

Supplementary File:

Phosphine oxide based semiconducting small molecule as an additive and an electron transport layer enables efficient and stable perovskite light emitting devices

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Keywords: Perovskite LED, PO-T2T small molecule additive, Methylammonium lead bromide (MAPbBr₃), Stable PL, Higher EL lifetime stability

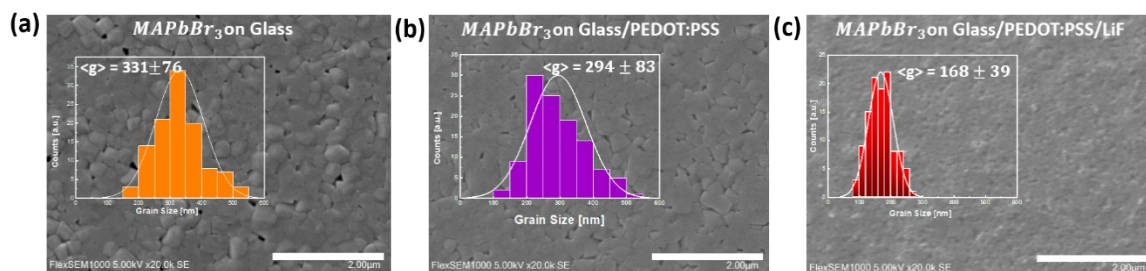
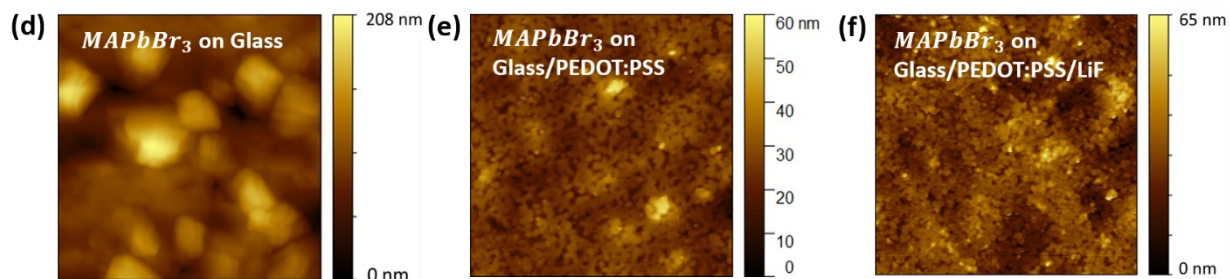


Figure S1. SEM images [Top row] and AFM images [Bottom Row] of MAPbBr₃ films on top of Glass,



Glass/PEDOT: PSS and Glass/PEDOT:PSS/LiF. The scale bar for SEM is 2 μm and AFM images are of scan size 5 × 5 μm

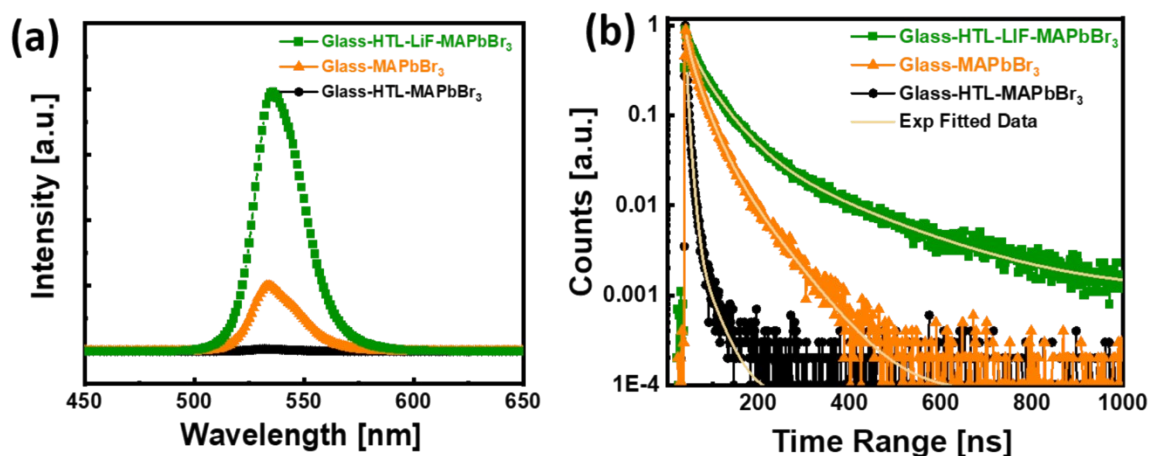


Figure S2. (a) Steady state photoluminescence spectra (b) Time resolved photoluminescence spectra of MAPbBr₃ films deposited on Glass, on Glass/PEDOT:PSS and on Glass/PEDOT:PSS/3 nm LiF.

	τ_1 [ns]	B_1 [%]	τ_2 [ns]	B_2 [%]	τ_3 [ns]	B_3 [%]	τ_{ave} [ns]	χ^2
Glass/ MAPbBr ₃	8.24	11.11	26.47	62.15	67.63	26.74	35.45	0.7850
Glass/PEDOT: PSS/ MAPbBr ₃	2.34	47.71	6.63	48.02	31.86	4.27	5.66	0.6284
Glass/PEDOT: PSS/LiF/ MAPbBr ₃	15.88	12.22	51.93	60.09	194.80	27.70	87.10	0.9513

Table S1. TRPL decay curve fitting and average lifetime calculated from tri-exponential fit for MAPbBr₃ films on 1) Glass 2) Glass/PEDOT:PSS 3) Glass/PEDOT:PSS/LiF .

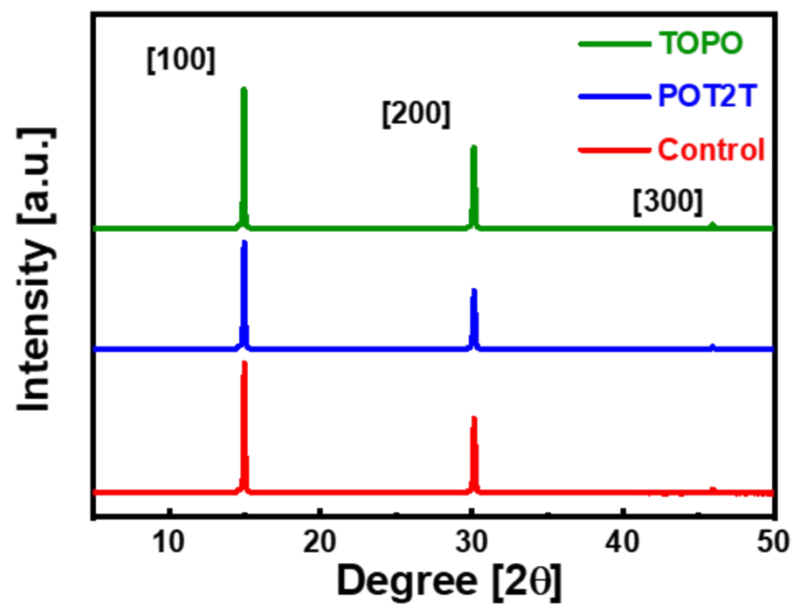


Figure S3. XRD spectra of MAPbBr₃ polycrystalline films with different additives (films made on HTL [Glass/PEDOT:PSS/3 nm LiF])

