

ELECTRONIC SUPPLEMENTARY INFORMATION

Finally, inkjet-printed metal halide perovskite LEDs – utilizing seed crystal templating of salty PEDOT:PSS

Felix Hermerschmidt,^{†a} Florian Mathies,^{†b} Vincent R. F. Schröder,^a Carolin Rehermann,^b Nicolas Zorn Morales,^a Eva L. Unger^{b,c} and Emil J. W. List-Kratochvil^{*a,d}

^aHumboldt-Universität zu Berlin, Institut für Physik, Institut für Chemie, IRIS Adlershof, Brook-Taylor-Straße 6, 12489 Berlin, Germany. Email: emil.list-kratochvil@hu-berlin.de

^bYoung Investigator Group Hybrid Materials Formation and Scaling, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Kekuléstraße 5, 12489 Berlin, Germany.

^cChemical Physics and NanoLund, Lund University, PO Box 124, 22100 Lund, Sweden.

^dHelmholtz-Zentrum, Berlin für Materialien und Energie GmbH, Brook-Taylor-Straße 6, 12489 Berlin, Germany.

ELECTRONIC SUPPLEMENTARY INFORMATION

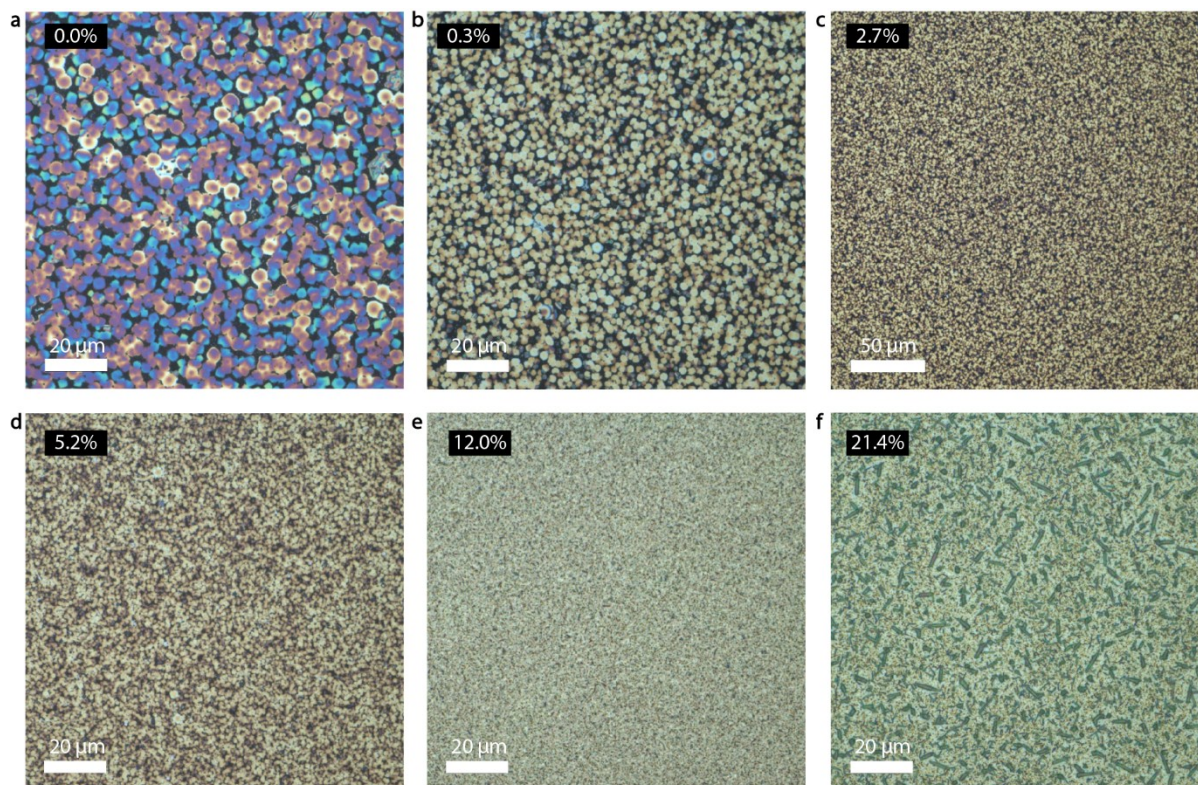


Figure S1: The optical microscopy images of MAPbBr₃:PEG composite blended with **a** 0.0%, **b** 0.3%, **c** 2.7%, **d** 5.2%, **e** 12.0% and **f** 21.4% PEG show optimum homogeneous morphology at 12.0% PEG addition.

