

## Article

# Highly Efficient and Stable CsPbBr<sub>3</sub>-Alginate Acid Composites for White Light-Emitting Diodes

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**Abstract:** All-inorganic perovskite nanocrystals (NCs) have attractive potential for applications in display and lighting fields due to their special optoelectronic properties. However, they still suffer from poor water and thermal stability. In this work, green CsPbBr<sub>3</sub>-alginate acid (CsPbBr<sub>3</sub>-AA) perovskite composites were synthesized by an in situ hot-injection process which showed a high photoluminescence quantum yield (PLQY) of 86.43% and improved moisture and thermal stability. Finally, white light-emitting diodes (WLEDs) were fabricated by combining the green CsPbBr<sub>3</sub>-AA perovskite composites with red K<sub>2</sub>SiF<sub>6</sub>:Mn<sup>4+</sup> phosphors and blue InGaN LED chips. The WLEDs show a relatively high luminous efficacy of 36.4 lm/W and a wide color gamut (124% of the National Television System Committee). These results indicate that the green CsPbBr<sub>3</sub>-AA perovskite composites have great potential applications in backlight displays.

**Keywords:** perovskites; in situ hot-injection process; luminescent materials; stability; light-emitting diodes



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## 1. Introduction

In recent years, we have witnessed a rapid development in the synthesis and application of all-inorganic CsPbX<sub>3</sub> (X = Cl, Br and I) perovskite NCs [1–3]. Compared with traditional semiconductor NCs (e.g., CdSe and InP-based NCs), they have better optical properties, such as broadband absorption, tunable emission wavelength, narrow full width at half maximum (FWHM), and high photoluminescence PLQY [4–6], which provides them with potential applications in light emitting diodes (LEDs) [7–9], solar cells [10–12], photodetectors [13–15], photocatalysis [16,17], lasers [18], displays [19], bioimaging [20], and anti-counterfeiting [21].

Although CsPbX<sub>3</sub> NCs possess many excellent properties, they are still far from practical applications due to their poor stability [21]. First, the ionic properties of the NCs make them easy to decompose when they are in contact with polar solvents. Second, organic ligands such as oleic acid and oleylamine are widely used in the synthesis of perovskite NCs, which are easily detached from the surface of NCs during the purification process, resulting in a significant decrease in colloidal stability and PLQY. Third, when the colloidal NC solution is solidified into powder (including solidification with encapsulant), the emission of aggregated fluorescence is quenched, and the PLQY decreases sharply.

To address these issues, matrix encapsulation has been carried out to enhance the photoluminescence (PL) stability by embedding perovskite NCs into transparent and stable

matrix materials to form composite luminescent materials [22]. For example, SiO<sub>2</sub> [23], Al<sub>2</sub>O<sub>3</sub> [23], KCl, KBr, ZIF-8 [24,25], UiO-67 [26], PMMA [27], and PMAO [27] have been employed as matrices to prepare perovskite composites with enhanced stability. Among them, organic polymers have unique advantages in improving the stability of perovskite NCs. The polymer can form chemical bonds with the surface ions of perovskite due to abundant groups of the polymer, passivating their surface defects and tightly wrapping the NCs to isolate water and oxygen [28–31].

Alginic acid (AA) is a naturally occurring edible polysaccharide [32,33]. It has many carboxyl and hydroxyl groups, and can easily combine with metal ions (such as sodium and calcium) to form the corresponding salt, called alginate. Thus, AA with abundant carboxyl and hydroxyl groups can be used as a surface ligand for perovskite NCs, while its long-chain structure helps to wrap perovskite NCs, forming a matrix structure. In addition, AA is easy to obtain, has abundant material sources and is of extremely low cost, which is also particularly important for late-stage WLEDs applications.

In this study, we successfully synthesized green CsPbBr<sub>3</sub>-AA perovskite composites using AA as matrix materials and surface ligands by a hot-injection process. The as-prepared perovskite composites not only show excellent optical properties with a high PLQY of 86.43% and a narrow FWHM of 23 nm, but also exhibit high stability against moisture and heat. Furthermore, WLEDs were fabricated by combining green CsPbBr<sub>3</sub>-AA perovskite composites with red K<sub>2</sub>SiF<sub>6</sub>:Mn<sup>4+</sup> phosphors and blue InGaN LED chips. The WLEDs show relatively high efficiency and a wide color gamut.

