Supporting Information:

Using two-step deposition technique to prepare perovskite ($CH_3NH_3PbI_3$) for thin film solar cells based on ZrO_2 and TiO_2 mesostructures

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Sample preparation:

Fluorine-doped tin oxide (F:SnO_2) coated glass (Pilkington TEC 15) 15 Ω/□ was patterned by etching with Zn powder and HCl diluted in distilled water. The etched substrate was then cleaned with Acetone, ethanol and then dried in air. A compact TiO_2 blocking layer was first deposited onto the surface of a pre-cleaned FTO substrate by spray pyrolysis on a hotplate at 450℃ using 0.2 M Ti-isopropoxide, 2 M acetylaceton in isopropanol. 0.5 μm thick mesoporous TiO_2 and ZrO_2 layer was deposited by spin-coating. The layers were then sintered in air at 500 ºC for 30 minutes. A PbI_2 layer is deposited from 0.4mol/L PbI_2 (Sigma-Aldrich) in Dimethylformamide solution with a spin-coating speed of 2500 rpm, $CH_3NH_3PbI_3$ (Sigma-Aldrich) was deposited by dipping the slide into a 10 mg/mL CH_3NH_3I in iso-propanol solution, as it is described in the reference.[1] The coated films were then placed on a hot plate set at 100ºC for 20 minutes in air. The composition of hole transport material (HTM) was 0.170 M 2,2′, 7,7′-tetrakis-(N,N-di-p-methoxyphenyl-amine)-9,99-spirobifluorene (spiro-OMeTAD, Lumtec.), 0.064 M bis(trifluoromethane)sulfonimide lithium salt (LiTFSI, 99.95%, Aldrich) and 0.198 M 4-tert-butylpyridine (TBP, 99%, Aldrich) in anhydrous chlorobenzene (99.8%, Aldrich). The (CH_3NH_3) PbI_3 sensitized TiO_2 films were coated with HTM solution using spin-coating method at 4000 rpm. 200 nm Ag electrodes was deposited onto the solar cell by thermal evaporation.

Current-voltage (J-V) characteristics were measured using a Keithley 2400 source/meter and a Newport solar simulator (model 91160) giving light with AM 1.5 G spectral distribution, which was calibrated using a certified reference solar cell (Fraunhofer ISE) to an intensity 1000 W/m^2. A black mask of 0.2 cm^2 was applied on top of the cell to avoid significant additional contribution from light falling on the device outside the active area.

Incident photon to current conversion efficiency (IPCE) spectra were recorded using a computer-controlled setup consisting of a xenon light source (Spectral Products ASBXE-175), a monochromator (Spectral Products CM110), and a potentiostat (EG&G PAR 273), calibrated using a certified reference solar cell (Fraunhofer ISE). Electron lifetime and transport times were performed using a white LED (Luxeon Star 1W) as the light source. Voltage and current traces were recorded with a 16-bit resolution digital acquisition board (National Instruments) in combination with a current amplifier (Stanford Research Systems SR570) and a custom-made system using electromagnetic switches. Transport time and lifetimes were determined by monitoring photocurrent and photovoltage transients at different light intensities upon applying a small square wave modulation to the base light intensity. The electron lifetime measured with transient photovoltage was calculated using from the following equation: $V_{oc}=V_{oc,0}+\Delta V \exp(-t/\tau)$, where ΔV is the change in open-circuit voltage (V_{oc}) due to the modulated small change in light intensity, $V_{oc,0}$ is the open-circuit voltage before the change in

light intensity, and τ is the electron lifetime. The photocurrent and photovoltaic responses were fitted using first-order kinetics to obtain time constants.

Optimization of spin-coating speed

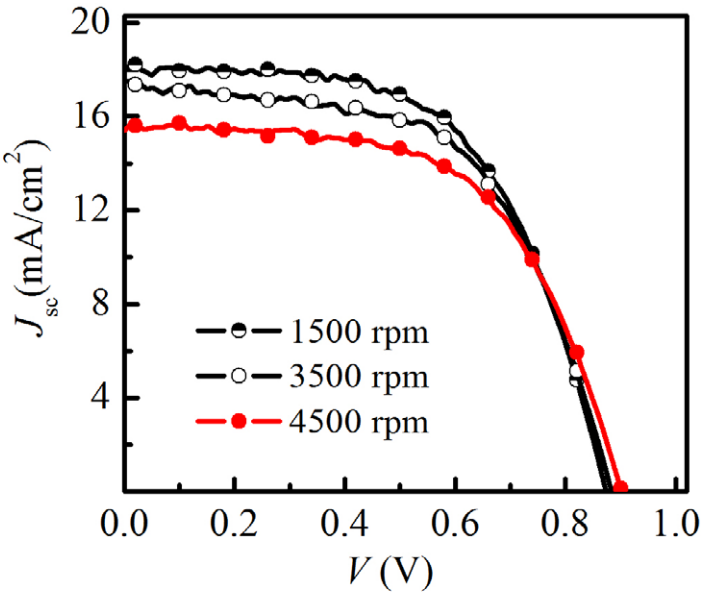

Fig.1 *J-V* curve under AM 1.5G illumination of 1000 W/m^2 for spiro-OMeTAD/$CH_3NH_3PbI_3$/TiO_2 solar cell using different spin-coating speed for PbI_2 deposition.

References

1. Liang, K. N.; Mitzi, D. B.; Prikas, M. T., Synthesis and characterization of organic-inorganic perovskite thin films prepared using a versatile two-step dipping technique. *Chem Mater* 1998,10, 403-411.