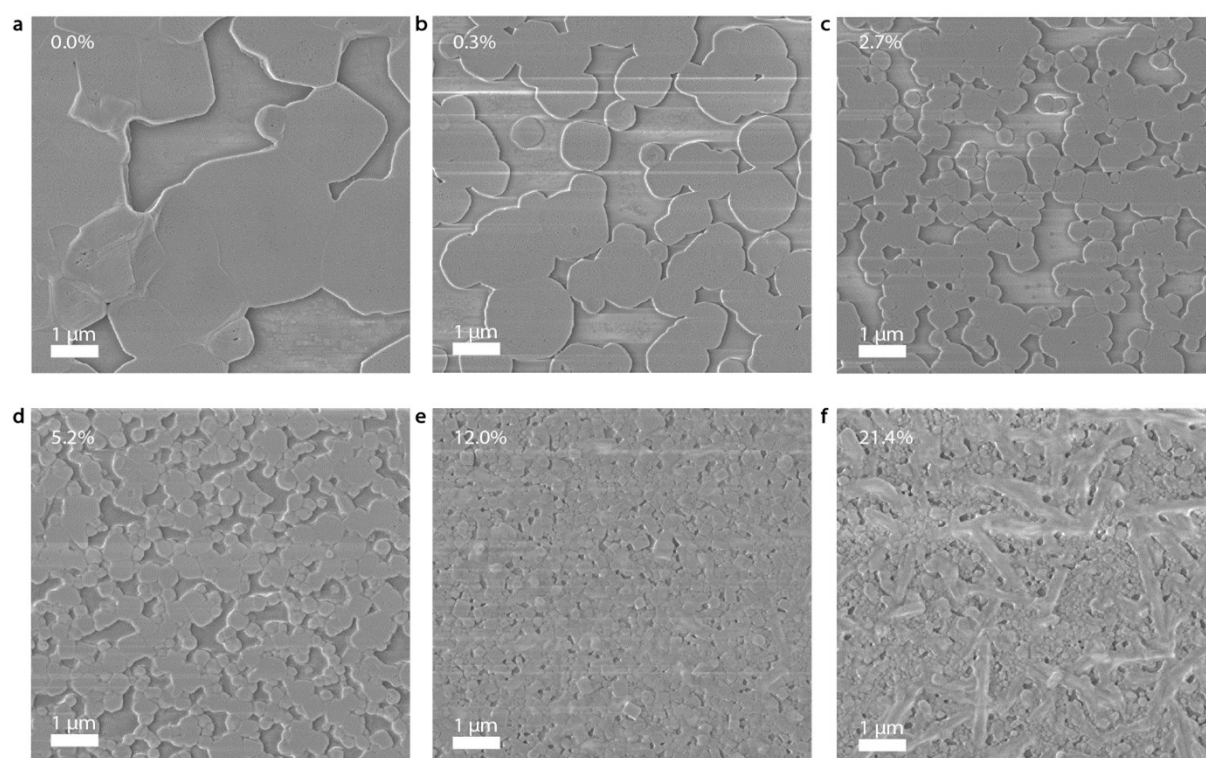


Figure S2: The scanning electron microscopy images of MAPbBr₃:PEG composite blended with **a** 0.0%, **b** 0.3%, **c** 2.7%, **d** 5.2%, **e** 12.0% and **f** 21.4% PEG confirm the optimum morphology for 12.0% PEG.

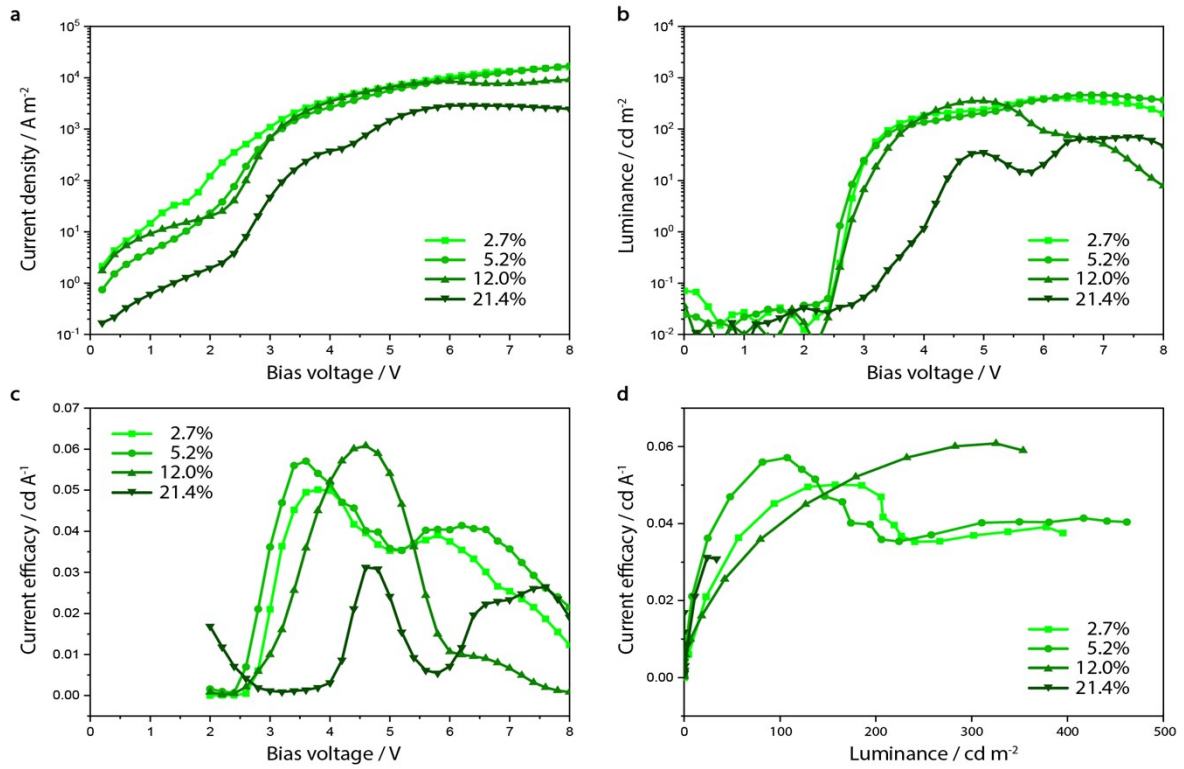


Figure S3: Also the device performance parameters for PeLEDs incorporating MAPbBr₃ perovskite active layer blended with 2.7%, 5.2%, 12.0% and 21.4% PEG, respectively, show optimum performance with 12% PEG content.

