Perovskite Light-Emitting Diodes with External Quantum Efficiency Exceeding 20%

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Metal halide perovskite materials are emerging solution-processed semiconductors with considerable promise in optoelectronic devices[1,2]. Metal halide perovskite-based light-emitting devices (pLEDs) have received extensive interest for applications in flat-panel displays and solid-state lighting owing to their promise of low cost, tunable colors with narrow emission bandwidths, high photoluminescence quantum yield (PLQY), and facile solution processing [3-7]. However, the highest reported external quantum efficiency (EQE) of green- and red-emitting pLEDs are 14.36%[6,8] and 11.7%[7], still far behind the performance of organic LEDs (OLEDs)[9-11] and inorganic quantum dot LEDs (QLEDs)[12]. Here we report visible perovskite LEDs that

surpass the 20% quantum efficiency milestone. This stems from a new compositional distribution management strategy that simultaneously provides high luminescence and balanced charge injection. We mix a pre-synthesized $CsPbBr_3$ perovskite with MABr (MA = CH_3NH_3) additive in the precursor; the different solubility of which yields sequential crystallization into a $CsPbBr_3$@MABr quasi-core-shell structure. The MABr shell passivates nonradiative defects otherwise present in $CsPbBr_3$ crystals, boosting their photoluminescence quantum efficiency; and the MABr capping layer enables balanced charge injection. We document as a result perovskite LEDs whose EQE reaches 20.3%, representing a significant step toward practical commercialization of pLED and application in lighting and display.

$MAPbI_{3-x}Cl_x$ and $MAPbBr_3$ (MA = CH_3NH_3) were used in early pLEDs that achieved EQEs of 0.76% and 0.1% for the near-infrared and green regimes, respectively [13]. Two strategies [4-6,14] subsequently led to notable improvements in pLED performance: i) direct spin-coating of colloidal perovskite nanocrystals [15-18], where the perovskite nanocrystals are highly luminescent with a PLQY of nearly 90%, and their optical properties can be tuned by compositional engineering and crystal size; ii) deposition of bulk perovskite films using perovskite precursor solutions whose composition was suitably engineered (*e.g.*, stoichiometrically-modified $MAPbBr_3$[5] and $MAPbX_3$ with the addition of long-chain ammonium halides[4] or 1-naphthylmethylamine halides[7]*).*

Here we built upon those prior works and pursued in addition a new strategy toward further-enhanced EQE. Our purpose was to combine high PLQY with balanced charge injection. Our strategy was to construct a compositionally-graded perovskite based on a quasi-core-shell structure: the bottom layer consists of perovskite light-emitting polycrystals capped using a

defect passivation layer that addresses the grain boundaries; and the top layer serves to passivate the surface and simultaneously balance charge injection into the pLED device.

We utilized the very different solubility and crystallinity of perovskite ($CsPbBr_3$) and passivant (MABr) in polar solvents and – using a one-step deposition method – fabricated *in-situ* the compositionally-graded perovskite layer. This consisted of a defect-passivated perovskite layer on the bottom ($CsPbBr_3$@MABr) and an electrical passivating layer (MABr) on top. The highly luminescent perovskite films and balanced charge injection enabled an initial EQE from a pLED device of 17%. We then further improved the device charge injection balance by inserting an insulating layer of PMMA between the perovskite layer and ETL, thereby maximizing the device efficiency to 20.3%.

We synthesized the $CsPbBr_3$ perovskite powder as a starting material, and then added MABr into the perovskite precursor. We engineered the amount of MABr additive to improve perovskite film formation and PLQY (Mixture-1.0 presents the molar ratio of MABr to $CsPbBr_3$ is 1). Fig. 1a shows three different perovskite structures fabricated using a strategy we term compositional distribution management, *i.e.*, single-layered $CsPbBr_3$ (prepared by one step spin-coating), bi-layered $CsPbBr_3$/MABr (prepared by coating another layer of MABr on the as-formed $CsPbBr_3$ layer) and quasi-core-shell $CsPbBr_3$@MABr (Mixture-1.0). Fig. 1b and Extended Data Fig. 1 indicate that the capping layer of MABr (bi-layered structure) only slightly enhances PL emission, while the Mixture-1.0 film with quasi-core-shell structure presents very bright PL emission (Movie S1). We found a PL enhancement proportional to the amount of MABr additive when the molar ratio of MABr to $CsPbBr_3$ was varied from 0.4 to 1.0, and PL began to drop after 1.0 (Extended Data Fig. 1).

