Supplementary Information

Fabrication of Pyramidal (111) $MAPbBr_3$ Film with Low Surface Defect Density Using Homogeneous Quantum-Dot Seeds

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Materials and methods

Synthesis of quantum dots (QDs): $MAPbBr_3$ QDs were synthesized as described by Feng Zhang et al. [*ACS Nano* **2015**, 9, 4533], with some modifications. $MAPbBr_3$ precursor solutions were prepared by dissolving MABr (45.79 mg, Ossila, >99.5%), $PbBr_2$ (146.80 mg, Sigma-Aldrich, ≥98%), oleic acid (1 mL, Sigma Aldrich, 90%), and oleylamine (0.1 mL, Sigma Aldrich, 70%) in dimethylformamide (DMF, 10 mL, Sigma-Aldrich, 99.8%) solution. The precursor solution was then added dropwise to toluene, with stirring at 500 rpm. To obtain pure QDs, the $MAPbBr_3$ QDs and nanocrystals in toluene were centrifuged for 10 min at 10 krpm.

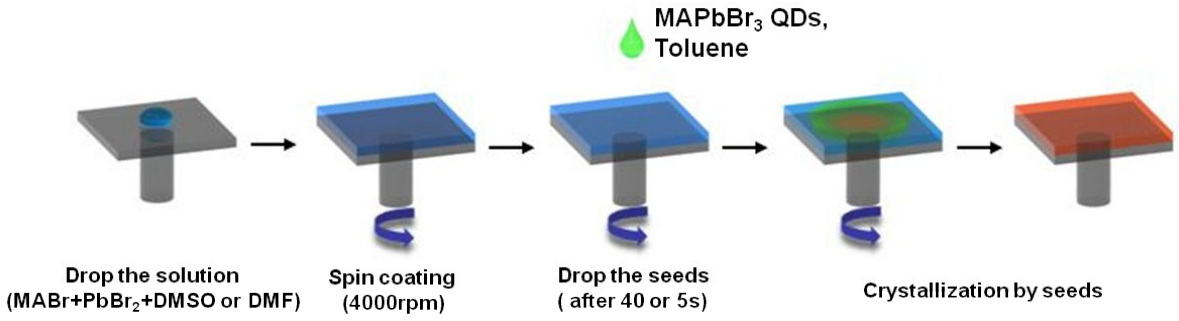
Preparation of $MAPbBr_3$ solution for films: MABr and $PbBr_2$ were dissolved in dimethyl sulfoxide (DMSO, Sigma-Aldrich, ≥99.9%) and DMF at a concentration of 40 wt.%.

Film and photodetector fabrication: Indium tin oxide, quartz, and silicon oxide substrates

