

Light-Emitting Diodes

Pattern-Matched Polymer Ligands Toward Near-Perfect Synergistic Passivation for High-Performance and Stable Br/Cl Mixed Perovskite Light-Emitting Diodes

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Abstract: Mixed Br/Cl perovskite nanocrystals (PeNCs) exhibit bright pure-blue emission benefiting for fulfilling the Rec. 2100 standard. However, phase segregation remains a significant challenge that severely affects the stability and emission spectrum of perovskite light-emitting diodes (PeLEDs). Here, we demonstrate the optimization of the spacing between polydentate functional groups of polymer ligands to match the surface pattern of CsPbBr_{1.8}Cl_{1.2} PeNCs, resulting in effective synergistic passivation effect and significant improvements in PeLED performances. The block and alternating copolymers with different inter-functional group spacing are facilely synthesized as ligands for PeNCs. Surprisingly, block copolymers with a higher functional group density do not match PeNCs, while alternating copolymers enable efficient PeNCs with the high photoluminescence intensity, low non-radiative recombination rate and high exciton binding energy. Density functional theory calculations clearly confirm the almost perfect match between alternating copolymers and PeNCs. Finally, pure-blue PeLEDs are achieved with the emission at 467 nm and Commission Internationale de l'Éclairage (CIE) coordinates of (0.131, 0.071), high external quantum efficiency (9.1 %) and record spectral and operational stabilities (~80 mins) in mixed-halide PeLEDs. Overall, this study contributes to designing the polymer ligands and promoting the development of

high-performance and stable pure-color PeLEDs towards display applications.

Introduction

Mixed-halide perovskite nanocrystals (PeNCs) have shown significant advantages in the field of light-emitting diodes (LEDs) due to their tunable band gap, high photoluminescence quantum yields (PLQY), excellent color purity, low cost, and facile synthesis, making them promising candidates for display and lighting applications.^[1] In particular, by mixing halogens in specific proportions, their luminescence can be easily achieved across the entire visible light range.^[2] The facile emission spectrum tuning characteristics, which are not dependent on strong quantum confinement effects, minimizing influences of the particle size distribution on the color purity, which is particularly advantageous for achieving high-quality and accurate color reproduction in electronic displays, as required by Rec. 2100 standard. For instance, mixed Br/Cl PeNCs enable the pure-blue luminescence at a peak wavelength of 467 nm and Commission Internationale de l'Éclairage (CIE) coordinates close to the standard (0.131, 0.046) without the need for additional size purification steps. However, their propensity for phase segregation during continuous operation seriously limits the spectral and operational stabilities of perovskite LEDs (PeLEDs), dominantly hindering their practical applications.^[3] Ligand passivation has been reported as a particularly effective strategy for suppressing halide segregation, and thus improving the efficiency and stability of PeLEDs.^[4] On the other hand, the length of ligand chains determines charge transfer characteristics which is particularly crucial for pure-blue PeLEDs with large band gaps.^[5]

Polymer ligands containing multi-dentate functional groups can form very strong interactions and passivation effects with the surface of PeNCs due to the synergistic effect between adjacent coordination bonds.^[6] Moreover, theoretical calculations have stated that the binding energy between polymer ligands and perovskite increases monotonically with the number of repeating units, indicating the potential for optimal passivation effects of polymer ligands.^[7] According to previous studies, the band edges of perovskite are primarily dependent on lead and halogen orbitals, and the main defect sites are often considered as

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uncoordinated Pb ions.^[8] Considering the neat surface of cubic mixed Br/Cl PeNCs with regular pattern arrangement of Pb²⁺ locations, the degree of spacing matching between the repeating functional groups of the polymer ligand and the uncoordinated ions at the surface of PeNCs is crucial for achieving perfect synergistic passivation effect. However, there are a few studies on designing the structure of polymer ligands, especially the spacing between functional groups, which results in the poor matching in PeNCs.^[9] Most polymers are reported as host materials for photoluminescence (PL) and their passivation effects are very limited.^[10]

