High stability and strong luminescence CsPbBr₃-Cs₄PbBr₆ thin films for all-inorganic perovskite light-emitting diodes

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Supplementary Table S1. Energy-dispersive spectrometer (EDS) results of Cs-Pb-Br films with different Cs/Pb ratios.

| Br atomic percentage (%) | Cs atomic percentage (%) | Pb atomic percentage (%) | Cs/Pb ratio |
|--------------------------|--------------------------|--------------------------|----------------|
| 62.88 | 18.55 | 18.57 | 1 |
| 62.16 | 20.72 | 17.12 | 1.21 |
| 62.45 | 21.47 | 16.08 | 1.34 |
| 63.94 | 22.12 | 13.94 | 1.58 |

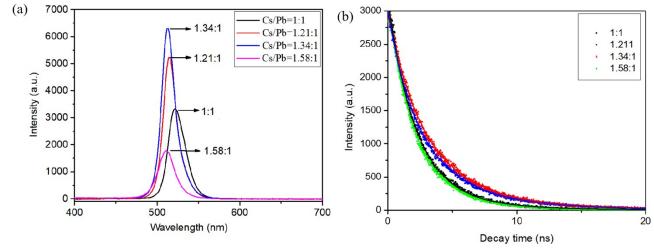


Figure S1 (a) PL spectra (b) Time-resolved photoluminescence (TRPL) spectra with different molar ratios

As the molar ratio of CsBr/PbBr₂ increased from 1 to 1.34, the lifetime was significantly prolonged. The strongest PL intensity and lifetime are observed at a Cs/Pb ratio of 1.34, which further confirms the superior optoelectronic quality and high radiative lifetime of the film.

Table S2. Fitting results of the time-resolved PL spectrum at different annealing temperatures based on dual-exponential fitting

| Sample | τ_1 (ns) | $\tau_2(ns)$ |
|------------|---------------|--------------|
| unannealed | 1.41 | 6.34 |
| 200 °C | 1.46 | 13.32 |
| 250 °C | 1.89 | 22.87 |
| 300 °C | 2.02 | 26.24 |
| 350°C | 1.80 | 10.66 |

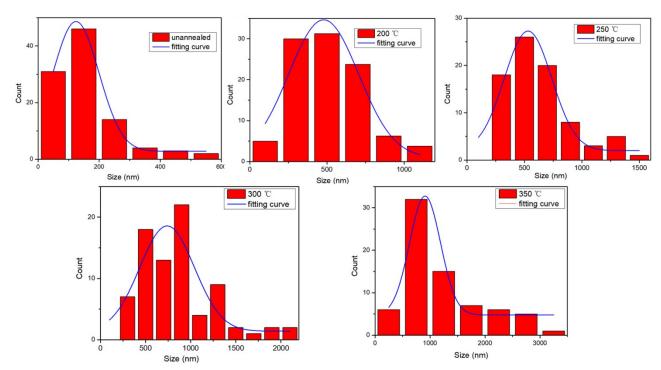


Figure S2 Domain size distribution histograms of films before and after annealing

As the annealing temperature increases, the grain size increases. The annealing process is always accompanied by the growth of CsPbBr₃ compared to unannealed, which is also consistent with our analysis.

Table S3. Fitting results of the time-resolved PL spectrum at different annealing times based on dual-exponential fitting

| Sample | τ_1 (ns) | $\tau_2(ns)$ |
|--------|---------------|--------------|
| 40 s | 1.06 | 6.94 |
| 60 s | 2.02 | 26.25 |
| 80 s | 1.57 | 15.44 |
| 100 s | 1.12 | 10.07 |

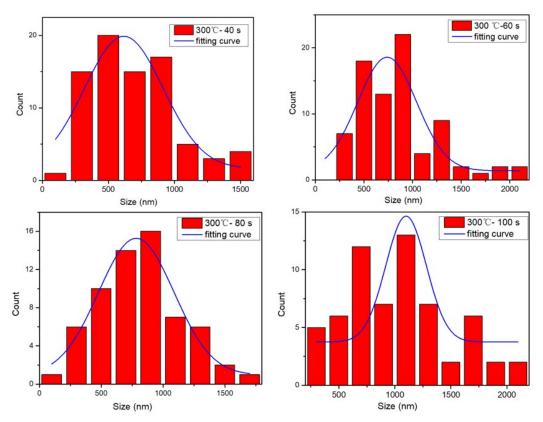


Figure S3 Domain size distribution histograms of different annealing time.

As the annealing time increases, the small grains corresponding to CsPbBr₃ grow together with Cs₄PbBr₆ grains and the grain size keeps increasing.

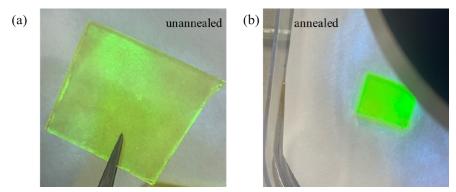


Figure S4 Images of the films after being placed in the air for 150 days (a) unannealed (b) annealed

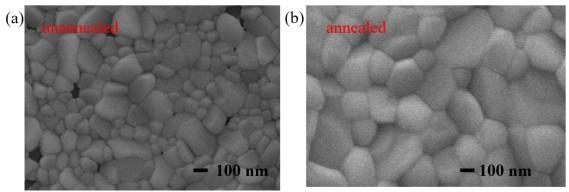


Figure S5 Surface morphology of films after being placed in the air for 150 days (a) unannealed (b) annealed

We can see that some white spots on the unannealed surface, while the surface of the annealed film is smooth. So we observed its surface morphology and found that holes appeared in the film, while the annealed film are uniform and dense, and the surface is fully covered, the grain size was increased.