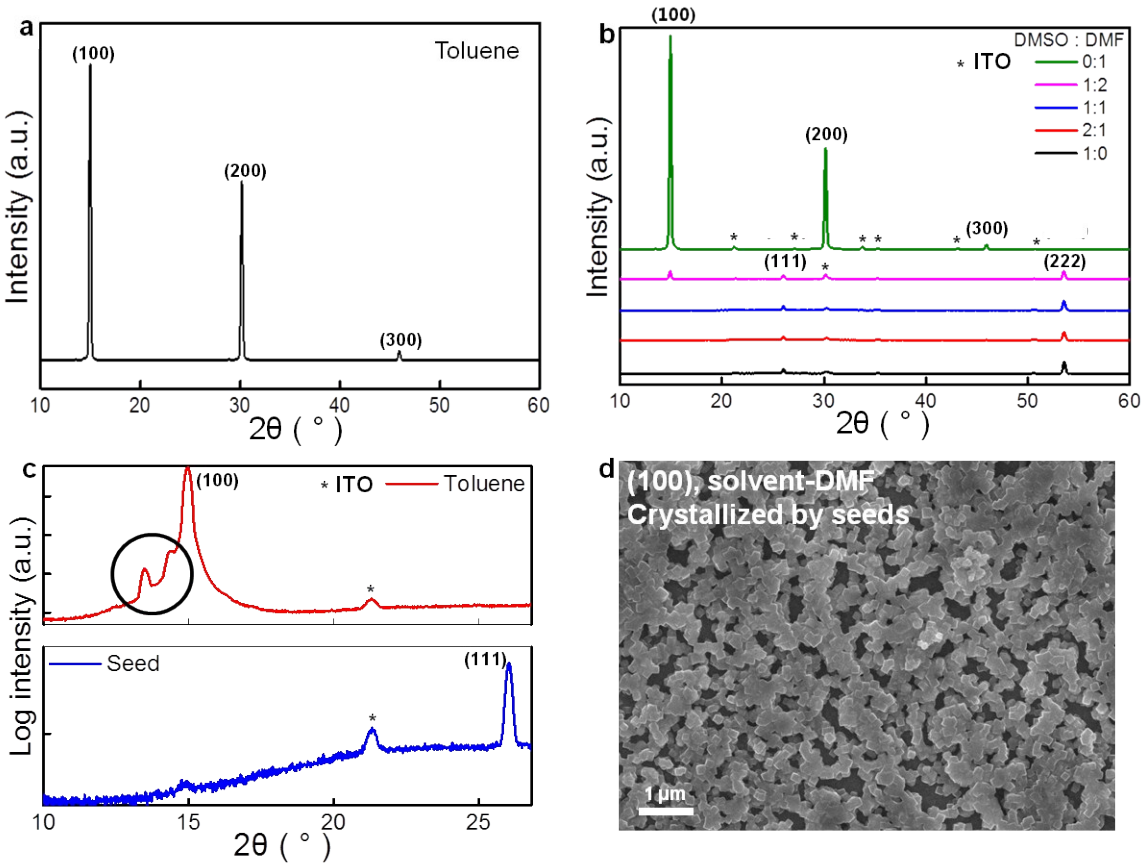
were cleaned sequentially in acetone, deionized water, and isopropyl alcohol for 10 min via sonication. The cleaned substrates were ultraviolet (UV) -ozone-treated for 5 min to make them hydrophilic. The $MAPbBr_3$ solution was spin-coated onto the substrates for 60 s at 4000 rpm in a glovebox. The seeds were dropped at 40 and 5 s for the DMSO and DMF solutions, respectively. To fabricate the photodetector, Cr (5 nm)/Au (50 nm) was deposited on a cleaned silicon oxide substrate using an electron-beam evaporator system, for forming the electrode.

Characterization: All absorption measurements were performed using a Jasco V-670 spectrometer. The UV–visible bandwidth and scan speed were 2.0 nm and 400 nm/min, respectively. Field-emission scanning electron microscopy images were obtained using a JEOL JSM-7600F. To prevent decomposition due to the electron beam, we used a C tape around the measurement region and an accelerating voltage of 5 kV. Transmission electron microscopy (TEM) images were obtained using a JEM-ARM200CF. X-ray diffraction (XRD) measurements were performed using a SmartLab (Rigaku) instrument with a Cu-Kα radiation source. The samples were scanned from 10° to 60° at a scan rate of 4°/min with a step size of 0.02°. Confocal scanning photoluminescence (PL) microscopy (CSPLM) images, the time-resolved PL (TRPL), and the excitation-power-dependent PL were measured using an NT-MDT NTEGRA instrument. An in-plane spatial resolution of ~380 nm was indicated by the numerical aperture (0.7) of the objective lens and the wavelength (405 nm) of the solid-state laser. The excitation power for the CSPLM was 0.077 μW cm^{-2}. To measure the TRPL, this machine included a time-correlated single-photon counting and high-speed photo-multiplier tube detector (PMC-100, Photonic Solution). The excitation source of the TRPL used a 405-nm pulse laser with a repetition rate of 50 MHz and excitation power of 0.025 μW cm^{-2}. The excitation source for the excitation-power-dependent PL was a 405-nm pulsed laser with a

