

Supplementary Materials for
**Tailoring crystallization kinetics for scalable and efficient large-area
perovskite light-emitting diodes**

Sung-Doo Baek *et al.*

Corresponding author: Yoon Ho Lee, yoohlee@sungshin.ac.kr; Letian Dou, letian.dou@emory.edu

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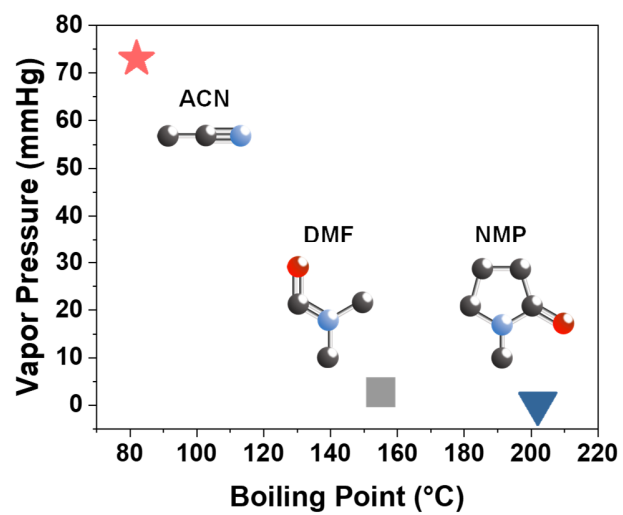


Fig. S1. Vapor pressures and boiling points of DMF, NMP, and ACN.

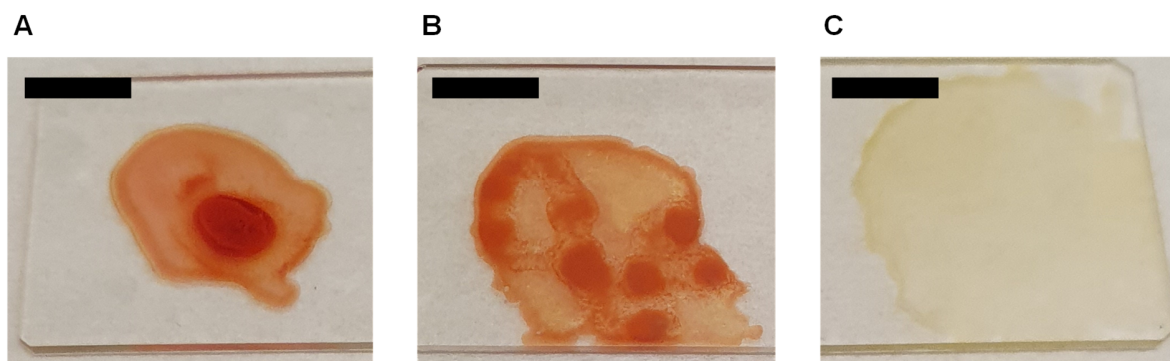


Fig. S2. Macroscopic appearance of drop-cast perovskite films from different solvent systems. Photographs of perovskite films prepared by drop-casting (A) single-, (B) binary-, and (C) ternary-solvent perovskite precursor solutions onto glass slides, followed by vacuum drying (scale bars: 1 cm).

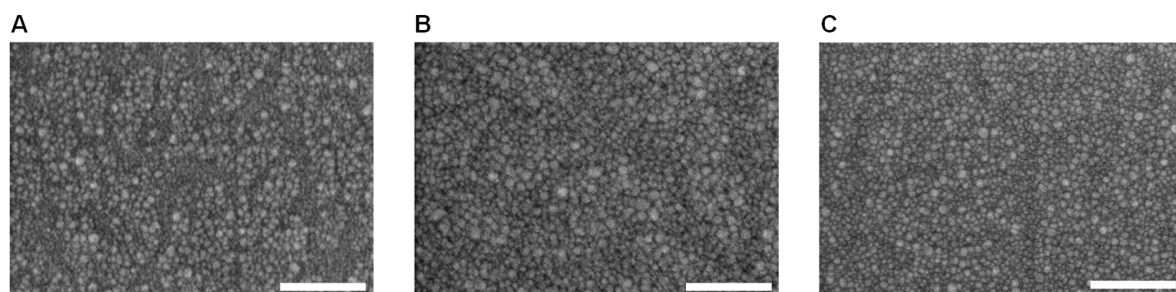


Fig. S3. Surface morphology of perovskite films prepared with different solvent systems. SEM images of perovskite films prepared with (A) single-, (B) binary-, and (C) ternary-solvent systems (scale bars: 1 μm).

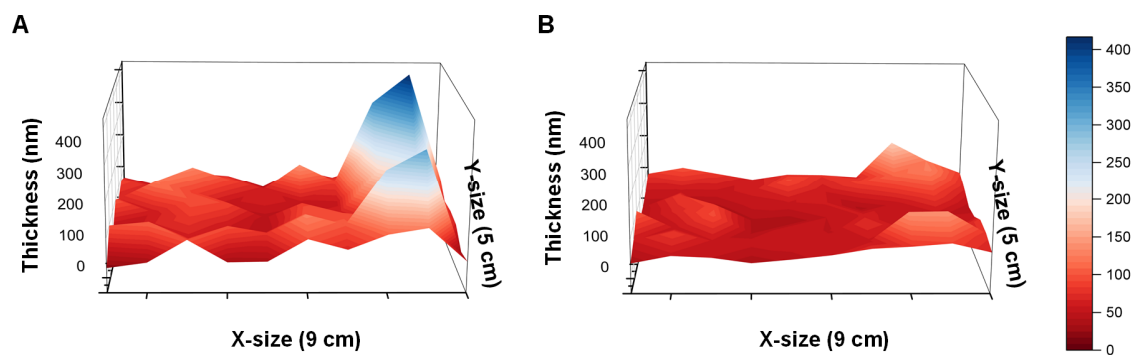


Fig. S4. Thickness uniformity across large-area films from binary- and ternary-solvent systems.