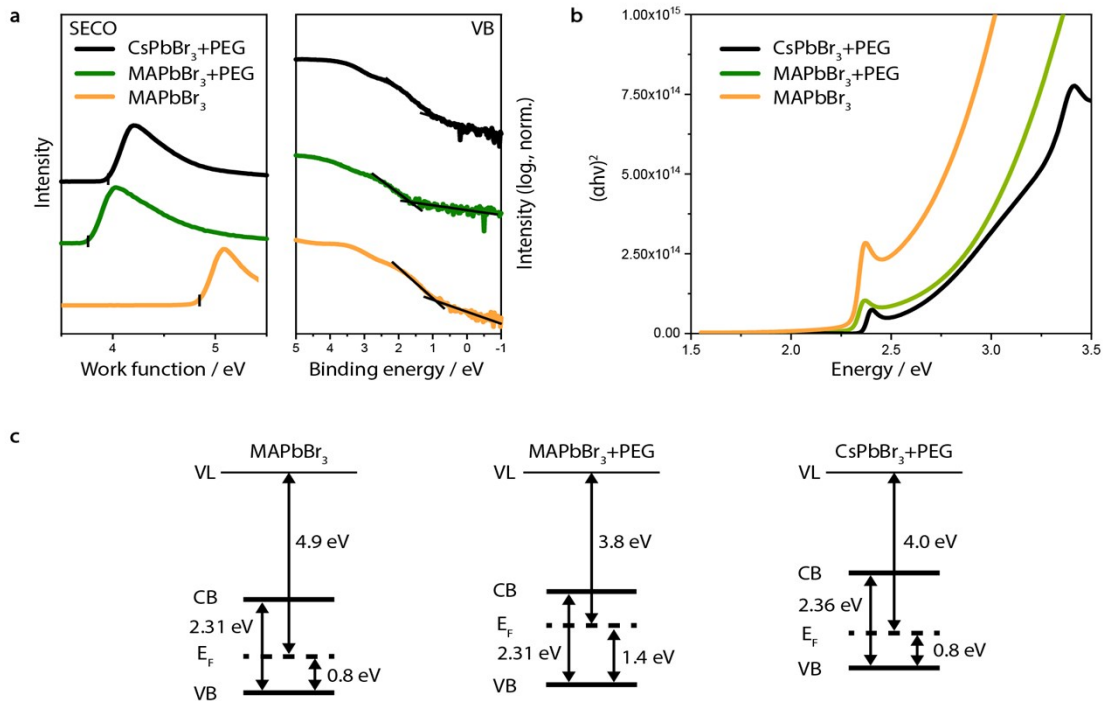


Figure S4: **a** The secondary electron cut-off and valence band on-set, **b** Tauc plots based on UV-vis absorption spectroscopy and **c** schematic of energy levels for MAPbBr₃, MAPbBr₃:PEG and CsPbBr₃:PEG resulting from the experimental data.

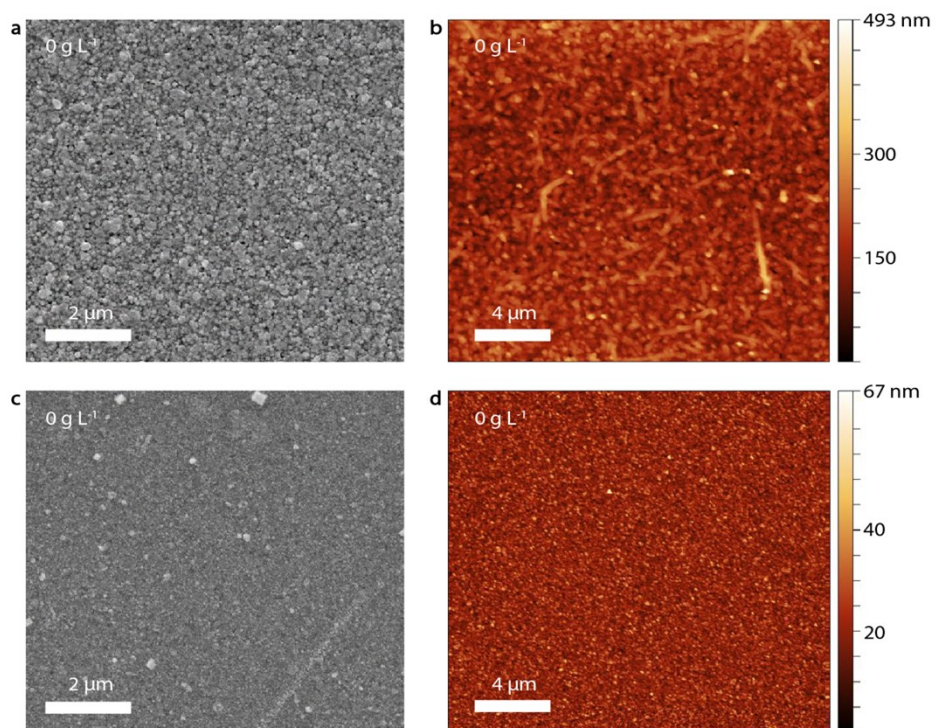


Figure S5: The SEM and SFM images of spin-coated (a, b) and inkjet-printed (c, d) MAPbBr₃:PEG composite films on pristine PEDOT:PSS (without KCl) confirm the pinhole-free active layer morphology achievable from the two processing methods.

