Novel ultra-stable and highly luminescent white light-emitting diodes from perovskite quantum dots—Polymer nanofibers through biaxial electrospinning

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Novel ultra-stable and highly luminescent white light-emitting diodes from perovskite quantum dots—Polymer nanofibers through biaxial electrospinning

**I. INTRODUCTION**

The efficient photovoltaic cells based on organometal halide perovskites were demonstrated for the first time in 2009. Since then, the organic/inorganic hybrid lead halide perovskites have attracted extensive attention worldwide for optoelectronic device applications. Organic/inorganic perovskite materials possess excellent optoelectronic and photovoltaic properties, such as a high photoluminescence quantum yield (PLQY), long lifetime, narrow emission bandwidth, and tunable light emission. Such advantages can be applied to light-emitting diode (LED) devices, transistor devices, solar cells, nanolasers, and bioimaging. However, perovskites suffer from poor stability due to their ionic nature that is sensitive to moisture and low melting temperature, which limits their practical applications.
has raised major concerns about their large-scale applicability.\(^8,9\) Therefore, the practical use of perovskites usually requires additional protective strategies to overcome degradation in the ambient atmosphere.

Numerous methods have been applied to enhance the stability of perovskites, such as the macroscale polymer matrix method,\(^1\) protective coatings,\(^4,11,12\) synthetic modification,\(^13\) and polymer blends.\(^14–16\) However, to date, only limited progress has been made in enhancing the stability. In practice, dense polymer matrices endow water-sensitive materials with hydrophobic characteristics. Encapsulation by a polymer matrix is an effective strategy to improve the stability of perovskite quantum dots (QDs), especially water resistance. For example, Wu et al.\(^1\) reported that a swelling-deswelling microencapsulation strategy could thoroughly disperse organic–inorganic perovskite nanoparticles. The PLQY could be as high as 48%, and the device could be immersed in boiling water and still maintain more than 65% of its initial brightness after 5 h. Chen et al.\(^1\) reported that the composite nano-/micrometer beads of perovskite quantum dots embedded in polystyrene showed similar phenomena and could be combined with cross-linked PS beads to retain 68% of the PLQY. The composites remained strongly luminescent after being immersed in water for over 9 months. Furthermore, even when immersed in acid/alkali aqueous solution, phosphate buffer solution, and Dulbecco’s modified eagle medium biological buffer, this type of device still emitted high luminescence. Allvisatos et al.\(^13\) revealed a ligand-assisted modification technique to produce perovskite QDs by grafting long alkyl ligand chains onto surfaces that interacted with hydrophobic polymers to enhance water stability. Moreover, the conditions of polarization and color tunable properties could be achieved by changing the morphology of the nanocrystalline component. Zhang et al.\(^10\) used a one-pot strategy to make perovskite-polymer composites that were stable and maintained a PLQY higher than 60%. A white device was also successfully prepared by the perovskite–polymer composites. Despite these advances, the search of other facile methods is ongoing in the hope of achieving a commercially viable preparation for perovskite materials that would be suitable for optoelectronic devices.

Different from typical thin-film preparation methods, electrospinning (ES) is a technique that has been widely used to prepare fibers because of its low-cost, simple operation, and capability of fine-tuning the morphology of the fibers in nano- to microscale. Because ES can produce fibers with high surface-to-volume ratios, the ES fibers are good candidates for sensory applications. For the past several years, diverse fluorescent sensory applications based on conjugated polymer nanofibers have been successfully prepared by our group.\(^18–21\) Another important effect of ES fibers is the geometrical confinement to the implants embedded in the fibers, which can adjust the orientation, crystallinity, stability, and optoelectronic properties of the implants. Furthermore, the diameter and morphology of the fibers can be easily controlled by ES parameters.\(^18–20\) Functional materials such as metallic nanoparticles,\(^18,21,22\) conjugated polymers,\(^23–25\) perovskite,\(^19\) and QDs\(^26\) can be incorporated into ES fibers to prepare composite fibers for various device applications. For example, Chen et al.\(^19\) exploited the one-step coaxial ES technique to fabricate the perovskite and polymer in the core/shell nanofibers. Owing to the hydrophobic characteristics of the shell polymer, the perovskite crystals in the core are