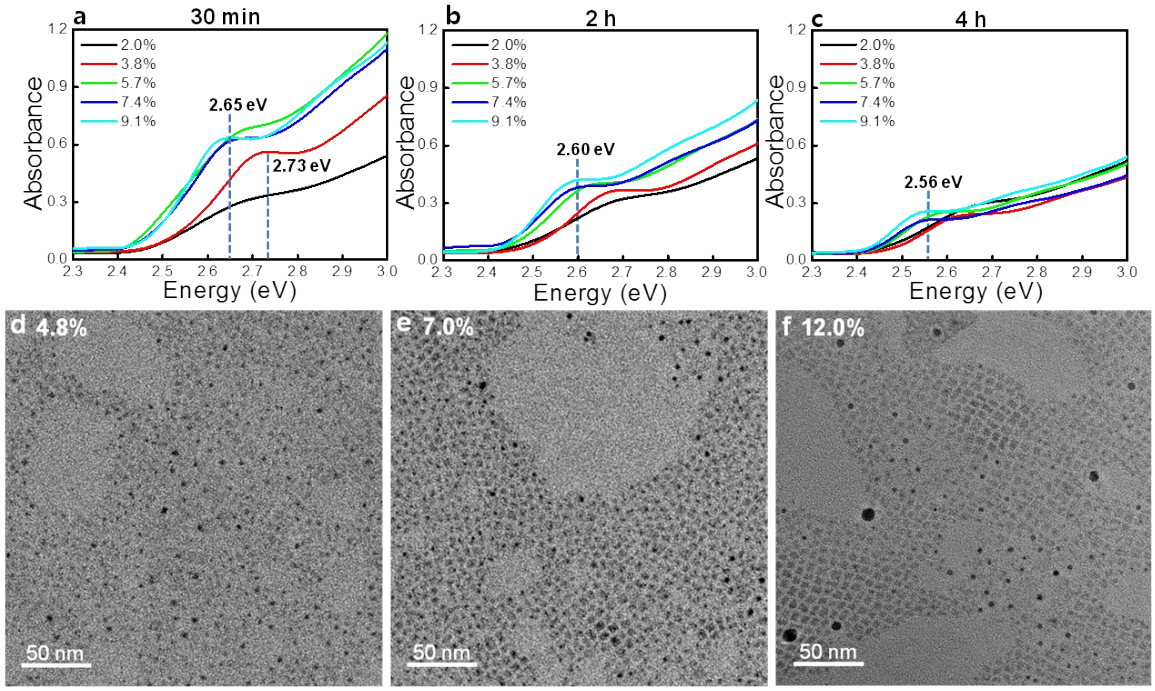
repetition rate of 20 MHz. The power-density range was 326–13038 mW/cm^2. The electrical properties were measured using a customized four-probe vacuum station system (2400, Keithley) at 10^{-6} Torr. For the time-dependent on/off photocurrent measurements, a homemade laser shutter system was used. The excitation source for the photocurrent measurements was a 458 nm solid-state laser. To examine the surface topography, atomic force microscopy images were obtained using an E-Sweep W/NanoNaviStation microscope.



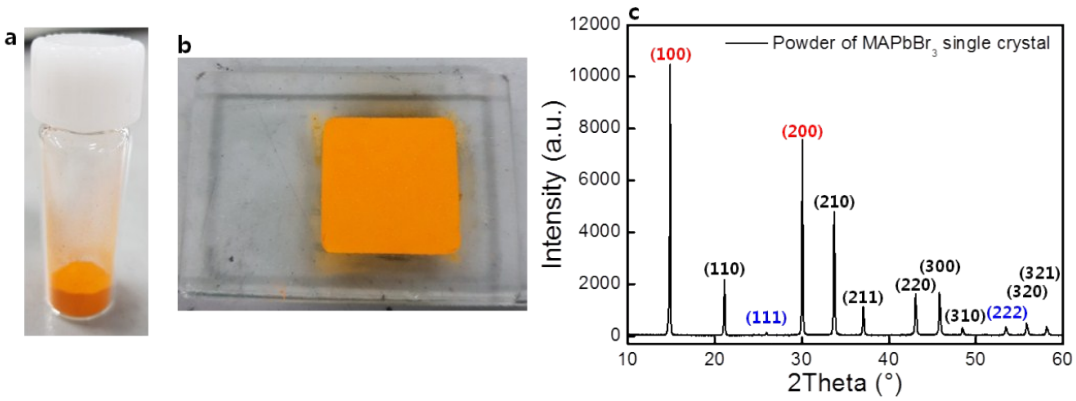
Supplementary Figure S1. Crystallization process with perovskite seeds.