We observed that, during film formation, $CsPbBr_3$ crystallized more rapidly, whereas MABr sequentially increased its crystallization rate after the $CsPbBr_3$ precursor was completely consumed (Extended Data Fig. 1d). We explain this by noting that the solubility of $CsPbBr_3$ (0.56 M, experimental result) is far below than that of MABr (5 M, experimental result) in DMSO.

Secondary ion mass spectrometry (SIMS) depth analysis (Fig. 1c) supports the $CsPbBr_3$-MABr gradient structure, from which one can see the top layer contains CH_4N^+ ions (from capping MABr, stage I), Pb^+ ions with few CH_4N^+ ions exist in the middle layer (from $CsPbBr_3$ and MABr, stage II) and In+ ions in the bottom layer (from ITO, stage III). To gain insight into the compositional distribution in the as-formed Mixture-1.0 films, we carried out cross-sectional SEM and TEM studies. The cross-sectional TEM samples were prepared using a focused ion beam (FIB), where C and Pt layers were pre-deposited to protect perovskite from possible ion beam induced damage. The cross-sectional SEM image (Extended Data Fig. 2a) shows a high quality perovskite film with obvious grain boundaries. The cross-sectional TEM and element mapping (Fig. 1d and Extended Data Fig. 2b) images show a well-defined layer-by-layer structure of ITO/PEDOT:PSS/$CsPbBr_3$-MABr/MABr/C. A MABr shell is observed in the grain boundaries of $CsPbBr_3$ (white arrows), and another layer of MABr caps the $CsPbBr_3$, forming the quasi-core-shell structure. We sought to estimate experimentally the trap state density of the three classes of perovskite samples. We find (Extended Data Fig. 3a) the MABr shell reduces defects in the Mixture-1.0 perovskite films by a factor of four compared to the single-layered $CsPbBr_3$ film.

To gain insight into the effect of thick top MABr layer on PL enhancement, we washed the bright perovskite film using anhydrous IPA solvent and observed that the PL gradually

decreased as the MABr was removed (Extended Data Fig. 3b). To make a direct comparison, we also prepared pure $CsPbBr_3$ and $MAPbBr_3$ perovskite films. As shown in Extended Data Fig. 3c, all perovskite films show transparent yellow color under room illumination. However, only the Mixture-1.0 perovskite films reveal high brightness under UV lamp excitation. UV-Vis absorbance spectra (Extended Data Fig. 3d) of the Mixture-1.0 film present a band edge absorbance at 531 nm, similar to $CsPbBr_3$ (528 nm), corresponding to a bandgap of 2.33 eV. The analysis of the PL spectra of the three perovskite samples indicates that Mixture-1.0 film emission is the closest to the emission of pure $CsPbBr_3$ films (Extended Data Fig. 3e). In order to quantify the PL enhancement induced by the MABr additive, we then measured the absolute PLQY according to a reported protocol[19]. The PLQY of the Mixture-1.0 perovskite film was determined to be ~80%, while the PLQY of $CsPbBr_3$ and $MAPbBr_3$ are not detectable (from an analysis of our system signal-to-noise, we conclude that their PLQY lies below 1%). Time-resolved PL spectra (Extended Data Fig. 3f) show that the Mixture-1.0 film has a 50% longer radiative lifetime than pure $CsPbBr_3$, and the longer lifetime of the PL transition is a direct evidence of decrease in the concentration of defects and increase in film crystallinity[1,6,8]. We attribute this to the fact that the compositionally graded structure combined with the MABr shell passivates the non-radiative defects in $CsPbBr_3$.

The crystal structure of the perovskite films was analyzed using XRD (Extended Data Fig. 4a). We conclude that the Mixture-1.0 perovskite exhibits the same crystal structure as monoclinic $CsPbBr_3$, instead of a mixture of $CsPbBr_3$ and $MAPbBr_3$ separated phases. The XPS data (Extended Data Fig. 4b-c) also indicates the existence of $CsPbBr_3$ and MABr in the Mixture-1.0 perovskite film.

