Supporting Information

Efficient Defect Passivation with Niacin for High-Performance and Stable Perovskite Solar Cells

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Methods

Materials

Unless stated otherwise, all materials were used as received without any further purification. Anhydrous ethanol (anhydrous, 98%) and niacin were obtained from Aladdin Inc. Titanium isopropoxide, N, N-dimethylformamide (DMF), chlorobenzene (CB), dimethyl sulfoxide (DMSO), and acetonitrile were purchased from Sigma-Aldrich Inc. PbI_2 was obtained from TCI Inc. MAI, spiro-OMeTAD, 4-tert-butylpyridine (TBP) and bis-(trifluoromethylsulfonyl) imide lithium (Li-TFSI) were purchased from Xi'an Polymer Light Technology Corp.

Device Fabrication

Fluorine-doped tin oxide (FTO) was pre-cleaned in detergent, distilled water, ethanol, acetone, 2-propanol and treated in ultraviolet-ozone for 20 min. A compact thin layer (~40 nm) of TiO_2 was spin-coated onto the FTO glass and then annealed at 500°C for 1 h. The compact TiO_2 precursor solution was prepared as follows: 350 μL titanium isopropoxide and 35 μL 2 M HCl were added into 5 mL anhydrous ethanol. Then the mesoporous TiO_2 layer was deposited on the compact TiO_2 layer via spin-coating at 5000 rpm for 30 s using commercial TiO_2 paste (Dyesol 18NR-D), which was diluted in ethanol with a weight ratio of 1:7. The samples were then dried at 125 °C for 10 min and sintered at 450 °C for 30 min. Subsequently, the perovskite film was prepared using a one-step procedure at 2500 rpm for 25 s, to which 200 μL of CB was quickly dropped onto the substrate in the last 15s. The as-prepared films were heat treated at 100 °C for 20 min. The perovskite precursor solution is prepared according to previous method.[1] For the niacin-containing perovskite, various amount of niacin was added into the solution. Afterward, the hole-transporting layer was deposited by spin-coating Spiro-OMeTAD solution, which was prepared by dissolving 72.3 mg Spiro-OMeTAD in CB, with the addition of 17.5 μL Li-TFSI solution (520 mg/mL in acetonitrile) and 28.8 μL TBP. Finally, the devices were completed by evaporating 80 nm Au electrode under high vacuum condition and the effective area of each device was 0.096 cm^2.

Characterization

X-ray photoelectron spectroscopy (XPS) experiments are carried out using monochromatic Al-Kα (1486.6 eV) as the radiation source (Shimadzu, Axis Supra). The X-ray diffraction (XRD) measurements are performed using a MiniFlex 600 (Rigaku) X-ray diffractometer with Cu Kα radiation (λ = 0.15406 nm, 40 kV, 100 mA). UV-2600 spectroscopy (Shimadzu) with an integration sphere is used to measure the UV–vis absorption. Scanning electron microscope (SEM) (FEI, Inspect F) is used to collect the morphology of perovskite layers. Steady-state photoluminescence (PL) and time resolved photoluminescence (TRPL) spectra were performed using an Edinburgh FLS980 fluorescence spectrometer. Keithley 2400 source and a solar simulator (San-EI Electric) with standard AM 1.5G (100 mW cm^{-2}) illumination are used to measure the *J-V* characteristics under ambient conditions. A standard silicon solar cell AK-200 (Konica Minolta) is used to calibrate the light intensity. The cells mask area was 0.096 cm2. The incident photon-to-electron conversion efficiency (IPCE) was recorded by QTest Station2000 IPCE Measurement System (CROWNTECH). Transient photocurrent decay (TPC), transient photovoltage decay (TPV) and Mott−Schottky plot are measured by Paios 2.0 system.