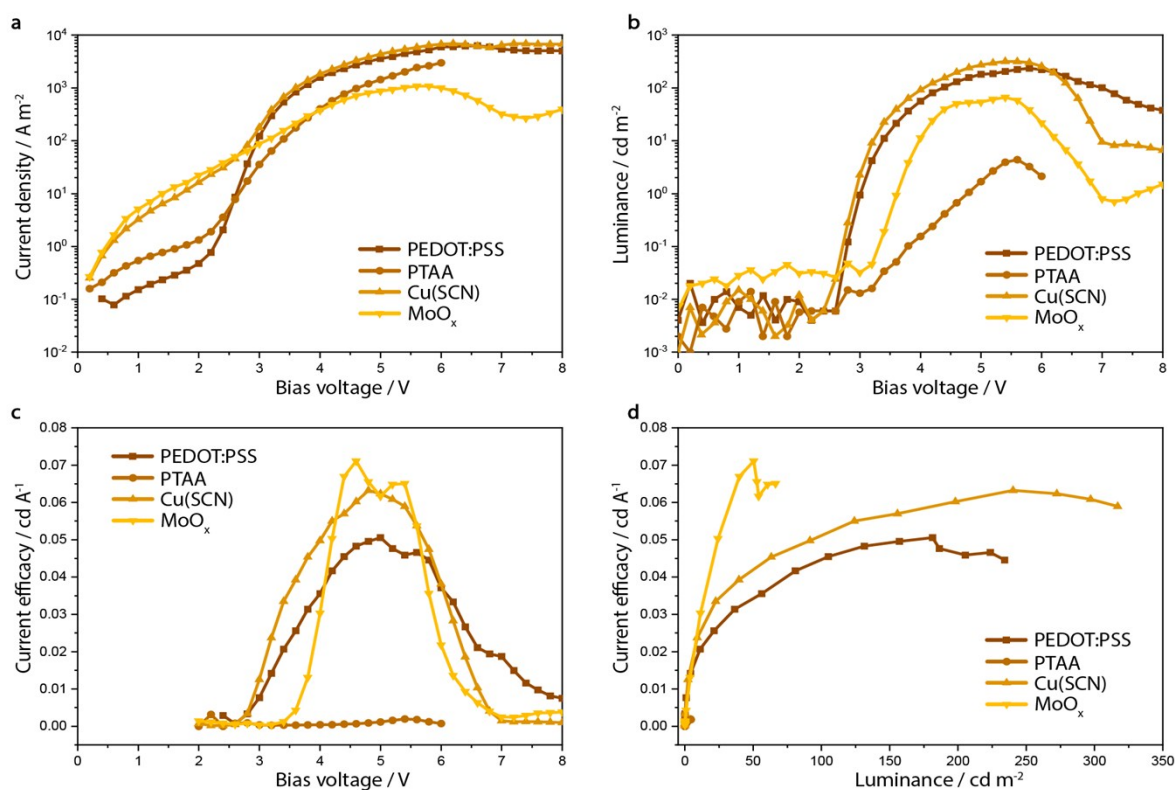


Figure S6: MAPbBr₃:PEG-based devices were produced on pristine PEDOT:PSS or molybdenum oxide (MoO_x) as HIL as well as PEDOT:PSS plus either poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA) and cuprous thiocyanate (CuSCN). Stark contrasts are seen, with only the processing on pristine PEDOT:PSS yielding devices that show the best combination of low leakage current and reasonable device luminance. Devices with MAPbBr₃:PEG active layer processed on pristine PTAA and CuSCN failed to function due to low wetting on PTAA and washing away of CuSCN during processing. PeLEDs including MoO_x layer showed degradation during operation due to interactions between MoO_x and the MAPbBr₃:PEG composite layer.

