Supporting Information

Amino acid salt Induced PbI_2 crystal orientation Optimization for High-Efficiency Perovskite Solar Cells with Long-Term Stability

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Materials

SnO_2 colloid precursor (tin (IV) oxide, 15% in H_2O colloidal dispersion) was purchased from Alfa Aesar. Dimethylformamide (DMF), dimethyl sulfoxide (DMSO), chlorobenzene (CB), isopropanol (IPA), lead iodide (PbI_2), rubidium chloride (RbI), bis(trifluoromethane)sulfonimide lithium salt (Li-TFSI) and acetonitrile and 4-tert-butylpyridine (tBP) were purchased from Sigma Aldrich. Formamidinium iodide (FAI), methylammonium chloride (MACl), and 1- Butylaminum iodide (BAI) were purchased

from Sigma Aldrich. 2,2′,7,7′-Tetrakis[N, N-di(4-methoxyphenyl)amino]-9,9′-spirobifluorene (spiro-OMeTAD) was purchased from Xi'an Polymer Light Technology in China. Except for SnO_2 solution stored in ambient air, all the materials are stored in the nitrogen-filled glove box to avoid the water.

Device fabrication

FTO substrates were washed with detergent, deionized water, acetone, isopropanol, and ethanol. The substrates were treated with ultraviolet ozone for 15 min. Then the diluted SnO_2 colloid solution (2.14 wt.%) was spin-coated at 3000 rpm for 30 s and annealed at 150 °C for 30 min. Then the PL-Glu solution (0.01 M ~ 0.03 M in water) was spin-coated onto the substrates at 3000 rpm for 30 s and annealed on a hot plate at 150 °C for 10 min under an ambient atmosphere. PbI_2 solution (1.3 M PbI_2, 13.8 mg RbI, and 11 mg CsI in DMF and DMSO with a volume ratio of 95:5) was dropped onto the ETL and then spin-coated at 2000 rpm for 50 s. At the end of 10 s of the spin coating process, organic salt solution (FAI/MACl (60:11.7 mg) in 1 mL IPA) was dropped rapidly onto the PbI_2 film and then annealed at 150 °C for 11 min. Upon cooling to room temperature, the 1- Butylaminum iodide (BAI, 2 mg mL^{-1} in IPA) was spin-coated on the perovskite at 5000 rpm for 30 s without annealing. The HTL solution, prepared by dissolving 72.3 mg Spiro-OMeTAD, 17.5 μL LiTFSI solution (520 mg Li-TFSI in 1 mL acetonitrile), and 28.8 μL tBP in 1 mL chlorobenzene, was spin-coated on the perovskite layer at 3000 rpm for 30 s. Finally, 100 nm Ag was thermally evaporated as an electrode using a shadow mask (0.04 cm^2).

PMMA is spin-coated onto the top surface of FTO/SnO_2/perovskite, and the

perovskite is bonded to the clean ITO using UV-curable glue. The perovskite film is then transferred to glass/ITO by forced peeling for the subsequent characterization (glass/ITO/PMMA/perovskite).

Measurements and characterization

X-ray diffraction patterns were acquired by the X-ray diffractometer (Rigaku SmartLab). Grazing incidence XRD was acquired by the X-ray diffractometer (R Rigaku Smartlab 9KW). The morphology of the perovskite surface and cross sections were obtained by Scanning electron microscopy (SEM, Hitachi SU8020). Surface roughness images were performed with an atomic force microscope (AFM, Bruker Nano Inc ICON2-SYS). Ultrahigh vacuum photoelectron spectroscopy (Thermo escalab 250Xi) was used for XPS measurements and UPS measurements. Steady-state PL spectra and TRPL transient decay were measured using a PL spectrometer (Edinburgh Instruments, FLS 1000). All absorption measurements were carried out by ultraviolet-visible spectrophotometer (TP-720 UV–VIS-NIR). The EQE curves were measured using an EnliTech EQE measurement system. J-V characteristics were measured using a Keithley 2450 source meter and solar simulator under standard simulated AM 1.5 illumination, scan rate is 0.4V/s.

