

Enhanced Crystallization in CsPbBr₃ All-Inorganic Perovskite via Advanced Nucleation Method

Yingke Ren,^a Delong Li,^{a,b} Jing Chen,^a Xinge Guo,^a Chao He,^a Zhaoqian Li,^{*c} and
Xingtao An^{*a}

^aHebei Provincial Key Laboratory of Photoelectric Control on Surface and Interface, College of
Science, Hebei University of Science and Technology, Shijiazhuang, 050018, China

^bYantai Yuhuangding Hospital, Yantai, 264000, China

^cKey Lab of Photovoltaic and Energy Conservation Materials, Institute of Solid State Physics,
HFIPS, Chinese Academy of Sciences, Hefei 230031, China

*Corresponding author

E-mail: anxt2005@163.com (X An)

E-mail: zqli@rntek.cas.cn (Z Li)

EXPERIMENTAL SECTION

Film Fabrication (TiO₂)

Fluorine-doped tin oxide (FTO, Pilkington, TEC15) glass was etched with HCl aqueous and Zn powder. The obtained glass was then cleaned with detergent, distilled water and ethanol, respectively. The blocking TiO₂ layers (bl-TiO₂) were deposited on the as-prepared FTO through spray pyrolysis method followed by calcining at 510 °C for 30 min. The mesoporous TiO₂ (mp-TiO₂) films were deposited on the above bl-TiO₂ layer by means of spin-coating the TiO₂ paste (Dyesol 30NR-T). The obtained layers were following by heating at 510 °C for 20 min.

Wide-bandgap Solar Cell Fabrications (CsPbBr₃)

An N,N-dimethylformamide (DMF) solution of 1.0 M PbBr₂ contain 0.1M CsBr was spin-coated onto the TiO₂ layer at 2000 rpm for 30 s. Then 90 mL of 0.07 M CsBr methanol solution was coated onto prepared substrate at 2000 rpm for 30 s. The CsBr was repeatedly spin-coated four times to form a CsPbBr₃ layer. Finally, a commercial carbon paste (Guangzhou Seaside Technology Co., Ltd) was used as carbon electrode, which was composed of carbon black and graphite. The carbon paste was painted on perovskite layer under ambient condition followed by heating at 100 °C for 30 min. For semitransparent CsPbBr₃ solar cells, all the processes were consistent with mentioned above until MoO_x/ITO electrode preparation. The process of semi-transparent MoO_x/ITO electrode was learned from previous work.[8, 20]

Low-bandgap Solar Cell Fabrications (FAPbI₃)

The PbI₂ solution (600 mg of PbI₂ and 93 μL of DMSO in 1 mL of DMF) was spin-coated on the TiO₂/ITO substrate. The obtained films were annealed at 70 °C for 10 min. Then 100 uL of the FAI solutions (80 mg of FAI and 10 mg of MAcl in 1 mL of isopropanol) were immediately spin-coated on the PbI₂ films at 5000 rpm for 20s. The intermediate films were immediately annealed at 150 °C for 20 minutes to form various perovskite films. The spiro-OMeTAD solution (25 μL), which consists 73 mg of spiro-OMeTAD, 28 μL of 4-tert-butyl pyridine and 17.5 μL of lithium bis (trifluoromethanesulfonyl) imide (Li-TFSI) solution (520 mg of Li-TSFI in 1 mL of acetonitrile) in 1 mL of CBZ, was spin-coated on the perovskite film at 3000 rpm for 20 s. Finally, Au electrode with a thickness of 60 nm was deposited by using thermal

evaporation under vacuum at a constant evaporation rate of 0.6 nm s^{-1} .

Characterization

Field emission scanning electron microscope (SEM) was used to study the top-morphology of the prepared film (FEI Sirion 200, Netherland). The crystal phase was obtained with X-ray diffraction (XRD) using Cu $K\alpha$ beam (X'Pert Pro, Netherland). The current-voltage curve was measured under one sun illumination (AM 1.5G) with a solar simulator (94043A, USA) equipped with Keithley 2400 source meter. Ultraviolet-visible (UV-vis) absorption spectroscopy was measured using the spectrophotometer (SOLID3700, Shimadzu Co. Ltd, Japan). Incident photon-to-electron conversion efficiency (IPCE) spectra were measured using a 300 W xenon lamp with a spectral resolution of 5 nm equipped with order sorting filters (Newport/Oriel).