In this work, we focus on tuning the functional group densities of polymer ligands to match the Pb-pattern of pure-blue mixed-halide PeNCs, and further investigate the influence of the structure of polymer ligands on optoelectronic properties and performances of pure-blue PeLEDs. Pattern matching was controlled by selecting alternating copolymers (ACP) and block copolymer (BCP) matrices with similar functional groups, and a length-optimized short-chain amine was grafted onto the main chain to enhance the oil solubility. Surprisingly, dense functional groups did not result in very strong interactions with PeNCs, whereas the functional group spacing of alternating copolymers better matched the ion distribution on the surface of CsPbBr_xCl_{3-x}, which was both experimental and theoretical verified. Due to the excellent passivation effect of the polymer ligands, the obtained PeNCs exhibited outstanding PL properties, and very low defect density and Pb⁰ content. In addition, the shortened alkyl chain improved electrical properties of the PeNCs film, including high mobility and low turn-on voltage of PeLEDs. Finally, the polymer ligands simultaneously exhibited effective synergistic passivation and improved charge transfer resulting in high performance pure-blue PeLEDs with an emission peak at 467 nm, CIE coordinates of (0.131, 0.071), high external quantum efficiency (EQE) of 9.1 %, and recorded spectral stability and operational lifetime (T_{50} ~80 mins).

Results and Discussion

The control Br/Cl-mixed PeNCs (C-PeNCs) were synthesized via a two-step approach. Initially, CsPbBr₃ was synthesized using hot injection method with traditional ligands, oleic acid (OA) and oleylamine (OAm), as depicted in Figure 1a.^[14] Subsequently, CsPbBr_xCl_{3-x} was obtained through halide exchange, using trioctylphosphine oxide (TOPO)-assisted dissolved ZnCl₂.^[11] This method effectively resolved issues such as PbCl₂ precipitation at high temperatures and differences in the crystallization rates of various halide perovskites. Regarding the polymer ligands, two polymers including a BCP of poly(styrene-co-maleic anhydride) and a ACP of poly(methyl vinyl ether-alt-maleic acid), as illustrated in Figure 1b and c, were chosen as polymer matrices to react with short-chain di-n-hexylamine to yield polymer ligands, named BCPN and ACPN, respectively. Notably, the aforementioned amidation reaction was essential to enhance the oil solubility, enabling the direct addition of the as-synthesized polymer ligands to the

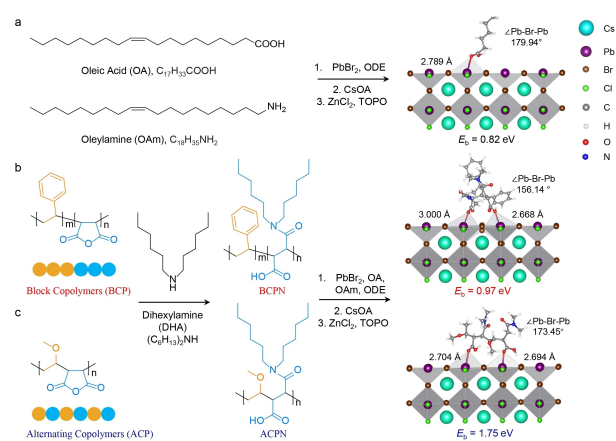


Figure 1. Synthesis process of (a) conventional C-PeNCs utilizing only OA and OAm as ligands; (b) B-PeNCs incorporating BCPN; and (c) A-PeNCs incorporating ACPN. The orange and light blue dots on the left panel represent different arrangements of monomers in the copolymer, which determine the spacing between carboxyl groups. The right panel present the optimized configuration structure of the ligands interacting with PeNCs based on DFT calculation, including the respective $\angle\text{Pb-Br-Pb}$, coordination bond lengths and E_b .

hot injection reaction solution. C₆H₁₃ group was optimized as the shortest alkyl chain due to the insolubility of the polymer ligands obtained by di-n-butylamine in the synthesis environment (Figure S1). The PeNCs synthesized by BCPN and ACPN ligands, were designated as B-PeNCs and A-PeNCs, respectively (see more details in Methods section).

Density functional theoretical (DFT) calculations were conducted to explore the optimal configuration of polymer ligands interacting with CsPbBr_xCl_{3-x}, and the associated binding energy (E_b).^[12] The binding energy (E_b) of each adsorbate was calculated using the formula: $E_b = E_{\text{ligand/surf}} - E_{\text{ligand}} - E_{\text{surf}}$, where $E_{\text{ligand/surf}}$ is energy of the surface with the bound polymer ligand, E_{ligand} is the energy of the isolated ligand polymer, and E_{surf} is the energy of the bare perovskite surface. As shown in the right panel of Figure 1 and Figure S2, the carboxyl groups of OA coordinate with the bare Pb in Br/Cl-mixed halide perovskites, resulting a E_b of -0.82 eV in C-PeNCs. In the case of B-PeNCs, despite there are two carboxyl groups, the E_b is only -0.97 eV. That low E_b can be attributed to two factors: 1) the carboxyl groups are too close together, resulting in mismatched distances between them and Pb, leading to restricted bond angles and elongated bond lengths; 2) the overcrowded carboxyl groups exert repulsive forces on the negatively charged Br⁻ ions, causing the displacement of Br⁻ and noticeable changes in the perovskite structure. Encouragingly, A-PeNCs exhibit a high E_b of -1.75 eV, even slightly higher than twice that of C-PeNCs. The two carboxyl groups in A-PeNCs coordinate with Pb in a nearly parallel manner, indicating that both carboxyl groups can freely interact with Pb, and this can be extrapolated to other units of the polymer ligand. Additionally, the perovskite structure of A-PeNCs remains in its energetically lowest and most stable form without significant deformations. All these results confirm that the functional groups in BCPN are too closely packed, while ACPN perfectly matches the surface pattern of