## 2. Materials and Methods

### 2.1. Chemicals

Cesium carbonate (Cs<sub>2</sub>CO<sub>3</sub>, Alfa Aesar (Shanghai, China), 99%), oleic acid (OA, Damas-beta (Shanghai, China), 90%+), oleylamine (OLA, Damas-beta (Shanghai, China), 90%+), 1-octadecene (ODE, Adamas, 90%), lead (II) bromide (PbBr<sub>2</sub>, Adamas (Shanghai, China), 99.9%), alginic acid ((C<sub>6</sub>H<sub>8</sub>O<sub>6</sub>)<sub>n</sub>, Adamas (Shanghai, China), 99%), ethanol (99%), and toluene (C<sub>7</sub>H<sub>8</sub>, SCRC (Shanghai, China), ≥99.5%) were purchased from SINOPHARM and used without further purification.

### 2.2. Synthesis of Cesium Oleate (Cs-OA) Precursor

A total of 0.420 g of Cs<sub>2</sub>CO<sub>3</sub>, 1.5 mL of OA, and 20 mL of ODE were added into a 50 mL three-necked flask, which was kept relatively sealed. The mixture was then vigorously stirred and dried under N<sub>2</sub> flow for 1 h at 120 °C. The reaction vessel was swiftly heated to 150 °C and stirred at this temperature for 5 h. After the reaction was completed, the precursor Cs-OA for hot injection were obtained, and were preheated to 100 °C when used.

### 2.3. Synthesis of CsPbBr<sub>3</sub> NCs

PbBr<sub>2</sub> (0.1835 g), 1.5 mL of OA, 1.5 mL of OLA, and 15 mL of ODE were added to a 50 mL three-necked flask, which was kept relatively sealed. The mixture was then vigorously stirred and dried under N<sub>2</sub> flow for 1 h at 120 °C. The reaction vessel was swiftly heated to 160 °C for 1 min under N<sub>2</sub> flow, and then Cs-Oleate (1 mL) was swiftly injected into the mixture solution. After 10 s, the reaction solution was cooled to room temperature in an ice water bath. The product solution was centrifuged at 11,000 rpm for 15 min, and the supernatant was subsequently discarded. The precipitated NCs were then redispersed in toluene for further centrifugation at 11,000 rpm for 10 min. The precipitate was dried in a vacuum drying oven at 36.5 °C for 24 h, and the solid samples were stored for characterization.

### 2.4. Synthesis of CsPbBr<sub>3</sub>-AA Perovskite Composites

PbBr<sub>2</sub> (0.1835 g), x mmol (x = 1, 1.5, 3, 4.5, 6, 7.5) of AA, 1.5 mL of OA, 1.5 mL of OLA, and 15 mL of ODE were added to a 50 mL three-necked flask, which was kept relatively sealed. The mixture was then vigorously stirred and dried under N<sub>2</sub> flow for

1 h at 120 °C. The reaction vessel was swiftly heated to 160 °C for 1 min under N<sub>2</sub> flow, and then Cs-Oleate (1 mL) was swiftly injected into the mixture solution. After 10 s, the solution was cooled to room temperature in an ice water bath. The product solution was centrifuged at 11,000 rpm for 15 min, and the supernatant was subsequently discarded. The precipitate was then redispersed in toluene for further centrifugation at 11,000 rpm for 10 min. The precipitate was dried in a vacuum drying oven at 36.5 °C for 24 h, and the solid samples were stored for characterization.