MAPbBr ₃ films made on Glass/PEDOT: PSS/3 nm LiF with below mentioned additive	[hkl]	Position [2 θ] (degree)	FWHM (degree)	Lattice Constant (Å)	Intensity (A.U)	Strain
Control (no additive)	[100]	14.96	0.11	5.91	135311	0.054
	[200]	30.14	0.15	5.92	77832	
	[300]	45.90	0.18	5.92	3913	
TOPO	[100]	14.94	0.11	5.92	145579	0.053
	[200]	30.14	0.15	5.92	85020	
	[300]	45.90	0.18	5.92	4595	
PO-T2T	[100]	14.96	0.13	5.91	111167	0.042
	[200]	30.14	0.16	5.92	61762	
	[300]	45.90	0.18	5.92	3224	

Table S2. Summary of XRD measurements on MAPbBr₃ films with different additives

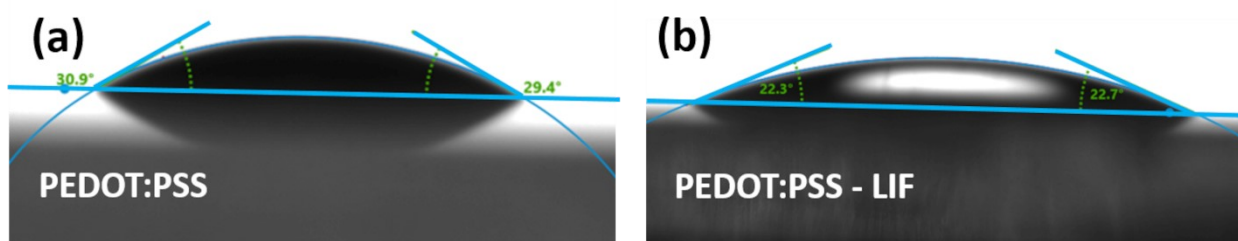


Figure S4. Contact angle of water measured (a) On glass/PEDOT:PSS layer (b) On glass/PEDOT:PSS/3 nm LiF layer

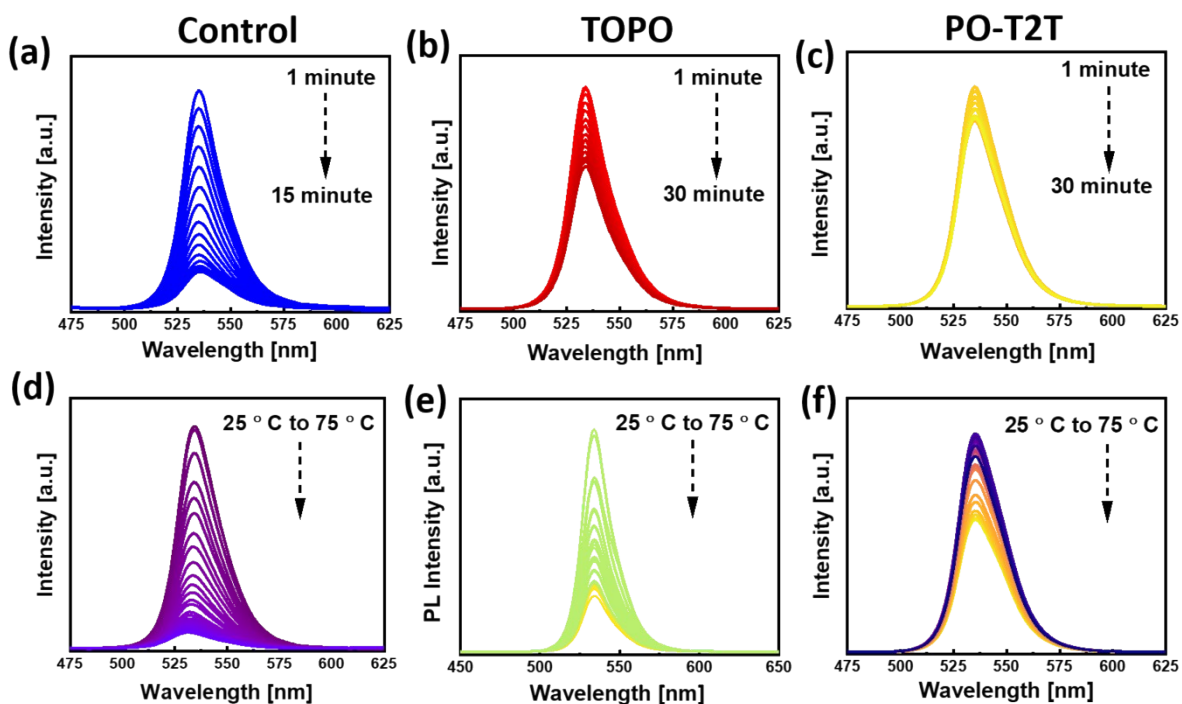


Figure S5. PL spectra recorded as a function of time (a-c) and temperature (d-f) for MAPbBr₃ films with control, TOPO and PO-T2T additives. Data is extracted from 2D PL maps shown in **Figure 5**. For control sample PL spectral intensity decreases by 83 %, for film with TOPO as additive there is decrease in PL spectral intensity by 40 % and for film with PO-T2T as additive the decrease in PL spectral intensity is significantly lower (20 %) as a function of time. Similarly, the decrease in PL Intensity for Control, TOPO as additive and PO-T2T as additive is 93 %, 75 % and 40 % respectively as a function of temperature. For all films we monitored the PL spectral intensity changes for 30 minutes and temperature is varied from room temperature to 75-degree C in 2-degree interval

