

Supporting Information

Polymer-complexed SnO₂ Electron Transport Layer for High-Efficiency *n-i-p* Perovskite Solar Cells

Zhenhua Xu^{1,2}, Chi Huey Ng³, Xinming Zhou², Xiaohui Li¹, Putao Zhang^{1*}, and Siow Hwa Teo⁴

¹Key Laboratory of Photovoltaic Materials, Henan University, Kaifeng, Henan, 475004, China

²Institute of New Energy Technology, College of Information Science and Technology, Jinan University, Guangzhou, 510632, China

³Chemical Engineering Program, Faculty of Engineering, Universiti Malaysia Sabah, 88400 Kota Kinabalu, Sabah, Malaysia

⁴Industrial Chemistry Program, Faculty of Science and Natural Resources, University Malaysia Sabah, 88400 Kota Kinabalu, Sabah, Malaysia

Materials

SnO₂ colloidal (15 wt% in H₂O), DMF(99.8%), DMSO (99.7%), Chlorobenzene (99.8%), 4-tBP (96%), LiTFSI (99.95%), IPA (99.99%) were obtained from Alfa Aesar. PbI₂ (99.99%), CsI (99.99%), PbBr₂ (98%), SpiroMeOTAD (99.5%), MAI, FAI, MACl, and MABr and ITO substrates were purchased from Preferred Tech Co., Ltd. Poly(amidoamine) (PM) was purchased from Chenyuan molecular New Mater. Co., Ltd.

Device Fabrication

ITO substrates were ultrasonically cleaned in acetone, ethanol, IPA and deionized water for 15 min, respectively. The cleaned ITO ware dried with air and treated with plasma for 15 min. Mix 0.5mL SnO₂, 2 mL deionized water and a certain amount of PM evenly (The final concentration of PM is 0~6 mg/mL). Then spin it onto the ITO substrate at 5000 rpm for 30 s then heated at 150°C for 30 min. Firstly, a PbI₂ solution (1.5M, PbI₂ in DMF/DMSO (9:1)) was spin coated on SnO₂ electron transport layer at 1500 rpm for 45s. After 5 min, place it on a 70 °C hotplate and heat it for 1 min. Secondly, FA/MA solution (FAI/MABr/MACl=90/6/9 mg in 1 mL IPA) drop on top of the PbI₂, by spin coating at 1600 rpm for 45s. The obtained perovskite film was put on a hotplate and heated at 150 °C for 15 min in open air (T=25°C, H=35%). The hole-transport layer was prepared by spin-coating a Spiro-OMeTAD on top of perovskite (72.3 mg Spiro-OMeTAD, 18.5 μL Li-TFSI (520 mg mL⁻¹ in acetonitrile), 28.8 μL 4-tBP in 1 mL of chlorobenzene). Finally, MoO₃ with a thickness of 5 nm and Ag with a thickness of 120 nm were evaporated successively to complete the device manufacturing.

Characterization

DLS measurements were carried out with alight-scattering apparatus (ALV/DLS/SLS- 5022F). The sample morphology were characterized by FE-SEM (JSM-7001F, Japan). Perovskite crystal structures were measured by X-ray diffraction (DX-2700). FTIR spectra were characterized by infrared spectrometer (Tensor 27, BRUKER). XPS test was measured by Thermo-Fisher system (ESCALAB 250Xi). PL and TRPL spectra were measured using a laser with wavelength of 514 nm (Renishaw inVia microRaman system, Hamamatsu C10910). The *J-V* curves were obtained using a

source-meter (Keithley 2400) and a solar simulator (Oriel 94023 A, Newport). The EQE measurement was conducted on an EQE measurement system (500ADX, USA).

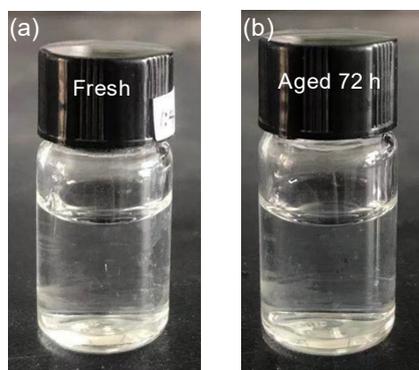


Figure S1. Photos of fresh (a) and aged SnO_2 (b) colloidal dispersions at 25°C for 72h. Commercial SnO_2 colloidal dispersion diluted in water (volume ratio, 1:4).