### 2.5. Characterization

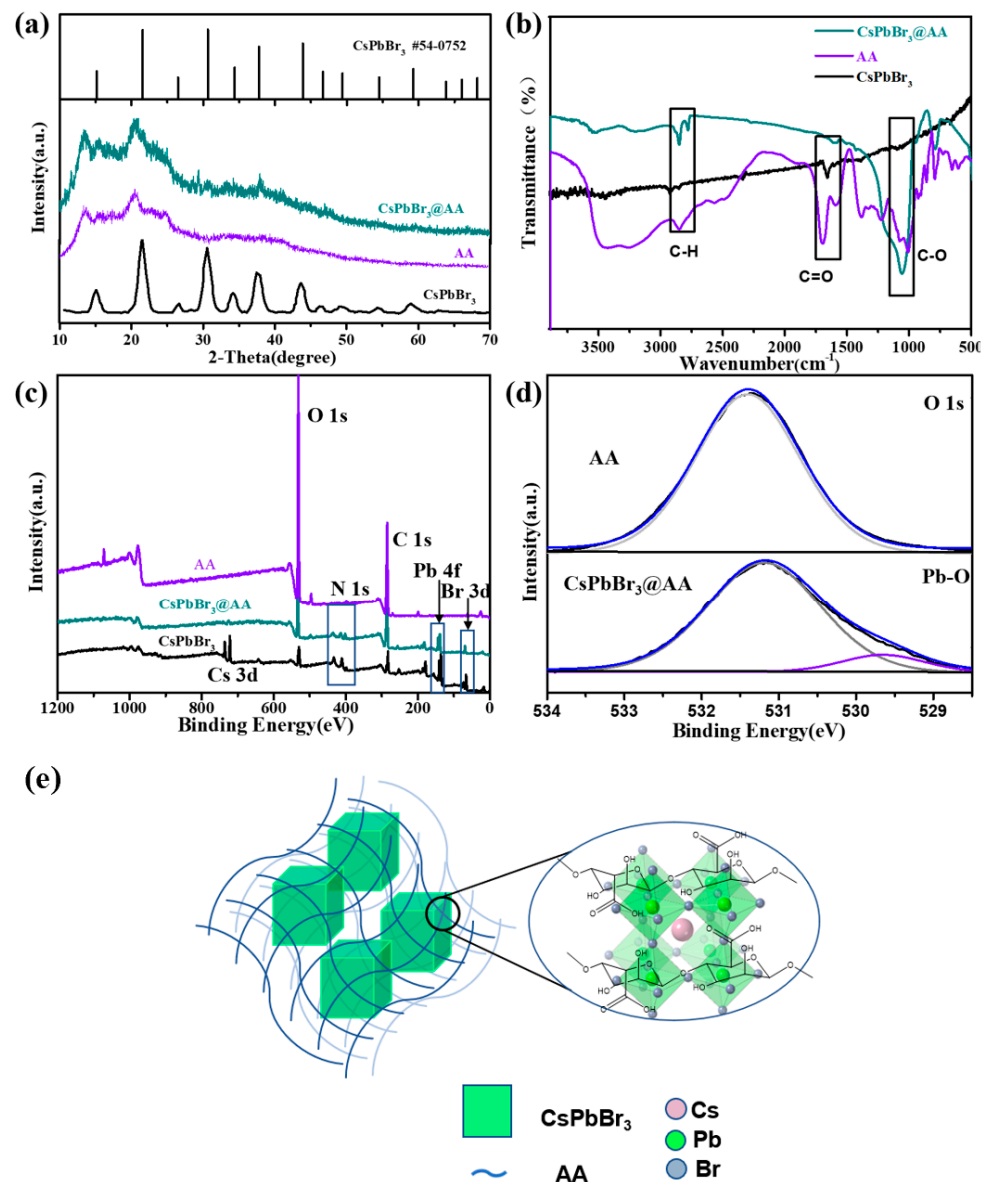
The XRD pattern was obtained from a Bruker D8 Advance X-ray diffractometer equipped with monochromatic Cu-K $\alpha$  radiation ( $\lambda = 0.154056$  nm) as an X-ray source. The X-ray photoelectron spectroscopy (XPS) spectra were acquired by an ESCALAB 250XI system produced by Thermo Scientific. The micromorphology of perovskite NCs was detected by transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM), and images were collected by a JEM2100F electron microscope with a 200 kV accelerating voltage. The optical properties were declared through PL and UV/visible absorption spectra using a FluoroMax-4HORIBAs spectrophotometer and a HITACHI U-3900 spectrophotometer, respectively.

## 3. Results and Discussion

As shown in Figure 1a, X-ray diffraction (XRD) was performed to confirm the successful synthesis of the CsPbBr<sub>3</sub>-AA composites. Compared with the XRD pattern of pure CsPbBr<sub>3</sub> NCs (54-0752), the XRD pattern of CsPbBr<sub>3</sub>-AA shows characteristic peaks consistent with pure CsPbBr<sub>3</sub> NCs. However, since the background peak of AA itself is particularly strong, this characteristic peak is not particularly pronounced, which may be because the wrap of AA is too thick.

From the FTIR spectra of the three samples (Figure 1b), it can be further determined that, compared with the original CsPbBr<sub>3</sub> NCs, the intensity of the infrared characteristic peaks corresponding to each organic functional group (C-H, C=O, C-O) in the CsPbBr<sub>3</sub>-AA composites has been markedly enhanced, which apparently results from the addition of AA. Moreover, we find that after encapsulation, the stretching vibration of -O-H shifts to a low wavenumber by 34 cm<sup>-1</sup> (3310 cm<sup>-1</sup> to 3276 cm<sup>-1</sup>), which indicates that the oxygen atoms are involved in bonding with Pb<sup>2+</sup>, resulting in an increase in the bond length of the O-H bond and a redshift in the stretching vibration peak. Therefore, a combination of AA and the metal ions on the surface of CsPbBr<sub>3</sub> NCs in the CsPbBr<sub>3</sub>-AA composites can be found.