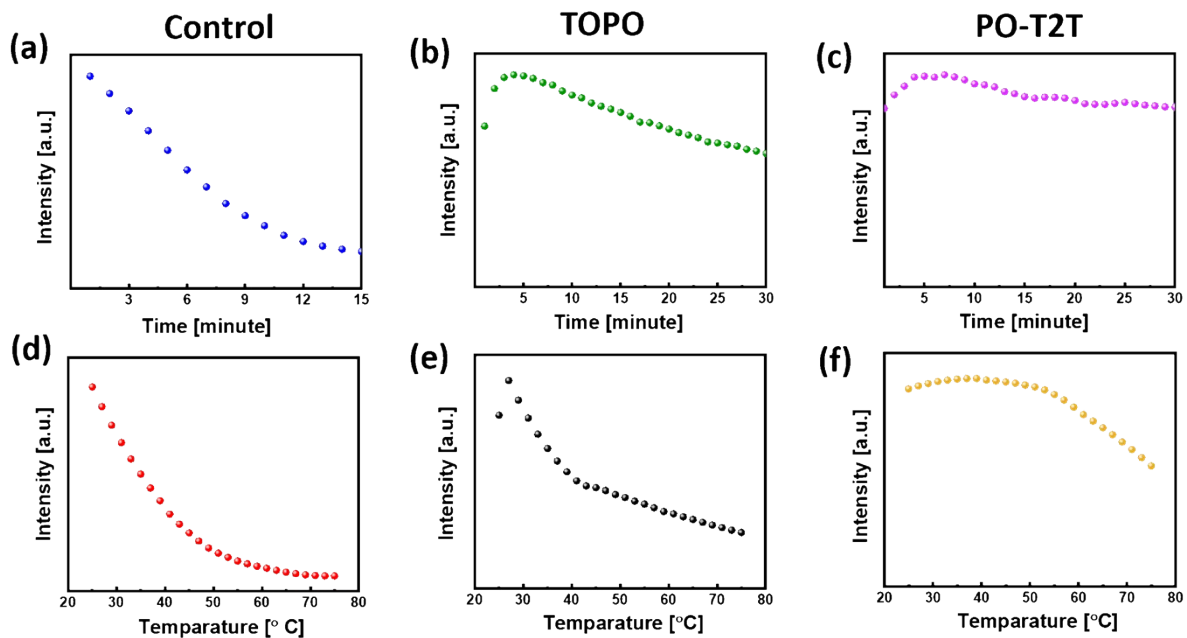


Figure S6. Integrated PL spectra as a function of time (a-c) and temperature (d-f) for MAPbBr₃ films with control, TOPO and POT-2T additives. Data is extracted from 2D PL maps shown in **Figure 5**.

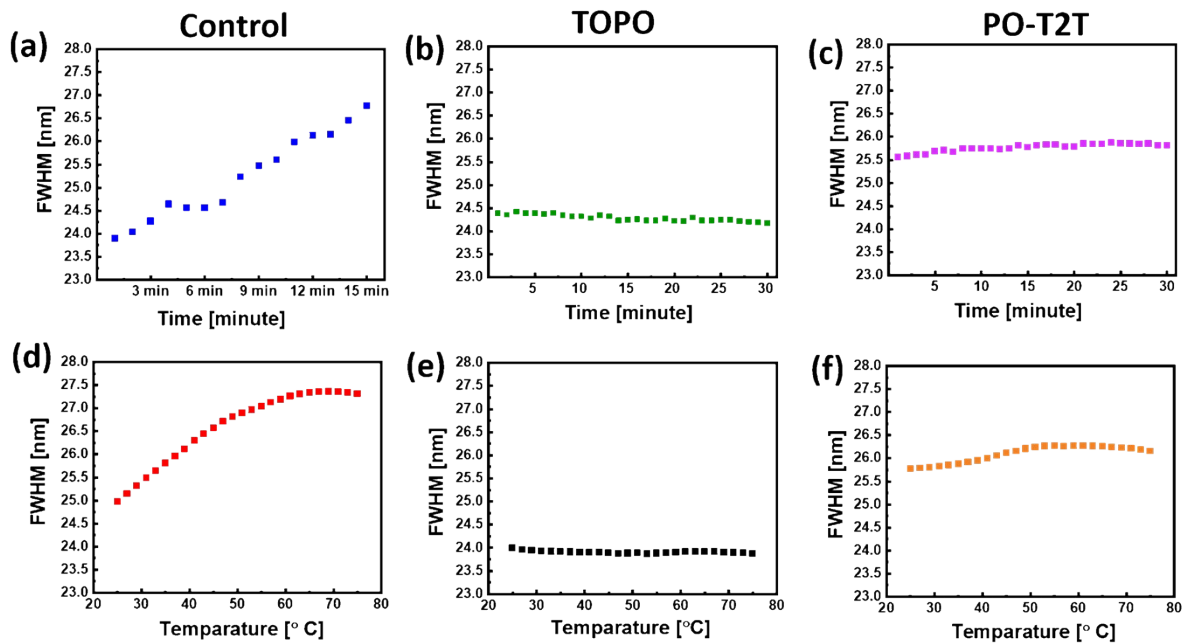
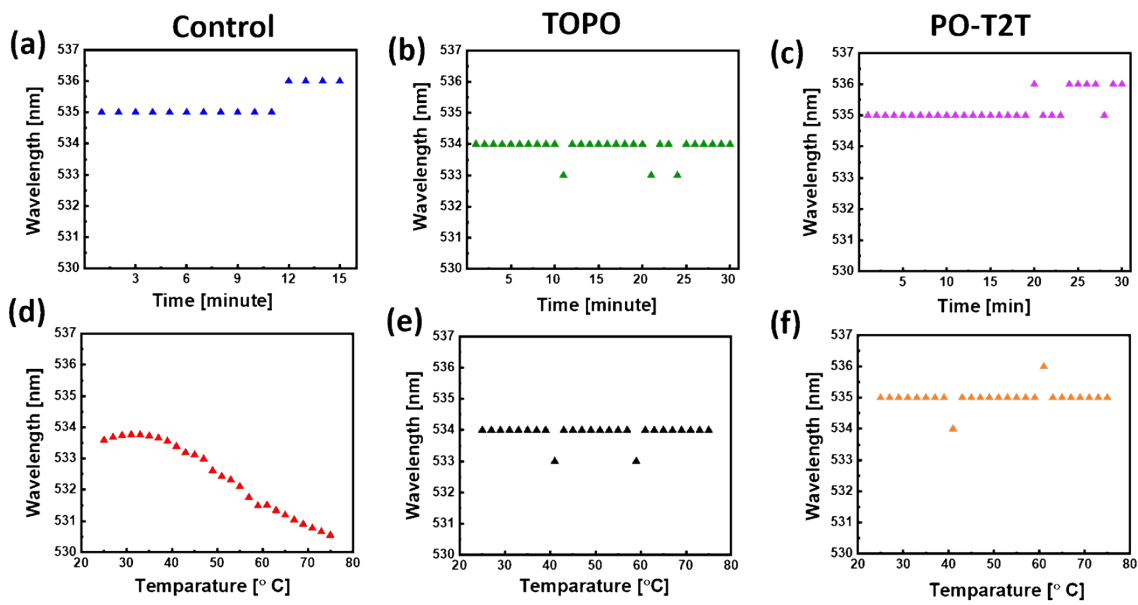


Figure S7. FWHM as a function of time (a-c) and temperature (d-f) for MAPbBr₃ films with control, TOPO and PO-T2T additives. Data is extracted from 2D PL maps shown in **Figure 5**.