![FIG. 1. (a) and (b)](Schematic of the CsPbX\(_3\) (X = Cl, Br, I) nanofiber fabrication process. (c) Optical image of blue, green, and red composite nanofibers under UV-light (365 nm) excitation. (d) Schematic for the configuration of the prototype WLED device.)
highly stable in the atmosphere and the aggregation is mitigated due to geometrical confinement. The PLQY of perovskite nanofiber is 30.9\%, and the photoluminescence intensity can retain 50\% of the initial intensity when it is immersed in water for 48 h. Furthermore, our group successfully used the fibrous mats packaging approach in bright perovskite LEDs. The correlated color temperatures (CCT) of white LEDs (WLEDs) can be easily controlled by adjusting the thickness of the nanofibers and the voltage. Given these advantages, we proposed that ES is a suitable method to stably maintain the pristine optical properties of perovskites. We thus investigated the effects of polymers on perovskites embedded in fibers, which is a topic that has not been thoroughly explored.

In this study, we exploited a simple strategy to prepare perovskite–polymer fibers, that is, the electrospinning of the composite fibers from the solutions where the perovskite QDs and polymer are mixed. We used three polymers, including poly(styrene-butadiene-styrene) (SBS), which is a rubber and can be easily prepared into ES fibers, poly(methyl methacrylate) (PMMA), which is a highly transparent polymer with high glass transition temperature, and polystyrene (PS), which is highly stable when immersed in water due to its hydrophobic characteristics. We further used the biaxial electrospinning method to create the core/shell structure in the fibers, where red perovskite materials serve as the core and green materials serve as the shell. The fibrous mats were used to package the blue LED chips to obtain efficient perovskite-based WLEDs. Unexpectedly, the luminance and efficiency of the core/shell WLED devices are as high as 430\% and 420\% of the values of the multilayered devices, respectively. The approach reported herein provides a promising avenue to fabricate stable and highly efficient perovskite-based WLEDs.

![Figure 2](image-url)
II. RESULTS AND DISCUSSION

The solutions for electrospinning were prepared from a stock solution of colloidal CsPbX₃ perovskite QDs in toluene (200 μl, ~20 mg ml⁻¹) mixed with polymers (SBS, PMMA, or PS) in dichloromethane (DCM) (1 ml, ~15 wt. %), as illustrated in Fig. 1(a). The QDs can be well dispersed, and polymers can be dissolved in the mixed solutions without precipitation. Figure 1(b) shows the schematic of the uniaxial ES setup with the mixed solutions. The feed rate and voltage for the ES process were 0.8–1.0 ml h⁻¹ and 12.4 kV, respectively. The colors of the perovskite–polymer fibers were tuned by adjusting halide ratios. The red, green, and blue fibrous mats are displayed in Fig. 1(c), which, as the primary colors, can mix to produce other colors. Subsequently, the perovskite-polymer fibrous mats were applied in blue LED chips to radiate while light, as shown in Fig. 1(d).

A. Characterization of perovskite fibers

The SEM images of the fibers with CsPbBr₃ perovskite QDs embedded are shown in Figs. 2(a)–2(c). The average diameters of the fibers were approximately 1 μm. The elemental analyses of the perovskite-polymer fibers are provided in Fig. S1. The TEM images that reveal the distribution of the perovskite QDs in the fibers are shown in Figs. 2(d)–2(f). The perovskite crystals can be clearly distinguished from the polymer matrices owing to the higher electron density. Among the fibers, CsPbBr₃@SBS exhibited less aggregation of the QDs, and we speculated that the better dispersion is because SBS contains polybutadiene hydrocarbon segments that are more compatible with the ligands (oleylamine and oleic acid) on the QD surface, similar to the phenomena reported previously. The severest aggregation occurred in CsPbBr₃@PS fibers, indicating the low compatibility between the ligands and PS.

High-resolution TEM (HRTEM) was used to investigate the perovskite crystals, as shown in Figs. 2(g)–2(i) and S2. The insets in Figs. 2(g)–2(i) are the fast Fourier transformation patterns from the HRTEM images. The CsPbBr₃ crystals were cubic phase with an average size of 9 ± 1 nm and a d-spacing of 0.58 nm for the (100) plane. After electrospinning, the particle size was similar to the size of the original perovskite QDs. The broad humps below 2θ = 40° found in the profiles of the fibers are the scattering halos produced by the amorphous polymers. We further conducted optical imaging for green CsPbBr₃ and red CsPbBr₃@PS, and CsPbBr₃ NPs standard JCPDS. The contact angles of neat polymer and CsPbBr₃-polymer in the forms of films and fibrous mats are listed in Table I. A significant increase in the hydrophobicity of fibrous mats compared to the corresponding films is attributed to the voids between fibers that can trap air.