Thickness profiles measured across $90 \times 50 \text{ mm}^2$ perovskite films prepared with (A) binary- and (B) ternary-solvent systems.

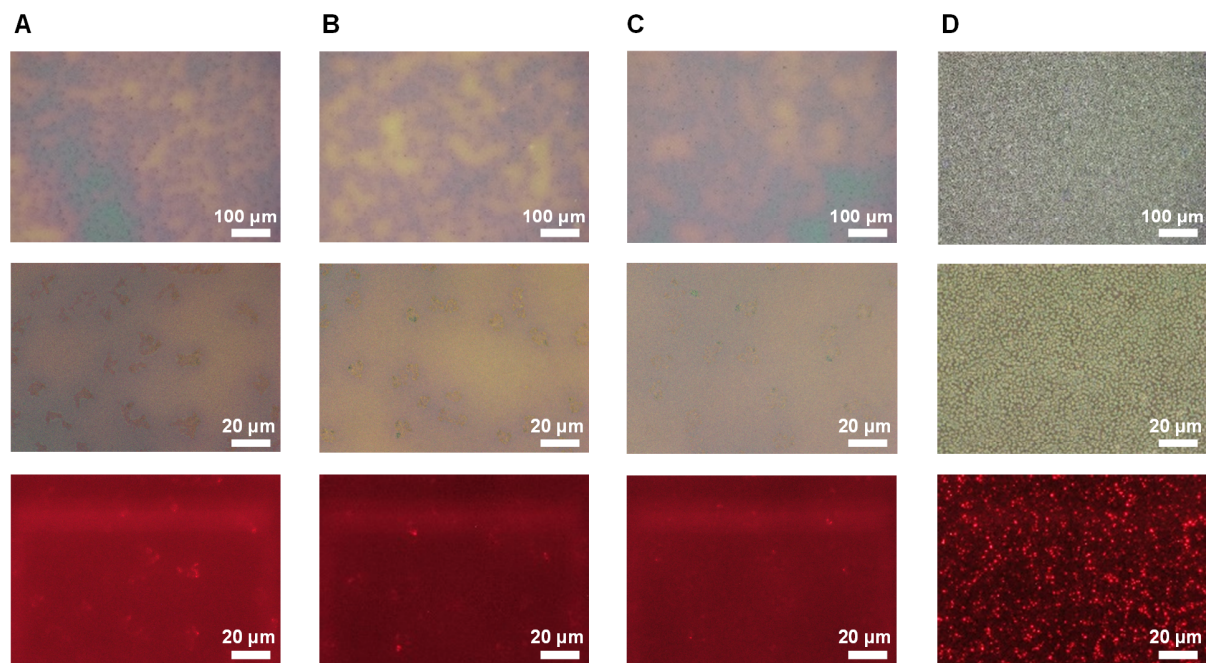


Fig. S5. Microscale morphology and emission homogeneity as a function of DMF:NMP volume ratio.

OM images (top), enlarged OM images (middle), and corresponding PL-OM images (bottom) of perovskite films prepared with **(A)** the single-solvent system (DMF-only) and the binary-solvent system (DMF/NMP) at DMF:NMP volume ratios of **(B)** 8:1, **(C)** 6:1, and **(D)** 4:1. The top-row OM images in panels (A) and (C) are reproduced from Fig. 1H top and middle panels for systematic comparison.

