

Supporting Information for

**Solvent-Free Synthesis of Stable Heterostructured-CsPbBr₃/Cs₂PbBr₅ Assisted
by SiO₂ for White Light-Emitting Diodes**

Jisong Yao,* Tianliang Yao, Kaishuai Zhang, Wenxuan Fan, Zhi Yang, Leimeng Xu, Shalong Wang,
Jizhong Song*

Key Laboratory of Materials Physics of Ministry of Education, School of Physics and Microelectronics,
Zhengzhou University, Daxue Road 75, Zhengzhou 450052, China.

E-mail: yaojisong@zzu.edu.cn; songjizhong@zzu.edu.cn

Experimental Section

Materials: Guanidine carbonate (Gu_2CO_3 , 99.9%), octylamine (OTA, 99.0%) and triphenylphosphine dibromide (TPDB, 95%) were purchased from Macklin. Cesium stearate, lead stearate and nickel stearate were purchased from Shanghai Dibo Biotechnology. Silicon dioxide (SiO_2) was purchased from Aladdin. All reagents were used without further purification.

H-CPB synthesis: Typical synthesis of H-CPB (2) composite was conducted as follows. The raw materials were weighted and mixed according to the desired molar ratio of Gu_2CO_3 /cesium stearate/lead stearate/nickel stearate/OTA/ $\text{SiO}_2 = 1:3:5:1:1:72$ and mixed in a mixer at high speed for 2 min under atmospheric environments. Subsequently, a certain amount of TPDB was added into the above reaction system, and stirred rapidly for 3 min to obtain the composites of LHPs and SiO_2 . For other H-CPB (1) and H-CPB (3) composites were following the same synthesis procedure, just changed the ratio of SiO_2 to Gu_2CO_3 /cesium stearate/lead stearate/nickel stearate/OTA/ $\text{SiO}_2 = 1:3:5:1:1:166$ and Gu_2CO_3 /cesium stearate/lead stearate/nickel stearate/OTA/ $\text{SiO}_2 = 1:3:5:1:1:28$, respectively. For other H-CPB (4) and H-CPB (5) composites were following the same synthesis procedure, just exchanged 100 m^2/g of SiO_2 with 170 m^2/g and 230 m^2/g of SiO_2 , respectively.

Fabrication of the H-CPB composites-based WLEDs: The H-CPB composites were mixed with a commercial red-emitting $\text{CaAlSiN}_3:\text{Eu}^{2+}$ (CASN) phosphor at a different weight ratio in UV-curing resin. The mixture was then deposited on a blue LED chip (wavelength 455 nm). It was exposed to UV light for 5 mins and then cured. All the processes of fabrication were carried out at ambient condition.

Characterizations: XRD patterns were detected by using a Philips X'Pert PRO SUPER X-ray diffractometer using Cu K α radiation ($\lambda = 1.54178 \text{ \AA}$). Morphologies were measured by field-emission SEM (GeminiSEM 500). The HRTEM and HAADF-STEM images were taken by a was taken on Tecnai G2 F30 S-TWIN with accelerating voltage of 300 kV. The energy dispersive X-ray spectra (EDS) were collected on Tecnai G2 F30 S-TWIN with an energy dispersive detector. Fourier-transform infrared (FTIR) spectra were collected on Nicolet IS10. The X-ray photoelectron spectroscopy (XPS) characterizations were conducted on a Thermo ESCALAB 250 spectrometer using a monochromatic Al Kr radiation source (1486.6 eV). The PL spectra were measured by a fluorescence spectrometer (Hitachi F-4600). The PLQYs were collected on FLS1000 fluorescence spectrometer with an integrating sphere, exciting the powders at 365 nm. Time resolved PL-decay spectra was collected using FLS1000 fluorescence spectrometer with an excitation wavelength at 365 nm. The EL properties of fabricated WLEDs were collected using a Keithley 2680 source meter and a Minolta CS2000 system.