PeNCs. DFT calculations indicate that further expanding the distance between functional groups will result in pattern mismatch, and there is no maleic anhydride-based ternary alternating copolymer to support for further expansion of the distance between functional groups. Therefore, the four C–C single bonds in ACPN provide the optimal spacing between functional groups.

The total reflection Fourier transform infrared spectroscopy (ATR-FTIR, Figure S3) was utilized to verify the completion of the amidation reaction, which was indicated by the disappearance of the anhydride signals (~ 1850 and 1774 cm^{-1}) and the emergence of amide groups (amide I, 1710 cm^{-1}).^[13] The amide I peak also indicated the presence of BCPN and ACPN in the purified PeNCs (Figure S4).^[14] Scanning transmission electron microscopy (STEM) was employed to investigate the morphology of the C-, B-, and A-PeNCs, as illustrated in Figure 2a–c. All samples exhibited a typical cubic shape with an average size of $\sim 12\text{ nm}$ which is determined by size distribution analysis (Figure S5). It has been reported that the introduction of strong coordinating ligands can inhibit the growth of PeNCs and improve size control.^[15] On the other hand, the hydrophobic chain length of the ligand also has a significant influence on the size and morphology of PeNCs during their nucleation and growth process.^[16] Therefore, the similar average sizes of C-, B-, and A-PeNCs are due to the combined effect of the strong synergistic coordination effect of the polymer ligands and the short alkyl chain that respectively inhibit and promote the growth of nanocrystals. In BCPN, which contains two blocks rich in benzene rings and carboxyl groups, respectively, with different interaction with PeNCs, leading to different growth environments for B-PeNCs and the broad size distribution.^[17] In contrast, the narrow distribution of A-PeNCs suggests that the ACPN with a

uniform structure contributes positively to the regulation of crystal growth. The elemental composition of all PeNCs was analyzed using energy dispersive X-ray spectroscopy (EDS, Figure S6), and the chemical formula was determined to be approximately $\text{CsPbBr}_{1.8}\text{Cl}_{1.2}$. The detailed crystal structure was characterized by high-resolution STEM (insert of Figure 2a–c) and X-ray diffraction (XRD, Figure S7), demonstrating the successful synthesis of thermodynamically stable β -phase PeNCs utilizing different polymer ligands.^[6,18]

X-ray photoelectron spectroscopy (XPS) was employed to study the chemical environment of Pb, as shown in Figure 2d and S8. The primary defects site of PeNCs are the exposed Pb^{2+} ions, as halide vacancies on the surface having the lowest defect formation energy.^[19] The Pb 4f signal was deconvoluted into three components, and the proportion of metallic Pb^0 in C-PeNCs reached $\sim 10\%$, which possibly due to the decomposition of incomplete perovskite surfaces and halide lead. In contrast, there was undetectable Pb^0 in A-PeNCs, and the proportion of Pb-COO^- increased considerably, indicating that the polymer ligand eliminated the easily decomposable Pb compounds and coordinated with the PeNCs surface through Pb–O bonds.^[20] Correspondingly, the proportion of O coordinated to Pb in the O 1s peak (Figure S9) also increased significantly.^[21] In addition, the $-\text{NH}_3^+$ signal in the N 1s peak (Figure S10) increased due to the incorporation of acidic polymer ligands, which could be another effective way to passivate PeNCs.^[22]