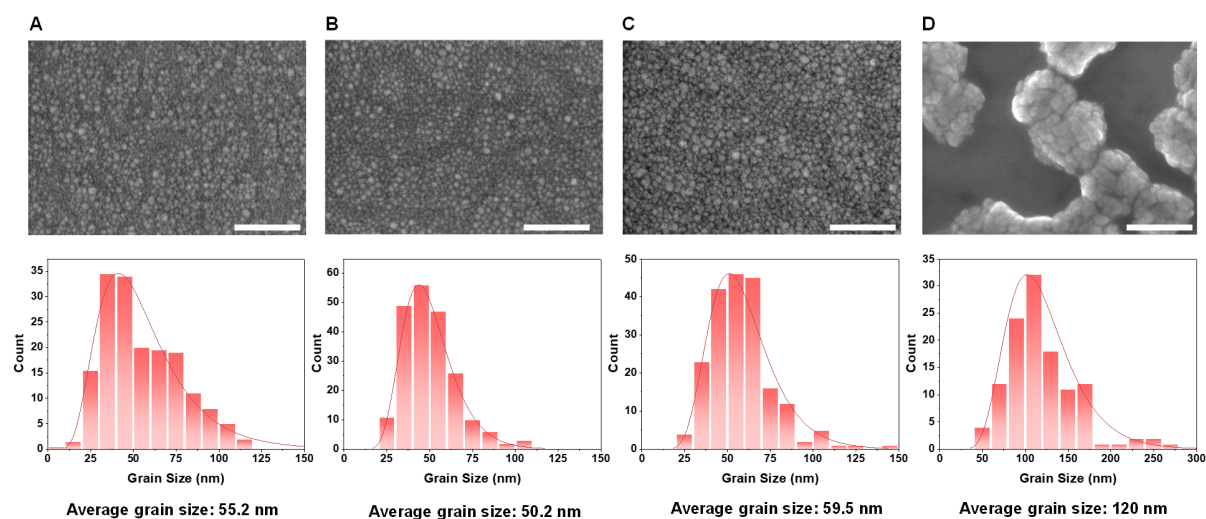


Fig. S6. Surface morphology and grain-size distribution as a function of DMF:NMP volume ratio.

SEM (top) and grain-size distributions (bottom) of perovskite films prepared with (A) the single-solvent system (DMF-only) and the binary-solvent system (DMF/NMP) at DMF:NMP volume ratios of (B) 8:1, (C) 6:1, and (D) 4:1 (scale bars: 1 μm). The SEM images in panels (A) and (C) are reproduced from Fig. S3A and S3B respectively, for systematic comparison across DMF:NMP volume ratios alongside the corresponding grain-size distributions.

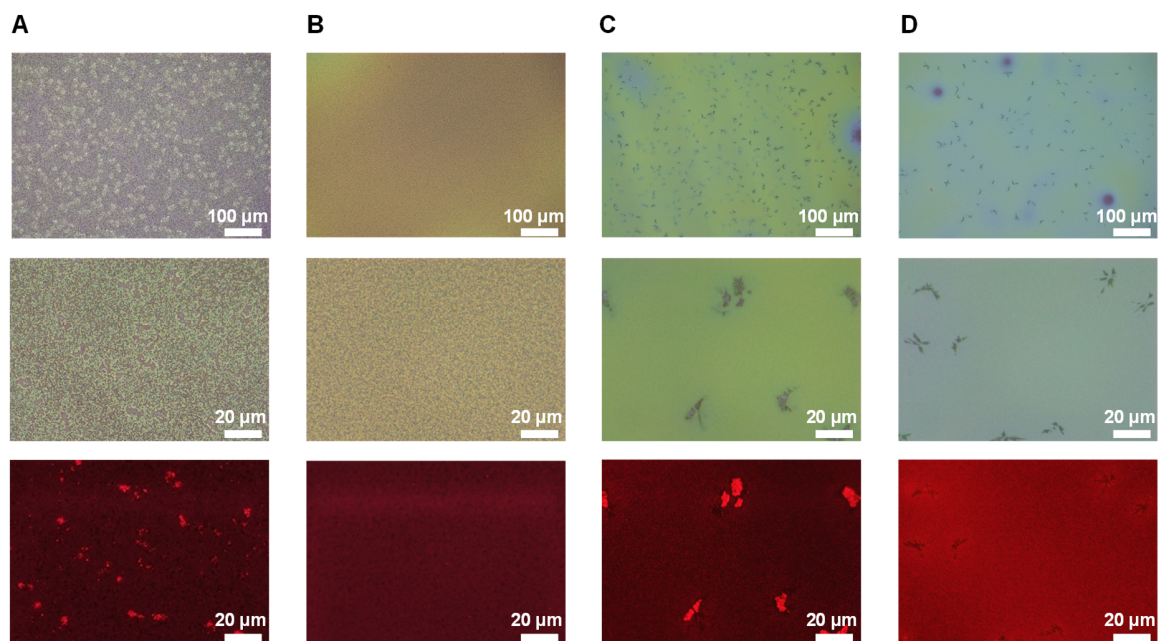


Fig. S7. Microscale morphology and emission homogeneity as a function of ACN volume fraction.

OM images (top), enlarged OM images (middle), and corresponding PL-OM images (bottom) of perovskite films prepared with the ternary-solvent system (DMF/NMP/ACN) at a fixed DMF:NMP volume ratio of 6:1 and ACN volume fractions of **(A)** 10%, **(B)** 20%, **(C)** 30%, and **(D)** 40%. The top-row OM image in panel (B) is reproduced from Fig. 1H bottom panel for systematic comparison.

