

Supporting Information

The effect of ALD-ZnO layer on the formation of $\text{CH}_3\text{NH}_3\text{PbI}_3$ with different perovskite precursors and sintering temperatures

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Experimental section

1. Transparent Conducting Substrate and the Compact ZnO Layer

Fluorine-doped transparent conducting SnO_2 -coated glass substrates (FTO) were cleaned using ultrasonic washing in ethanol for 30 min and were subsequently treated in an oxygen plasma cleaning machine for 10 min. A 30 nm thick compact ZnO layer was deposited by atomic layer deposition. ALD was performed using an ALD system (TFS200) with a typical procedure. Zinc oxide (ZnO) was deposited at 70 °C using high purity diethylzinc and H_2O as the Zn and O precursors, respectively. ZnO was deposited in pulse mode in a nitrogen flow of 100 sccm. At 70 °C, diethyl zinc was retained in the chamber for 210 ms, after which H_2O was retained for 100 ms. The waiting period for both precursors was 1.5 s. The ZnO growth rate per cycle (GPC) was 1.8 Å. The ZnO deposition was performed for 330 cycles to achieve a layer thickness of 30 nm.

2. Solar Cell Fabrication

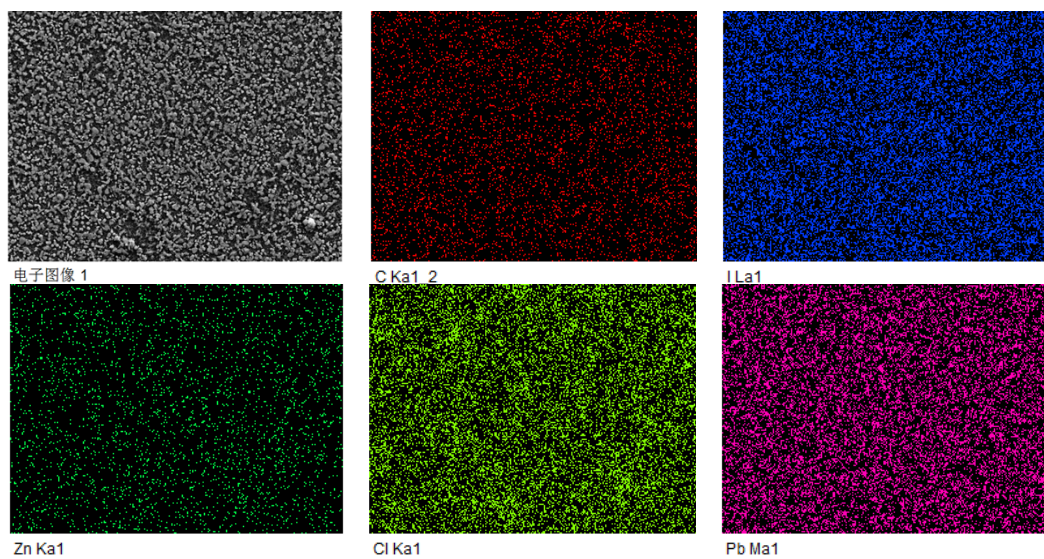
Solar cells were fabricated on precleaned FTO-coated glass substrates with a sheet resistance of $18\Omega\text{ sq}^{-1}$. First, a thin compact ZnO layer was deposited onto the substrate by ALD at 70 °C. A 350 nm thick mesoporous Al_2O_3 layer composed of 30 nm sized particles was then deposited by spin-coating. The $\text{CH}_3\text{NH}_3\text{PbI}_3$ solution (40wt%) prepared by reacting the $\text{CH}_3\text{NH}_3\text{I}$ and PbI_2 in N, N-dimethylformamide

(DMF) was spin-coated onto the substrate, and anneal at 100 °C for 10 min. Or the initially optimized 350 mg ml⁻¹ CH₃NH₃I and PbCl₂, 3: 1 molar ratio in DMF precursor solution was spin-coated onto the substrate at room temperature, followed by standing without heating for 30 min. Subsequently, the spiro-OMeTAD-based hole-transfer layer (170 mg spiro-OMeTAD, 28.5 ul 4-tert-butylpyridine and 20 mg lithium-bis(trifluoromethanesulfonyl) imide (Li-TFSI) all dissolved in 1 ml chlorobenzene) was deposited by spin coating at 4000 r min⁻¹ for 30 s. Finally, a 120 nm thick silver layer was deposited by vacuum evaporation at a pressure of 1.2×10^{-3} Pa. Prior to the evaporation of the Ag top contact, all device fabrication steps were carried out in a N₂-purged glove box (<0.1 ppm O₂ and H₂O).