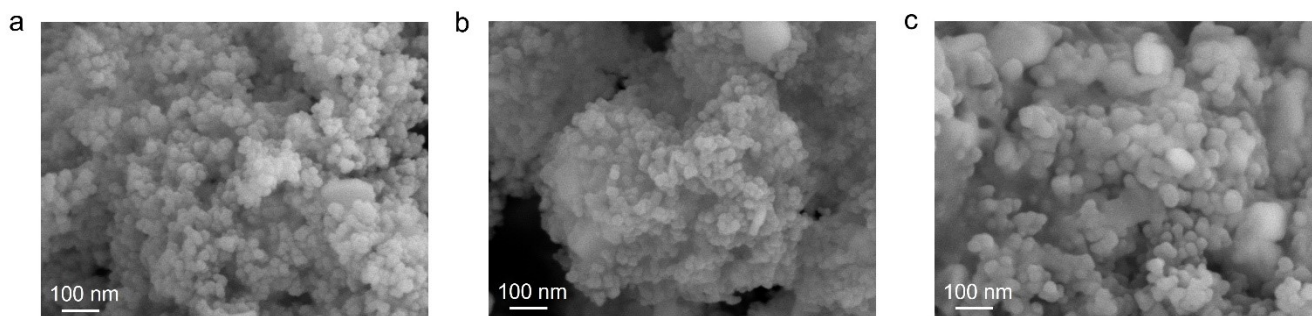


Fig. S1 SEM images of H-CPB (1) (a), H-CPB (2) (b), and H-CPB (3) (c), respectively.

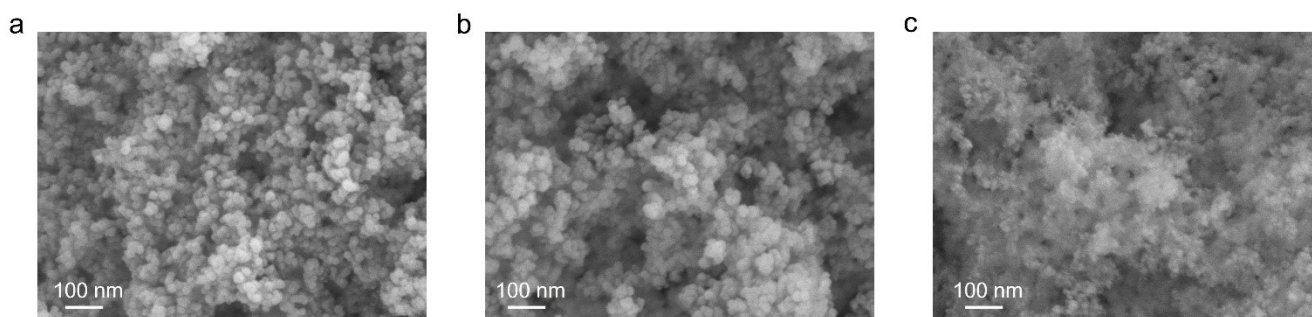


Fig. S2 SEM images of SiO₂ nanospheres with specific surface areas of 100 m²/g (a), 170 m²/g (b), and 230 m²/g (c).

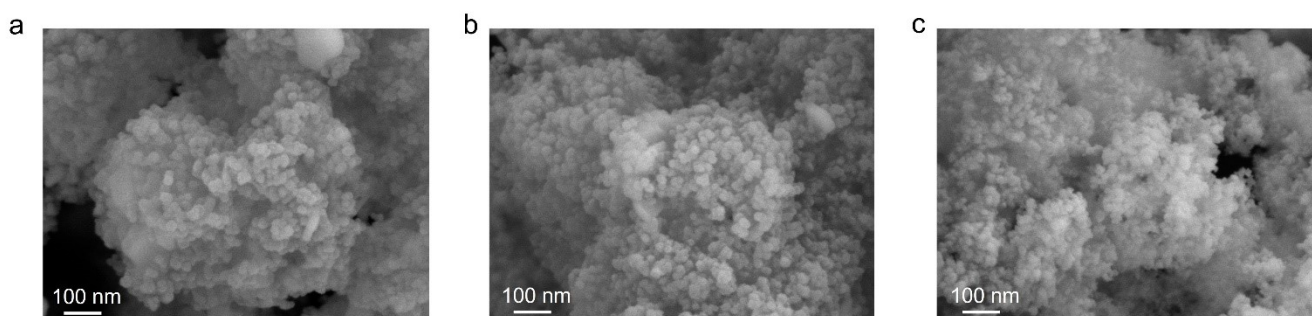


Fig. S3 SEM images of H-CPB (2) (a), H-CPB (4) (b), and H-CPB (5) (c), respectively.