The perovskite layer also requires a high surface coverage and low roughness to achieve high performance LEDs. The surface morphology of the pure $CsPbBr_3$, $MAPbBr_3$ and Mixture-1.0 perovskite films were characterized using SEM and AFM (Extended Data Fig. 5). Small particles and pinholes were observed in the $CsPbBr_3$ and $MAPbBr_3$ films; whereas in the Mixture-1.0 film, smooth and well-packed micrometer-sized cuboids were combined with good crystallinity.

We fabricated pLEDs with single-layered $CsPbBr_3$, $MAPbBr_3$, bi-layered $CsPbBr_3$/MABr and Mixture-1.0 perovskites with quasi-core-shell structure based on a device structure consisting of ITO/PEDOT:PSS/perovskite/B3PYMPM/LiF/Al (Fig. 2a). PEDOT:PSS served as the HTL, B3PYMPM as the ETL, LiF as an electron injection layer and Al as the cathode (Extended Data Fig. 6a). Photographs of pLEDs devices with six uniform and bright green-emitting pixels (2 mm × 1.5 mm) are shown in Fig. 2b. Larger-area devices (6 mm×20 mm (Extended Data Fig. 6b) showcase uniform and bright emission.

The device performance of bi-layered $CsPbBr_3$/MABr perovskite is quite limited (Extended Data Fig. 6c), slightly better than the single-layered $CsPbBr_3$, maybe due to their low PLQY and poor surface morphology. The Mixture-1.0 devices display an emission peak at 525 nm with a FWHM of 20 nm (Extended Data Fig. 6d-e), corresponding to a CIE coordinate of (0.18, 0.75). Current density-voltage (*J*-*V*) and luminance-voltage (*L*-*V*) curves were collected to evaluate the LED performance (Extended Data Fig. 6f-h). Devices fabricated using Mixture-1.0 exhibited the lowest current density simultaneously with the highest luminance, indicating the best performance among the three classes of pLEDs. The Mixture-1.0 devices gave a maximum current efficiency (CE) of 23 $cd \cdot A^{-1}$ at 3.8 V, fully 3 orders of magnitude higher than the pure $CsPbBr_3$ and $MAPbBr_3$-based LEDs[5,14].

We posited that the superior LED performance of the Mixture-1.0 could arise not only from the high PLQY, but also from its combination with improved charge injection balance. To quantify charge injection, we measured current density-voltage (*J-V*) characteristics of electron (ITO/B3PYMPM/perovskite/B3PYMPM/Al) and hole (ITO/PEDOT:PSS/perovskite/Au) only devices (Fig. 2d). We conclude that electrons dominate injection into $CsPbBr_3$ devices, whereas a more balanced charge injection occurs in the case of Mixture-1.0. After optimization, we obtained an average CE of 35 $cd \cdot A^{-1}$ over 20 devices, with the best CE reaching 65 $cd \cdot A^{-1}$ (Fig. 2e). The *EQE-V* characteristic of the best-performing device shows a maximum EQE of 17% at 4.2 V (Fig. 2f), a record for green-emitting perovskite LEDs.

Fig. 2d shows that the capping MABr layer in the quasi-core-shell structure helps to reduce electron injection and improve charge balance. Further improvement is available if we can further optimize charge balance. We did so by depositing a thin PMMA layer on the as-formed perovskite. We then tested electron-only and hole-only device performance again and found the PMMA layer further helps in balancing charge injection (Fig. 3a). We therefore inserted a thin PMMA layer between the perovskite and ETL (Fig. 3b and Extended Data Fig. 7a). The PMMA layer was found to be continuous and smooth (Extended Data Fig. 7b-c), enabling charge injection into perovskite through PMMA via tunneling.[12] After we optimized the PMMA layer thickness and molar ratio between MABr and $CsPbBr_3$ in mixed perovskite precursor (Extended Data 7d-e), the devices reached a higher CE of 78 $cd \cdot A^{-1}$ (Fig. 3c).

Fig. 3d presents the *J-V* and *L-V* curves of the best-performing Mixture-1.0 device, showing a low driving current density and high luminance of 14,000 cd m^{-2}. A low turn on voltage of 2.7 V, just slightly higher than the bandgap of the Mixture-1.0 perovskite, is obtained as a consequence of the high-quality perovskite thin film and more efficient carrier injection

from HTL and ETL. It is also found that the EL spectra at various applied voltage remain the same and the maximum power efficiency is 69 lm W^{-1} at 3.6 V (Extended Data Fig. 8). A maximum EQE value of 20.3% is achieved with a luminance of 3,400 cd m^{-2} (Fig. 3e, Movie S2 and S3).