The contact angles of neat polymer and CsPbBr₃-polymer in the forms of films and fibrous mats are listed in Table I. A significant increase in the hydrophobicity of fibrous mats compared to the corresponding films is attributed to the voids between fibers that can trap air. The contact angles of the films with perovskites are not significantly different from those of neat polymers, indicating that perovskites tend to be buried inside the polymers due to their high surface tension. For the neat polymer and perovskite-polymer

| Table I: Contact angle values of the samples |
|---------------------------------------------|
|                                        |
| SBS film | PMMA film | PS film | SBS fiber | PMMA fiber | PS fiber |
| Contact angle (deg) | 81.29 | 67.98 | 85.2 | 104.35 | 117.35 | 134.66 |
| CsPbBr₃@SBS film | CsPbBr₃@PMMA film | CsPbBr₃@PS film | CsPbBr₃@SBS fiber | CsPbBr₃@PMMA fiber | CsPbBr₃@PS fiber |
| Contact angle (deg) | 79.71 | 67.92 | 84.4 | 103.41 | 122.99 | 141.78 |

...
CsPbBr\textsubscript{3} was found to be in the order of CsPbBr\textsubscript{3} due to the reaction with CO\textsubscript{2} for 6 h. The PL of the perovskites degraded under UV illumination. The redshift in the order of PS > PMMA > SBS. The PL peaks of CsPbBr\textsubscript{3} fibrous mats, the contact angle followed the order PS > PMMA > SBS. We used the high-resolution SEM (HRSEM) to observe the fibers, as illustrated in Figs. 4(a)–4(c), and found that the surfaces of the CsPbBr\textsubscript{3}@PS and CsPbBr\textsubscript{3}@PMMA fibers are porous, whereas the CsPbBr\textsubscript{3}@SBS fibers are smooth. The porous structure may be responsible for the further enhancement of the hydrophobicity for PS and PMMA fibers.

B. Photoluminescence and stability

Figure S4 displays the photoluminescence (PL) spectra of the CsPbBr\textsubscript{3} QD-based fibers, and the data are summarized in Table II. The PL peaks of CsPbBr\textsubscript{3} in polymers were slightly redshifted from that of CsPbBr\textsubscript{3} QDs in nanofiber film, from 511 nm to 513, 515, and 516 nm for SBS, PMMA, and PS, respectively, as illustrated in Fig. S4. The redshift can be attributed to the aggregation of perovskite QDs, which facilitates nonradiative recombination.\textsuperscript{31} The redshift in the order of PS > PMMA > SBS and the photoluminescence quantum yield (PLQY) in reverse order (Table II) agree with the TEM images [Figs. 2(d)–2(f)], where CsPbBr\textsubscript{3}@PS fibers show the severest aggregation.

Figure 5(a) displays the time-dependent PL intensity of the CsPbBr\textsubscript{3} polymer fibers under UV LED chip (405 nm) excitation for 6 h. The PL of the perovskites degraded under UV illumination due to the reaction with CO\textsubscript{2}, O\textsubscript{2}, and H\textsubscript{2}O\textsuperscript{+} and the UV stability was found to be in the order of CsPbBr\textsubscript{3}@PS > CsPbBr\textsubscript{3}@PMMA > CsPbBr\textsubscript{3}@SBS. To examine the water resistance, we immersed the samples in a water bath, and the PL intensity as a function of immersing time is shown in Fig. 5(b). The PL intensities dropped to 0%, 50%, and 83% of the initial values for SBS, PMMA, and PS, respectively, after 3 months. Note that after 3 months, the PL peaks red-shifted possibly because of the faster degradation of the smaller QD populations that emit lights of shorter wavelengths. This is accompanied by a decrease in the full width at half maximum (FWHM) of the emissions as shown in Fig. S5. Both the above tests suggests that the rigid, hydrophobic PS can effectively avoid the diffusion of active gases and water into fibers to prevent the perovskites from degradation, while the SBS with soft polybutadiene segments provides less protection against those molecules. The superior UV and water stabilities of the CsPbBr\textsubscript{3}@PS fibers are due to the fact that the higher hydrophobicity lowers the affinity between PS and the polar molecules, and the high rigidity at room temperature retard the motion of the molecules inside PS fibers.