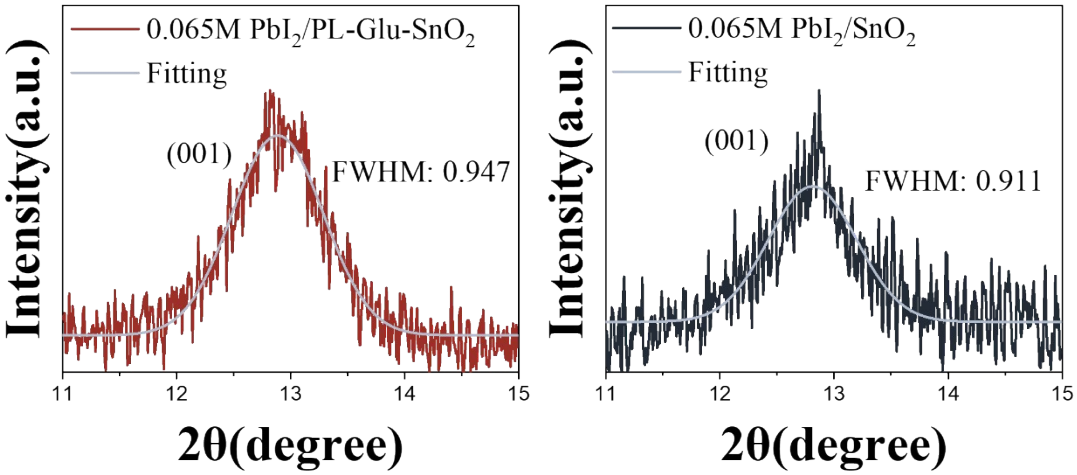

Figure S1. The FWHM of the PbI_2 (001) peak of 0.065 M PbI_2 films upon the Control SnO_2 and the PL-Glu-SnO_2.

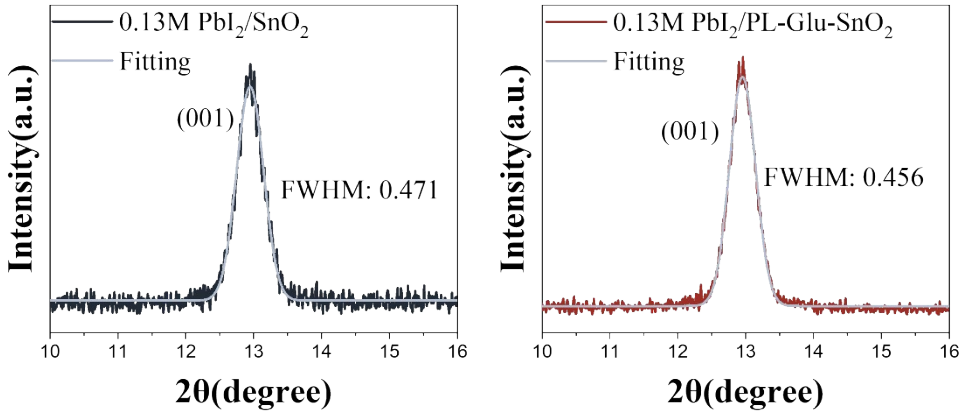

Figure S2. The FWHM of the PbI_2 (001) peak of 0.13 M PbI_2 films upon the Control SnO_2 and the PL-Glu-SnO_2.