The XPS energy spectra of CsPbBr<sub>3</sub> NCs, AA, and CsPbBr<sub>3</sub>-AA composites were tested and compared (Figure 1c,d). It is not difficult to find that the oxygen peak (530 eV) and carbon peak (284.8 eV) of the CsPbBr<sub>3</sub>-AA composites are significantly enhanced compared with those of the original CsPbBr<sub>3</sub> NCs, which results from the large amount of organic functional groups from AA. At the same time, it can be seen that the spectra of the CsPbBr<sub>3</sub>-AA composites show N peaks, Pb peaks, and Br peaks that are consistent with the original CsPbBr<sub>3</sub> NCs. These peaks are not available in AA, which indicates the existence of CsPbBr<sub>3</sub> NCs in the CsPbBr<sub>3</sub>-AA composites. However, the Cs peak of the original CsPbBr<sub>3</sub> NCs in the spectrum could not be found in the CsPbBr<sub>3</sub>-AA composites, which may be attributed to the fact that Cs<sup>+</sup> is usually located in the interior of the nanocrystals, and the coating of AA is too thick, so its signal is not obvious. From Figure 1d a new peak appears near 529.5 eV after the high-resolution spectrum of the O 1s orbital is processed by peak separation, which is presumed to be the Pb-O peak. It is further confirmed that the carboxyl and hydroxyl groups in AA are chemically bonded with the metal ions on the surface of the nanocrystals. This phenomenon passivates mezigzag surfacetal ion defects on the surface of nanocrystals, thereby achieving high luminescence performance.

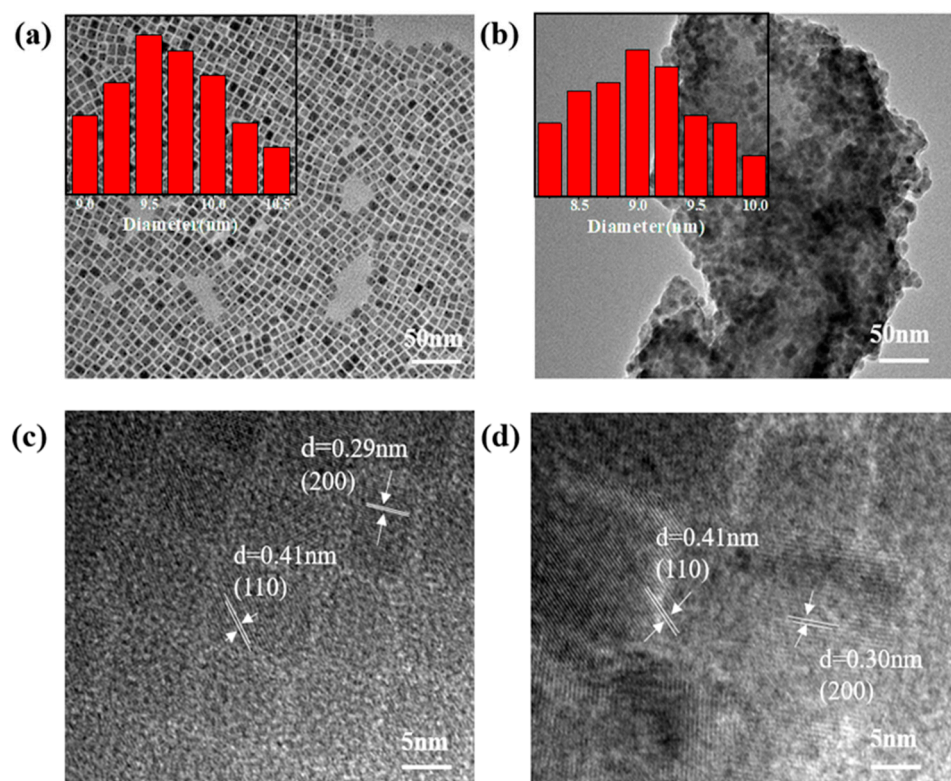


**Figure 1.** (a) XRD patterns of CsPbBr<sub>3</sub> NCs and CsPbBr<sub>3</sub>-AA composites. (b) FTIR spectra of AA, CsPbBr<sub>3</sub> NCs and the CsPbBr<sub>3</sub>-AA composite. (c) XPS spectra of CsPbBr<sub>3</sub> NCs, AA, and CsPbBr<sub>3</sub>-AA composites. (d) High-resolution XPS spectra of O 1s of AA and CsPbBr<sub>3</sub>-AA composites. (e) Schematic diagram of CsPbBr<sub>3</sub>-AA composites.