Thermal stability is also a crucial index of perovskite-polymer composites, which was tested in the present study by monitoring the PL spectra under thermal circles. Figure 5(c) compares the PL intensity of the samples heated and then cooled between 20 and 120 °C. The PL intensity for all the samples decreased with increasing temperature and then increased upon cooling. The CsPbBr\textsubscript{3}@PMMA fibers show the highest reversibility with nearly overlapped heating and cooling curves while the PL intensity of the CsPbBr\textsubscript{3}@PS fibers is less than 40% of the initial value when cooling back to 20 °C after heating to 120 °C. Such a polymer-dependent thermal stability is correlated with the glass transition temperature $T_g$ of the polymers. The $T_g$s of the fibers were determined using a dynamic mechanical analyzer (DMA) shown in Fig. S6. The $T_g$ of CsPbBr\textsubscript{3}@PMMA is 157 °C below which the PMMA chains are frozen and the

| TABLE II. Preparation conditions and characteristics of the samples. |
|---------------------------------------------------------------|
| Sample | Polymer concentration (wt. %) | Diameter (μm)\textsuperscript{a} | PL peak position (nm) | PL FWHM (nm)\textsuperscript{b} | PLQY value (%) | CIE (X, Y) |
|--------|-------------------------------|------------------|-------------------|-----------------|----------------|------------|
| CsPbBr\textsubscript{3} QDs | NA | NA | 511 | 20 | 70 | (0.0623, 0.7155) |
| CsPbBr\textsubscript{3}@SBS fiber | 15 | ~1.5 | 513 | 21 | 29 | (0.0736, 0.7116) |
| CsPbBr\textsubscript{3}@PMMA fiber | 15 | ~1.5 | 515 | 22 | 27 | (0.0898, 0.7498) |
| CsPbBr\textsubscript{3}@PS fiber | 15 | ~1.5 | 516 | 23 | 23 | (0.0873, 0.7373) |
| CsPbBr\textsubscript{3}@SBS film | 15 | NA | 515 | 22 | 26.7 | (0.0777, 0.7398) |
| CsPbBr\textsubscript{3}@PMMA film | 15 | NA | 515 | 22 | 23.5 | (0.0736, 0.7383) |
| CsPbBr\textsubscript{3}@PS film | 15 | NA | 515 | 21 | 22 | (0.0791, 0.7381) |

\textsuperscript{a}Diameter of the ES nanofiber.

\textsuperscript{b}The line broadening at half the maximum intensity of the PL peak.
morphology inside the fibers is nearly fixed, which explains why the decay rate of the PL intensity is low upon heating and the reversibility is high below 120 °C. The $T_g$ of CsPbBr$_3$@PS is 109 °C. In other words, the PS chains become mobile at 120 °C, which along with the low compatibility between PS and QDs causes the QDs to merge irreversibly, thus leading to the rapid decay of the PL intensity and the low reversibility. To confirm this argument, the CsPbBr$_3$@PS fibers were heated to 80 °C instead of 120 °C and the PL profiles are shown in Fig. 5(d). The reversibility can indeed be greatly improved when the thermal cycle was performed below $T_g$: CsPbBr$_3$@SBS fibers show two $T_g$: −91 °C from polybutadiene segments and 112 °C from PS segments. The highly mobile polybutadiene segments that allow a faster diffusion of molecules are responsible for the low UV and water stability of CsPbBr$_3$@SBS fibers at room temperature as described above. However, the higher compatibility between polybutadiene and the QDs impart a better thermal stability and reversibility to CsPbBr$_3$@SBS fibers in comparison with CsPbBr$_3$@PS, even though SBS also bears PS segments.

Figure S7 shows the temperature-dependent PL data within the reversible temperature range from 78 K to 300 K, which were fitted by

$$I(T) = \frac{I_0}{1 + Ae^{-E_b/k_BT}},$$

where $I_0$ is the intensity at 0 K, $E_b$ is the exciton binding energy, $k_B$ is Boltzmann constant, and higher $E_b$ indicates a better dispersion of QDs in polymers. The $E_b$ values for CsPbBr$_3$@SBS, CsPbBr$_3$@PMMA, and CsPbBr$_3$@PS are 55, 46, and 39 meV, respectively. The low $E_b$ for CsPbBr$_3$@PS fiber implies a poor dispersion of QDs in PS, consistent with the TEM images [Figs. 2(d)–2(f)].

