

## Supplementary Information

# SnO<sub>x</sub>-Brookite TiO<sub>2</sub> Bilayer Electron Collector for Hysteresis-less High Efficiency Plastic Perovskite Solar Cells Fabricated at Low Process Temperature

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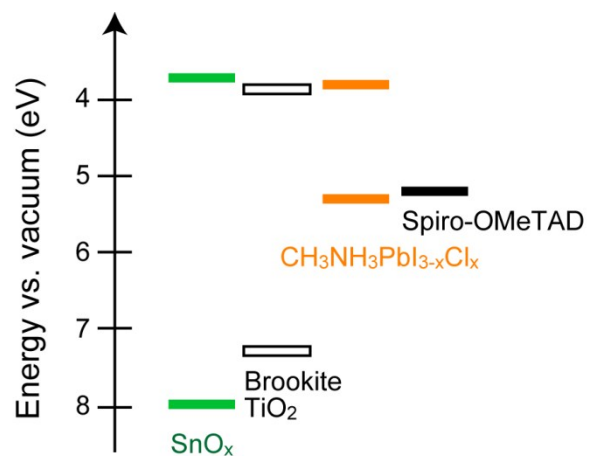
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## Experimental details

A SnO<sub>x</sub> hole-blocking layer was formed on the surface of an ITO-PEN films (Pecell Technologies, Inc.) by spin-coating 1-butanol solution of 0.1 M SnCl<sub>2</sub>·2H<sub>2</sub>O and dried at 150 °C for 1 h. A binder-free brookite slurry (TiO<sub>2</sub> 3.5wt% in ethylene glycol monopropyl ether), which was prepared by solvent replacement of brookite aqueous sol. (PECC-B01, Pecell Technologies, Inc, particle size 10–20 nm), was spin-coated and dried for further 3 h. For planar-structured cells, SnCl<sub>2</sub>·2H<sub>2</sub>O solution was dried for 4 h without deposition of brookite.

After UV-ozone treatment for 5 min, a mixed solution of CH<sub>3</sub>NH<sub>3</sub>I (2.7 M) and PbCl<sub>2</sub> (0.9 M) in *N,N*-dimethylformamide were spin-coated on the substrate at 2000 rpm for 30 s and annealed at 105 °C for 90 min in dry air to form perovskite CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3-x</sub>Cl<sub>x</sub> layer. A 0.12 M 2,2',7,7'-tetrakis-(*N,N*-di-4-methoxyphenylamino)-9,9'-spirobifluorene (spiro-OMeTAD) solution in chlorobenzene containing 0.034 M bis(trifluoromethane) sulfonamide lithium salt and 0.098 M 4-*tert*-butylpyridine was spin-coated at 4000 rpm for 30 s and aged overnight in dry air. Finally, Au was vapor-deposited on the top of spiro-OMeTAD as a counter electrode. The device size with ITO-PEN substrate was 1.2 cm × 1.2 cm on which three cells were fabricated by depositing Au counter electrode (3.2 mm × 8.0 mm). Photovoltaic characteristics of all the devices were measured with a black mask (aperture area of 0.09 cm<sup>2</sup>, thickness of 400 μm) with a Keithley 2400 source meter under 1 sun illumination in ambient air at room temperature. Pecell Technologies PEC-L01 solar simulator (AM 1.5 G, 100 mW cm<sup>-2</sup>) was employed with a reference crystalline Si cell (BS-520, calibrated and certified by Bunkou Keiki) for correction of incident intensity. Photocurrent density-voltage curves were measured with scan speed and dwell time of 0.125 V s<sup>-1</sup> and 0.05 s, respectively. The IPCE action spectra of device were measured in ambient air at room temperature on Pecell Technologies, PEC-S20 action spectrum measurement setup. For characterization, an UV spectrophotometer (UV-1800, SHIMADZU), an X-ray diffractometer (D8 Discover, Bruker) with CuKα radiation source, ~~and~~ a scanning electron microscope (SU8000, HITACHI) and an atomic force microscope (SPM-9500J3, SHIMADZU) were employed. Photoelectron yield spectroscopy system (BIP-KV201, Bunkou Keiki) was employed to measure work function of materials.



**Figure S1.** Energy levels of the materials used in this study. Values of  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  and spiro-OMeTAD are obtained from literature.<sup>1</sup>

## References

1. J. T.-W. Wang, J. M. Ball, E. M. Barea, A. Abate, J. A. Alexander-Webber, J. Huang, M. Saliba, I. Mora-Sero, J. Bisquert, H. J. Snaith and R. J. Nicholas, *Nano Lett.*, 2014, **14**, 724–730.