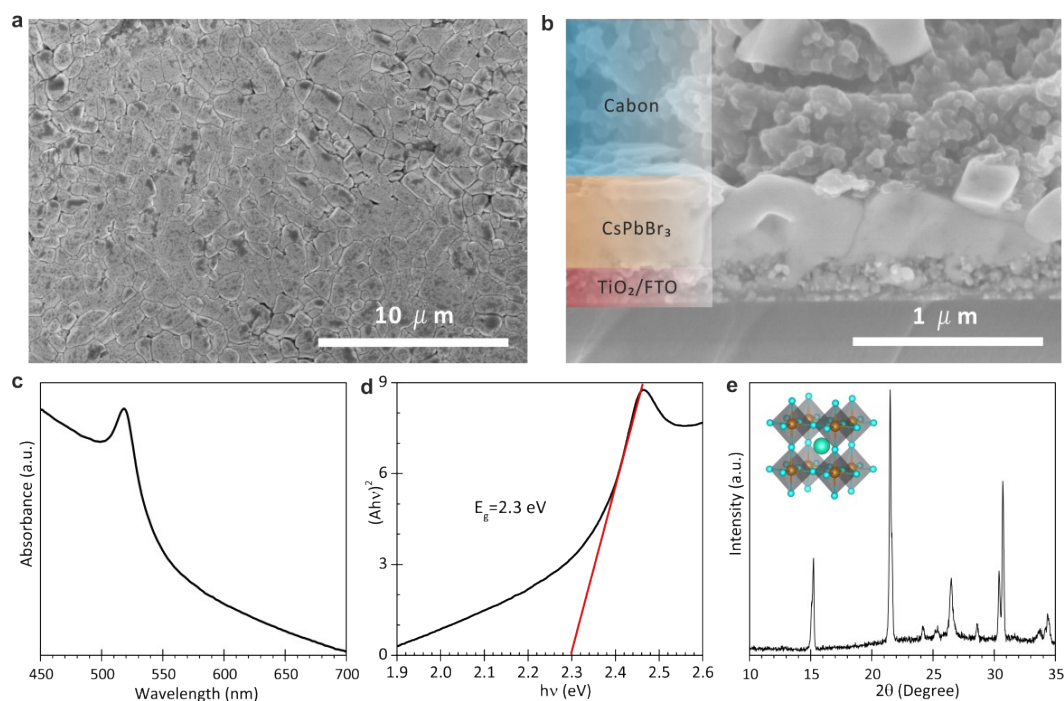


Figure S1 (a) Top-view SEM images of prepared CsPbBr₃ layer. (b) Cross-sectional SEM images of the prepared device based on FTO/TiO₂/CsPbBr₃/carbon structure. (c)

UV-vis spectra of CsPbBr₃ layer prepared on the substrates. (d) The $(Ah\nu)^2$ vs. energy $(h\nu)$ curve of the CsPbBr₃ layer. The optical band gap of CsPbBr₃ is about 2.3 eV. (e) X-ray diffraction (XRD) of prepared CsPbBr₃ film.