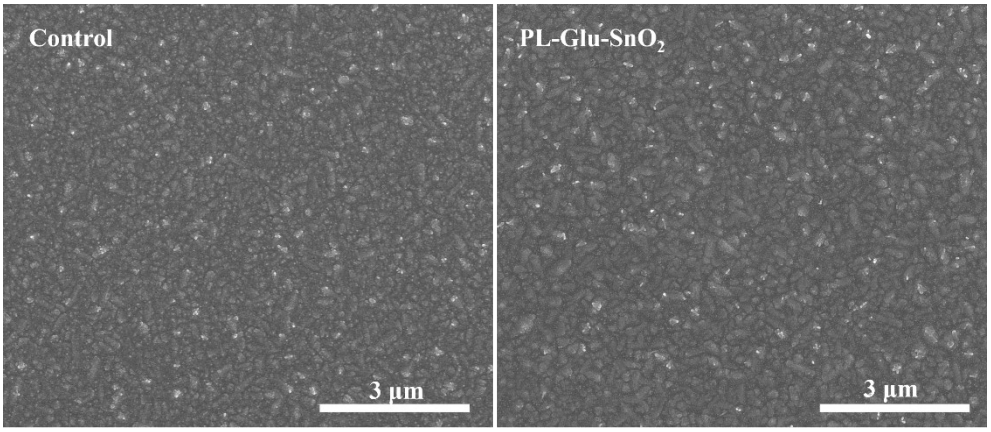

Figure S3. SEM image of 0.13 M PbI_2 films upon the Control SnO_2 and the PL-Glu-SnO_2.

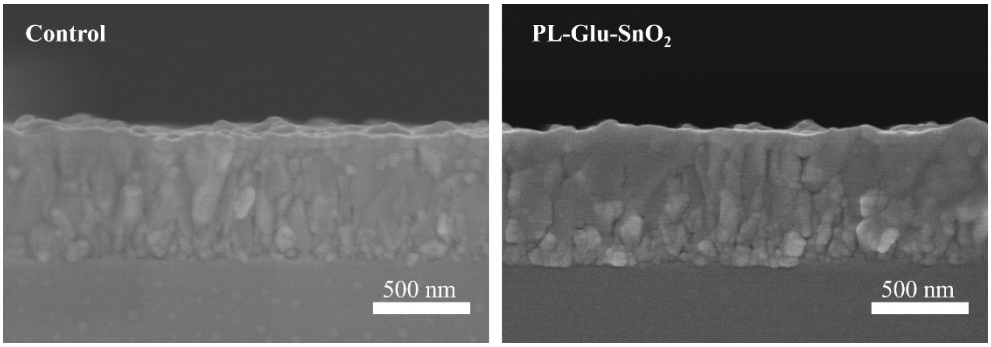

Figure S4. Cross-section SEM image of 0.13 M PbI_2 films upon the Control SnO_2 and the PL-Glu-SnO_2.

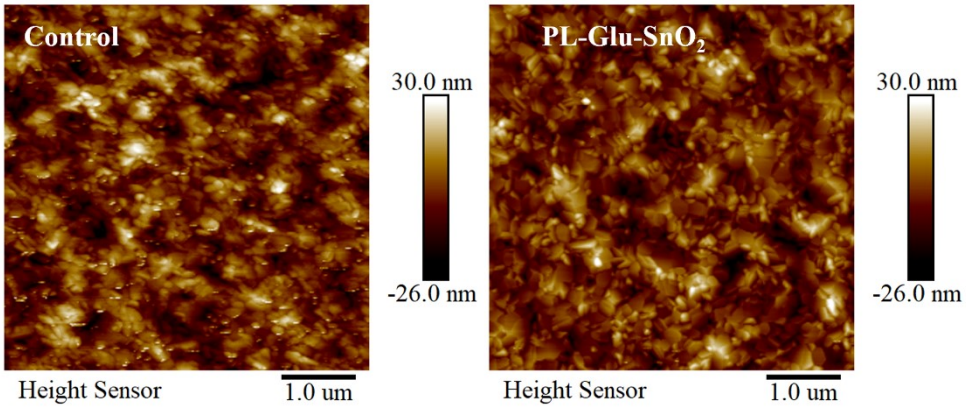

Figure S5. AFM image of 0.13 M PbI_2 films upon the Control SnO_2 and the PL-Glu-SnO_2.