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Figure S8. Peak wavelength as a function of time (a-c) and temperature (d-f) for MAPbBr₃ films with control, TOPO and PO-T2T additives. Data is extracted from 2D PL maps shown in **Figure 5**.

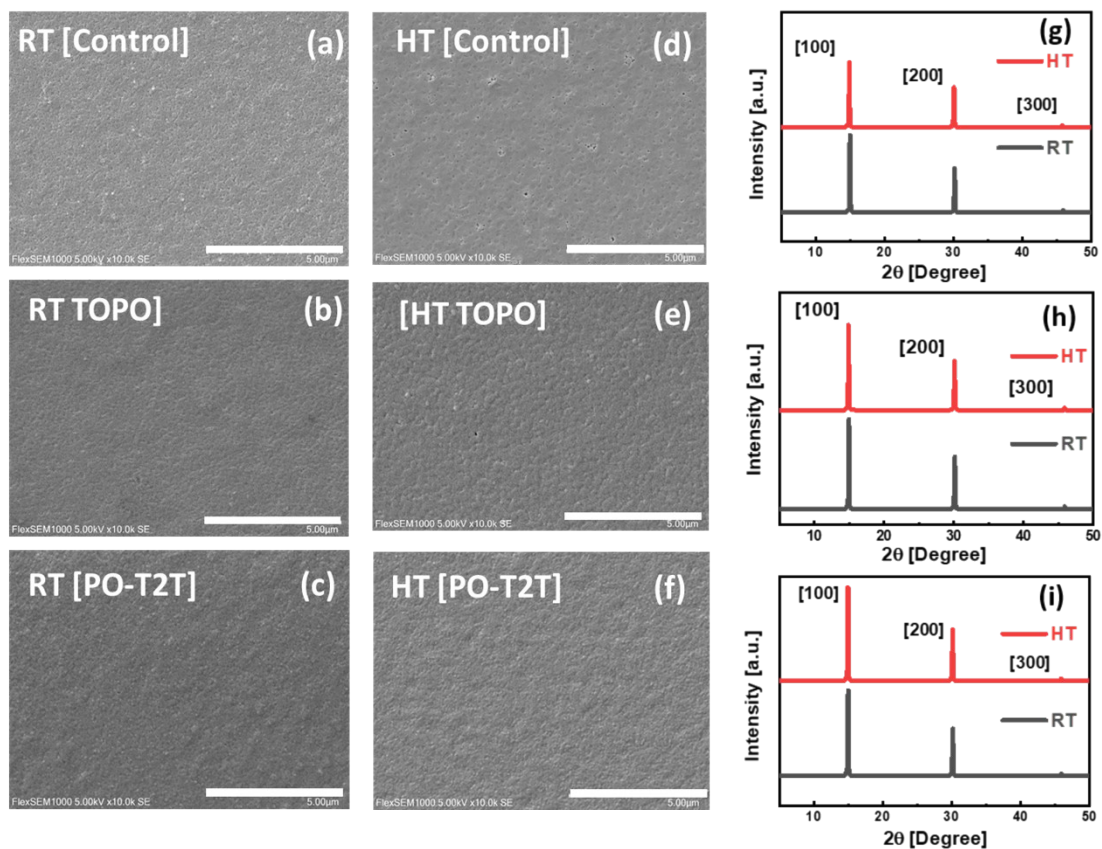


Figure S9. SEM images of MAPbBr₃ films at room temperature (a-c) and high temperature (75 deg) (d-f) for control, TOPO and POT-2T additives. XRD spectra were recorded at room and high

temperature (75 deg) for control (g), TOPO (h) and PO-T2T(i) additives. Enlarged version of XRD spectra on top of HTL for Control and PO-T2T treated films are shown in Figure (j) and (k).

Composition	[hkl]	Position [2 θ] (degree)	Lattice Constant (Å)
Control [RT]	[100]	14.96	5.91
	[200]	30.14	5.92
	[300]	45.90	5.92
PO-T2T [RT]	[100]	14.96	5.91
	[200]	30.14	5.92
	[300]	45.90	5.92
Control [HT]	[100]	14.86	5.95
	[200]	30.06	5.94
	[300]	45.82	5.93
PO-T2T [HT]	[100]	14.92	5.93
	[200]	30.12	5.92
	[300]	45.88	5.92

Table S3: Summary of XRD measurements on MAPbBr₃ films with different compositions at different temperatures.