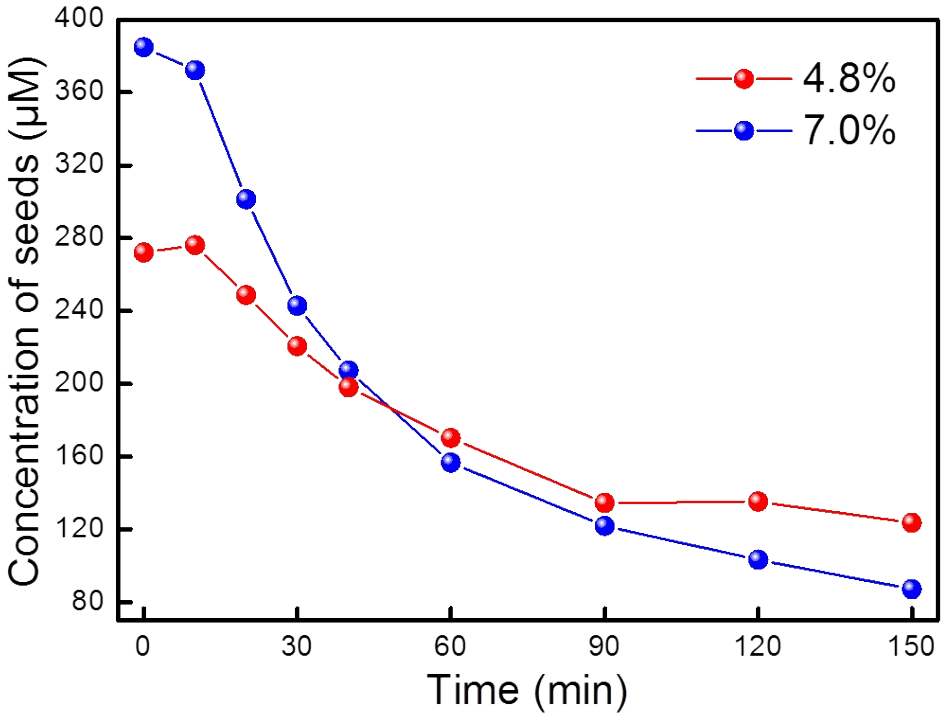
Supplementary Figure S2. XRD pattern of a) the film formed by toluene and b) the films formed by seeds with different ratios of DMSO to DMF in the perovskite solution. c) Log-scale XRD pattern of the (100) plane for the film formed by toluene and the (111) plane for the film formed by seeds. d) SEM image of the film formed by seeds. The solvent of the perovskite solution was DMF. Owing to the rapid evaporation of DMF, it was difficult to produce full-coverage films.