ELECTRONIC SUPPLEMENTARY INFORMATION

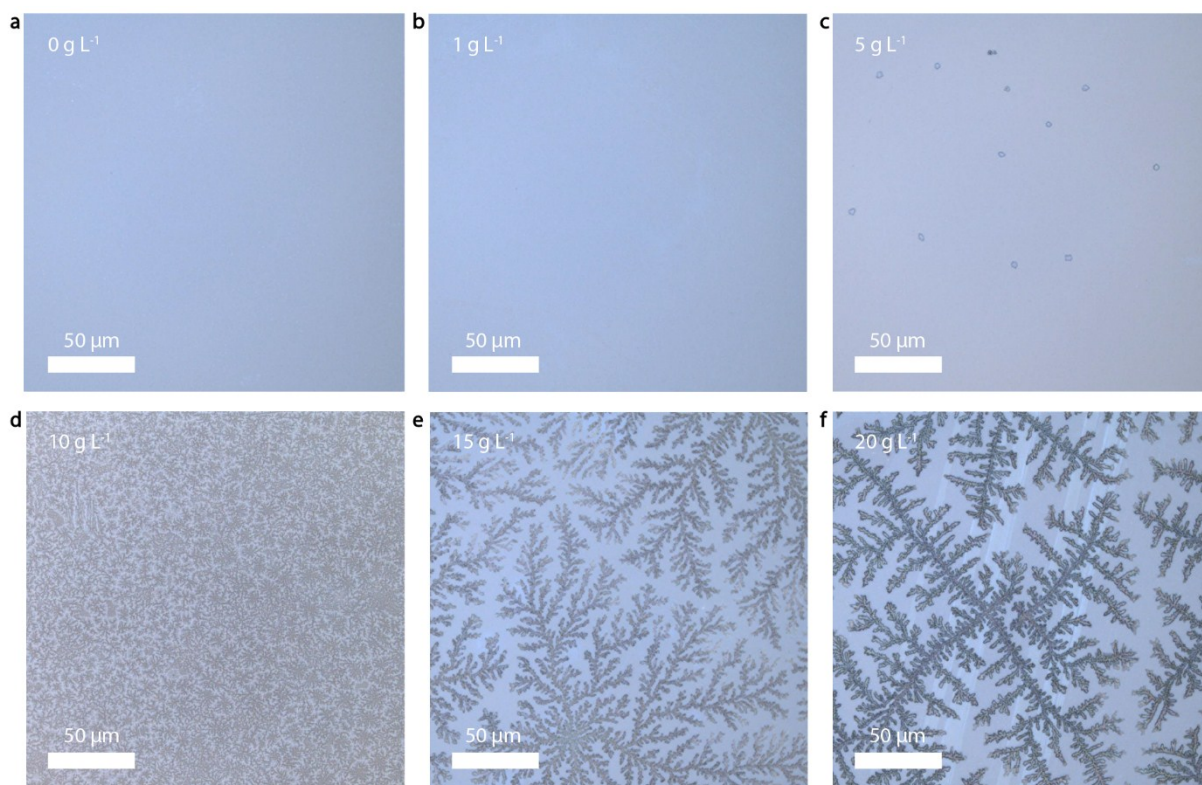


Figure S7: The series of optical microscope images of the PEDOT:PSS layer with increasing amounts of KCl shows the increasingly large dendritic structures of the KCl.

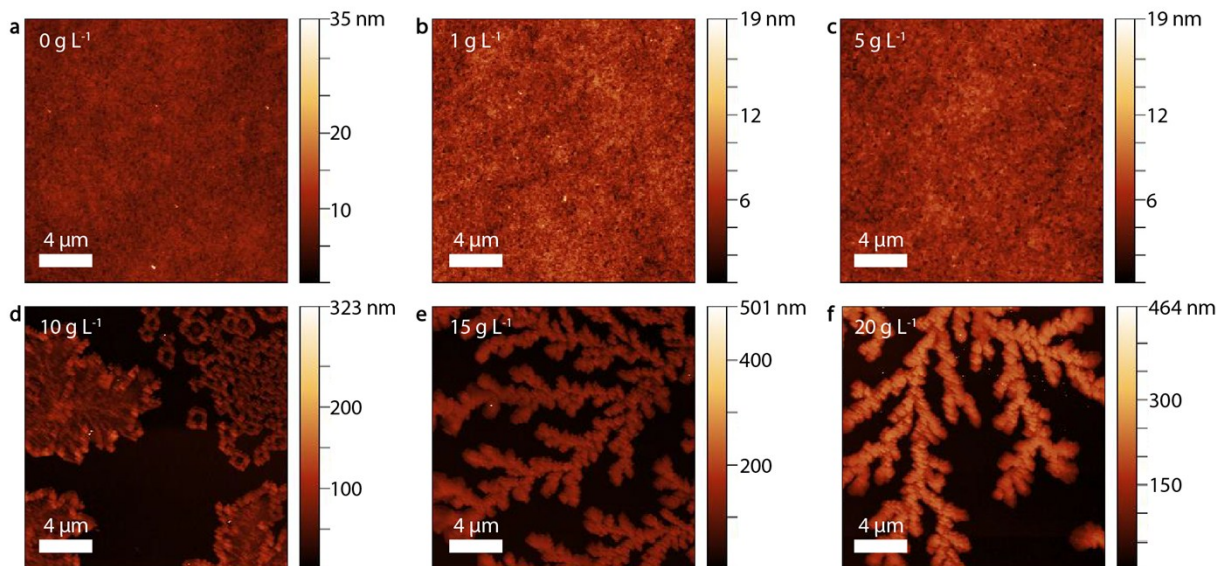


Figure S8: The dendritic structures are confirmed in SFM images of PEDOT:PSS incorporating increasing amounts of KCl.