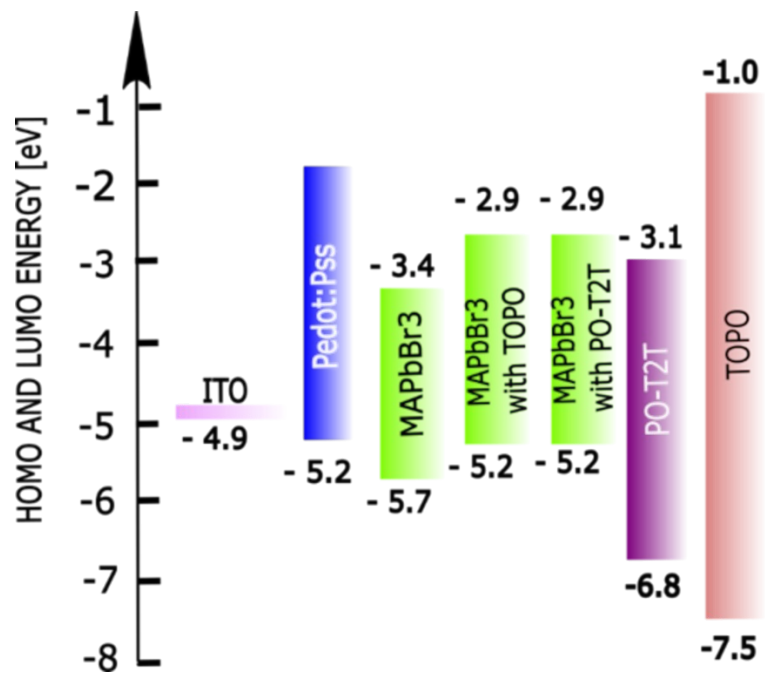


Figure S10. Energy level diagram for Pe-LED device and its components

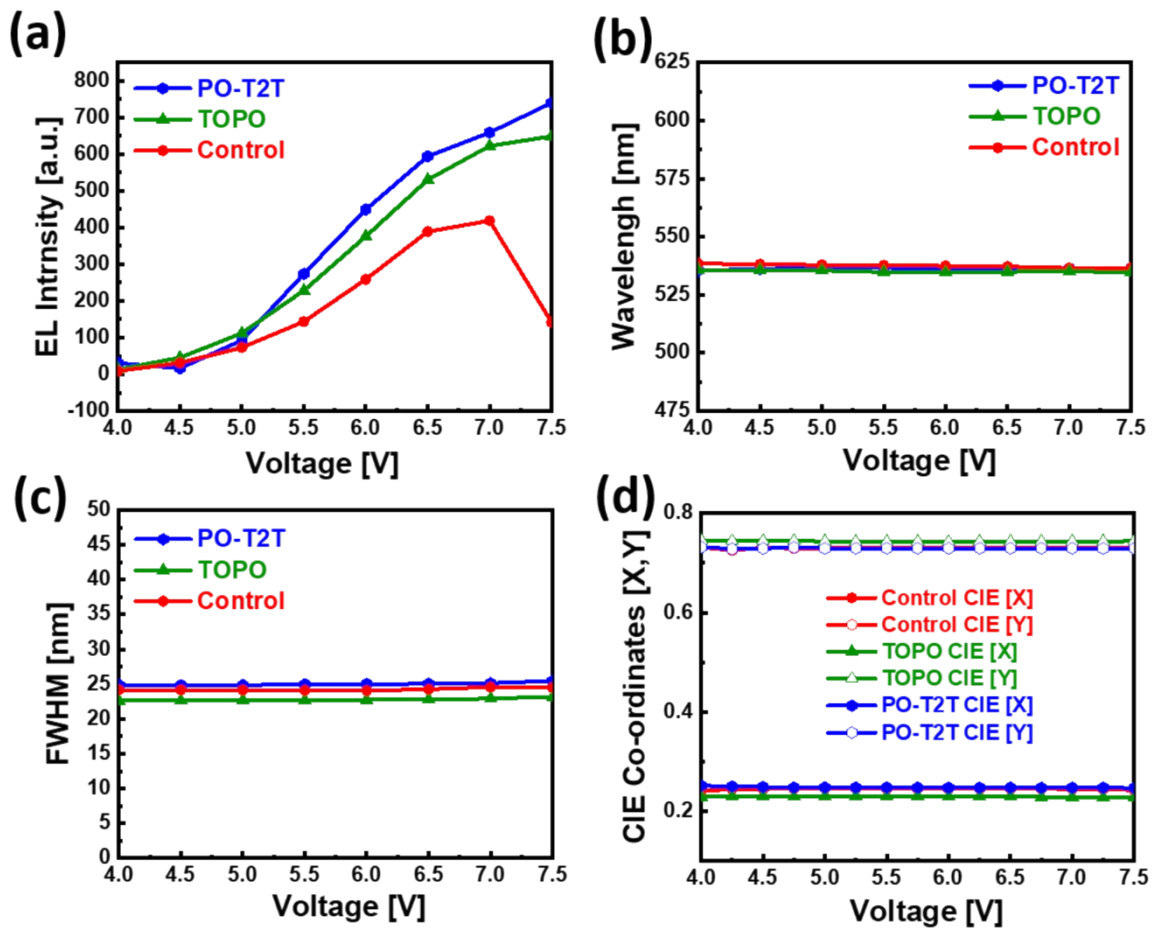


Figure S11. (a) EL intensity (from EL Spectrum), (b) Wavelength, (c) FWHM and (d) CIE coordinate (0.2, 0.7) changes plotted as a function of voltage bias for Pe-LED devices made with control, TOPO and PO-T2T additives

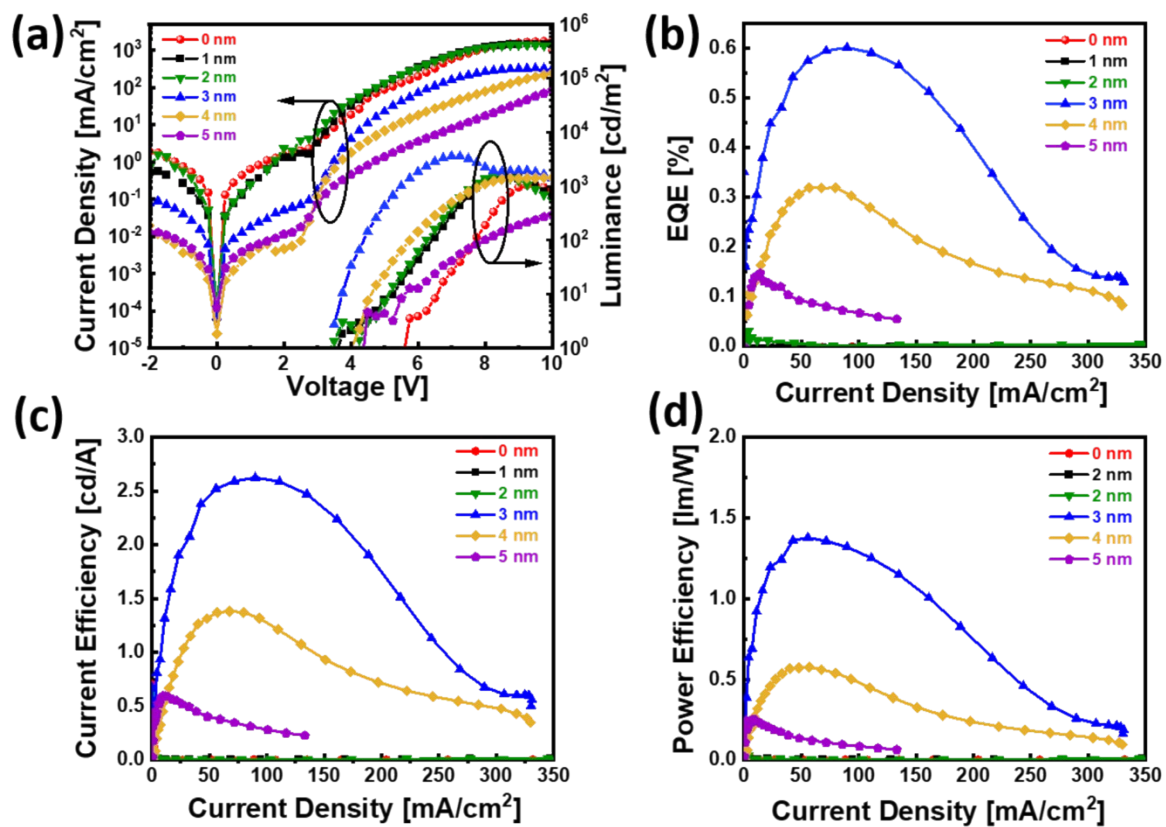


Figure S12. LiF layer optimization: Current Density-Voltage-Luminance [J-V-L] characteristics, EQE vs Current Density, Current Efficiency vs Current Density and Power Efficiency vs Current Density of Perovskite LEDs with different LiF layer thickness from 0-5nm.