Subsequently, the PL characteristics of PeNCs were investigated to elucidate the passivating effect of polymer ligands on defects. Figure 2e displays the corresponding UV/Visible absorption and PL spectra, showing that the PL peaks remaining in the pure-blue range of $\sim 465\text{ nm}$ with polymer ligands which is consistent with the particle size and atomic ratio characterization. Further, PLQY and time-resolved PL (TRPL, Figure S11) were conducted to investigate the defect passivation and carrier recombination dynamics.^[23] As shown in Figure 2f, the excitation density-dependent PLQY revealed that the average PLQY of C-, B-, and A-PeNCs were $\sim 20\%$, 40% , and 60% , respectively. In addition, the PLQY, as an important parameter reflecting the luminescence performance of PeNCs, is used as one of the indicators to evaluate the long-term storage stability of PeNCs. As shown in Figure S12, A-PeNCs exhibited excellent dispersion (the illustration) and PL properties even after six months of storage, with the PLQY retaining approximately 70% of its initial value. This result suggests that the introduction of ACPN improved the stability of the sample, while the use of double short alkyl chains ensured the excellent dispersion of the PeNCs. Combined the PLQY value with the significantly prolonged amplitude-weighted average carrier lifetime (T_{average}) by ACPN, which is extracted by the tri-exponential fitting of TRPL, the as-calculated nonradiative recombination rate (k_{nr}) dramatically decreased from $\sim 3.4 \times 10^8\text{ s}^{-1}$ for C-PeNCs to $\sim 1.0 \times 10^8\text{ s}^{-1}$ for A-PeNCs, indicating that ACPN effectively suppressed defect-assisted nonradiative recombination. Previous research has shown that due to the soft ionic nature, light radiation or electrical field-induced ion migration is common in perovskites, which is manifested as phase

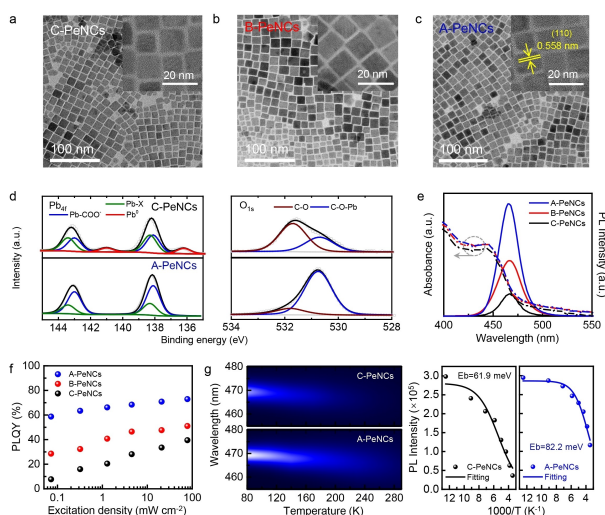


Figure 2. (a–c) The STEM image of C-, B-, and A-PeNCs. (d) XPS Pb 4f and O 1s spectra of C- and A-PeNCs. (e) UV/Vis absorption and PL spectra, as well as (f) excitation density-dependent PLQY of A-, B-, and C-PeNCs. (g) Temperature-dependent PL spectra (left panel) and corresponding PL intensity integrated band fitting curves (right panel) of C- and A-PeNCs.

segregation in mixed halide perovskites.^[24] Phase segregation seriously affects the spectral stability of PeNCs determining the feasibility of final applications, but in turn spectral stability serves as a reliable indicator for studying ion migration and phase segregation.^[25] Eliminating surface defects is one of the effective methods to suppress the segregation of halogens in PeNCs.^[1a] Encouraged by the suppression of defects by ACPN, we further investigated the impact of ACPN on phase segregation by monitoring the PL spectra stability (Figure S13) and electroluminescence (EL) spectrum stability (discussed later in device part). Within 10 mins continuous laser soaking (373 nm, $\sim 78 \text{ mW cm}^{-2}$), the PL peak of C-PeNCs obviously shifted from 465 to 474 nm, while only 1 nm shift is observed in A-PeNCs film, confirming the significant effect of ACPN on suppressing phase segregation.

Furthermore, we investigate the exciton binding energy (E_{eb}) by measuring temperature-dependent PL from 80 K to 290 K (Figure 2g).^[26] The PL peak shifts with temperature, which may be due to lattice thermal expansion with a positive temperature coefficient resulting in the change of the band gap.^[26–27] As the temperature increased, the PL intensity monotonically decreased, indicating thermal quenching related to non-radiative trapping at defect sites after exciton dissociation. The introduction of ACPN significantly mitigated this thermal quenching by passivating defects. Meanwhile, the integrated PL intensity was fitted using the Arrhenius equation: $I(T) = I_0/[1 + A \exp(-E_{\text{eb}}/k_{\text{B}}T)]$, where $I(T)$ and I_0 are the PL intensities at temperatures T and 0 K, respectively, A is the pre-exponential coefficient, and k_{B} is the Boltzmann constant. The extracted E_{eb} were 61.9 and 82.2 meV for C- and A-PeNCs, respectively, indicating that the coordination of ACPN also contributes to inhibiting exciton dissociation, which is beneficial for achieving good luminescent properties.