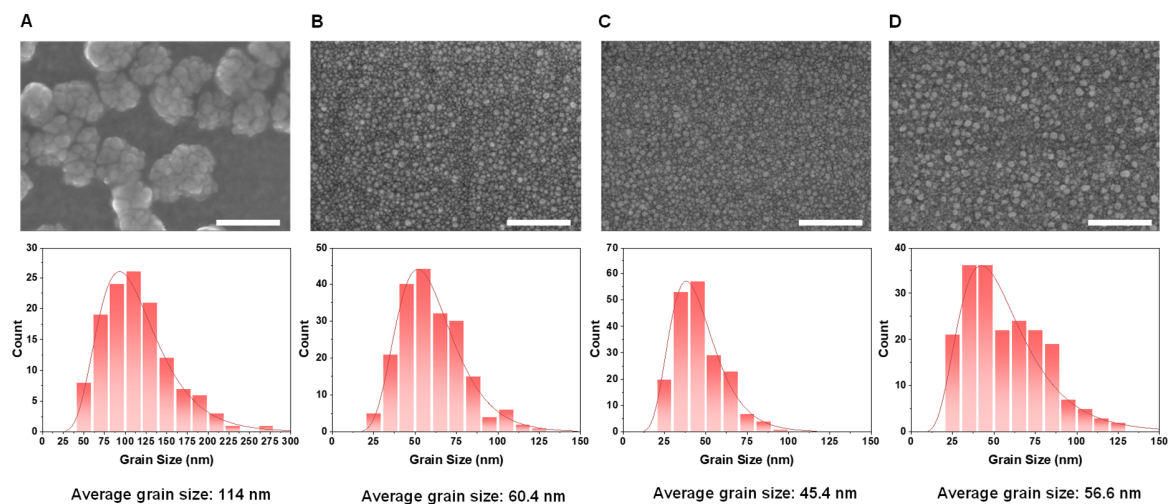


Fig. S8. Surface morphology and grain-size distribution as a function of ACN volume fraction.

SEM (top) and grain-size distributions (bottom) of perovskite films prepared with the ternary-solvent system (DMF/NMP/ACN) at a fixed DMF:NMP volume ratio of 6:1 and ACN volume fractions of (A) 10%, (B) 20%, (C) 30%, and (D) 40% (scale bars: 1 μm). The SEM image in panel (B) is reproduced from Fig. S3C for systematic comparison across ACN volume fraction alongside the corresponding grain-size distributions.

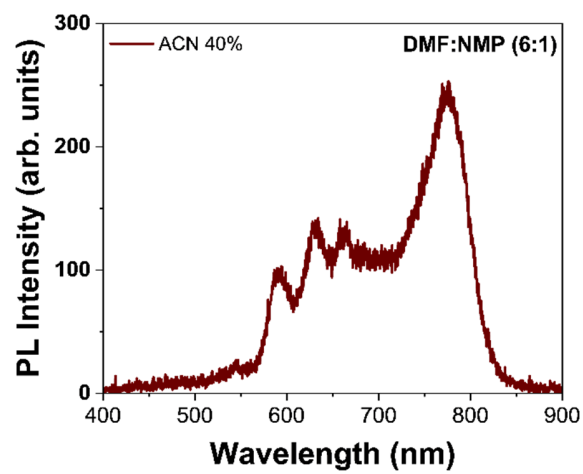


Fig. S9. Enlarged PL spectrum of a ternary-solvent film at 40% ACN volume fraction.

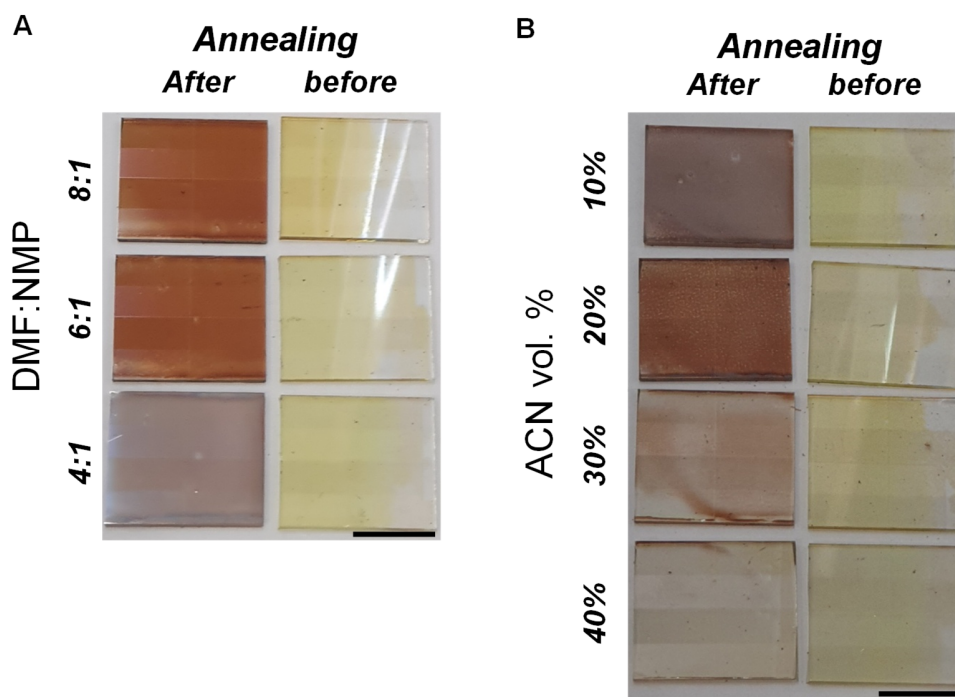


Fig. S10. Macroscopic film appearance before and after thermal annealing.

Photographs of perovskite films prepared using **(A)** the binary-solvent system and **(B)** the ternary-solvent system (DMF:NMP = 6:1), before and after thermal annealing (scale bars: 1.6 cm).