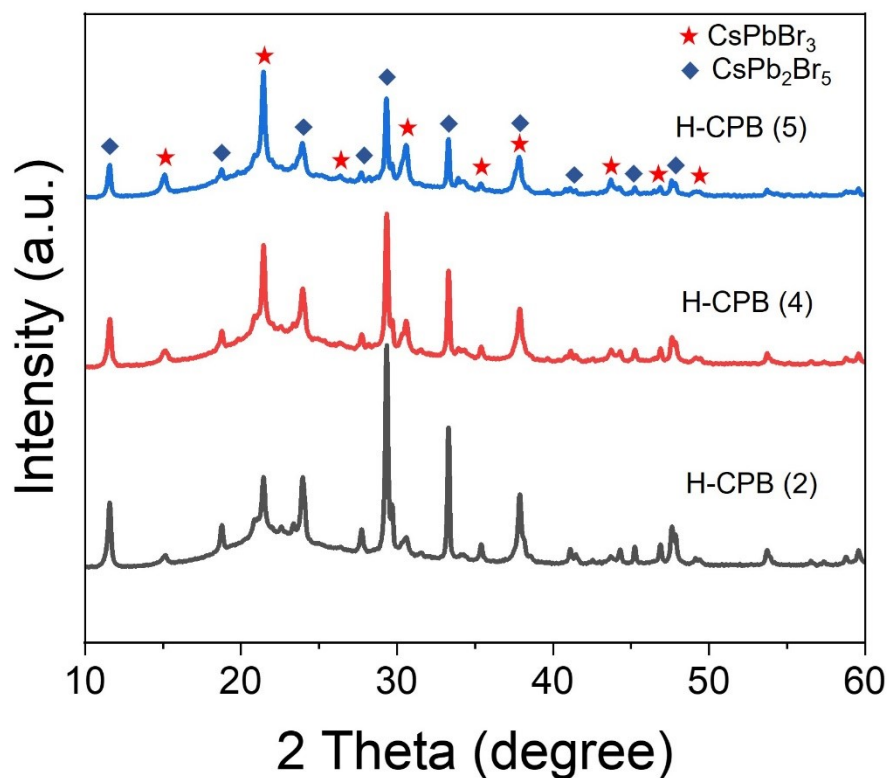


Fig. S4 Powder XRD patterns of the corresponding H-CPB composites.