DFT calculations

The calculation was performed by Vienna Ab-initio Simulation Program (VASP) package employing with a PAW-PBE pseudopotentials.[2-4] For the perovskite slabs, to simplify the calculations, the $CH_3NH_3PbI_3$ perovskite[5] was employed slab for the calculation of traps states. The 20 Å vacuum was added on top of the slabs surface to minimize the interaction with a Monkhorst-Pack k-point mesh of 2 × 2 × 1, and the plane-wave cutoff energy was set as 600 eV. The optimizations of the lattice constants and the atomic coordinates are made by the minimization of the total energy and all the atoms were allowed to relax until all internal atomic forces were within 0.005 eV/Å.
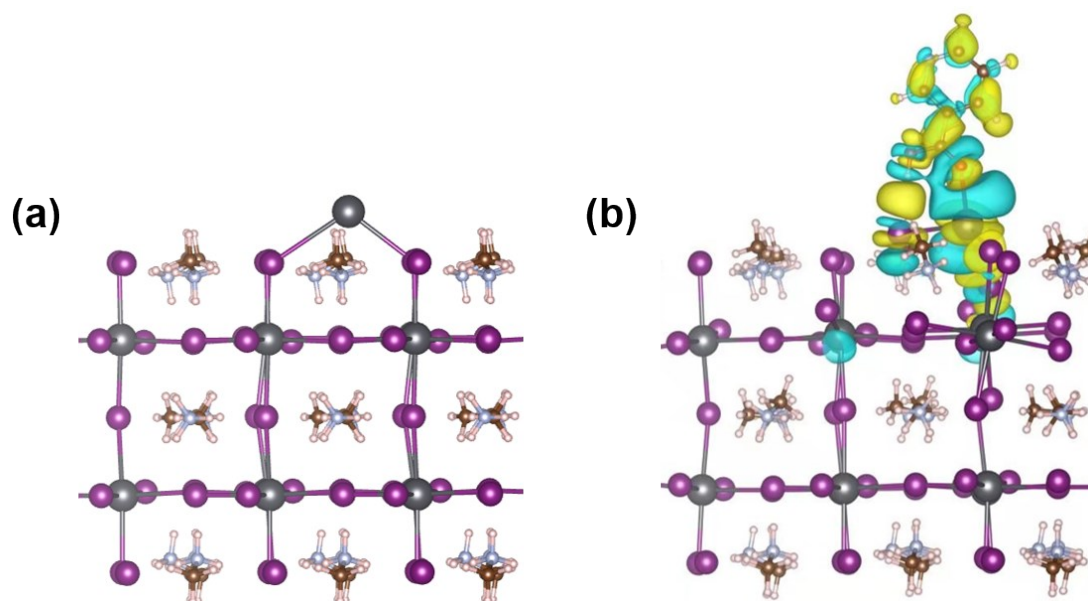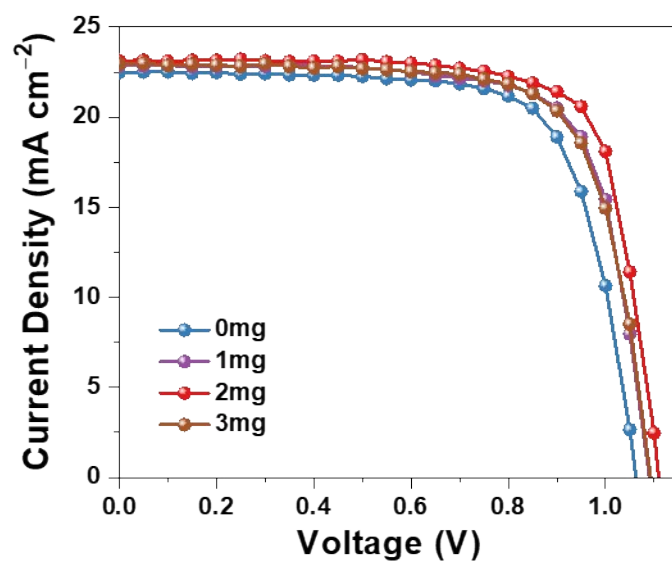
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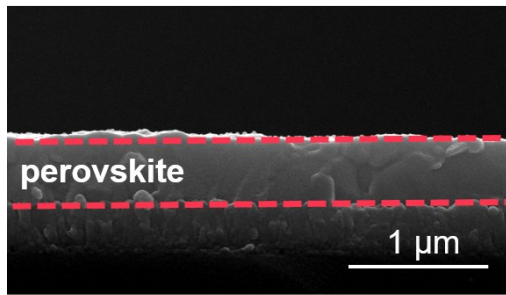
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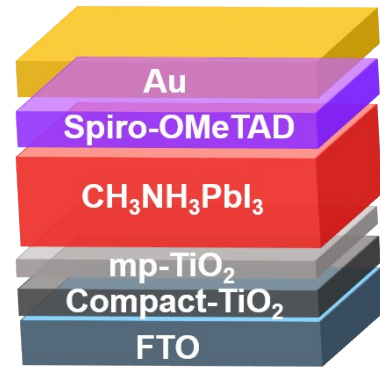
Fig. S1. (a) Cross-sectional SEM of perovskite film with niacin. (b) Schematic illustration of normal mesoporous PSCs.

Fig. S2. (a) Optimized geometry of (001) perovskite surface with Pb-dimer defect. (b) Optimized geometry and charge density difference of defected perovskite surface adsorbed with niacin.

