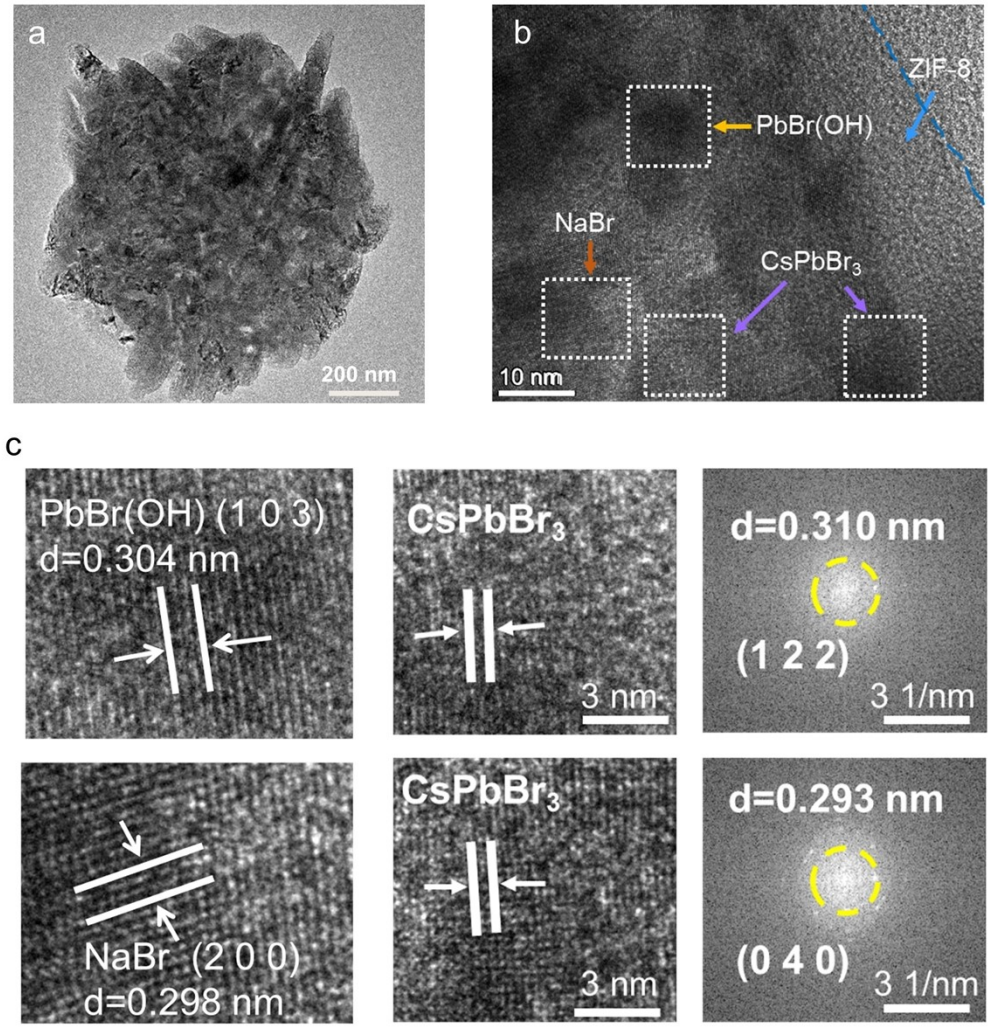

Figure S7. (a) A TEM image, (b) and (c) high-resolution TEM images and FFT patterns of CPB-DDAB.