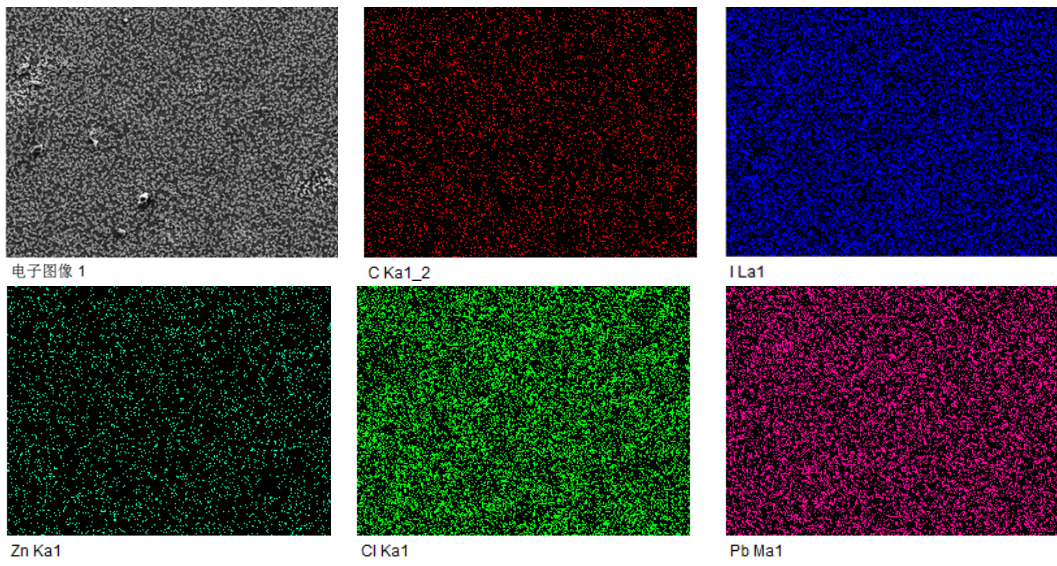
3. Device Characterizations

The current-voltage (I-V) curves of the solar cells were measured under room conditions using a Keithley 2400 source meter (Keithley Instruments Inc., Cleveland, OH, USA). The cells were illuminated by a 450 W Class AAA simulator equipped with an AM1.5G filter (XES-40S1) at a calibrated intensity of 100 mW cm⁻², as determined by a standard silicon reference cell. The effective area of the cell was defined as 0.0405 cm² using a non-reflective metal mask. The crystallographic structure of the perovskite was analyzed by X-ray diffraction (XRD) (D/MAX Ultima III, Rigaku Corporation, Tokyo, Japan) using Cu K α radiation. The morphology was determined by scanning electron microscopy (SEM). Specific surface areas of the perovskite and ZnO film were measured with a Quanta-chrome Autosorb-3b static volumetric instrument (Quanta-chrome Instruments, Boynton Beach, FL, USA).

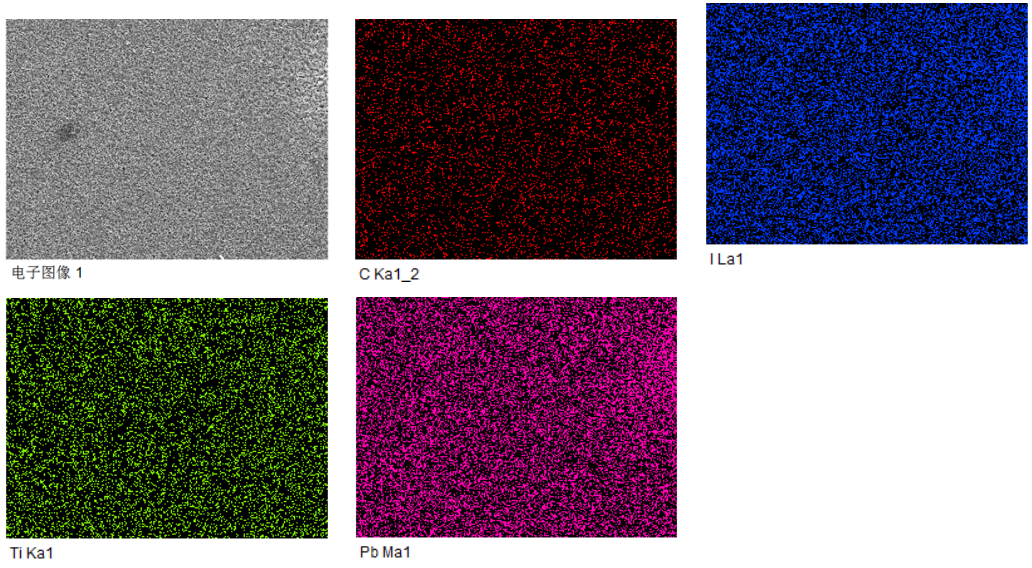
Distribution of chloridion on the different substrate was investigated using an energy-dispersive X-ray spectroscopy (EDS) combined with a field-emission scanning electron microscope (FE-SEM, Jeol JSM 6700 F). UV-visible (UV-vis) spectra were carried out on a Hitachi U-3010 spectrophotometer (Hitachi, Ltd., Chiyoda, Tokyo, Japan).



EDS maps of the $\text{CH}_3\text{NH}_3\text{PbI}_3$ on $70\text{ }^\circ\text{C}$ -ALD-ZnO annealing at $200\text{ }^\circ\text{C}$ for 3h



EDS maps of the $\text{CH}_3\text{NH}_3\text{PbI}_3$ on ALD-ZnO sintered at 450 °C annealing at 200 °C for 3h



EDS maps of the $\text{CH}_3\text{NH}_3\text{PbI}_3$ on ALD- TiO_2 annealing at 200 °C for 3h