Supporting Information

Low-Temperature Solution Processed Tin Oxide as an Alternative Electron Transporting Layer for Efficient Perovskite Solar Cells

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Methods

Materials

4-tert-butylpyridine (TBP), and Li-bis(trifluoromethanesulfonyl) imide (Li-TFSI) were purchased from Aladdin-reagent. PbI₂ (99.99%), hydroiodic acid (57 wt.% in water, 99.99%), and methylamine (33 wt.% in absolute ethanol) were purchased from Sigma-Aldrich. Ethanol, diethyl ether, acetonitrile, and dimethylformamide were purchased from Sinopharm Chemical Reagent Corporation Co., Ltd. 2,2',7,7'-tetrakis-(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene (spiro-OMeTAD) (\geq 99.0%) was purchased from Shenzhen Feiming Science and Technology Co., Ltd. Fluorine-doped tin oxide (FTO) glass with a sheet resistance of 14 Ω sq⁻¹ was purchased from Asahi Glass (Japan). The purity of gold wire is 99.99%. All of the used reagents were analytical grade.

Devices fabrications

To prepare the TiO₂ ETLs a precursor solution of 18 ml ethanol, 1.8 ml tetrabutyl titanate, and 0.38 ml diethanolamine was stirred at 40 °C for 2 h.¹⁻² The solution was aged for 24 h to form a sol and then spin-coated on FTO substrates. Finally, the TiO₂ compact films were heated in air at 500 °C for 0.5 hour.

 SnO_2 ETLs were prepared by spin-coating precursor solutions of $SnCl_2 \cdot 2H_2O$ in ethanol with various concentrations on clean FTO substrates. The SnO_2 thin films were finally heated in air at 180 °C for 1 hour.

The perovskite $CH_3NH_3PbI_3$ films were prepared by a two-step method.²⁻³ Firstly, 462 mg PbI_2 dissolved in 1 mL dimethylformamide was stirred at 70 °C for 12h. The solution was spin-coated on the FTO substrates with either TiO_2 ETLs or SnO_2 ETLs at a low speed of 500 r.m.p. for 3s and followed a high speed of 2000 r.m.p for 30s. Secondly, the substrates were dipped into 10 mg/mL CH₃NH₃I dissolved in isopropanol for 8 mins and then dipped into clean isopropanol at room temperature. Finally, the films were heated in air at 70 °C for 0.5 hour.

To complete the devices, the perovskite films was spin-coated with HTLs using a solution composed of 68 mM spiro-OMeTAD, 26 mM Li-TFSI, and 55 mM TBP dissolved in acetonitrile and chlorobenzene (V/V=1:10) and then evaporated with thin gold electrodes.

Characterization

Transmission spectra were characterized by an ultraviolet–visible (UV–vis) spectrophotometer (CARY 5000, Varian, Australia) at room temperature. The compositions of the low-temperature prepared SnO_2 nanocrystalline films were characterized by a XPS system (Thermo Scientific, Escalab 250Xi). The crystalline structures of perovskite films were examined by an X-ray diffraction (XRD, Bruker AXS, D8 Advance). Transmission electron microscopy (TEM) was conducted by a JEOL-2010 TEM. The morphologies of the SnO_2 nanocrystalline and perovskite films and completed devices were imaged by a high-resolution field emission SEM (JSM 6700F). The morphologies of the surfaces of bare FTO and the SnO_2 coated on the FTO substrates were characterized by atomic force microscopy (AFM, SPM-9500j3). Impedance spectra were performed on a CHI660D electrochemical workstation (ShangHai, China) with a 10 mV AC amplitude, a frequency ranging from 100 kHz to 0.1 Hz. The current-voltage (*J-V*) characteristics were performed on a CHI660D electrochemical workstation (ShangHai, China). The area of the Au electrode was 0.09 cm², which defines the active area. All the cells were measured under a 100 mW cm⁻² (AM1.5 simulated irradiation) illumination with a standard ABET Sun 2000 Solar Simulator. A standard silicon solar cell was used to calibrate the light intensity.