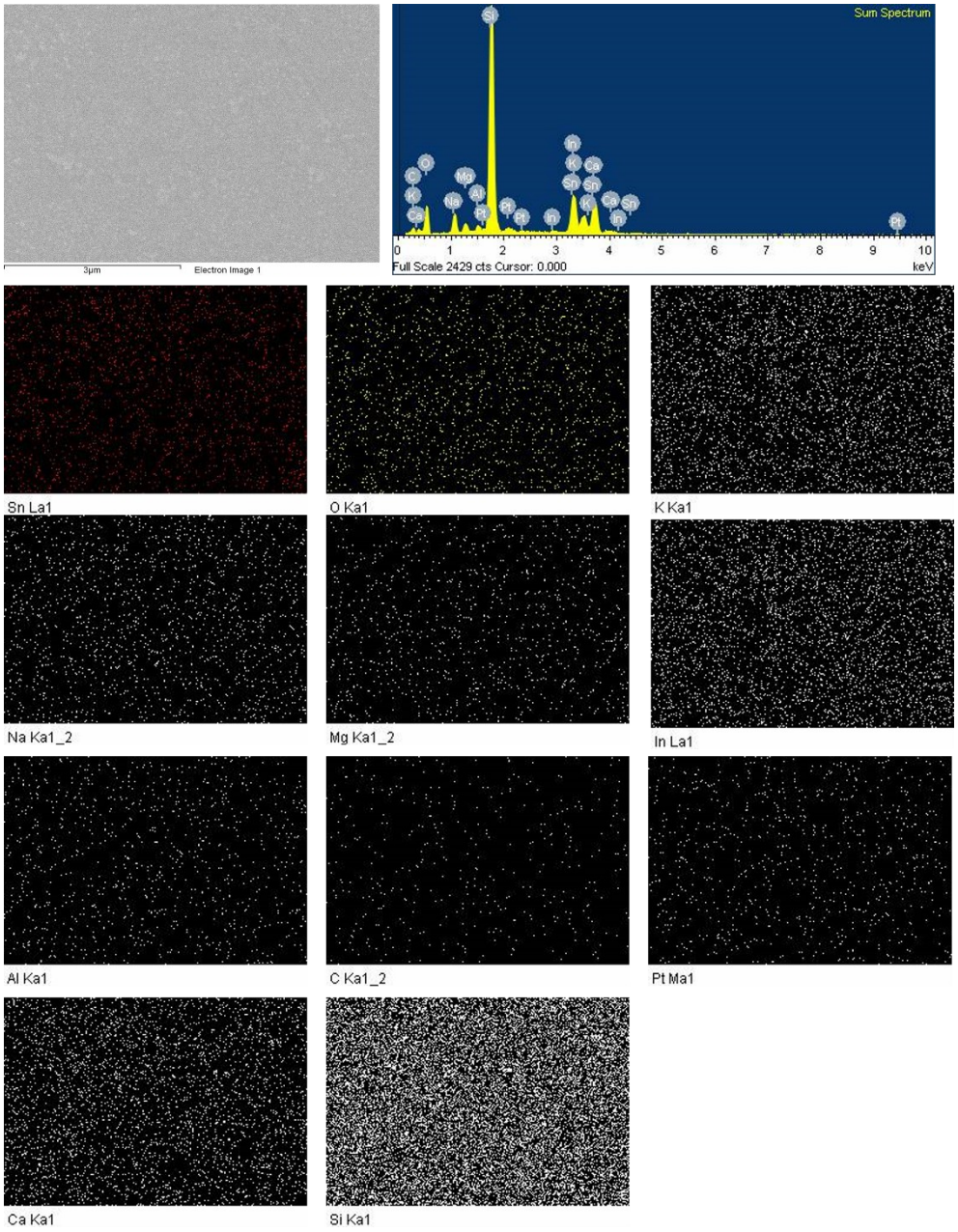

Figure S6. SEM-EDX elemental mapping image of surface SnO_2 by forced peeling.