ELECTRONIC SUPPLEMENTARY INFORMATION

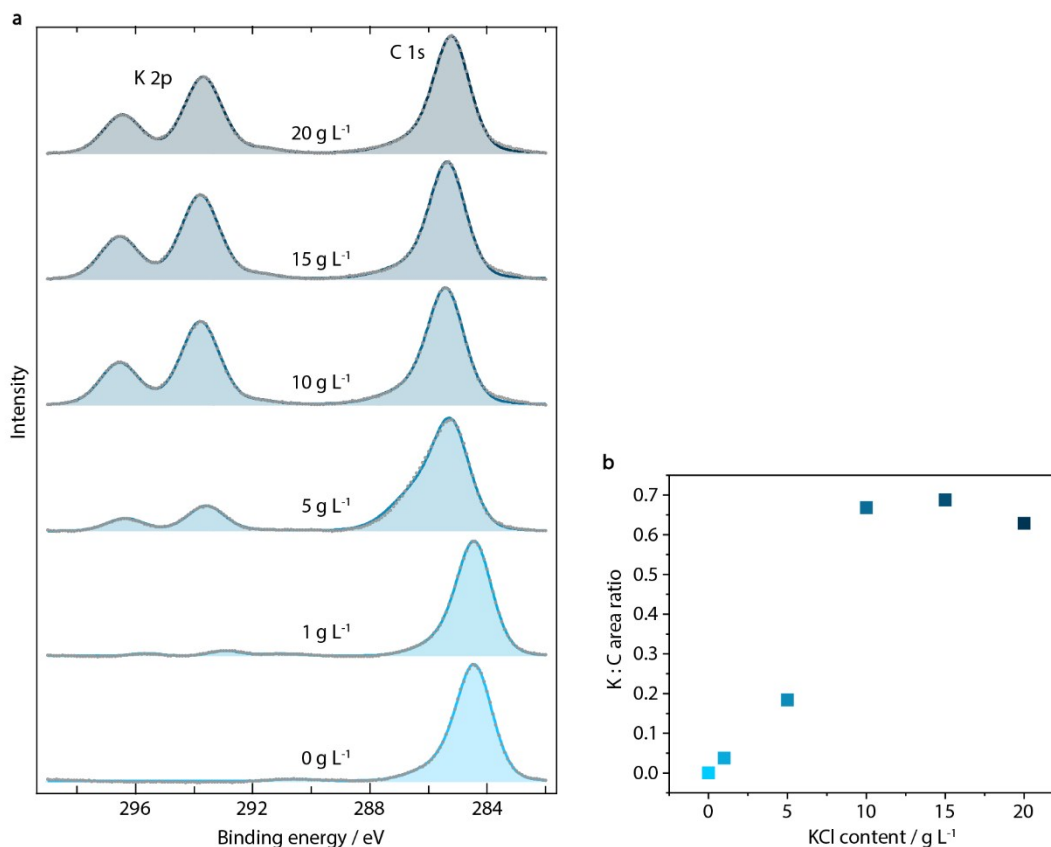


Figure S9: **a** XPS spectra show the increasing K 2p peak (doublet at 295 eV, corresponding to the added KCl) and the C 1s peak (285 eV, corresponding to the PEDOT:PSS surface), **b** the K:C area ratios show the saturation at high KCl content.

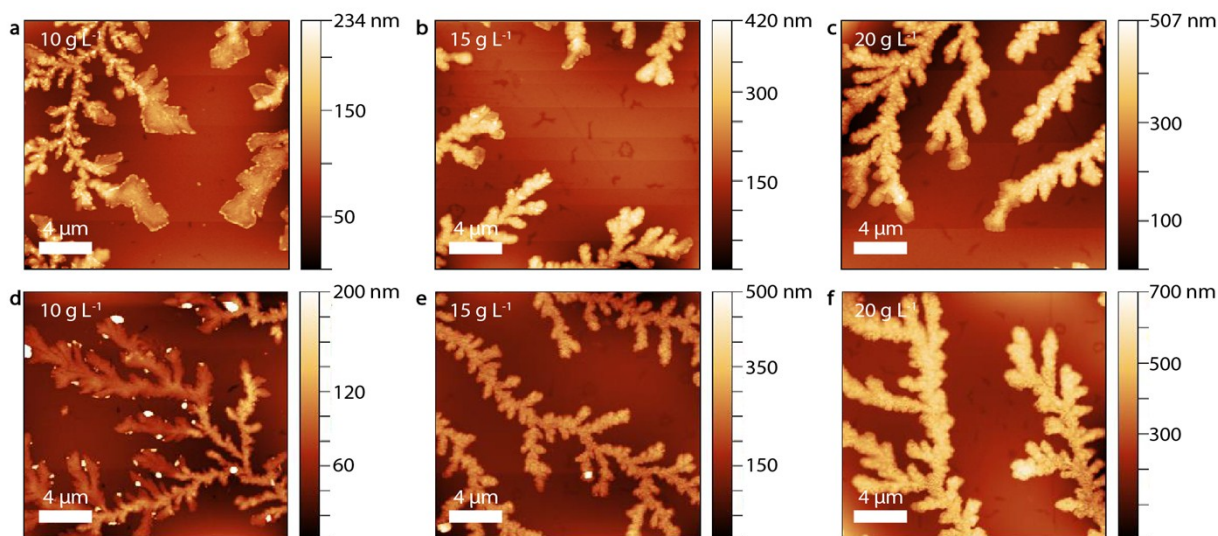


Figure S10: The SFM images of PEDOT:PSS layers containing KCl before and after washing with the solvent used for perovskite deposition show no noticeable change in the dendritic structures visible.

Table S1: This is confirmed in the feature heights measured before and after washing.

	0 g L ⁻¹	1 g L ⁻¹	5 g L ⁻¹	10 g L ⁻¹	15 g L ⁻¹	20 g L ⁻¹
pre washing	-	-	-	~100 nm	~250 nm	~330 nm
post washing	-	-	-	~50 nm	~210 nm	~300 nm