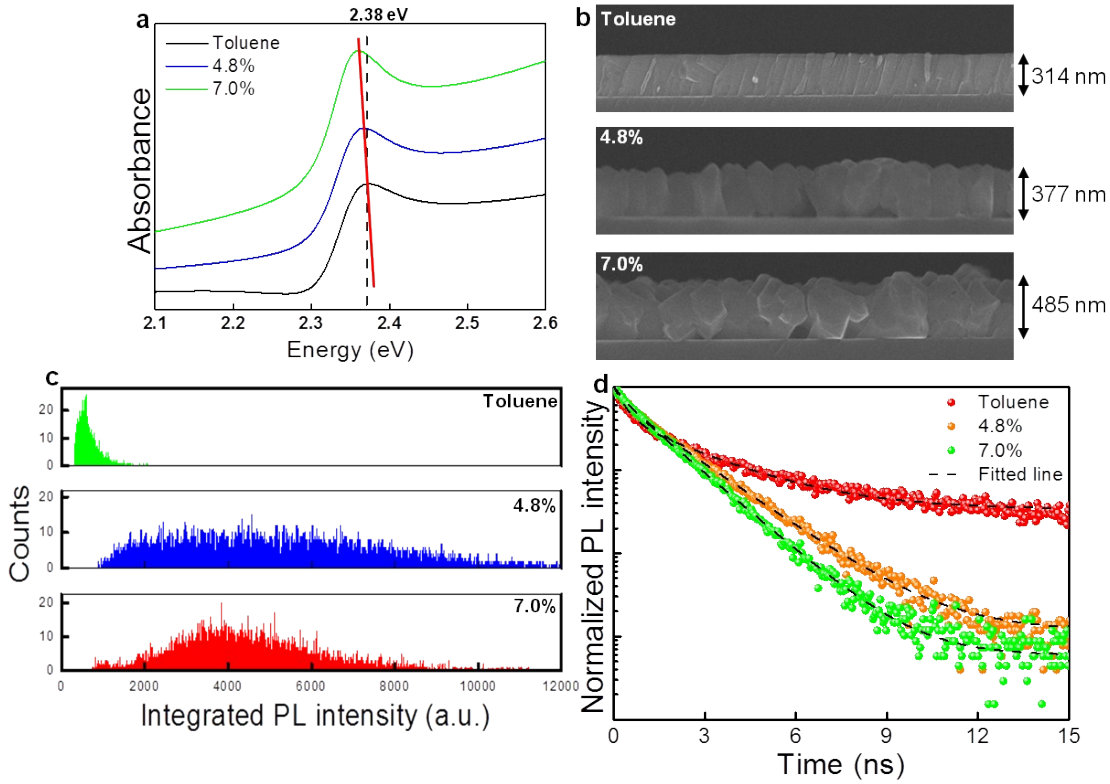
Supplementary Figure S3. UV–visible absorption spectra of the $MAPbBr_3$ QDs with the increasing concentration of the precursor solution after a) 30 min, b) 2 h, and c) 4 h. TEM image of the $MAPbBr_3$ QDs with a concentration of d) 4.8%, e) 7.0% and f) 12.0%.



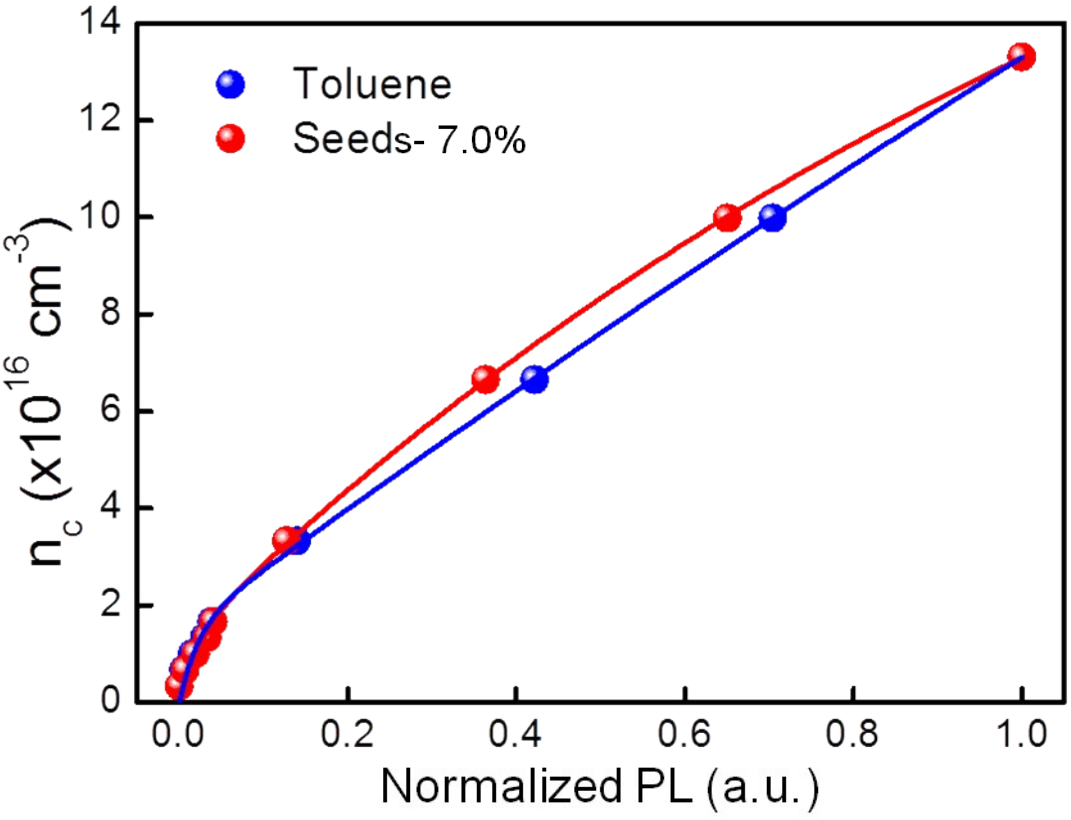
Supplementary Figure S4. Photographs of a) the powder of the $MAPbBr_3$ single crystal and b) the uniform setting of the powder. c) XRD pattern of the powder of the $MAPbBr_3$ single crystal for the calculation of the texture coefficient.