We measured the device lifetime by applying a constant current to the device and monitoring the luminance evolution[12,20]. After we apply a constant drive current of 5 mA (167 mA cm^{-2}) to the device, the luminance increases from 3,800 cd m^{-2} to 7,130 cd m^{-2} (L_0) in 0.66 min and then starts to diminish (Fig. 3f). The half-lifetime (T_{50}), defined as the time for the luminance to decrease to $L_0/2$, is ~10 min. By using a calculation of L_0^n T_{50} = constant and assuming an acceleration factor of n = 1.5[12] (Extended Data Fig. 9a), we estimate T_{50} for this device at 100 cd m^{-2} to be ~100 h, the highest value estimated to date in high performance pLEDs, paving a bright future in daily application. We also measured stability under continuous operation with luminance maintained at a constant value of ~100 cd m^{-2}, achieved by tuning the applied current to maintain luminance. The results (Movies S4 and Extended Data Fig. 9b) show that the device operated continuously for ~46 h, of similar order to the extrapolated value from the accelerated aging test. The corresponding EQE decreased from 13% to 5.6% over these 46 h of continuous operation, indicating that the device did degrade even at this low luminance. The stability performance shown here is 2-3 orders of magnitude higher than previous reports[6,8,21-24] (Extended Data Table 1); and the topic of stability clearly remains a crucial challenge for further progress in pLEDs.

In summary, we demonstrated a new strategy to realize compositionally-graded perovskite devices that simultaneously achieve high PLQY and balanced charge injection. Our approach exploited the different solubility of perovskite precursors to control the crystallization

of $CsPbBr_3$-MABr gradient compounds in a single step. The MABr shell passivates non-radiative defect sites in the $CsPbBr_3$ crystals and the MABr capping layer balances charge injection for pLEDs applications. These effects combined allow us to achieve pLEDs with a narrow green emission exhibiting a new record EQE above 20%. The high EQE of these devices is for the first time on par with more mature established technologies such as OLEDs. Improvements in device stability remain an important challenge, and strides in that direction can be envisaged by suppression of ion migration with additives or blocking layer, fabrication of high quality perovskite layer and further optimizing the perovskite/ETL and perovskite/HTL interfaces[25-27].

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Acknowledgments: The work was supported by the Scientific Research Funds of Huaqiao University (600005-Z16J0038) and National Natural Science Foundation of China (U1705256). Z.H. Wei thanks Prof. Ying Wang (Technical Institute of Physics and Chemistry, Chinese Academy of Sciences) and Prof. Jianpu Wang (Institute of Advanced Materials, Nanjing Tech University) for helpful discussion on how to accurately measure pLEDs device performance. Z.H. Wei thanks Prof. Shihe Yang (The Hong Kong University of Science and Technology) for TOF-SIMS test. Q.X. gratefully acknowledges the support from Singapore National Research

Foundation through NRF Investigatorship award (NRF-NRFI2015-03), Ministry of Education via AcRF Tier2 grant (MOE-2015-T2-1-047) and Tier1 grants (RG 113/16 and RG 194/17).

Author Contributions

Z.H. Wei, Q.H. Xiong and E.H. Sargent conceived the idea. K.B. Lin, J. Xing and W.J. Zhao prepared the samples, carried out optical spectroscopy characterizations. L.Q. Xie and Y. Lu carried out XPS, AFM and some other materials characterizations. K.B. Lin, J.X. Lu, D. Zhang, C.Z. Yan, W.Q. Li and X.Y. Liu conducted pLED device fabrication and characterization. Z.H. Wei supervised the pLED fabrication and characterization. L.N. Quan, X.W. Gong, J. Kirman and F.P.G. Arquer assisted in device fabrication, measurement and manuscript writing. K.B. Lin, Z.H. Wei, Q.H. Xiong and E.H. Sargent carried out data analysis and wrote the manuscript. All authors discussed the results and commented on the manuscript.

Author Information

Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to Z.H. Wei (weizhanhua@hqu.edu.cn).