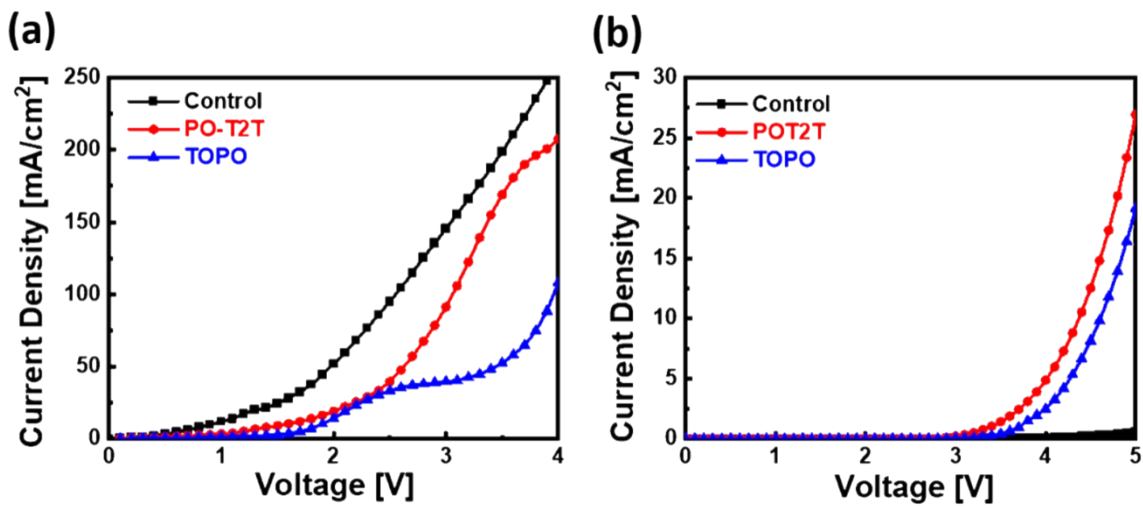


Figure S13. Current density-voltage plots for (a) Glass/ITO/PEDOT:PSS:LiF/ MAPbBr₃ with different Additives/LiF/Al (b) Glass/ITO/ MAPbBr₃ with different Additives/TPBi/LiF/Al.

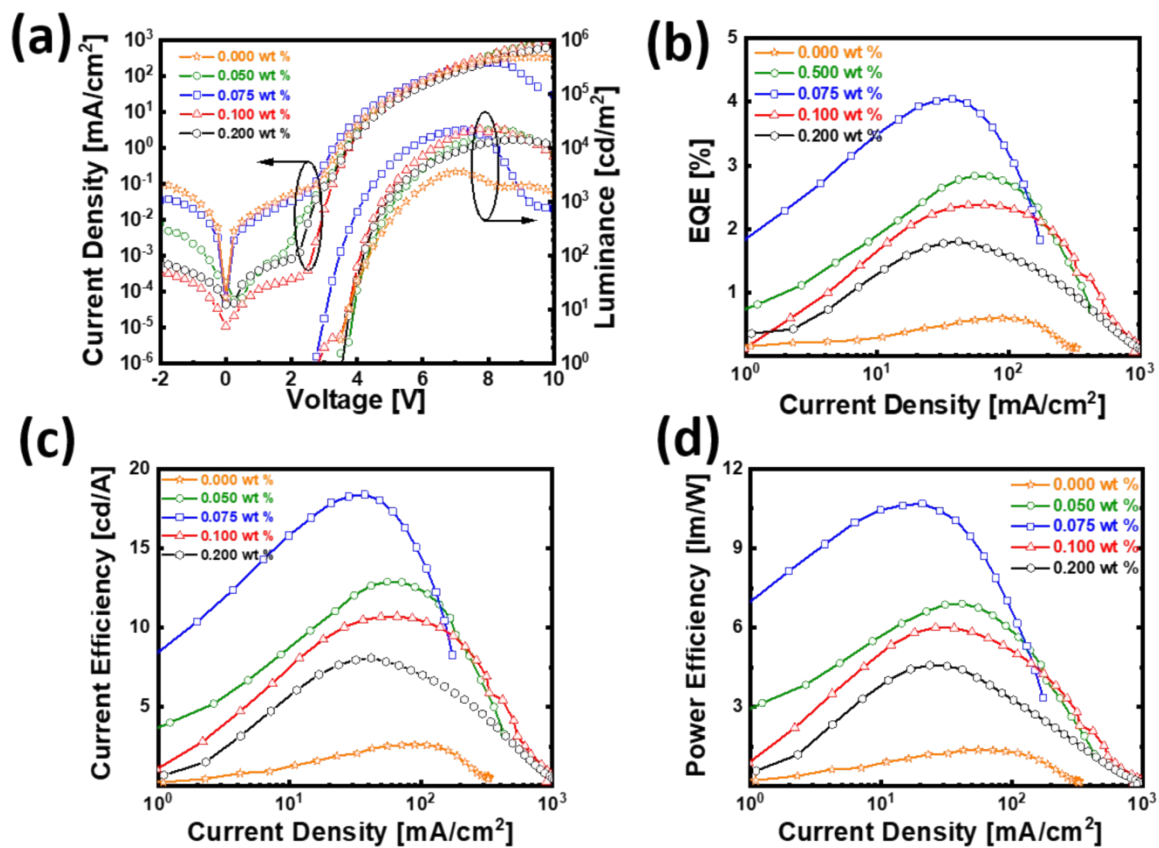


Figure S14. Optimization of PO-T2T in anti-solvent treatment for forming MAPbBr₃ films