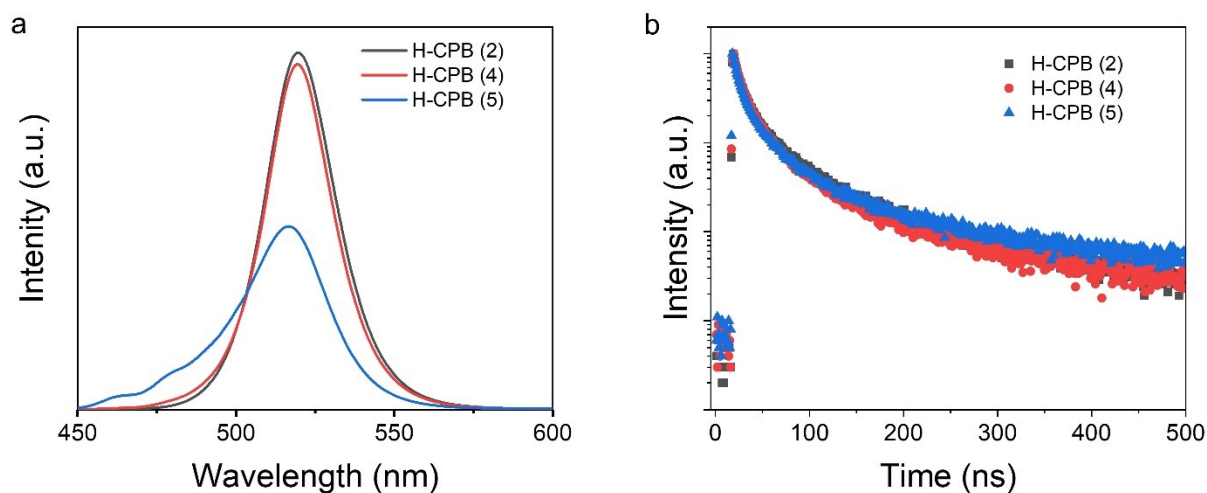


Fig. S5 The PL spectra (a) and PL decay kinetics (b) (excitation at 365 nm) of the corresponding H-CPB composites.

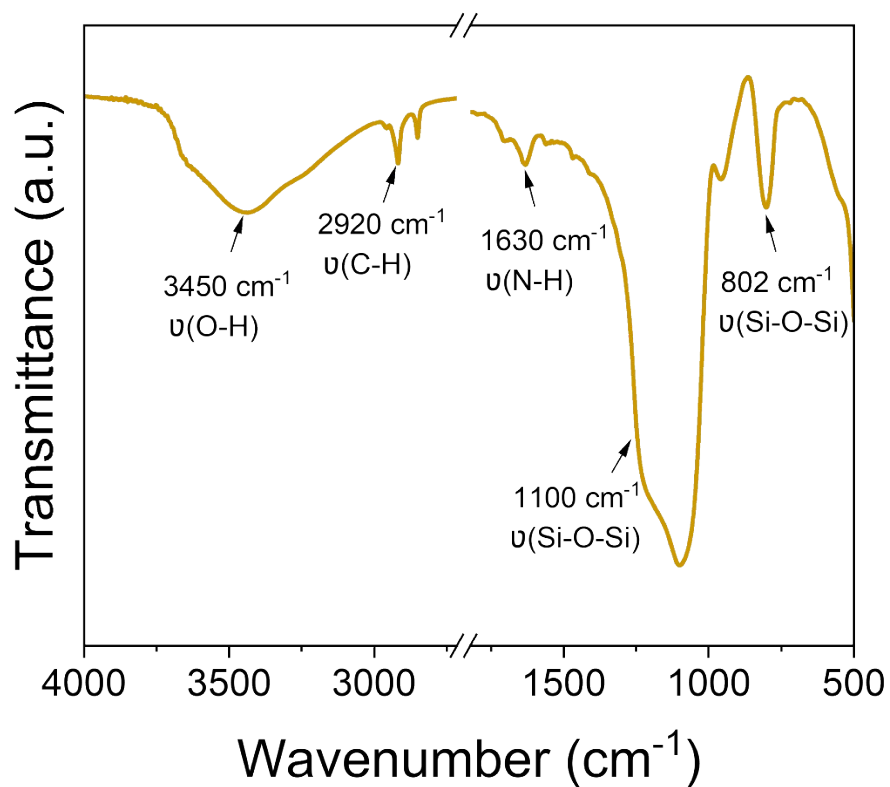


Fig. S6 FTIR spectra of pure SiO₂ nanospheres.