Main Figure Legends

Figure 1 PL enhancement via compositional distribution management. a, Schematic illustration of single-layered $CsPbBr_3$, bi-layered $CsPbBr_3$/MABr and quasi-core-shell $CsPbBr_3$@MABr structures; **b,** Photo of the as-prepared three perovskite films under UV light; **c,** Secondary ion mass spectrometry (SIMS) depth analysis of the $CsPbBr_3$@MABr structure; **d,** Cross-sectional TEM image of the $CsPbBr_3$@MABr structure (the white arrow indicates MABr shell in the grain boundary). The sample was prepared using focused ion beam and the top C layer was pre-deposited to protect the perovskite.

Figure 2 Fabrication and performance evaluation of the pLEDs. a, Cell configuration diagram of the pLEDs with PEDOT:PSS and B3PYMPM as HTL and ETL, respectively; **b,** Photographs of pLEDs devices fabricated with the Mixture-1.0 perovskite, displaying six uniform and bright pixels and a logo of "Pero-LED"; **c,** Typical current efficiency-voltage (*CE*-*V*) curves of the three pLEDs without optimization; **d,** Current density-voltage (*J*-*V*) curves of electron-only and hole-only devices; **e,** Current efficiency statistic histogram of the Mixture-1.0 perovskite LED; **f,** *EQE*-*V* characteristic of the best-performing Mixture-1.0 perovskite LED.

Figure 3 pLED performance enhancement by inserting a thin PMMA layer between perovskite and ETL. a, *J*-*V* curves of the electron-only and hole-only devices with PMMA layer; **b,** Cell configuration diagram of the pLEDs with a very thin PMMA layer inserted between perovskite and ETL; **c,** Current efficiency statistical histogram of pLEDs devices with PMMA layer; **d,** *L*-*J*-*V* and **e,** *EQE*-*L* characteristics of the best-performing pLEDs; **f,** Lifetime measurement of the pLEDs.

METHODS

Unless otherwise stated, all chemicals were purchased from Sigma-Aldrich and used as received.

Preparation of perovskite precursor: $CsPbBr_3$ powder was firstly synthesized and used as starting materials for precursor preparation. $PbBr_2$ (10 mmol, 3.67 g) was dissolved in hydrobromic acid (8 mL) firstly, then CsBr (10 mmol, 2.12g, dissolved in 3 mL of water) was added drop by drop and formed an orange precipitate. The precipitate was filtered, washed twice using ethanol and dried at 60 ^oC in a vacuum oven for 12 h before use.

CH_3NH_3Br (abbreviated to MABr) was prepared by reacting 12 mL of methylamine (33 wt. % in absolute ethanol) and 11 mL of hydrobromic acid (48 wt. % in H_2O) in ice bath for 2 h with continuous stirring. The solvent was removed using rotary evaporation at 50 °C to get white MABr powder. For purification, the as-prepared MABr powder was re-dissolved in ethanol and precipitated with diethyl ether. Finally, the white powder was collected by filtration and dried at 60 °C in a vacuum oven for at least 12 h before use.

$MAPbBr_3$ precursor was prepared by dissolving $PbBr_2$ and MABr (1:1, molar ratio) in DMSO solvent to make a 0.5 M solution. $CsPbBr_3$ powder can be fully dissolved in DMSO solvent to make a 0.5 M starting solution. Then, different amount of MABr was added to the as-prepared $CsPbBr_3$ solution to make a mixture perovskite precursor. And the mixture perovskite precursor was named by the molar ratio of MABr to $CsPbBr_3$. For example, to prepare a precursor of Mixture-1.0, 55.98 mg of MABr (0.5 mmol) was added to 1 mL of the as-prepared $CsPbBr_3$ (0.5 mmol) solution.