According to the above analysis, we analyzed the encapsulation mechanism of CsPbBr<sub>3</sub>-AA composites, as shown in Figure 1e. After encapsulation, AA first participates in the formation of nanocrystals through the carboxyl groups contained in it instead of oleic acid, and then a large number of carboxyl groups and hydroxyl groups on the surface of AA are coordinately bonded to the surface of CsPbBr<sub>3</sub> NCs, resulting in chemical coordination and the encapsulation of nanocrystals. At the same time, the defects on the nanocrystal surface are passivated, the probability of nonradiative recombination is reduced, and the luminous intensity and quantum efficiency of the composites are significantly improved. At the same time, the flexible long-chain structure of AA can effectively slow the effect of heat on the nanocrystals and enhance their stability.

To further characterize the morphology and structure of the prepared nanocrystals, CsPbBr<sub>3</sub> and CsPbBr<sub>3</sub>-AA were tested by TEM. We can see in Figure 2 that the CsPbBr<sub>3</sub> NCs synthesized by the hot-injection process are cubic crystal phases with good dispersibility, and the average particle size is approximately 9.81 nm. After encapsulation with AA,

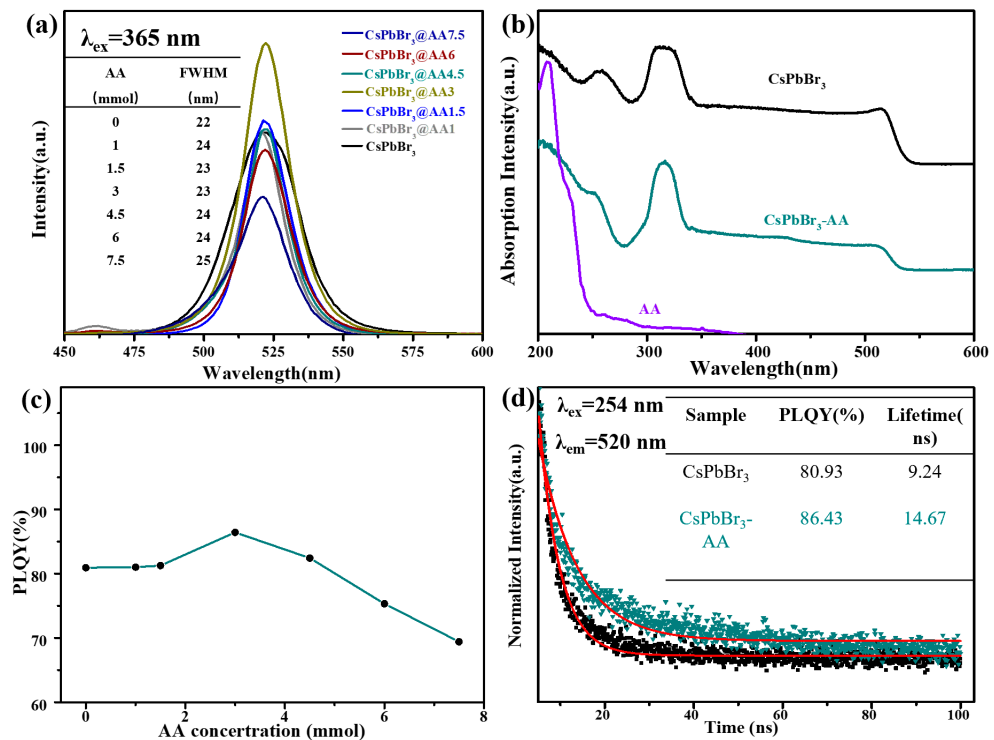
the CsPbBr<sub>3</sub> NCs were embedded in the agglomerated and indistinct organic polymer with stacking, the size distribution of the grains decreased, and the average particle size decreased to 9.57 nm (Figure 2b). The occurrence of this phenomenon may be due to the lack of ligands in the grains, which makes nucleation and growth insufficient and the grains smaller. The further magnification of the local area of the samples, HRTEM, is shown in Figure 2c,d. The measured interplanar spacings of the nanocrystals are 0.29 and 0.41 nm (Figure 2c) and 0.30 and 0.41 nm (Figure 2d), respectively, corresponding to the (200) and (110) planes of the cubic phase CsPbBr<sub>3</sub>. This shows that in the CsPbBr<sub>3</sub>-AA composites, the CsPbBr<sub>3</sub> NCs are successfully encapsulated by AA. Therefore, the CsPbBr<sub>3</sub>-AA composite should have significantly improved environmental stability.



**Figure 2.** TEM and the corresponding histogram of the particle size distribution (b) of the CsPbBr<sub>3</sub> NCs (a) and CsPbBr<sub>3</sub>-AA composite (b). HRTEM images of the CsPbBr<sub>3</sub> NCs (c) and CsPbBr<sub>3</sub>-AA composite (d).