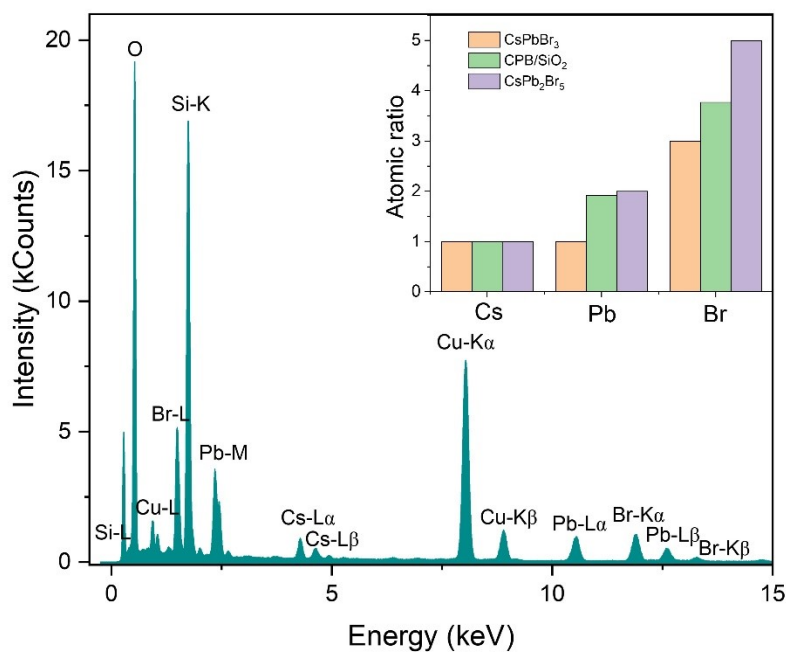


Fig. S7 Energy Dispersive X-ray Spectroscopy of H-CPB. Inset: quantification of the EDS spectrum showing a Cs:Pb:Br atomic ratio of 1:1.92:3.77.

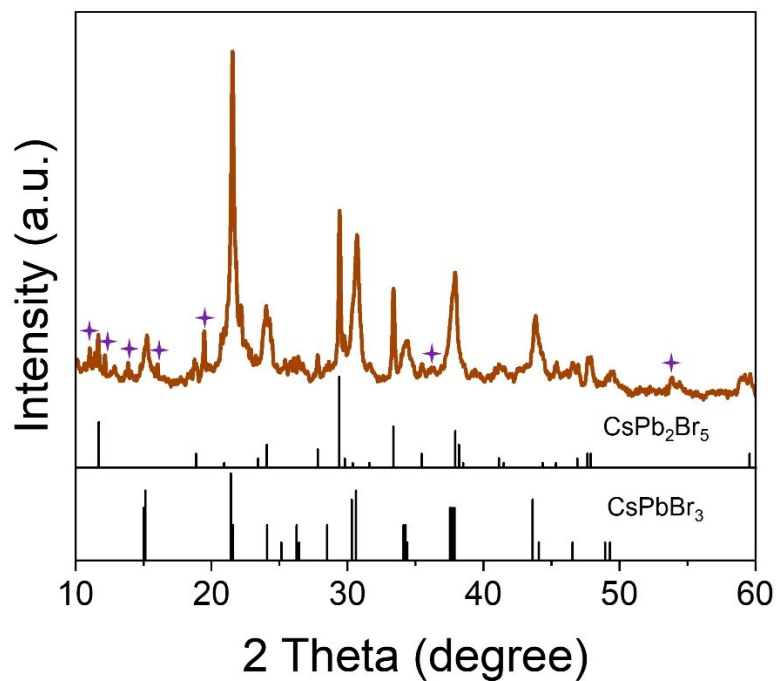


Fig. S8 Powder XRD patterns of P-CPB.