Fabrication of pLEDs: Pre-patterned ITO glasses (20 mm * 20 mm) were ultrasonically washed sequentially in detergent solution, deionized water, acetone, and ethanol, then dried with compressed N_2. The substrates were further cleaned with UV-Ozone cleaner (Novascan, PSD) for 30 min before spin-coating. A 40 nm of hole-transporter layer was prepared by spin-coating using PEDOT:PSS (Clevios PV P AI4083) at 4,000 r.p.m for 60 s and baked at 150 ^{o}C for another 15 min. After cooling to room temperature, the substrates were transferred into a nitrogen-filled glove box (H_2O < 1 p.p.m, O_2 < 1 p.p.m) for perovskite layer deposition. The perovskite layers were prepared in same spin-coating procedures but use different precursor solution. Specifically, 30 uL of perovskite precursor was dropped onto the substrates and spun at 2,000 r.p.m for 60 s, during which 500 uL of toluene was dropped quickly onto the surface at 30 s. Another thin layer of PMMA blocking layer was prepared if needed, 50 uL of PMMA solution (0.5 mg mL^{-1}, acetone) was spin-coated onto the as-prepared perovskite layer at 4,000 r.p.m for 60 s. There is no annealing process and the as-prepared substrates were transferred into a thermal evaporator. The chamber was vacuum pumped down to 5.0 * 10^{-4} Pa, and a 40 nm of B3PYMPM (Lumtec, Taiwan), 2 nm of LiF and 100 nm of Al layers were sequentially evaporated. The pero-LED device area was defined by overlap area between the ITO and Al electrode, and it is 3 mm^2 (2 mm * 1.5 mm). We also used CCD camera to measure the actual device area.

Material characterization: Surface morphologies (secondary electron image) were characterized by field-emission scanning electron microscopy (SEM, Hitachi S-8000). Time-of-flight secondary ion mass spectrometry (TOF-SIMS, ION-TOF GmbH, ToF SIMS V) was used to determine ion distribution in the perovskite film. Bright filed, high angle annular dark field and element mapping images were characterized by FEI Talos F200S transmission electron

microscope (TEM). The perovskite sample for TEM observation was prepared using FIB (FEI Scios), while two protective layers of C and Pt were deposited before ion beam cutting and etching. X-ray diffraction (XRD) patterns were recorded using D8 Advance (Bruker AXS). UV-Vis and steady-state photoluminescence (PL) spectra were acquired by Flame spectrometer (Ocean Optics) in glovebox. PL quantum yield (PLQY) was measured using blue excitation laser (405 nm), integrating sphere and Flame spectrometer. PL decay curves were measured using a fluorescence lifetime imaging microscope (FLIM, Leca TCS SP8) with a pulsed excitation laser of 405 nm.

Performance evaluation of pLEDs: The as-fabricated pLED devices were inserted into a home-made test socket and measured in the glovebox (Movie S2-S4). Current density-Voltage (*J-V*) curves were measured by Keithley 2400 from 0 V to 5 V with a step voltage of 0.2 V and delay time of 3s, and luminance was measured using a luminance meter (Konica Minolta, LS-160 or CS-200) simultaneously. The electroluminescence (EL) characteristics were recorded with Flame spectrometer (Ocean Optic). External quantum efficiency (EQE) was calculated using Lambertian emission profiles or measured using an integrated sphere (FOIS-1, Ocean Optics) and the Flame spectrometer. Before measurement, the integrated sphere was firstly calibrated using a standard light source (HL-3-INT-CAL, Ocean Optics). As the substrate size (20 mm * 20 mm) was larger than input port (9.5 mm in diameter) of the integrating sphere, the pLED can be put onto the input port and make a close contact easily. Meanwhile, the device area (2 mm * 1.5 mm) is much smaller than the input port, the emission light is supposed to be fully collected by the integrating sphere. Then an *I-V* sweep was conducted by Keithley 2400 and EL characteristics (including EL spectra, photon flux and etc.) were recorded using Spectra Suite software (Ocean Optics). Finally, the EQE can be calculated using the photon flux and current

value. The initial high performance device with an EQE of 16% was first observed in Nanyang Technological University group, while the EL characteristics were further studied there as well.

Accelerated aging device lifetime measurement: Prior studies show that the product of the initial luminance (L_0) of the lifetime measurement, and the T_{50} lifetime (defined as the time when the luminance drops to 50% of L_0), is a constant: $L_0^n \times T_{50} = constant$, where the n is acceleration factor. The factor can be determined experimentally by running lifetime tests for different L_0 values. The equation is rewritten in the form: $LogT_{50} = K - nLogL_0$. In this way, n is obtained as the slope of the linear fitting curve of the various measured T_{50} and L_0.

Data availability statement: The data that support the findings of this study are available from the corresponding author (Z.H. Wei) upon reasonable request.

