Supporting information for

Sprayed P25 scaffolds for High-Efficiency Mesoscopic Perovskite Solar Cells

Experimental details

Spray TiO_2 mesoscopic film

A spray system as shown in Figure S1 was set up for the deposition of mesoscopic TiO_2 films. The commercial P25 nanoparticles were dispersed in the ethanol with rigorous stirring and ultrasonic. The concentration of the TiO_2(P25) dispersion has been optimized, and 0.0014 M was determined as the best condition.

Before spray, the FTO/compact TiO_2 substrate was preheated to 100 °C for the fast evaporation of the ethanol solvent in the P25 source. The work of this spray system can be controlled with a digital controller, and the movement of the spray gun is also controlled by the stepping motor. When digitally turn on the spray gun, the P25 dispersion will be sprayed out and deposited onto the substrates. After 1 s, the spray gun will be automatically turned off. This is a spray circle. For each circle, 2 nm-thick TiO_2 film can be deposited. After the spray process, the TiO_2 film is annealing at 500 °C for 30 min. For the addition of titanium diisopropoxide bis(acetylacetonate) (TiAcAc), a certain amount of TiAcAc is added into the P25 dispersion with continuous stirring. The optimal molar ratio of TiO_2(P25)/TiAcAc is about 4.

Cell fabrication

Firstly, a dense TiO_2 underlayer with a thickness of about 20 nm was spin coated onto the pre-cleaned laser patterned FTO glass, which was then sintered at 500 °C for 30 min. The TiO_2 mesoscopic film was then sprayed onto the FTO/compact TiO_2 substrates, and annealed at 500 °C for another 30 min. After that, TiO_2 films were treated with 25 mM $TiCl_4$ solution at 70 °C for 40 min and sintered at 500 °C for 30 min.

$CH_3NH_3PbI_3$ absorber layer was deposited by a two-step deposition method in the N_2-glove box. Firstly, 1.3 M PbI_2 dissolved in DMF was spin coated onto the TiO_2 film twice at a speed of 3500 rpm for 30 s and then heat treated at 90 °C for 2 min to remove the DMF solvent. After the PbI_2 film cooling to room temperature, it was dipped into 10 mg/ml CH_3NH_3I solution with isopropanol as the solvent. After immersion in the solution (70 °C) for about 2 min, the $CH_3NH_3PbI_3$ film was obtained. The obtained $CH_3NH_3PbI_3$ films were then heat treated at 115 °C for 30 min in air with humidity of about 20% on a hotplate. After heat treatment, the films were kept in dark in air for overnight. A 300 nm hole transport layer was then deposited by spin-coating Spiro-OMeTAD solution in chlorobenzene on the $CH_3NH_3PbI_3$ layer. Finally, Au was thermal evaporated (Kurt J. Lesker) as electrode (80 nm) for heterojunction solar cells at an atmospheric pressure of 10^{-7} Torr.

Characterizations

Current-voltage (*I-V*) characteristics were measured by an additional voltage from the Keithley 2602 source meter together with a sunlight simulator (Oriel Solar Simulator 91192, AM 1.5 100 mW/cm^2) calibrated with a standard silicon reference cell. The solar cells were masked with a black aperture to define the active area of 0.1 cm^2. Impedance spectra were measured on IM6ex electrochemical workstation in the dark under different DC positive biases, in which scan frequency was in the range of 100 K ~ 100 mHz with the amplitude of the perturbation bias of 10 mV. X-ray diffraction (XRD) was measured with a Bruker X-ray diffractometer with Cu Kα as the radiation source. The scanning electron microscopy images of the films were obtained with FEI-SEM (XL 30 S-FEG). The external quantum efficiency of the cell was obtained with a lab-made IPCE spectrometer. The transient photoluminescence were measured with the PL spectrometer (Edinburgh Instruments, FLS 920) together with a pulsed diode laser (445 nm, 0.8 μJ/cm^2). An optical filter at 590 nm was used to filter out the excitation light during measurements. Transient photocurrent were measured with a pulsed red-light laser (1 KHz, pulse width 35 ns) at 660 nm and a sub-nanosecond resolved digital oscilloscope (Tektronix DPO 7104) with a sampling resistance of 50 Ω.

Figures

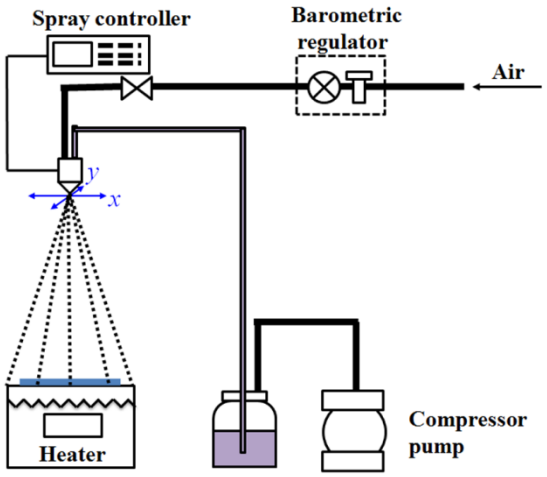
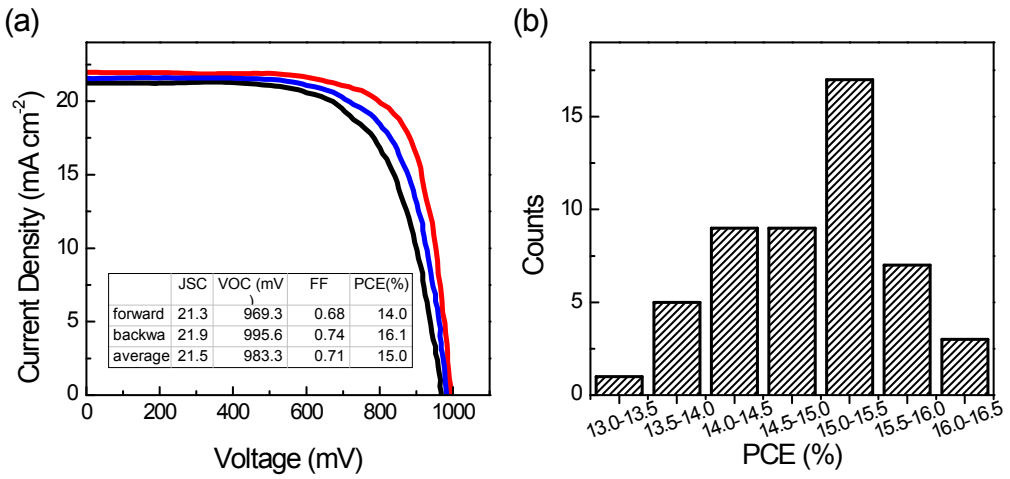


Figure S1. Schematic diagram of the spray system we set up.



	JSC	VOC (mV)	FF	PCE(%)
forward	21.3	969.3	0.68	14.0
backwa	21.9	995.6	0.74	16.1
average	21.5	983.3	0.71	15.0

Figure S2. (a) Forward, backward and the average current-voltage curves of one of the best performed perovskite solar cells based on the spray P25 film, and (b) the statistic histogram of the cell efficiency (backward scanning).