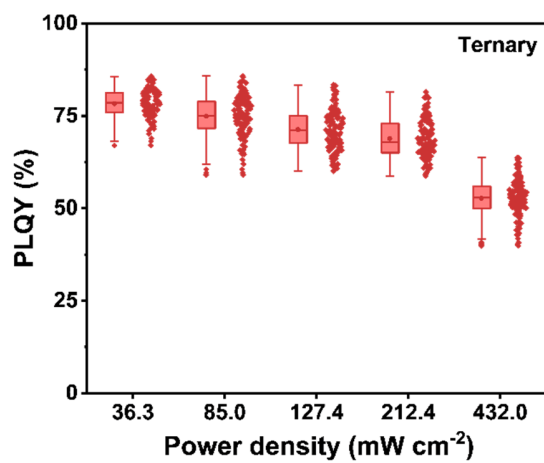


Fig. S11. Excitation-power-dependent PLQY distributions of the optimized ternary-solvent film.

Each distribution includes 120 measurement points per power level, displayed as box plots with overlaid data points.

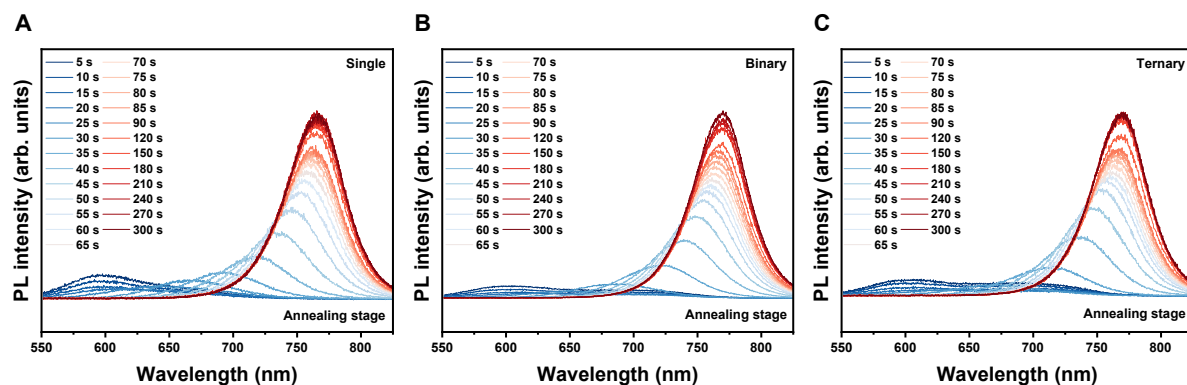


Fig. S12. In situ PL evolution during thermal annealing.

Spectra were continuously acquired at 100 °C for films prepared with (A) single-, (B) binary-, and (C) ternary-solvent systems.

Note: The PL peak blue-shifts to ~770 nm during thermal annealing due to the temperature dependence of the bandgap. At 100 °C, the bandgap increases relative to room temperature, resulting in emission at shorter wavelengths.

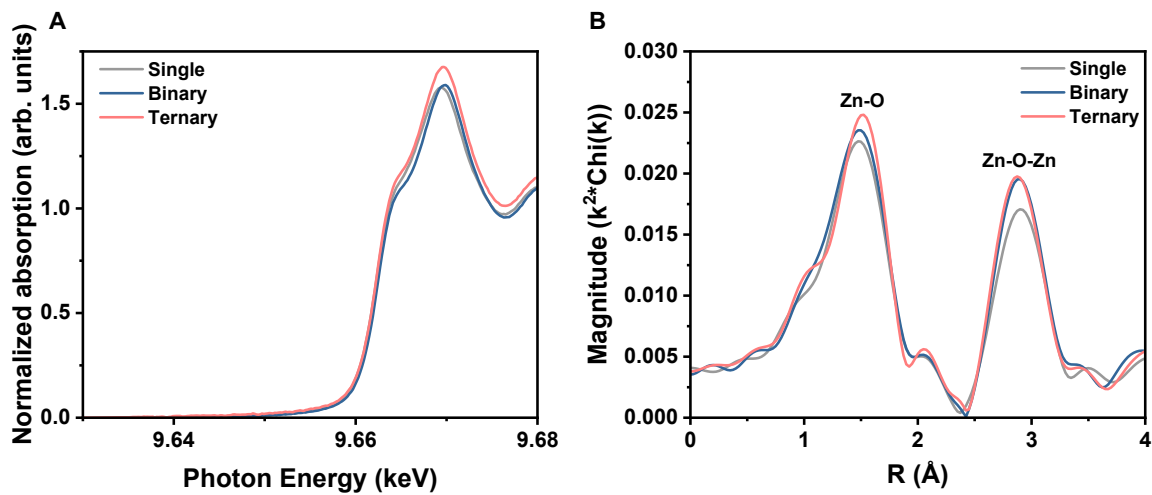


Fig. S13. Solvent-dependent local Pb coordination probed by Pb L3-edge X-ray absorption spectroscopy.

(A) XANES and **(B)** EXAFS spectra of perovskite films prepared with single-, binary-, and ternary-solvent systems on ZnO-coated substrates.

Note: Control measurements verified that the ZnO-coated substrate remains unaffected by the solvents, confirming that the observed spectral features originate exclusively from the perovskite precursors.