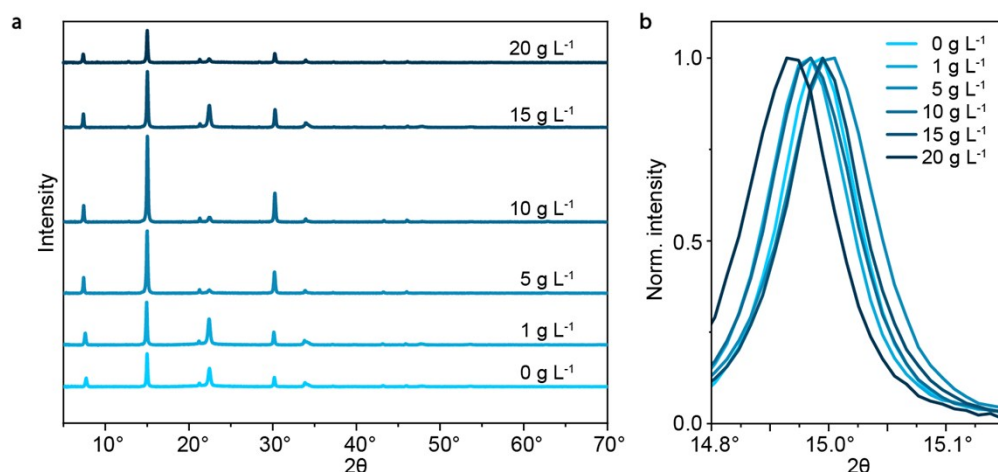


Figure S11: a XRD diffraction patterns of the full series of MAPbBr₃:PEG-composite spin-coated on PEDOT:PSS/KCl with increasing KCl concentration indicate no apparent shift in the diffraction peaks. The diffraction peaks at 7.5° and 22.5° most likely result from the PEG. b No trend in shift of the magnified predominant [100] MAPbBr₃ diffraction peak is visible as a function of KCl content.

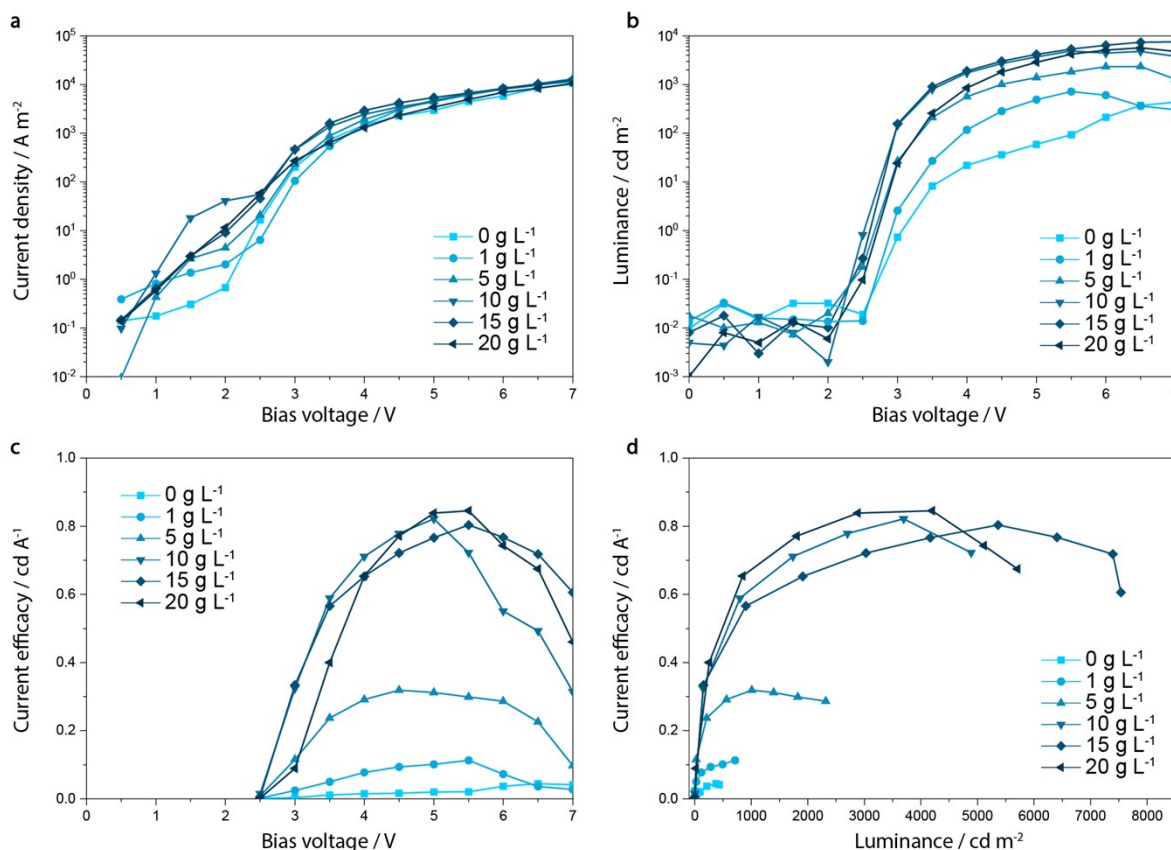


Figure S12: The current density / voltage / luminance curves of spin coated devices incorporating varying amounts of KCl within the PEDOT:PSS HIL show the rising luminance and current efficacy (cd A⁻¹) as a function of incorporated KCl. While the absolute maximum efficacy is achieved by devices incorporating 20 g L⁻¹ KCl, this maximum is only marginally greater than that obtained from devices incorporating 15 g L⁻¹ KCl, but at the expense of a much lower luminance.