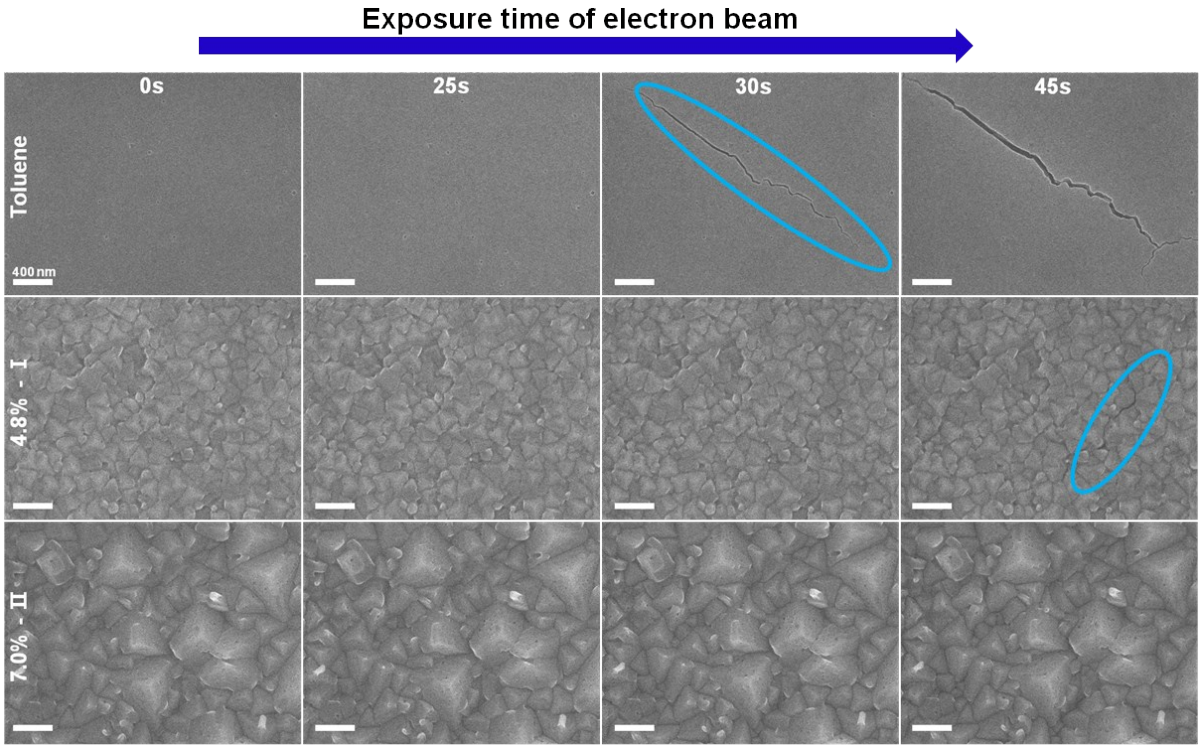
Supplementary Figure S5. Calculated concentration of seeds with respect to the aggregation time (4.8% and 7.0% seeds). It was calculated using the reported molar extinction coefficient [*ACS Energy Letters* **2017**, 2, 88] and lattice constant [*Nano Letters* **2015**, 15, 5191] of $MAPbBr_3$ QDs.