Finally, we investigated the influence of pattern-matched polymer ligands on the electrical properties and performance of pure-blue PeLEDs. The space-charge limit current (SCLC) method was applied to analyze both the trap density (N_{trap}) and carrier transport characteristics by fabricating electron-only devices (Figure S14).^[28] By fitting the current density-voltage curves of double-exponential form into three regions of Ohmic ($n=1$), trap-filled limited (TFL, $n>3$), and Child ($n=2$), the trap density was extracted as 5.93×10^{17} for C-PeNCs, 5.13×10^{17} for B-PeNCs, $4.26 \times 10^{17} \text{ cm}^{-3}$ for A-PeNCs, according to the following equation: $N_{\text{trap}} = 2\epsilon_0\epsilon V_{\text{TFL}}/eL^2$, where ϵ_0 and ϵ (~ 28) are the vacuum and relative permittivity, respectively. V_{TFL} is the trap-filled limit voltage. e is the elementary charge. L is the thickness of PeNCs film.^[29] Subsequently, by substituting the current density (J) and voltage (V) in the Child region into the Mott-Gurney law equation: $\mu = 8L^3J/9\epsilon_0\epsilon V^2$, the mobility (μ) was calculated to be 3.82×10^{-4} , 5.16×10^{-4} , $5.51 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ based on C-, B-, A-PeNCs, respectively.^[30] These results indicate that the application of short-chain alkanes effectively improved the charge transport properties. Previous theoretical studies have indicated that chlorine-containing perovskites are more prone to forming deep-level defects, which act as non-radiative recombination centers and severely affect the PLQY.^[8b,31] To further

verify the passivation effect of ACPN on deep-level defects, trap density of states (tDOS) was measured in devices fabricated based on C- and A-PeNCs.^[32] The tDOS (NT) is derived from the angular frequency-dependent capacitance with the equation: $\text{NT} = -(V_{\text{bi}}/qW) \times (dC/d\omega) \times (\omega/k_{\text{B}}T)$, where C is the capacitance, ω is the angular frequency, q is the elementary charge, k_{B} is the Boltzmann constant, T is the temperature, V_{bi} is the built-in potential, and W is the depletion width, respectively. As shown in Figure S15, the defect density of A-PeNCs is significantly lower than that of C-PeNCs within entire energy ranges, especially for energy above 0.4 eV, suggesting the incorporation of ACPN effectively passivates the deep-level defects.

The pure-blue PeLEDs with optimized p-i-n structure were fabricated utilizing as-synthesized PeNCs as electro-luminescent active layer. The upper panel of Figure 3a presents the energy level alignment, which consisted of indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS)/poly-TPD/poly(9-vinylcarbazole) (PVK)/PeNCs/tris-(1-phenyl-1H-benzimidazole) (TPBi)/LiF/Al from bottom to top. The bottom panel of Figure 3a shows the typical cross-sectional scanning electron microscope (SEM) image of PeLEDs. Figure 3b depicts the current density-voltage-luminance (J - V - L) curves, in which the devices incorporating polymer ligands with short side-chain exhibited higher brightness and a slightly improved current. Specifically, the turn-on voltage decreased from $\sim 3.7 \text{ V}$ in C-PeLEDs to 3.3 and 3.0 V in B- and A-PeLEDs, respectively. More importantly, the champion EQE significantly increased from 1.1 % in C-PeLEDs to 3.3 % in B-PeLEDs and 9.1 % in A-PeLEDs (Figure 3c). The statistical EQE of 30 devices was summarized in Figure S16. These significant enhancements of PeLEDs performance were attributed to the effective passivation of PeNCs defects and improved charge transport properties,