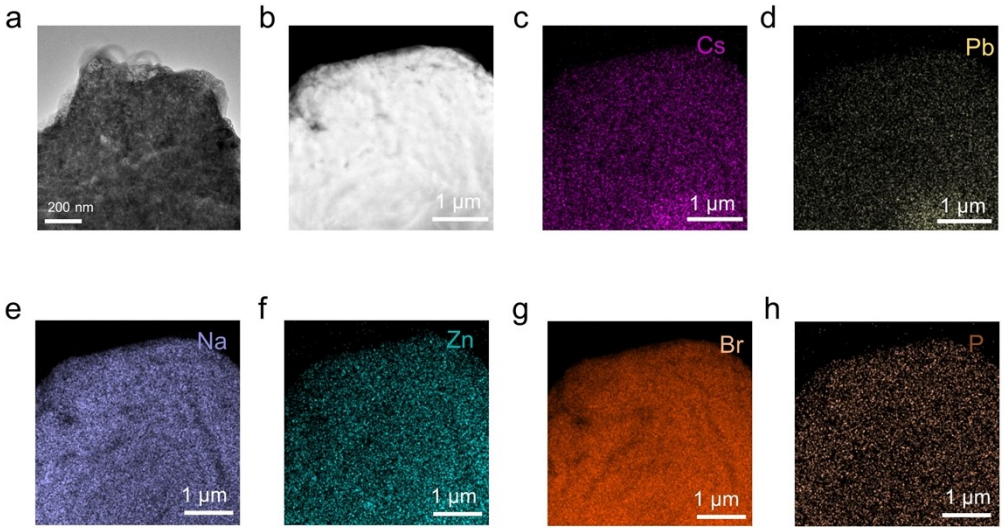

Figure S8. EDS elemental distribution mappings of CPB-TBPO.

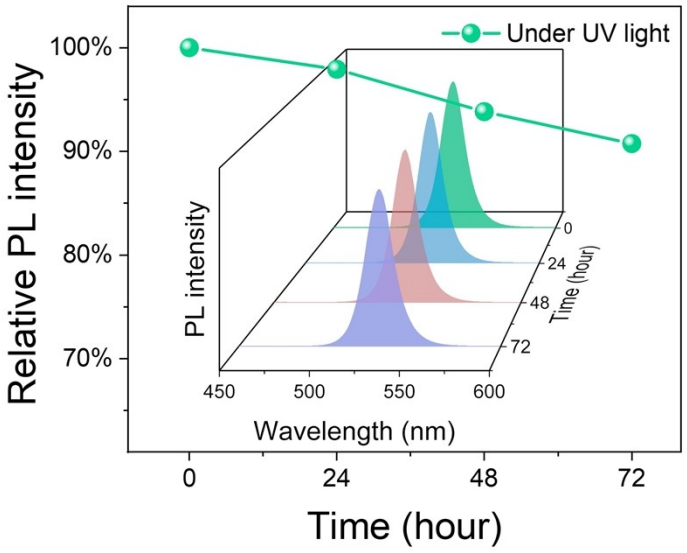

Figure S9. Photostability of perovskite phosphors under UV light.

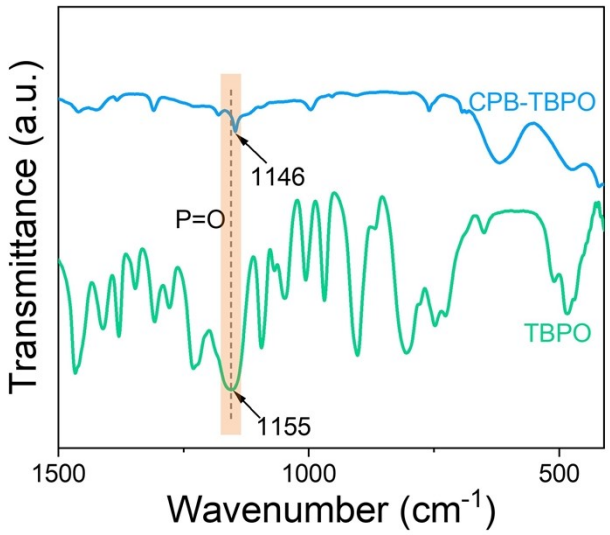

Figure S10. Infrared spectra of TBPO and CPB-TBPO.

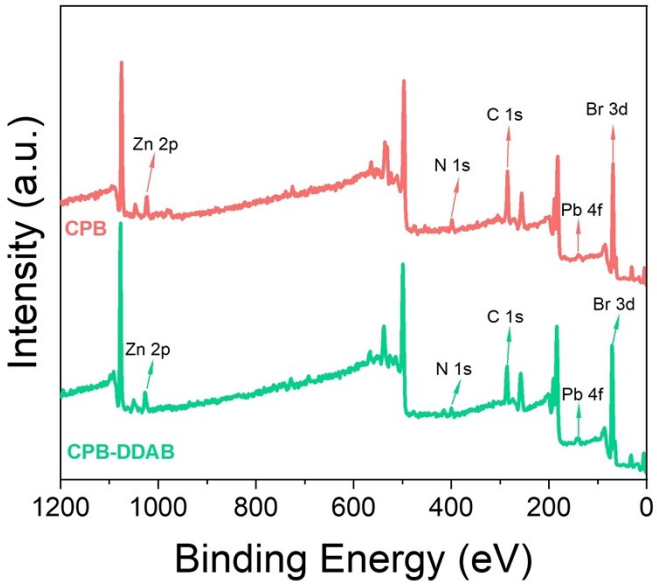

Figure S11. XPS survey spectrum of perovskite phosphors.