We then characterized the fluorescence emission spectra of the samples, as shown in Figure 3a. Compared with the original CsPbBr<sub>3</sub> NCs, the fluorescence intensity of the CsPbBr<sub>3</sub>-AA composites first gradually increased with an increasing AA doping amount and then decreased, which is consistent with the trend shown by the results of the PLQYs test that followed. Moreover, when the doping amount was 3 mmol, the intensity reached the maximum value. Correspondingly, the PLQY increased from 80.93% for the original CsPbBr<sub>3</sub> NCs to 86.43% for the CsPbBr<sub>3</sub>-AA composites. At the same time, a change in the half-peak width was observed. The overall half-peak width of the AA-encapsulated perovskite NCs was wider, and the minimum was 23 nm when the addition amount was 3 mmol, which was still higher than that of the original NCs. It is preliminarily speculated that the encapsulation matrix of AA has a large number of hydroxyl and carboxyl groups. When the concentration is low, the nucleation and growth of NCs is not sufficient due to the absence of oleic acid, resulting in low luminescence performance. When the molar amount of AA increases to a certain extent, this effect is eliminated. At the same time, a large number of carboxyl and hydroxyl groups in the encapsulation matrix can chemically coordinate with the metal ions on the surface of CsPbBr<sub>3</sub> NCs, thereby passivating the ionic defects on the surface of the NCs

and finally improving the fluorescence intensity and quantum efficiency of the composites. With a further increase in the amount of AA, too much AA covers the surface of the NCs, and its luminescence performance decreases. In addition, in the fluorescence test, it was found that the emission peaks of the CsPbBr<sub>3</sub>-AA composites have a redshift compared with the original CsPbBr<sub>3</sub> NCs, which is inconsistent with the change in the average particle size. This may be due to the lack of ligands on the surface of NCs with small particle sizes, resulting in increased surface defects and reduced luminescence performance. Although the average particle size is lowered, the luminescent host is still large NCs. As a result, the average particle size decreases while the emission peaks redshift.



**Figure 3.** (a) PL spectra of pure CsPbBr<sub>3</sub> NCs and CsPbBr<sub>3</sub>-AA composites. (b) Absorption spectra of AA, pure CsPbBr<sub>3</sub> NCs, and the CsPbBr<sub>3</sub>-AA composite. (c) PLQYs of CsPbBr<sub>3</sub> NCs and CsPbBr<sub>3</sub>-AA composites with different concentrations of AA. (d) PL decay curves of CsPbBr<sub>3</sub> pure NCs and CsPbBr<sub>3</sub>-AA composites.

CsPbBr<sub>3</sub> NCs, AA and CsPbBr<sub>3</sub>-AA composites were characterized by UV-Vis absorption spectroscopy and FTIR to reveal the real bonding mode between AA and CsPbBr<sub>3</sub> NCs. As shown in Figure 4, compared with the original CsPbBr<sub>3</sub> NCs, the absorption spectrum of the CsPbBr<sub>3</sub>-AA composites is steeper than that of the original CsPbBr<sub>3</sub> NCs in the 200–210 nm range, and their position corresponds to the absorption peaks of AA in the same range, which may be due to the addition of AA. At the same time, it was determined that the absorption peaks of the CsPbBr<sub>3</sub>-AA composites at approximately 510 nm have a redshift compared with the original CsPbBr<sub>3</sub> NCs, which is consistent with the emission spectrum.

To verify the above hypothesis, tests for the fluorescence lifetime of CsPbBr<sub>3</sub> NCs and CsPbBr<sub>3</sub>-AA composites were performed. The fluorescence lifetime of perovskite NCs usually exhibits a double exponential decay mode, which can be divided into two parts: a long-lived part ( $\tau_1$ ) and a short-lived ( $\tau_2$ ) part, which correspond to radiative and non-radiative transitions, respectively. The fitted mean life can be based on the following formula [34]:

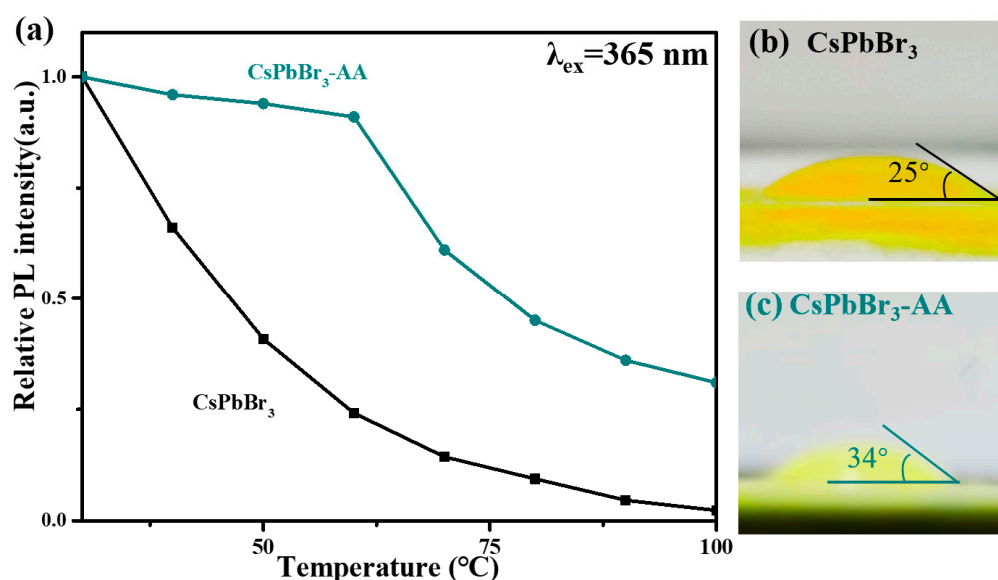
$$y = A_1 \exp\left(-\frac{x}{\tau_1}\right) + A_2 \exp\left(-\frac{x}{\tau_2}\right) + y_0 \quad (1)$$

$$\tau_{av} = \frac{\sum A_i \tau_i^2}{\sum A_i \tau_i} \quad (2)$$

Under UV excitation at 254 nm, the fluorescence lifetimes of the original CsPbBr<sub>3</sub> NCs and CsPbBr<sub>3</sub>-AA composites at an emission wavelength of 520 nm were tested and fitted according to Formula (1). The fitting results of  $\tau_1$ ,  $\tau_2$  and  $\tau_{av}$  for all doping concentrations are listed in Table 1. After AA encapsulation, the fluorescence lifetime of the CsPbBr<sub>3</sub>-AA composites is increased to 14.67 ns. The longer fluorescence lifetime indicates that the defects of the NCs are indeed passivated, and that the nonradiative recombination process of defect-related fast exciton trapping in the pristine CsPbBr<sub>3</sub> NCs is effectively eliminated through AA encapsulation. Eventually, the fluorescence intensity and quantum efficiency of the composites increase.

**Table 1.** PL lifetimes of CsPbBr<sub>3</sub> NCs with different contents of AA.

	$A_1$	$\tau_1$ (ns)	$A_2$	$\tau_2$ (ns)	$\tau_{av}$ (ns)
CsPbBr <sub>3</sub>	0.64	4.73	0.36	12.27	9.24
CsPbBr <sub>3</sub> -AA	8.36	0.77	22.51	0.23	14.67

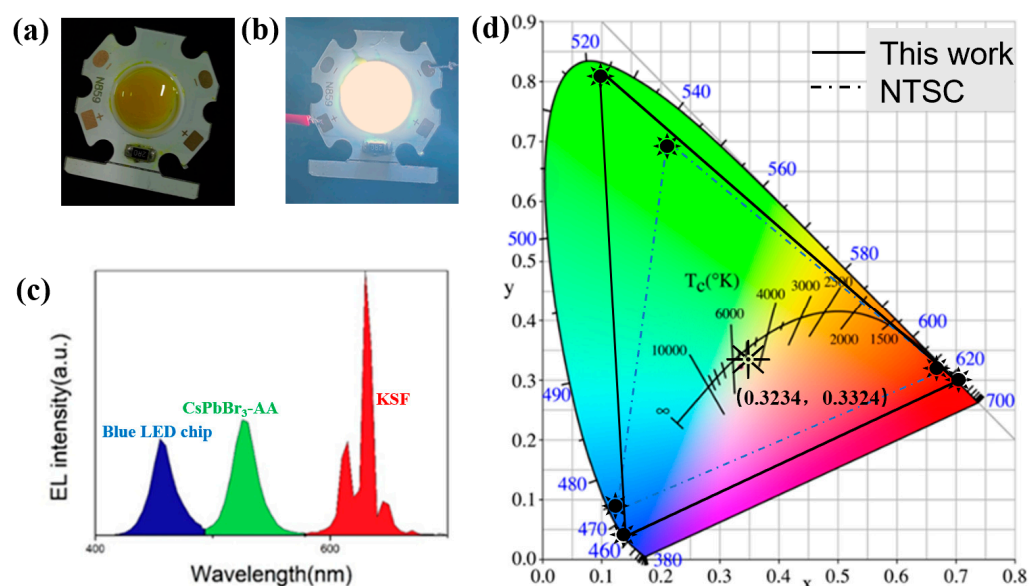


**Figure 4.** (a) Relative PL intensity of the pure CsPbBr<sub>3</sub> NCs and CsPbBr<sub>3</sub>-AA contact angle of CsPbBr<sub>3</sub> NCs (b) and CsPbBr<sub>3</sub>-AA (c) with water.