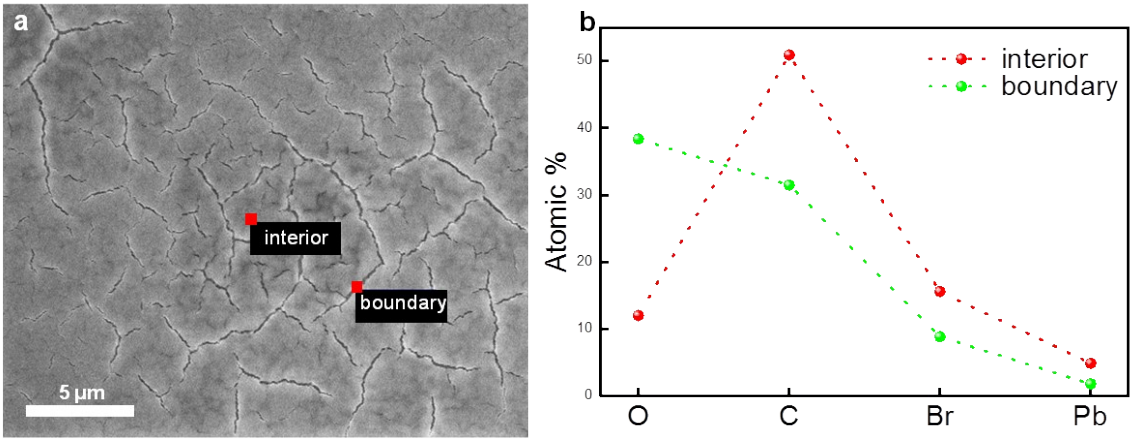
Supplementary Figure S6. a) Absorption spectra of $MAPbBr_3$ films formed by toluene and with seed concentrations of 4.8% and 7.0%. The dashed line indicates the exciton peak position for the film formed by toluene. The red line corresponds to the exciton peak position. b) Cross-sectional SEM images, c) integrated PL intensity histograms and d) TRPL spectra of the $MAPbBr_3$ films formed by toluene and with seed concentrations of 4.8% and 7.0%.



Supplementary Figure S7. Initial generated charge carrier density with respect to the normalized PL intensity measured via excitation-power-dependent PL.



Supplementary Figure S8. Time-dependent SEM images of MAPbBr3 films formed by toluene and with seed concentrations of 4.8% and 7.0% under irradiation by an electron beam.



Supplementary Figure S9. a) SEM image of the film formed by toluene exposed to an electron beam for 2 min. b) Atomic percentage of O, C, Br, Pb in the interior and at the boundary (crack).

	τ_1 (ns)	Fraction f_1 (%)	τ_2 (ns)	Fraction f_2 (%)	$\tau_{average}$ (ns)
Toluene	0.36	59	2.54	41	1.25
4.8% - I	0.54	38	1.82	62	1.33
7.0% - II	0.55	25	1.46	75	1.23

Supplementary Table S1. Bi-exponential decay fitted values and the average lifetime for Figure S5d.

	O	C	Pb	Br
Interior	12.00	50.90	4.84	15.56
Boundary	38.33	31.48	1.76	8.84

Supplementary Table S2. Atomic percentages for Figure S7b.