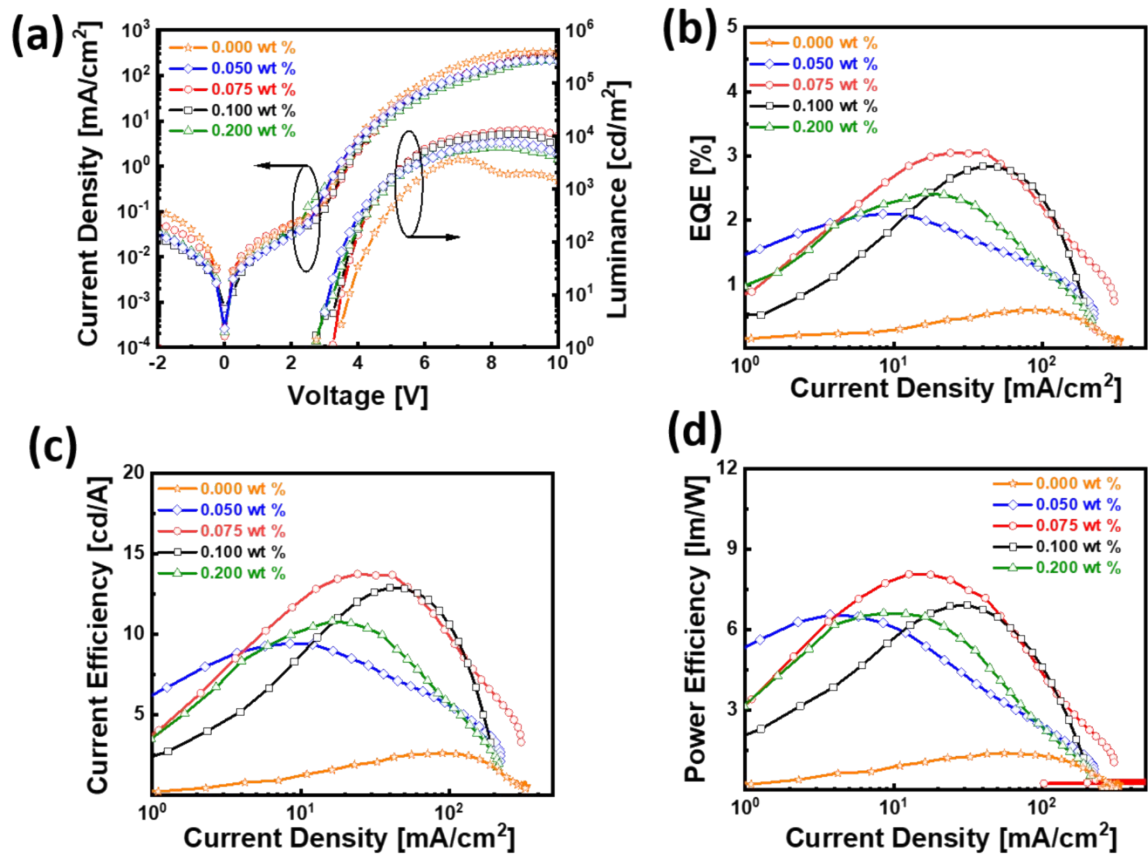


Figure S15. Optimization of TOPO in anti-solvent treatment for forming MAPbBr₃ films

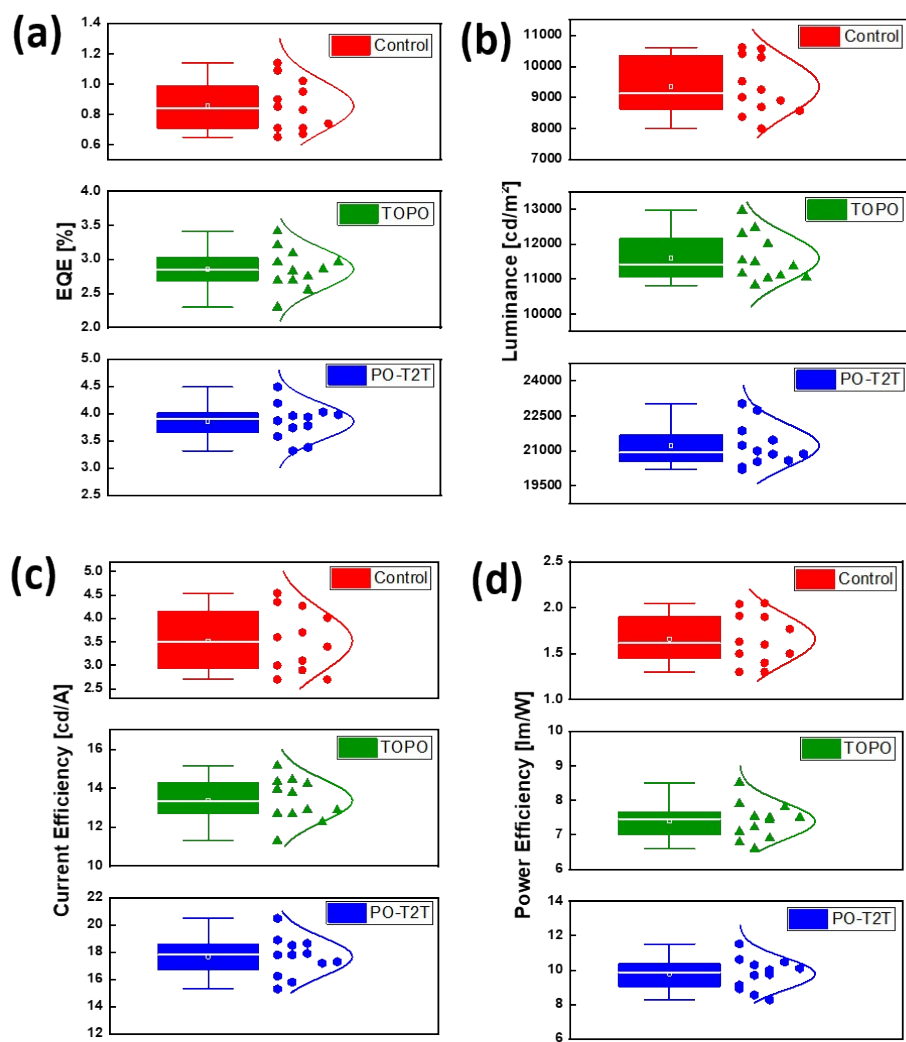


Figure S16: Histograms for (a) EQE (b) Luminance (c) Current Efficiency and (d) Power Efficiency of 12 Pe-LED devices.

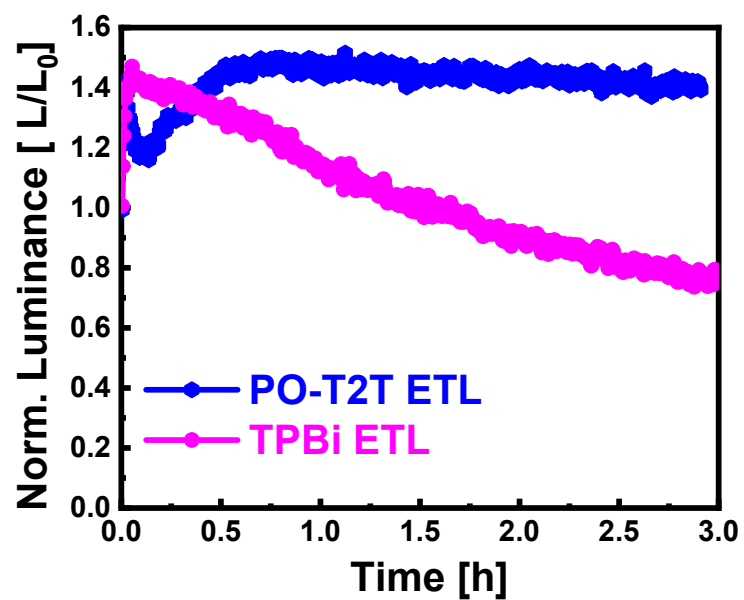


Figure S17. Comparison of Pe-LED lifetime with TPBi ETL conventionally used and PO-T2T ETL used in this work. The MAPbBr₃ films had PO-T2T as additive in antisolvent in both cases.