To evaluate the thermal stability of the as-prepared composites, we tested the temperature-dependent PL spectra of the CsPbBr<sub>3</sub> NCs and the CsPbBr<sub>3</sub>-AA composites with an excitation wavelength of 365 nm. As shown in Figure 4a, the relative PL intensity of the green CsPbBr<sub>3</sub>-AA composites maintained 90% of the initial intensity when the environmental temperature was increased to 60 °C, which is higher than that of the pure CsPbBr<sub>3</sub> NCs (~15%). Even as the temperature increased to 100 °C, the perovskite composites maintained 30% of the initial value, while the PL of the pure NCs almost disappeared. Furthermore, the contact angle test of water was performed on the CsPbBr<sub>3</sub> NC and CsPbBr<sub>3</sub>-AA composite films. As shown in Figure 4b,c, the CsPbBr<sub>3</sub>-AA composite films show a contact angle 34° higher than that of the pure CsPbBr<sub>3</sub> NC films (25°), which indicates that the composites exhibit better hydrophobicity than the pure NC films. However, the small increase in hydrophobicity is mainly limited by the large number of hydroxyl groups on the surface of AA, and the full consumption of hydroxyl groups cannot be guaranteed during the synthesis process. Due to the excessive hydroxyl groups absorbing water, the hydrophobicity of the composite material is not particularly good.

Finally, we applied the synthesized CsPbBr<sub>3</sub>-AA composites to white LEDs. A certain amount of CsPbBr<sub>3</sub>@AA<sub>3</sub>, K<sub>2</sub>SiF<sub>6</sub>:Mn<sup>4+</sup> and AB glue (the mass ratio of a glue to B glue is 1:2) are added into the agate mortar and ground evenly until the bubbles disappear completely. The obtained product was uniformly coated on a gallium nitride (GaN) blue light chip ( $\lambda_{em} = 460$  nm), which was heated and cured in an oven at 120 °C for 2 h, and the optical properties of the obtained white LED were tested.

As shown in Figure 5, the WLEDs show bright white emission at an operating voltage and current of 3.7 V and 20 mA. The electroluminescence spectrum (EL) contains three characteristic emission peaks, located at 460 nm (blue), 521 nm (green) and 631 nm (red), which belong to the blue LED chips, the green CsPbBr<sub>3</sub>-AA composites, and red K<sub>2</sub>SiF<sub>6</sub>:Mn<sup>4+</sup> phosphor, respectively. The WLEDs exhibit CIE color coordinates of (0.335, 0.344) with a maximum luminous efficiency of 36.4 lm/W. More importantly, the color gamut of the WLEDs can cover over 124% of the NTSC standard, which is much higher than 86% of the traditional phosphor-based WLEDs and 104% of the CdSe-based WLEDs [35–39], which indicates that the green CsPbBr<sub>3</sub>-AA composites have potential applications in LCD devices.



**Figure 5.** Photographs of (a) unlighted and (b) lighted of the WLEDs operated at 3.7 V and 20 mA. (c) EL spectrum of the WLEDs at 20 mA and 3.7 V, as well as (d) the CIE coordinates.

#### 4. Conclusions

In summary, we demonstrated a facile strategy for the synthesis of green CsPbBr<sub>3</sub>-AA composites using AA as the surface ligand and matrix by a hot-injection method. The green CsPbBr<sub>3</sub>-AA composites not only preserve a high PLQY of 86.43% and a narrow FWHM of 23 nm, but also show enhanced stability. At 60 °C, the CsPbBr<sub>3</sub>-AA composites still maintained 90% of the initial intensity, which is much higher than that of the pure CsPbBr<sub>3</sub> NCs. Finally, WLEDs were fabricated by combining the green CsPbBr<sub>3</sub>-AA composites with commercial red K<sub>2</sub>SiF<sub>6</sub>:Mn<sup>4+</sup> phosphors and blue LED chips, which exhibited a luminous efficiency of 36.4 lm/W and an ultrawide color gamut of 124% NTSC.

**Author Contributions:** Conceptualization: M.W., S.W. and T.X.; Methodology: M.W., S.W., R.C., M.Z., Y.L., H.D., J.R., T.X., H.L.; formal analysis: M.W. and S.W.; Investigation: M.W. and S.W.; Resources: All materials, devices and laboratories were provided by the Engineering Research Center for Nanophotonics and Advanced Instrument; Writing—Original Draft: M.W.; Writing—Review & Editing: M.W., T.X. and H.L.; Supervision: T.X. and H.L.; Project Administration: H.L.; Funding Acquisition: T.X. and H.L. All authors have read and agreed to the published version of the manuscript.



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**Conflicts of Interest:** The authors declare that they have no conflict of interest.

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