Figure 6 shows the time-resolved PL of CsPbBr$_3$-based QDs and fibers. The curves were fitted with the following equation:

$$A(t) = A_1 \exp\left(\frac{-t}{\tau_1}\right) + A_2 \exp\left(\frac{-t}{\tau_2}\right),$$

where $\tau_1$ and $\tau_2$ are the lifetime parameters. The PL profiles of CsPbBr$_3$@SBS, CsPbBr$_3$@PMMA, CsPbBr$_3$@PS, and CsPbBr$_3$ NPs are shown in Fig. 6.
TABLE III. Time-resolved PL spectra of CsPbBr$_3$@SBS, CsPbBr$_3$@PMMA, CsPbBr$_3$@PS fiber, and CsPbBr$_3$ NPs.

|                  | $\tau_1$ (ns) | $A_1$ (%) | $\tau_2$ (ns) | $A_2$ (%) | $\tau_{avg}$ |
|------------------|--------------|-----------|--------------|-----------|--------------|
| CsPbBr$_3$@SBS fiber | 1.061 01    | 76        | 5.007 9      | 24        | 3.431 296   |
| CsPbBr$_3$@PMMA fiber | 0.947 23    | 78        | 4.709 87     | 22        | 3.169 109   |
| CsPbBr$_3$@PS fiber  | 0.900 87    | 77        | 4.095 33     | 23        | 2.740 836   |
| CsPbBr$_3$ film    | 0.850 17    | 77        | 3.772 5      | 23        | 2.518 881   |

where $\tau_1$ is the short-lived lifetime attributed to the intrinsic recombination, $\tau_2$ is the long-lived lifetime from the surface-state recombination, and $A_1$ and $A_2$ are the fractions of the two different contributions. The fitting parameters are listed in Table III. $\tau_1$s were 0.8, 1.1, 0.9, and 0.9 ns and $\tau_2$s were 3.7, 5.0, 4.7, and 4.1 ns for CsPbBr$_3$ QDs, CsPbBr$_3$@SBS fiber, CsPbBr$_3$@PMMA fiber, and CsPbBr$_3$@PS fiber, respectively. $\tau_1$ has been reported to reflect the trapping of excitons by crystal defects. The higher $\tau_1$ for the CsPbBr$_3$–polymer fibers suggested that less nonradiative recombination occurred in the perovskite embedded in the fibers. $\tau_2$ corresponds to the surface defects of the perovskite crystals, and the perovskite-polymer fibers show greater probability of radiative recombination than the neat QDs, as implied by the higher $\tau_2$. As previously discussed, the polymers form a protection layer for the embedded perovskites such that the surface defects caused by CO$_2$, O$_2$, and H$_2$O in the ambient condition are reduced.

In sum, the above results suggest that the polymers can impart different characteristics to the perovskite-polymer fibers. CsPbBr$_3$@SBS fibers demonstrated the highest PLQY and the longest PL lifetime, which could be useful for applications in...
high-efficiency optoelectronic devices, and CsPbBr$_3$@PMMA exhibited excellent thermal stability, which could be utilized in high-voltage LEDs. Although CsPbBr$_3$@PS exhibited the shortest PL lifetime and the largest PL shift, it possessed the highest UV and water stability, which could provide constant fluorescent intensity under atmosphere.

C. White-light-emitting applications

We have demonstrated that the perovskite-polymer fibers are color-tunable and stable. We further developed white-light-emitting fibers prepared by the core/shell biaxial electrospinning in which the red and green perovskite materials served as the core and the shell, respectively. Figure 7(a) illustrates the setup and the process of the biaxial electrospinning. The Taylor cone with red CsPb(Br$_{0.4}$I$_{0.6}$)$_3$QDs @PS in the core and green CsPbBr$_3$ QDs @PS in the shell could be seen. The confocal image clearly shows the distinct red and green perovskites inside the fiber. The WLEDs were fabricated by using a single layer of core/shell fibrous mats on a commercial blue chip ($\lambda_{\text{max}} = 450$ nm), which were then compared with the devices with multilayers of single-colored green and red fibers, as illustrated in Fig. 7(d). The single layer of the core/shell fibers markedly enhanced the WLED performance and exhibited the luminescence and efficiency of 400% higher than those of the multilayer structures, as shown in Figs. 7(b) and 7(c). The great improvement is attributed to the efficient energy transfer of photoluminescence between the perovskites in the core and shell inside the fibers [Fig. 7(d)]. Note that in Fig. 7(d), the multilayer means using two kinds of fiber mats, red “CsPb(Br$_{0.4}$I$_{0.6}$)$_3$ QDs @PS” and “green CsPbBr$_3$ QDs @PS,” which were separated and collected on the LED chip. However, single layer means just using one kind of fiber mat, red CsPb(Br$_{0.4}$I$_{0.6}$)$_3$ QDs @PS (core), and green CsPbBr$_3$ QDs @PS (shell), which were directly collected on the LED chip. Moreover, the fiber mat thickness of single-layered and multilayered WLED device is the same (0.3 mm $\pm$ 0.05 mm) by controlling the collecting time of ES process.