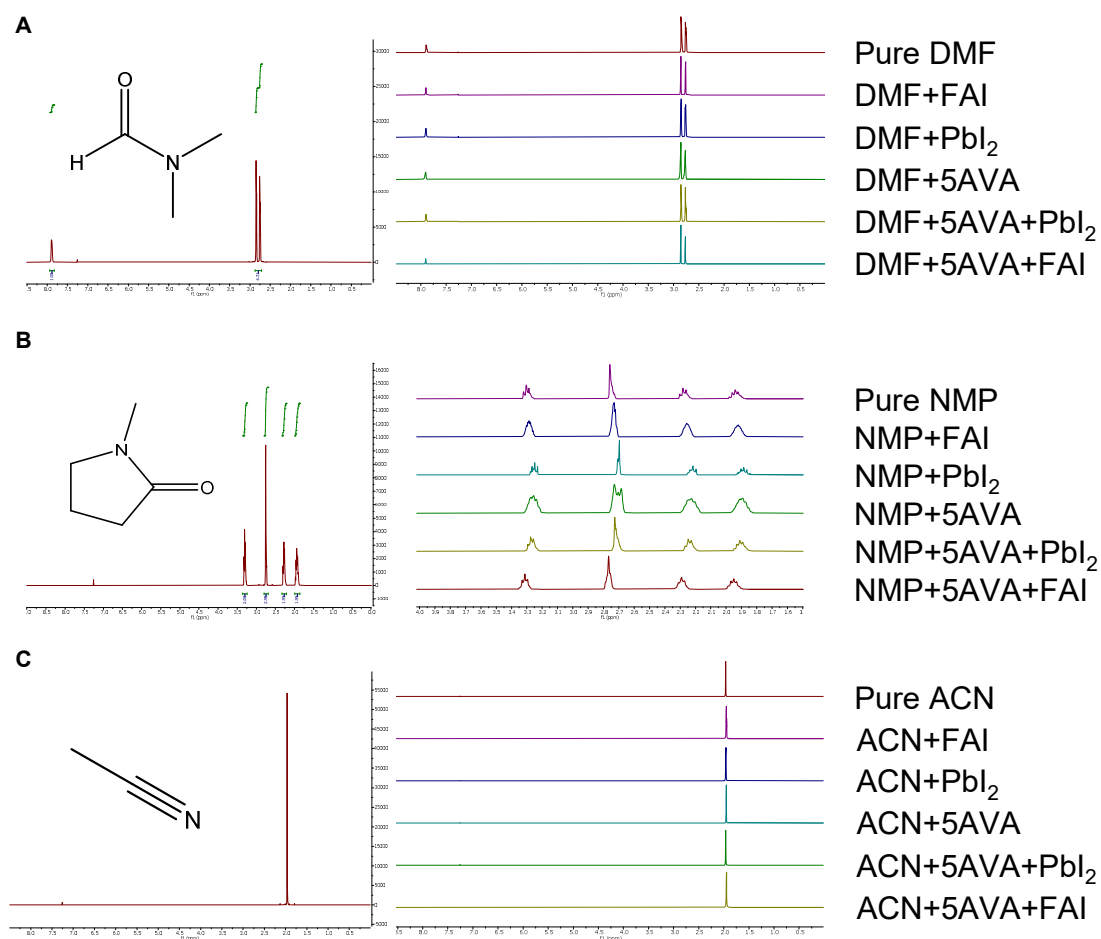


Fig. S14. Solvent-dependent ¹H-NMR chemical shifts of perovskite precursors.

Spectra were acquired with PbI₂ and FAI dissolved in (A) DMF, (B) NMP, and (C) ACN.

Note: Consistent with its higher Kamlet–Taft β value (strong hydrogen-bond accepting ability) (20), NMP interacts more strongly with FAI than DMF. Accordingly, DMF and ACN show negligible changes in ¹H resonances upon addition of the precursor solutes, whereas NMP exhibits pronounced chemical shifts in the presence of PbI₂ and FAI, indicating strong coordination and hydrogen bonding.

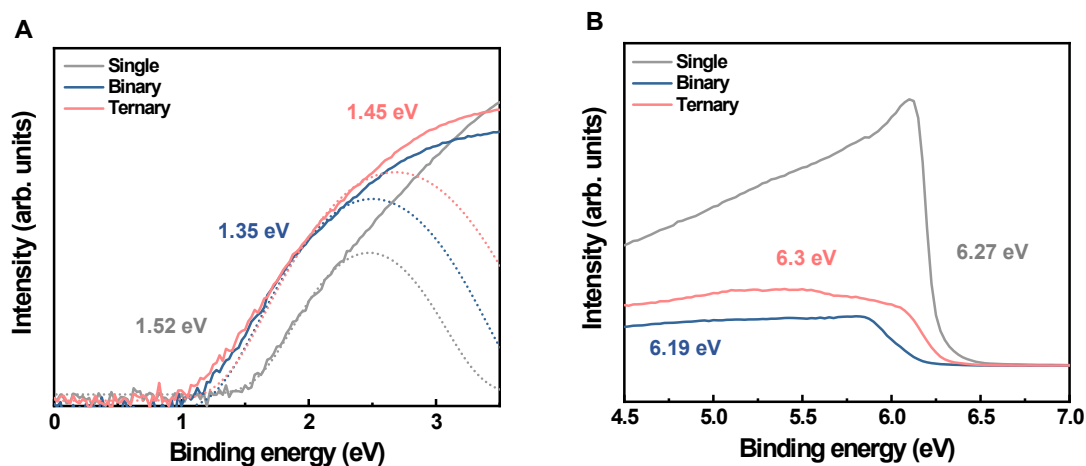


Fig. S15. UPS analysis of perovskite films prepared with different solvent systems.
(A) Secondary electron cutoff spectra and (B) valence-band spectra of perovskite films fabricated with single-, binary-, and ternary-solvent systems.