Supporting information: Movie S1 (Preparing a highly luminous film using the Mixture-1.0 precursor), Movie S2 (Real-time measurement of a pLED device), Movie S3 (Measurement of a high performance pLED device with CE of 74.2), Movie S4 (Operation lifetime measurement of a pLED device working in continuous mode with luminance of around 100 cd m^{-2}).

Extended Data Legends

Extended Data Figure 1 | Emission properties and formation mechanism of Mixture perovskites. a, PL spectra of single-layered $CsPbBr_3$, bi-layered $CsPbBr_3$/MABr and quasi-core-shell structured Mixture-1.0 films; **b,** PL spectra of various Mixture perovskite films with different amount of MABr; **c,** Photograph of perovskite films under UV light and **d,** Schematic illustration of formation process of the $CsPbBr_3$-MABr quasi-core-shell structure.

Extended Data Figure 2 | SEM and TEM images of the Mixture-1.0 films. a, Cross-sectional SEM images of the as-prepared Mixture-1.0 film and **b,** Cross-sectional TEM and elemental mapping images of the Mixture-1.0 film, where the sample was prepared using focused ion beam, and the top C and Pt layers were pre-deposited to protect perovskite film.

Extended Data Figure 3 | Comparison between $CsPbBr_3$, $MAPbBr_3$ and the Mixture-1.0 perovskite films. a, Photograph of a pristine Mixture-1.0 perovskite film (left) and a film treated using iso-propanol washing (right, to remove the MABr capping layer) under UV light; **b,** Trap state density estimation in the SCLC method obtained using dark *I-V* curves of perovskite devices with the structure of ITO/perovskite/Au; **c,** Photograph of the three perovskite films under UV light; **d,** UV-Vis absorbance spectra of the $CsPbBr_3$ and different Mixture perovskite films; **e,** PL spectra and **f,** time-resolved PL decay curves (excitation source: 400 nm, 4 μW) of the $CsPbBr_3$, $MAPbBr_3$ and Mixture-1.0 films.

Extended Data Figure 4 | XRD and XPS characteristic of the perovskite films. a, Original and magnified XRD patterns of the $CsPbBr_3$, $MAPbBr_3$, Mixture-1.0 and MABr+$PbBr_2$+CsBr mixture perovskite films; **b,** XPS results of the $CsPbBr_3$, $MAPbBr_3$ and Mixture-1.0 perovskite films, indicating that there are $CsPbBr_3$ and MABr phases in the mixture-1.0 film, not $MAPbBr_3$.

Extended Data Figure 5 | Morphology comparison of the three perovskite films. Top-view SEM image (left), AFM topography (middle) and root mean square roughness (right) of the **a,** $CsPbBr_3$, **b,** $MAPbBr_3$ and **c,** Mixture-1.0 perovskite films.

Extended Data Figure 6 | pLED device fabrication and evaluation. a, Energy level diagram of the as-fabricated pLED; **b,** Photograph of a large area pLED device (6 mm * 20 mm); **c,** Device performance of the bi-layered $CsPbBr_3$/MABr pLEDs; **d,** EL spectra and **e,** CIE Chromatic diagram of the as-fabricated three pLEDs; **f,** Current density-voltage (*J-V*) and **g,** luminance-voltage (*L-V*) curves of the as-fabricated three pLEDs and **h,** *L-J-V* curves of the best-performing Mixture-1.0 perovskite LED without PMMA layer.

Extended Data Figure 7 | pLED device performance optimization. a, Cross-sectional SEM image of a pLED device with PMMA blocking layer; **b,** AFM topography image and **c,** thickness measurement of the optimized PMMA layer; Device performance optimization by tuning **d,** PMMA thickness and **e,** amount of MABr additives in the perovskite precursor;

Extended Data Figure 8 | EL spectra and power efficiency of the best-performing Mixture-1.0 pLED devcie. a, EL spectra at various applied voltage and **b,** power efficiency curve of the champion Mixture-1.0 pLED device.

Extended Data Figure 9 | Operation lifetime measurement. a, Lifetime extrapolation from accelerated aging tests, from which one can see that the acceleration factor n is ~1.5 in the devices investigated herein; **b,** Operational lifetime measurement of a pLED working in continuous luminance mode, by careful tuning the applied current, we kept the luminance output of ~ 100 cd m^{-2}.

Extended Data Table 1 | Summary of stability performance of other reported highly efficient green pLEDs (EQE>10%).