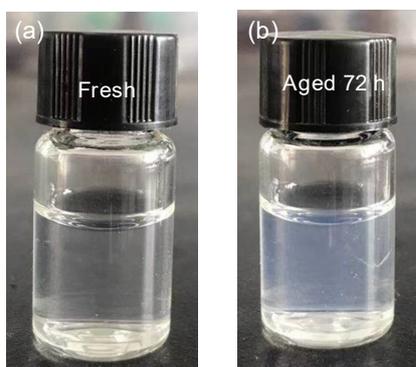


Figure S2. Photos of fresh and aged SnO_2 colloidal dispersions at 35°C for 72h. (a) control and (b) aged- SnO_2 (Commercial SnO_2 colloidal dispersion diluted in water with volume ratio, 1:4).

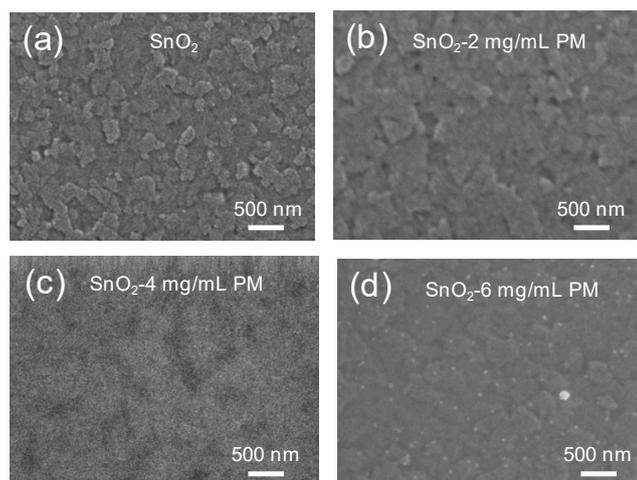


Figure S3. Top-view SEM images of SnO₂ films with different PM additions (a), (b), (c), and (d) control, 2 mg mL⁻¹, 4 mg mL⁻¹, and 6 mg mL⁻¹.

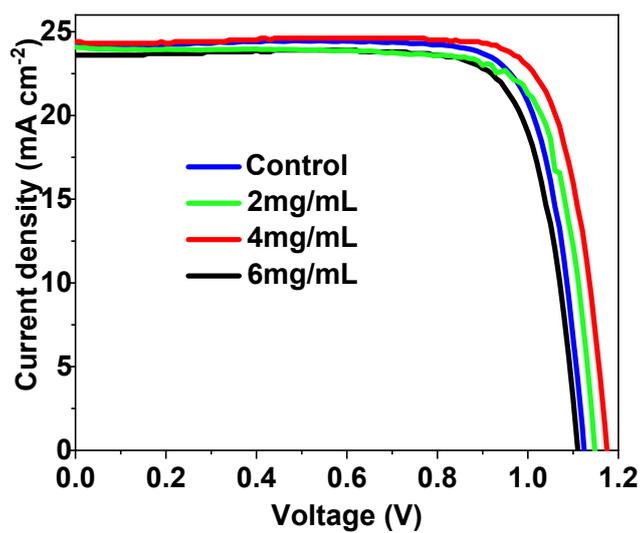


Figure S4. J - V curves of devices based on SnO₂ ETL with different PM additions (control/2/4/6 mg mL⁻¹).

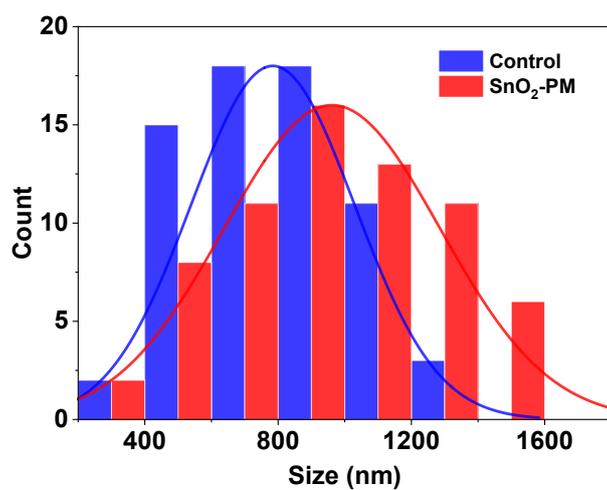


Figure S5. Distribution of perovskite grain size deposition on control and SnO₂-PM ETLs.