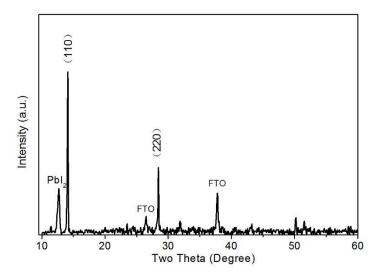


Figure S1. XRD pattern of a perovskite CH₃NH₃PbI₃ film coated on a FTO substrate coated with a SnO₂ ETL.

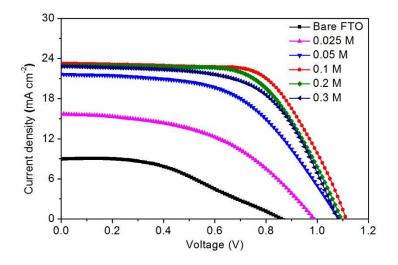


Figure S2. *J-V* curves of the cells made on bare FTO substrate and FTO substrate coated with SnO₂ films prepared by using different SnCl₂•2H₂O solution concentrations.

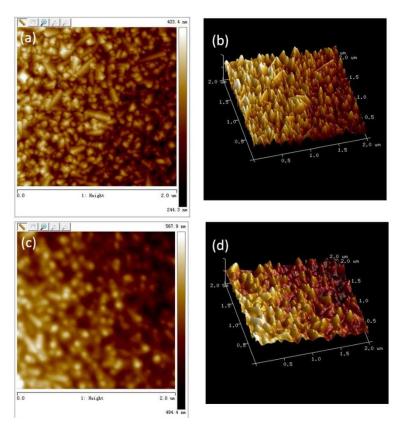


Figure S3. AFM images of (a, b) a bare FTO and (c, d) a FTO substrate coated with SnO_2 nanocrystalline film.

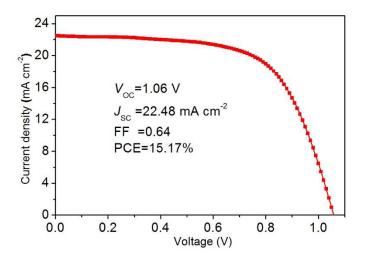


Figure S4. *J-V* curve of our best-performing perovskite solar cell using a TiO₂ ETL.

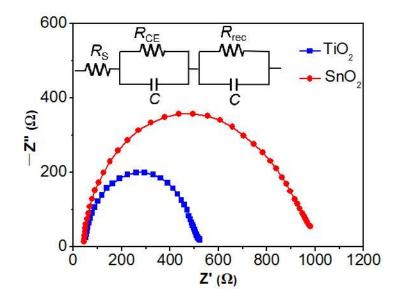


Figure S5. Nyquist plots of the perovskite solar cells using the SnO_2 and TiO_2 ETLs under illumination at 1.0 V biases.

Table S1. Photovoltaic parameters of the cells based on bare FTO substrate and the SnO_2 ETL coated onFTO substrates. The data are received from Figure S2.

	V _{oc} (V)	$J_{\rm sc}$ (mA cm ⁻²)	FF (%)	PCE (%)
Bare FTO	0.87	9.15	41.72	3.32
0.025 M	0.99	16.07	47.66	7.58
0.050 M	1.09	22.11	53.45	12.88
0.100 M	1.11	23.24	65.28	16.84
0.200 M	1.09	22.92	63.25	15.80
0.300 M	1.08	22.8	60.61	14.93

References

1. Ke, W. J., Fang, G. J., Tao, H., Qin, P. L., Wang, J., Lei, H. W., Liu, Q., Zhao, X. Z. ACS Appl. Mater. Interfaces 2014, 6, 5525-5530.

2. Ke, W. J., Fang, G. J., Wang, J., Qin, P. L., Tao, H., Lei, H. W., Liu, Q., Dai, X., Zhao, X. Z. ACS Appl. Mater. Interfaces 2014, 6, 15959-15965.

3. Burschka, J., Pellet, N., Moon, S. J., Humphry-Baker, R., Gao, P. Nazeeruddin, M. K.; Grätzel, M. *Nature* **2013**, *499*, 316-319.