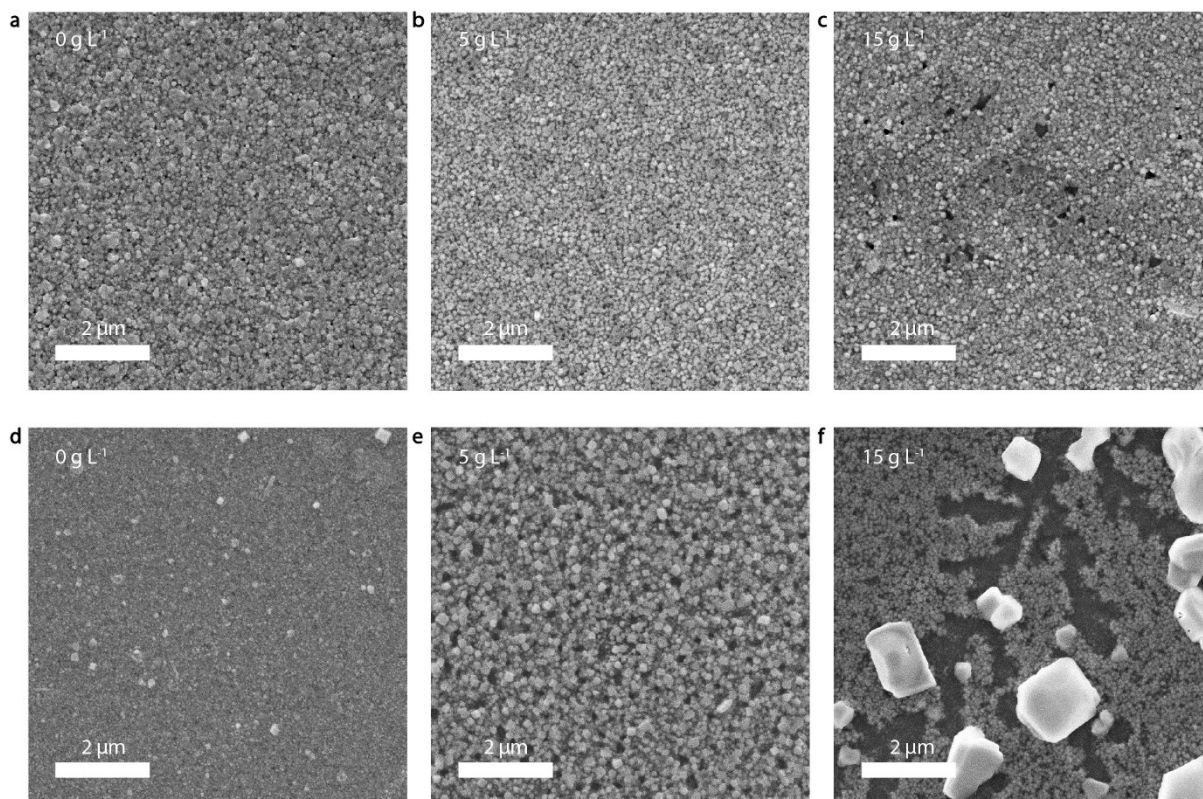


Figure S13: The SEM images of MAPbBr₃:PEG-composite spin-coated (top row **a-c**) and inkjet-printed (bottom row **d-f**) on PEDOT:PSS/KCl show different effects when increasing KCl concentration from 0 g L⁻¹ to 5 g L⁻¹ to 15 g L⁻¹. While similar morphology is seen for spin-coated and inkjet-printed layers at KCl concentrations of 15 g L⁻¹ (**c**) and 5 g L⁻¹ (**e**), respectively, a substantial difference in morphology is observed for layers inkjet-printed on PEDOT:PSS containing 15 g L⁻¹. This leads to the two different optimums achieved in PeLED device performance described in the main text.

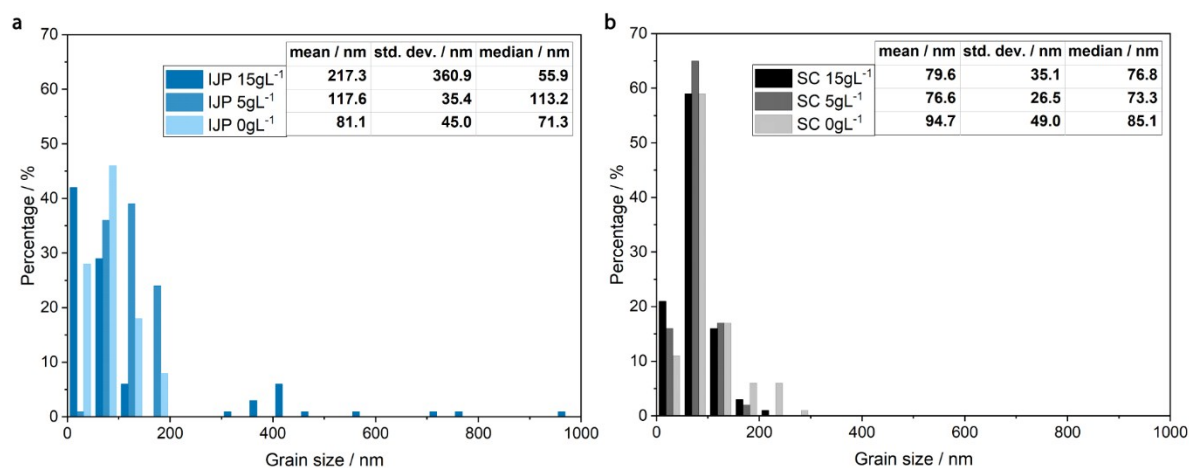


Figure S14: Upon increasing the KCl content, smaller average grains are seen for spin-coated devices. Additionally, the standard deviation decreases, signifying a more homogeneous film. For inkjet-printed films, the mean grain size increases with increasing KCl content. But while at 5 g L⁻¹ the standard deviation is also lower, indicating a more homogenous film, for the high KCl content of 15 g L⁻¹ these values have to be taken with care, as some micrometer large grains distort the calculations.