Fig. S3. *J-V* curves of the PSCs with different concentrations of niacin in the reverse scan direction.
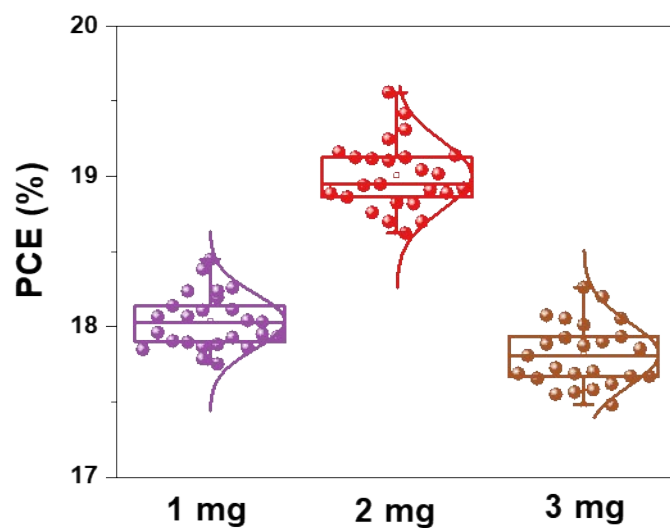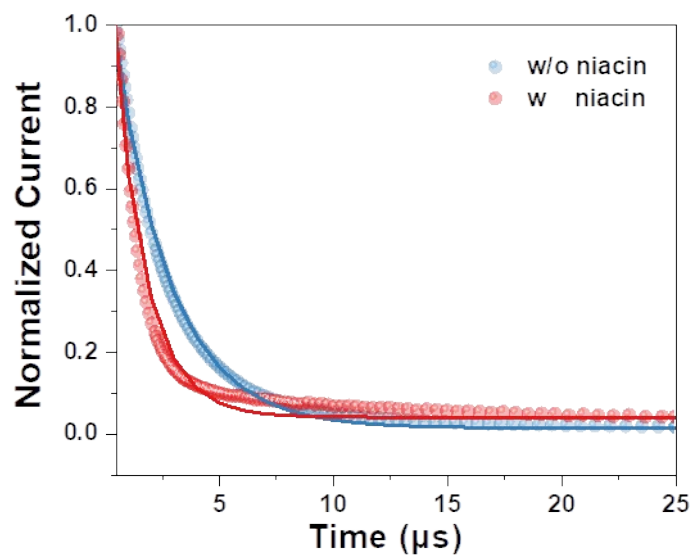
Fig. S4. A statistical distribution of PCE of 25 independent devices with various concentration of niacin.
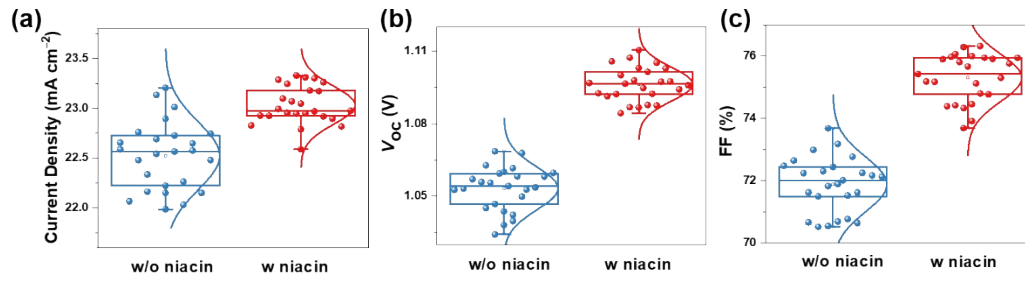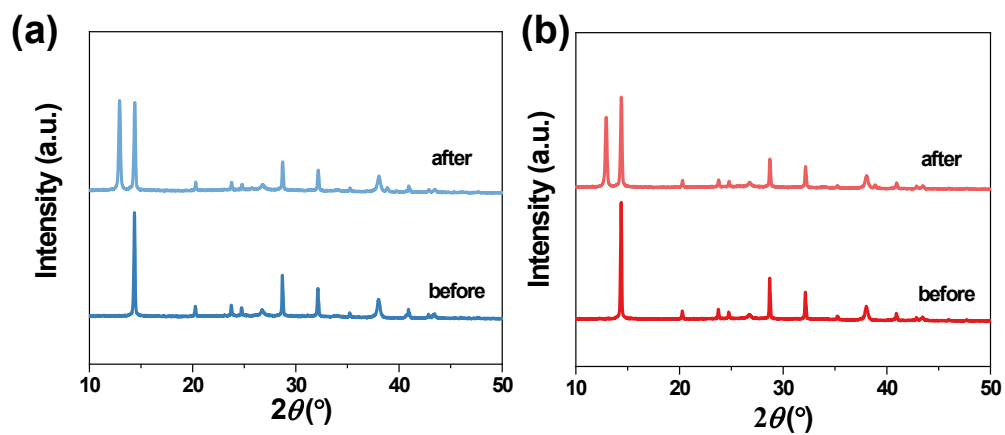
Fig. S5. The statistic distribution of (a), J_{sc} (b), V_{oc} (c) FF for a batch of 25 independent devices.
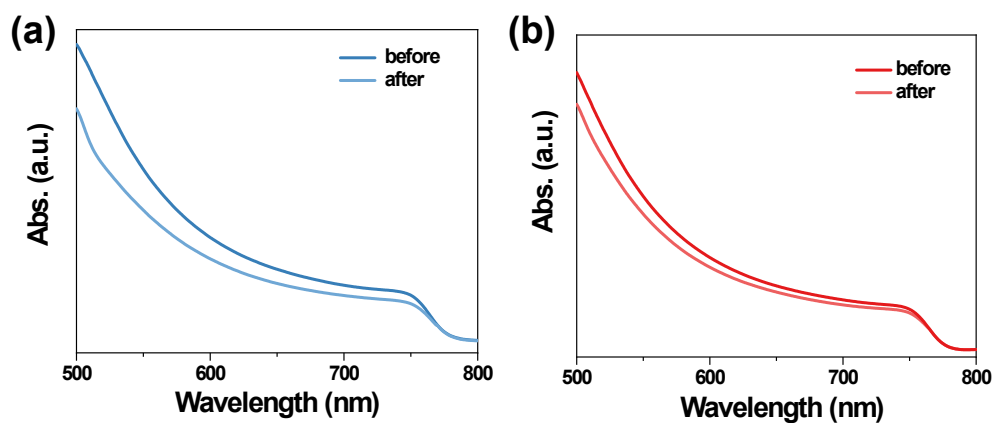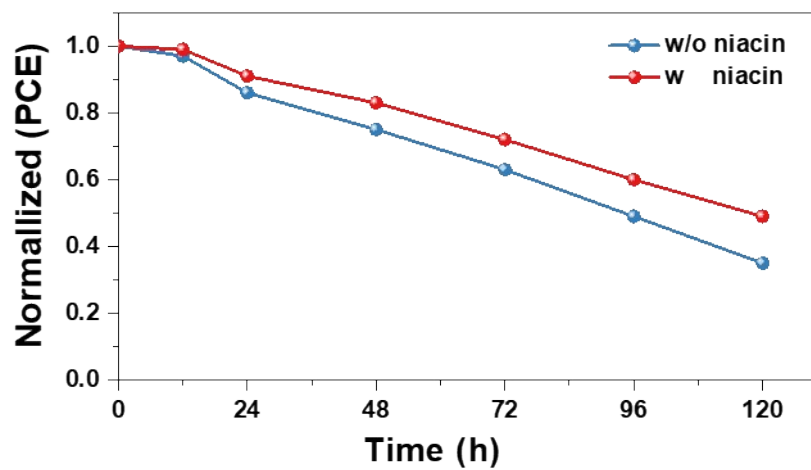
Fig. S6. Transient photocurrent (TPC) decays for PSCs devices with and without niacin.

Fig. S7. XRD patterns of perovskite films without niacin (a) and with niaicn (b) under 52 mW cm^{-2} UV irradiation for 100 minutes.

Fig. S8. UV-vis absorption spectra of perovskite films without niacin (a) and with niaicn (b) under 52 mW cm^{-2} UV irradiation for 100 minutes.

Fig. S9. (a) Devices thermal stability upon 85°C continuous annealing in ambient air atmosphere with relative humidity of 60% ± 10%

Table S1. Summary of devices parameters under different scan directions.

Niacin Concentration (mg mL^{-1})	Scan directions	J_{sc} (mA cm^{-2})	V_{oc} (V)	FF (%)	PCE (%)	HI (%)
0	forward	22.46	1.05	66.47	15.69	9.93
	reverse	22.48	1.06	73.18	17.42	
2	forward	23.12	1.10	72.21	18.36	5.94
	reverse	23.19	1.11	75.83	19.52	

Table S2. Flat band potentials and slopes obtained from the Mott-Schottky Plots.

Samples	Solpe	V_{bi} (V)	N (× 10^{15} cm^{-3})
Control	-4.20×10^{16}	0.93	5.20
Niacin	-3.56×10^{16}	0.98	6.14

References:

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