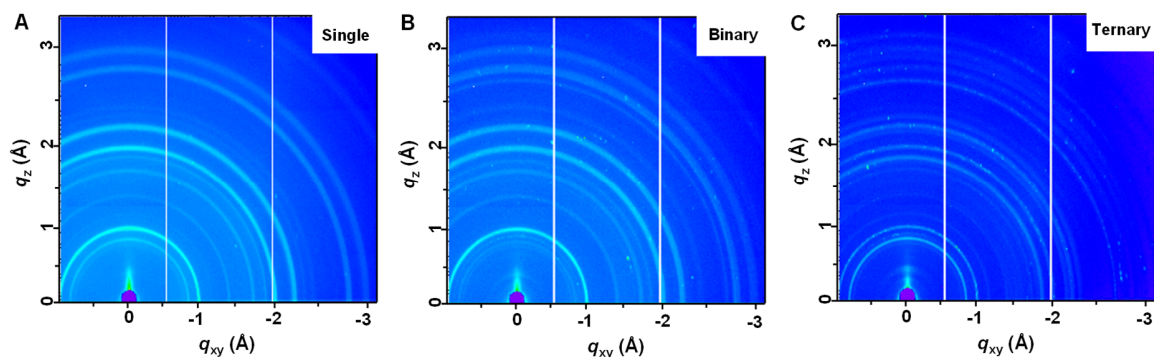


Fig. S16. Two-dimensional GIWAXS patterns of perovskite films from different solvent systems.

Films were prepared with (A) single-, (B) binary-, and (C) ternary-solvent systems.

Note: While the 3D α -FAPbI₃ phase constitutes the dominant crystalline framework across all solvent systems, additional diffraction features characteristic of low-dimensional phases emerge in the binary system and become more pronounced in the ternary-solvent system.

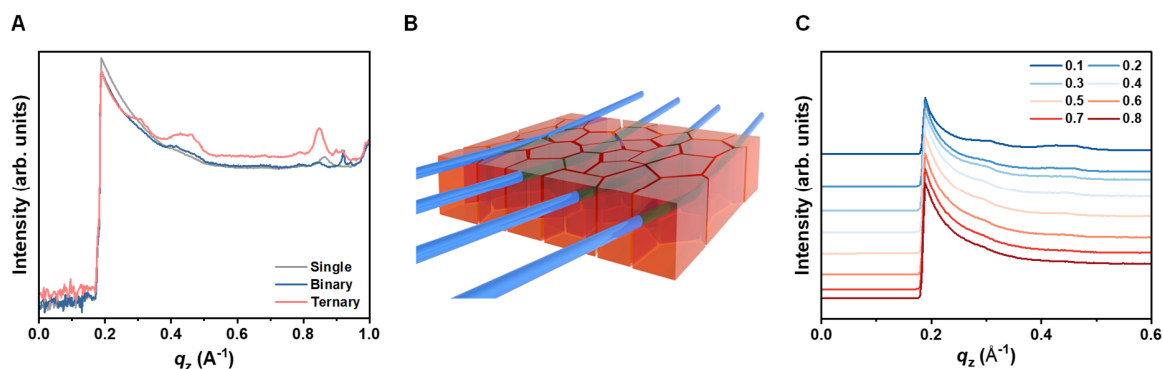


Fig. S17. Depth-resolved GIWAXS analysis via variable incidence-angle measurements.

(A) Out-of-plane GIWAXS line profiles of perovskite films prepared with single-, binary-, and ternary-solvent systems. (B) Schematic of the incidence-angle geometry. (C) Angle-dependent line profiles of the ternary-solvent film.

Note: Low-dimensional phases are attributed to quasi-2D 5AVA-based perovskites. Angle-dependent GIWAXS analysis indicates that these phases are predominantly localized at the film surface.

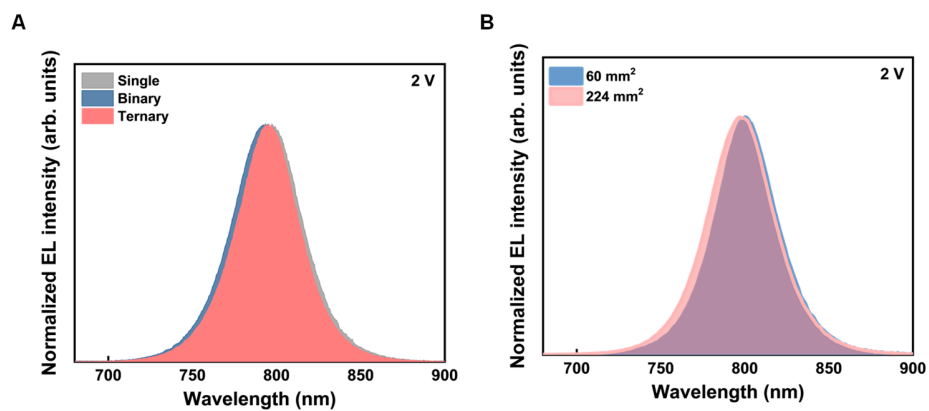


Fig. S18. EL spectra of PeLEDs with different solvent systems and active areas.