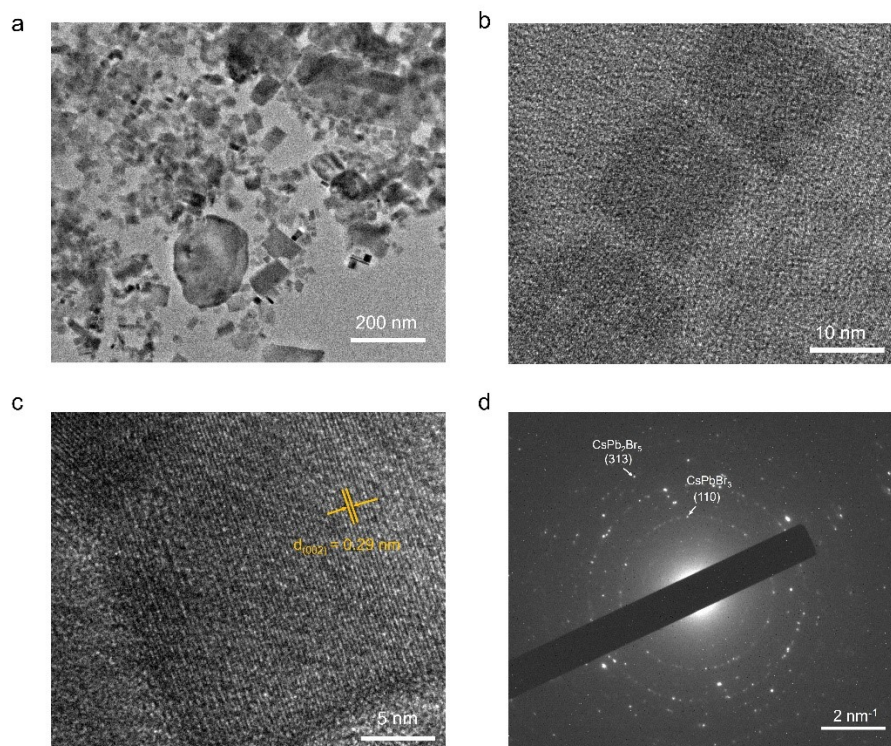


Fig. S9 (a) Typical TEM image of P-CPB. (b,c) HRTEM image of CsPbBr₃ NCs embedded in CsPb₂Br₅. (d) FFT pattern for the pure CPB which is derived from the region in (b).

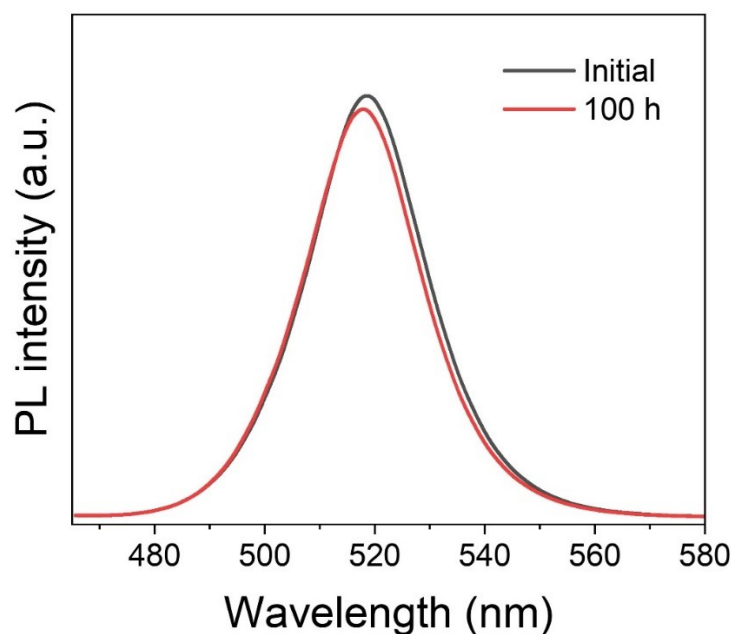


Fig. S10 The PL spectra of H-CPB composites before and after continuous UV light irradiation for 100 h.

Table S1. The PL lifetimes for the the corresponding H-CPB composites.^a

Samples	H-CPB (1)	H-CPB (2)	H-CPB (3)	H-CPB (4)	H-CPB (5)
τ_1 (ns)	0.66 [34%]	8.2 [33%]	3.8 [25%]	8.3 [36%]	8.3 [37%]
τ_2 (ns)	11.7 [66%]	57.8 [67%]	41.5 [75%]	49.4 [64%]	59.4 [63%]
τ_{avg} (ns)	7.92	41.5	32.0	34.7	40.5

^aThe PL was excited at 332 nm and monitored at maximum emission peak. The average PL lifetimes (τ_{avg}) were obtained by $\tau_{\text{avg}} = (A_1 \times \tau_1^2 + A_2 \times \tau_2^2)/(A_1 \times \tau_1 + A_2 \times \tau_2)$, where A_1 and A_2 stand for the statistical weights of the corresponding lifetime components (given in the square parentheses). The errors for PL lifetimes are given in parentheses in units of the last digits quoted.