The CIE color coordinates and CCT of WLEDs with a single layer of the core/shell fibers can be controlled by adjusting the voltages, which are shown in Fig. 8(a) and listed in Table S1. Furthermore, warm WLED, natural WLED, and cold WLED devices were successfully fabricated by adjusting the thicknesses of the fibrous mats under blue LED excitation [Figs. 8(b)–8(d)]. The insets in Figs. 8(b)–8(d) display the WLED devices with different CCTs. By controlling both the spacing between the LED chip and the fibrous mat and the porosity of the fibrous mats, the degradation of the temperature-sensitive perovskites can be reduced. The packaging method developed in this work can enhance the device performance as well as improve the stability.

III. CONCLUSION

In summary, we adopted a simple electrospinning method by mixing cesium lead halide perovskite quantum dots and polymers to prepare stable luminescent fibers. The color can be tuned by changing the halide ratios in the perovskites, and the properties of the fibers are affected by the polymers. The higher compatibility
between QDs and SBS rubber leads to a better dispersion of the QDs in the fibers, the higher hydrophobicity of PS imparts higher UV and water stability to the perovskites, and the higher glass transition temperature of PMMA gives superior thermal stability of the fibers. The optimized fibers with perovskite QDs in PS yielded a high PLQY of 23.3%, which surpassed the value of the thin-film counterpart. The PL intensity of the fibers retained 83% of initial intensity for 3 months when immersed in water. Further we demonstrated a new biaxial electrospinning strategy to prepare core/shell fibers with red and green colors, respectively, which produce white light with high luminance and high efficiency when irradiated by a blue LED due to the efficient energy transfer of photoluminescence. These findings reveal the potential of the electrospinning techniques to fabricate stable and efficient luminescent perovskite fibers.

IV. EXPERIMENTAL SECTION

A. Materials

Oleylamine (primary amine, >98%), oleic acid (90%), cesium carbonate (99.995%), 1-octadecene (90%), lead(II) bromide, poly(styrene-butadiene-styrene) (SBS) (Mw = 142 000), poly(methyl methacrylate) (PMMA) (Mw = 350 000), polystyrene (PS) (Mw = 192 000), and dichloromethane (99.5%) were purchased from Sigma Aldrich.

B. Preparation of CsPbX₃ (X = Cl, Br, and I)

For the preparation of the Cs-oleate precursor, CsCO₃ (0.2035 g) and oleic acid (0.625 ml) were dissolved in 1-octadecene (10 ml) and added to a 25-ml two-neck flask. The solution was heated under vacuum at 120 °C for 1 h under stirring. After 1 h, the reaction flask was stored with N₂, and the temperature was heated to 150 °C. The Cs-oleate in 1-oleylamine solution was stored in N₂ until required for QD synthesis.

PbBr₂ (0.13818 g) and 1-octadecyl (ODE) (10 ml) were added in a 25-ml two-neck flask under vacuum at 120 °C for 1 h under stirring. After 1 h, the reaction flask was stored with N₂ and oleylamine (1 ml) and oleic acid (1 ml) were added to stabilize the lead precursors and as-synthesized NCs. Subsequently, the flask was heated to 170 °C, followed by rapid injection of 0.8 ml of Cs-oleate solution. The solution was then quenched in an ice bath after 5 s. After the temperature of the green colloidal solution was cooled to 25 °C, it was centrifuged at 4000 rpm for 10 min to separate the upper solution and precipitates. Toluene (0.6 ml) was added to the solid precipitates to be stored under vacuum.