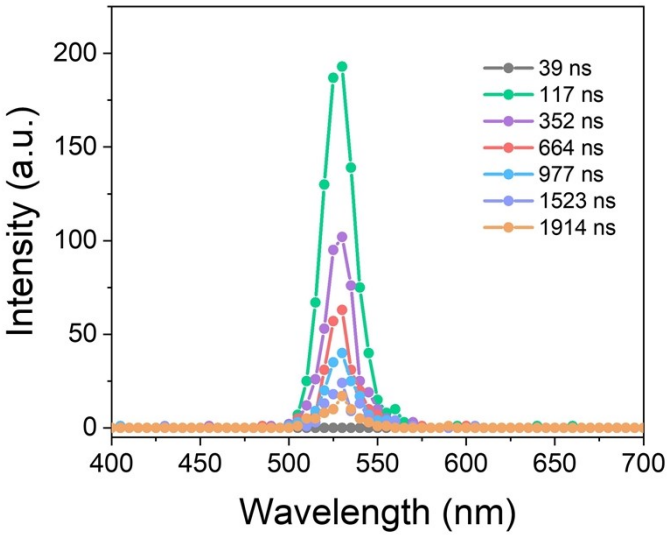

Figure S12. PL spectra extracted from measurements in TRPL spectra at a varied time after photoexcitation.

Table S1. PLQY performance comparison of the reported literatures.

Materials	PLQY (%)	Solvent	Synthesis temperature	Ref.
CPB-DDAB	31.5	Water/Ethanol	room temperature	**This work**
$CsPbBr_3$	16.7	Water	40 °C	Chem. Eng. J. 2021, 425,131456
$CsPb_2Br_5$/ Pb(OH)Br	27.14	Water/Ethanol	room temperature	ACS Appl. Mater. Interfaces 2021, 13, 23960−23969
$CsPbBr_3$	31.2	Water	room temperature	J. Mater. Chem. C, 2020, 8, 5594−5599
$CsPbBr_3$/ ZIF-62	33.6	Water	350 °C	ACS Appl. Nano Mater. 2023, 6, 1808−1816
$CsPbBr_3$@ PbBr(OH)	56.1	Water/DMF/ toluene	60 °C	Adv. Optical Mater. 2024, 12, 2400333
$CsPbBr_3$@PbBrOH	52	DMF/Water/ $NH_3{\cdot}H_2O$	room temperature	J. Mater. Chem. C, 2022, 10, 15843
$CsPbBr_3$	44.4	/	700 °C	Nano Lett. 2024, 24, 16400−16407

Table S2. The fitting of the PL decay lifetime for phosphors.

	$CsPbBr_3$	$CsPbBr_3$-DDAB	$CsPbBr_3$-TBPO
τ_1 (ns)	0.918501	584.969	39.6926
τ_2 (ns)	4.86984	2371.76	321.333
τ_3 (ns)	169.141	/	/
τ_{avg} (ns)	91.973	1576.668	235.707
τ_{rad} (ns)	505.35	5005.3	876.23
τ_{nonrad} (ns)	112.44	2301.0	322.47
k_r (s^{-1})	1.979×10^6	1.997×10^5	1.141×10^6
k_{nr} (s^{-1})	8.893×10^6	4.346×10^5	3.101×10^6
$k_{rad}:k_{nonrad}$	0.22	0.45	0.36
PLQY (%)	0.182	0.315	0.269

τ_1, τ_2, and τ_3 represent the decay lifetimes corresponding to the intrinsic exciton relaxation, the interaction between excitons and phonons, and the interaction between excitons and defects, respectively. The τ_{rad} and τ_{nonrad} were calculated by using the following two functions:

$$\frac{1}{t_{avg}} = \frac{1}{t_{rad}} + \frac{1}{t_{nonrad}}, \qquad (1)$$

$$PLQY = \frac{\frac{1}{t_{rad}}}{\frac{1}{t_{rad}} + \frac{1}{t_{nonrad}}}, \quad (2)$$

Radiative rate (k_r) and non-radiative rate (k_{nr}) were estimated by equations:

$$PLQY = \frac{k_r}{k_r + k_{nr}} \quad (3)$$

$$T_{ave} = \frac{1}{k_r + k_{nr}} \quad (4)$$