All spectra are normalized to peak intensity. **(A)** Devices with a 10 mm² active area fabricated using single-, binary-, and ternary-solvent systems. **(B)** Ternary-solvent devices scaled to active areas of 60 mm² and 224 mm².

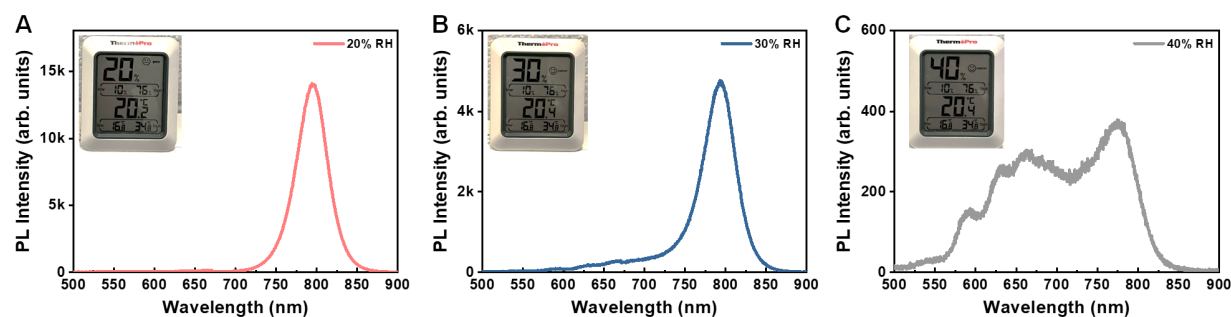


Fig. S19. Humidity dependence of blade-coated ternary-solvent films under ambient conditions.

Spectra were recorded on films blade-coated at (A) 20% RH, (B) 30% RH, and (C) 40% RH. Insets display photographs of the temperature and RH readings captured during deposition.

Note: Relative to 20% RH, films prepared at 30% RH show a marked decrease in NIR PL intensity and an additional emission feature in the 550–750 nm range. At 40% RH, the degradation becomes more severe. These trends suggest moisture-enhanced nonradiative losses and/or humidity-induced film heterogeneity during crystallization under ambient conditions.

Table S1. EXAFS fitting parameters for Pb L3-edge XAS of precursor solutions prepared with the single-, binary-, and ternary-solvent systems.

Pb L3-edge XAS						
Sample	Edge energy (keV)	Scattering path	CN	R (Å)	$\Delta\sigma^2$ (Å ²)	ΔE_0 (eV)
Single	13.033	Pb–I	2.8	2.96	0.013	–5.4
		Pb–O	0.3	2.28	0.005	–6.8
Binary	13.035	Pb–I	3	2.97	0.013	–3.3
		Pb–O	0.2	2.4	0.005	0.8
Ternary	13.0338	Pb–I	3.1	2.95	0.013	–6.3
		Pb–O	0.3	2.41	0.005	7.7

Table S2. EXAFS fitting parameters for Zn K-edge XAS of ZnO layers coated with perovskite films prepared with the single-, binary-, and ternary-solvent systems.

Zn K-edge XAS						
Sample	Edge energy (keV)	Scattering path	CN	R (Å)	$\Delta\sigma^2$ (Å ²)	ΔE_0 (eV)
Single	9.6621	Zn–O	3.9	1.96	0.004	0.94
Binary	9.6624	Zn–O	3.9	1.96	0.004	0.87
Ternary	9.6624	Zn–O	4	1.96	0.004	1.3

Table S3. Reported device characteristics of large-area PeLEDs.

EL peak (nm)	Fabrication method	Area (mm ²)	Peak EQE (%)	Operational stability (<i>T</i> ₈₀ ; driving condition)	Device structure	Year	Ref. number
795	Blade coating	10	25.2	23.4 h at 10 mA cm ⁻² 3 h at 50 mA cm ⁻²	ITO/ZnO/FAPbI ₃ /TFB/MoO _x /Au	2026	This work
800		60	22.1	-			
799		224	19.0	-			
650		4	15.3	~16 s	ITO/PEDOT:PSS/Poly-TPD/CsPbI ₃ QD/TPBi/Al	2024	33
489		4	10.3	~230 s at 100 cd m ⁻²	ITO/PVK/CsPb(Br _{0.84} Cl _{0.16}) ₃ /TPBi/LiF/Al	2022	14
515		12	8.2	~2.6 min at 10 mA cm ⁻²	ITO/NiO _x /(PEA) ₂ Cs _{<i>n</i>-1} Pb _{<i>n</i>} Br _{3<i>n</i>+1} /TPBi/LiF/Al	2022	34
		100	6.1	-			
732		4	16.1	~9.3 h at 3 mA cm ⁻²	ITO/poly-TPD/MAPbI ₃ /TPBi/LiF/Al	2021	13
		100	12.7	-			
769	Inkjet printing	4	14.3	~1 h at 10 mA cm ⁻²	ITO/PEDOT:PSS/Poly-TPD/FA _{0.8} CS _{0.2} PbI ₃ /TPBi/LiF/Al	2024	18
520		225	2.8	~1 min at 90 cd m ⁻²	ITO/PEDOT:PSS/Poly-TPD/FA _{0.3} CS _{0.7} PbBr ₃ /TPBi/LiF/Al	2020	35

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