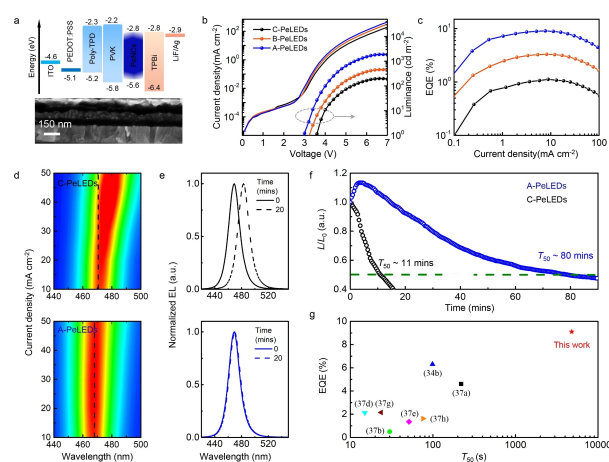


Figure 3. (a) The energy band alignment diagram and cross-sectional SEM image of PeLEDs. (b) J - V - L curves and (c) EQE curves as a function of current density. (d) EL spectra at different current densities. (e) EL spectra of C- and A-PeLEDs working at 0 and 20 minutes. (f) T_{50} of C- and A-PeLEDs. (g) Summarization of the reported EQE of pure-blue (460–470 nm) PeLEDs based on mixed-halide perovskite nanocrystal with weak quantum confinement as a function of T_{50} .

facilitated by polymer ligands with short side chains that possess pattern-matching characteristics.^[33]

For mixed-halide PeLEDs, the spectral and operational stability of the devices are profoundly affected by the electrically driven phase segregation during prolonged operation.^[34] Figure 3d illustrates the evolution of EL spectra of PeLEDs with increasing current density. Notably, a distinct red shift of the EL peak position was observed in C-PeLEDs when the current density reached 50 mA cm^{-2} , while the emission peak of A-PeLEDs remained largely unchanged. Furthermore, we investigated the stability of the devices under continuous operation at a typical luminance of 100 cd cm^{-2} . Figure 3e presents the EL spectra at 0 and 20 minutes during continuous operation, and Figure S17 further shows the evolution of EL peak position over time. Encouragingly, a significant red shift of 16 nm was observed in C-PeLEDs, while A-PeLEDs showed only a negligible shift of 3 nm from pure-blue emission at 467 nm. At 0 mins, all PeLEDs emitted pure-blue light with corresponding CIE coordinates of (0.131, 0.071) as shown in Figure S18, that are close to the Rec. 2100 standard of (0.131, 0.046).^[35] After continuous operation for 20 mins, a significant shift of CIE coordinates to (0.096, 0.185) was observed in C-PeLEDs, while the ACPN ligands effectively maintained the stability of the coordinates. Consistent with the spectral stability, the T_{50} as presents in Figure 3f, which represents the time required for the initial EL intensity to decrease by half,^[36] was dramatically increased from ~11 mins in C-PeLEDs to ~80 mins for A-PeLEDs. Figure 3g and Table S1 present a summary of the reported EQE and T_{50} values for pure blue (460–470 nm) PeLEDs based on mixed-halide PeNCs with weak quantum confinement, highlighting the advantages of pattern-matched polymer ligands in enhancing performance and operational lifetimes through their excellent passivation effect.^[25d,34b,37]

Conclusion

In summary, we have investigated the modulation of the spacing or density of functional groups in polymer ligands to match $\text{CsPbBr}_x\text{Cl}_{3-x}$ PeNCs for enhancing the performance and stability of pure-blue PeLEDs. Two polymers (BCP and ACP) with similar functional groups were selected as the matrices to synthesize suitable polymer ligands by grafting short-chain alkanes for direct addition to the PeNCs synthesis. Our theoretical and experimental results show that ACPN match with PeNCs well and effectively passivated PeNCs, even though BCPN has a higher functional groups density. The PL characteristics revealed that ACPN was most effective in suppressing nonradiative recombination and laser-induced phase segregation. Further exciton dynamics studies revealed that ACPN effectively suppressed defects and increased exciton binding energy. Furthermore, the optimized short alkyl chains improved charge transport properties, resulting in higher mobility and lower turn-on voltage of PeLEDs, while maintaining long-term stable dispersion of PeNCs. Finally, PeLEDs achieve high EQE (9.1 %) with CIE coordinates (0.131, 0.071) that meet the Rec. 2100 standard. Particularly, the introduction of ACPN

significantly improved the spectral and operational stability of PeLEDs, achieving a recorded T_{50} of up to 80 minutes. Overall, this study demonstrated that pattern-matched polymer ligands effectively suppressed phase separation and improved the spectral and operational stability of pure-blue PeLEDs contributing for the development of perovskite-based optoelectronic devices.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords: Perovskite nanocrystals • polymer ligands • pattern-matching • spectrum and operational stability • pure-blue light-emitting diodes

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