Table S4: A literature survey of device performance of 3D Bulk MAPbBr₃ Perovskite-based LEDs.

Serial Number	Composition of Perovskite	EQE [%]	Current Efficiency [mA/cm ²]	EL Stability	Reference Number
1.	MAPbBr ₃ [3D, Bulk]	8.53	42.9	-	[Ref] ¹
2.	MAPbBr ₃ [3D, Bulk]	2.96	14.31	-	[Ref] ²
3.	MAPbBr ₃ [3D, Bulk]	0.78	3.7	-	[Ref] ³
4.	MAPbBr ₃ [3D, Bulk]	-	0.05	-	[Ref] ⁴
5.	MAPbBr ₃ [3D, Bulk]	-	9.45	L ₅₀ ~ 0.31 hr @ 100 cd/m ²	[Ref] ⁵
6.	MAPbBr ₃ [3D, Bulk]	2.36	8.76	L ₅₀ ~ 120 s	[Ref] ⁶
7.	MAPbBr ₃ [3D, Bulk]	0.8	-	-	[Ref] ⁷
8.	MAPbBr ₃ [3D, Bulk]	0.07	-	L ₅₀ ~ 340 s	[Ref] ⁸
9.	MAPbBr ₃ [3D, Bulk]		13.23	L ₄₀ ~ 38 min	[Ref] ⁹
10.	MAPbBr ₃ [3D, Bulk]	1.03	-	L ₅₀ < 100 s	[Ref] ¹⁰
11.	MAPbBr ₃ [3D, Bulk]	0.71	-	L ₅₀ ~ 55 min @ 50 mA/cm ²	[Ref] ¹¹
12.	MAPbBr ₃ [3D, Bulk]	6.18	-	L ₇₀ ~ 14000 s	[Ref] ¹²
13.	MAPbBr ₃ [3D, Bulk]	0.38	1.72	-	[Ref] ¹³
14.	MAPbBr ₃ [3D, Bulk]	0.023	0.10	-	[Ref] ¹⁴
15.	MAPbBr ₃ [3D, Bulk]	-	8.22	-	[Ref] ¹⁵
16.	MAPbBr ₃ [3D, Bulk]	4.4	20.4	L ₅₀ > 3 h@	Our work

				100 cd/m ²	
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References:

- 1 H. Cho, S.-H. Jeong, M.-H. Park, Y.-H. Kim, C. Wolf, C.-L. Lee, J. H. Heo, A. Sadhanala, N. Myoung, S. Yoo, S. H. Im, R. H. Friend and T.-W. Lee, *Science (1979)*, 2015, **350**, 1222–1225.
- 2 S. Ahn, M. Park, S. Jeong, Y. Kim, J. Park, S. Kim, H. Kim, H. Cho, C. Wolf, M. Pei, H. Yang and T. Lee, *Adv Funct Mater*, , DOI:10.1002/adfm.201807535.
- 3 N. F. Jamaludin, N. Yantara, Y. F. Ng, A. Bruno, B. K. Chandran, X. Y. Chin, K. Thirumal, N. Mathews, C. Soci and S. Mhaisalkar, *J Mater Chem C Mater*, 2018, **6**, 2295–2302.
- 4 K. Lin, J. Xing, L. N. Quan, F. P. G. de Arquer, X. Gong, J. Lu, L. Xie, W. Zhao, D. Zhang, C. Yan, W. Li, X. Liu, Y. Lu, J. Kirman, E. H. Sargent, Q. Xiong and Z. Wei, *Nature*, 2018, **562**, 245–248.
- 5 A. Kanwat, E. Moyan, S. Cho and J. Jang, *ACS Appl Mater Interfaces*, 2018, **10**, 16852–16860.
- 6 Y. Shi, W. Wu, H. Dong, G. Li, K. Xi, G. Divitini, C. Ran, F. Yuan, M. Zhang, B. Jiao, X. Hou and Z. Wu, *Advanced Materials*, , DOI:10.1002/adma.201800251.
- 7 L. N. Quan, Y. Zhao, F. P. García de Arquer, R. Sabatini, G. Walters, O. Voznyy, R. Comin, Y. Li, J. Z. Fan, H. Tan, J. Pan, M. Yuan, O. M. Bakr, Z. Lu, D. H. Kim and E. H. Sargent, *Nano Lett*, 2017, **17**, 3701–3709.
- 8 C. T. Prontera, D. Taurino, A. Coriolano, A. Maggiore, M. Pugliese, R. Giannuzzi, F. Mariano, S. Carallo, A. Rizzo, G. Gigli, L. De Marco and V. Maiorano, *Nanoscale Adv*, 2023, **5**, 2508–2516.
- 9 H. Kim, J. S. Kim, J.-M. Heo, M. Pei, I.-H. Park, Z. Liu, H. J. Yun, M.-H. Park, S.-H. Jeong, Y.-H. Kim, J.-W. Park, E. Oveisi, S. Nagane, A. Sadhanala, L. Zhang, J. J. Kweon, S. K. Lee, H. Yang, H. M. Jang, R. H. Friend, K. P. Loh, M. K. Nazeeruddin, N.-G. Park and T.-W. Lee, *Nat Commun*, 2020, **11**, 3378.
- 10 J.-W. Lee, Y. J. Choi, J.-M. Yang, S. Ham, S. K. Jeon, J. Y. Lee, Y.-H. Song, E. K. Ji, D.-H. Yoon, S. Seo, H. Shin, G. S. Han, H. S. Jung, D. Kim and N.-G. Park, *ACS Nano*, 2017, **11**, 3311–3319.
- 11 J. C. Yu, D. W. Kim, D. Bin Kim, E. D. Jung, K.-S. Lee, S. Lee, D. Di Nuzzo, J.-S. Kim and M. H. Song, *Nanoscale*, 2017, **9**, 2088–2094.
- 12 S. Lee, J. H. Park, B. R. Lee, E. D. Jung, J. C. Yu, D. Di Nuzzo, R. H. Friend and M. H. Song, *J Phys Chem Lett*, 2017, **8**, 1784–1792.
- 13 S. Lee, J. H. Park, Y. S. Nam, B. R. Lee, B. Zhao, D. Di Nuzzo, E. D. Jung, H. Jeon, J.-Y. Kim, H. Y. Jeong, R. H. Friend and M. H. Song, *ACS Nano*, 2018, **12**, 3417–3423.
- 14 N. K. Kumawat, N. Jain, A. Dey, K. L. Narasimhan and D. Kabra, *Adv Funct Mater*, 2017, **27**, 1603219.

15 J. Liang, Y. Zhang, X. Guo, Z. Gan, J. Lin, Y. Fan and X. Liu, *RSC Adv*, 2016, **6**, 71070–71075.