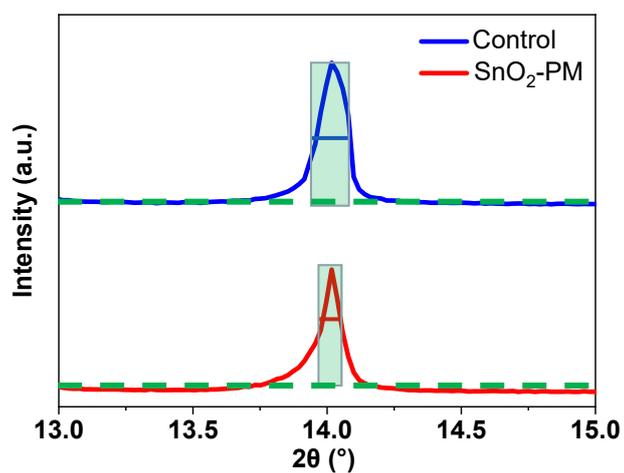


Figure S6. Full-width at half-maximum of perovskite characteristic peak at 14.05° based on control and SnO₂-PM, respectively

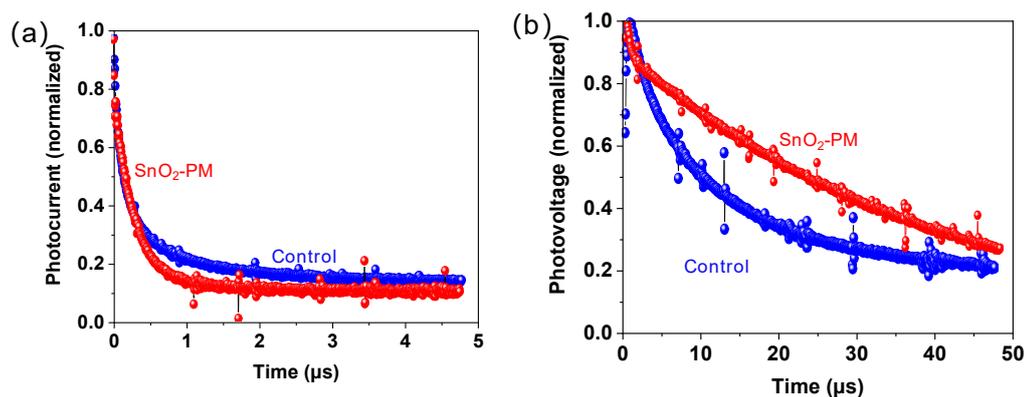


Figure S7. (a) TPC and (b) TPV for PSCs based on control and SnO₂-PM ETL.

Table. S1 Photovoltaic parameters of devices based on different SnO₂ ETL (PM concentration: 0/2/4/6 mg mL⁻¹).

Samples	$J_{SC}/ \text{mA cm}^{-2}$	V_{OC}/ V	FF	$PCE/ \%$
Control	24.17	1.13	0.769	21.01
SnO ₂ -PM-2	24.18	1.12	0.797	21.69
SnO ₂ -PM-4	24.41	1.17	0.803	22.93
SnO ₂ -PM-6	23.60	1.11	0.790	20.70

Table. S2 TRPL fitting results of control and SnO₂-PM based PSCs.

Samples	τ_1	τ_2
Control	13.76	31.09
SnO ₂ -PM	21.06	5.91

Table. S3 Fitting results of electrochemical impedance spectroscopy control and SnO₂-PM based PSCs.

Samples	$R_s (\Omega)$	$R_{rec} (\Omega)$
Control	36	331
SnO ₂ -PM	28	586