C. Preparation of the ES fibers and thin film

The polymer solution was fed into a plastic syringe and expelled using syringe pumps (KD Scientific Model 100, USA) at a feed rate of 0.8–1.0 ml h⁻¹. The tip of the metallic needle was connected to a high-voltage power supply (Chargemaster CH30P SIMCO, USA) that was set at 12.4 kV during ES. When the syringe pumps started, the polymer solution was expelled from a metallic needle and formed a nanoscale fiber at high voltage. A piece of aluminum foil or quartz was placed 15 cm below the tip of the needle for 1 h to collect the ES nanofibers. All experiments were performed at room temperature with a relative humidity of approximately 20%.

D. Preparation of the biaxial ES of core [CsPb(Br₀.₆I₀.₄) QDs]/shell (CsPbBr₃ QDs) nanofibers

Fabrication was similar to the uniaxial ES technique. Two syringes were used, and each syringe was connected to a separate needle; the diameters of the core and shell needles were 0.80 and 1.25 mm, respectively. The CsPbBr₃/α-oleate@polymer solution was fed into the core syringe, and the CsPbBr₃@polymer solution was fed into the shell syringe to form the two-fluid coaxial ES system, with feed rates of 0.6 and 0.8 ml h⁻¹, respectively. The tip of the metallic needle was connected to a high-voltage power supply that was set at 12.4 kV during ES. The same process condition was applied with a piece of aluminum foil or quartz placed 15 cm below the tip of the needle for 1 h to collect the ES nanofibers. All experiments were performed at room temperature with a relative humidity of approximately 20%. These processes were monitored by using a high-speed charge-coupled device camera (USA Xilix 3M USB 2.0) to ensure that the core solution was wrapped by the shell solution.

E. Characterization

Characterization XRD patterns were recorded using Cu Kα radiation (λ = 1.5418 Å) on a D2 Bruker diffractometer operating at 40 kV and 20 mA. The morphologies of the polymer@CsPbBr₃ nanofibers were characterized using a scanning electron microscope (Hitachi S-4700). Samples were coated with platinum prior to SEM characterization, and analysis was performed at an acceleration voltage of 15 kV. TEM images were obtained using a Hitachi H-7100 field emission electron microscope, operating at 75 kV. Fluorescence optical microscopy images were taken using a two-photon laser confocal microscope (Leica LCS SPS). PL spectra were measured by using a Fluorolog-3 spectrophotometer (Horiba Jobin Yvon), and the CsPbBr₃@polymer and CsPbBr₃@polymer samples were excited at wavelengths of 350 and 460 nm, respectively. The water-resistant properties of the CsPbBr₃@polymer fiber film were observed through contact angle measurement (Phoenix 300 Touch, GAT Scientific) of water droplets on the samples. The CIE color coordinates and CCT of the WLED spectra were measured by PR670 under ambient air conditions at 25 °C. The calibration method was standard sunset light, which correlated with a color temperature of 2856 K to confirm instrument correctness. The TR-PL spectra were coupled to a spectrometer (Horiba iHR320) with a Hamamatsu C10910 streak camera and an M10913 slow single sweep unit. Temperature-dependent photoluminescence was measured with a pulsed diode-laser (PicoQuant LDH-D-C-375) at a repetition rate of 1 MHz (power density: 3.63 μW cm⁻²). The excitation wavelength was 375 nm. The samples were mounted in a liquid nitrogen cryostat (Janis ST-500-UC) in vacuum (10⁻⁶ Torr), and the sample was measured from 300 to 78 K.

SUPPLEMENTARY MATERIAL

See the supplementary material for the energy dispersive spectroscopy (EDS) mapping that was conducted for the CsPbBr₃ and CsPb(Br₀.₆I₀.₄) fiber films, HRTEM of the CsPbBr₃ fiber film was conducted, and confocal images of the CsPbBr₃ and CsPb(Br₀.₆I₀.₄)
fiber films were recorded. The PL spectra of the samples, PL peak and FWHM of CsPbBr$_3$@PMMA fiber and CsPbBr$_3$@PS fiber, and DMA analysis of the CsPbBr$_3$-polymer fiber film were conducted. The low temperature-dependent PL was measured. Properties of WLED fabricated using core/shell biaxial perovskite@PS nanofibers with blue-chip LED ($\lambda_{\text{max}} = 450$ nm) excitation under the operating current 10 mA were also discussed.

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