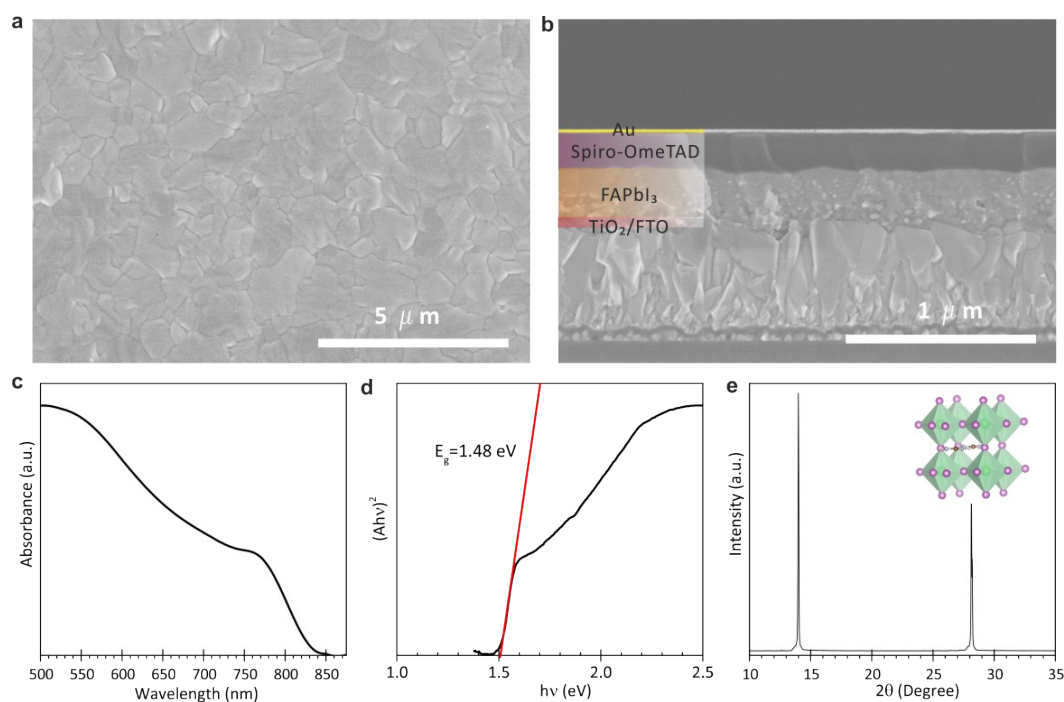


Figure S2 (a) Top-view SEM images of prepared FAPbI₃ layer. (b) Cross-sectional SEM images of the prepared device based on ITO/TO₂/ FAPbI₃/Spiro-OMeTAD/Au structure. (c) UV-vis spectra of FAPbI₃ layer prepared on the substrates. (d) The $(Ah\nu)^2$ vs. energy $(h\nu)$ curve of the FAPbI₃ layer. The optical band gap of CsPbBr₃ is about 1.48 eV. (e) X-ray diffraction (XRD) of prepared FAPbI₃ film.

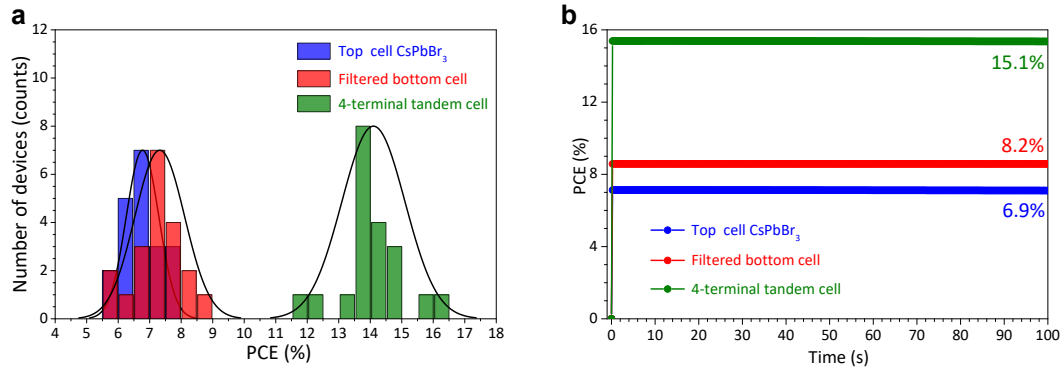


Figure S3: (a) Efficiency statistics shown as box plots for PEC distributions. (b) Steady-state efficiencies of the top cell CsPbBr₃, the filtered bottom cell, and the summed 4-terminal tandem cell.

Table S1 Parameters of current density–voltage curves of device.

Cell	J_{sc} (mAcm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
Top cell CsPbBr ₃	7.2	1.39	75.8	7.6
FAPbI ₃ (Unfiltered)	23.6	1.09	76.9	19.8
FAPbI ₃ (Filtered)	10.8	1.07	76.2	8.8
4-Terminal				16.4