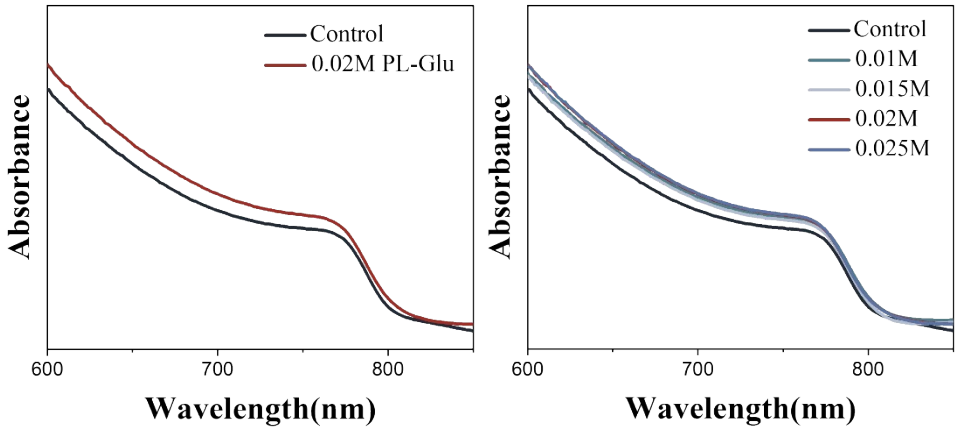

Figure S7. The Ultraviolet-visible (UV-vis) absorption spectra of perovskite films fabricated on SnO_2 substrates modified with different concentrations of PL-Glu.

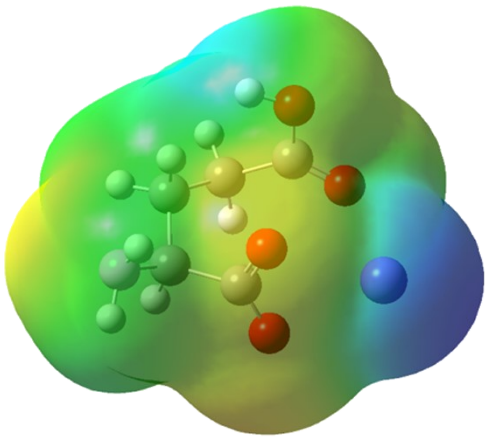

Figure S8. ESP map of PL-Glu.

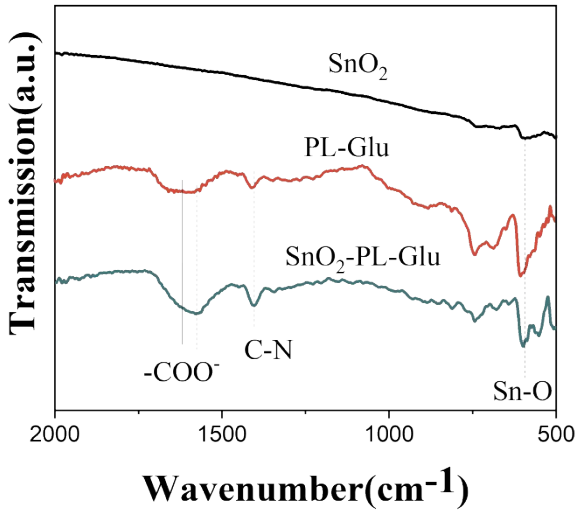

Figure S9. The Fourier Transform Infrared (FTIR) spectroscopy measurements for the SnO_2, PL-Glu and SnO_2- PL-Glu films.

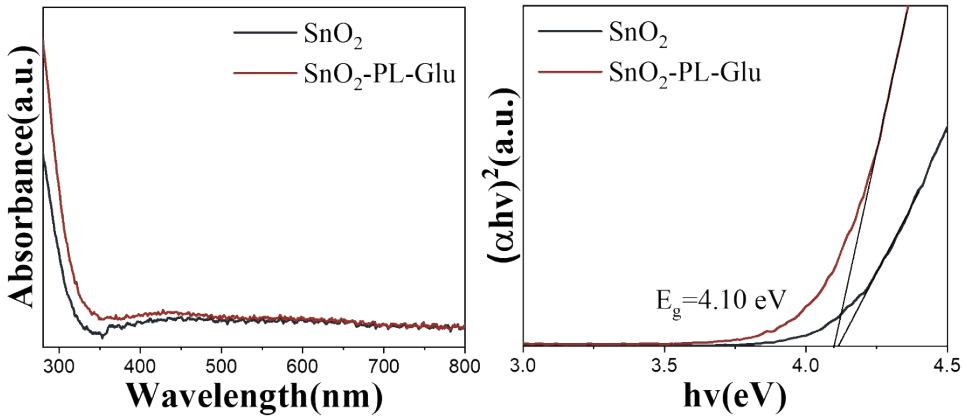

Figure S10. The UV-vis absorption spectra of the Control SnO_2 and the PL-Glu-SnO_2.

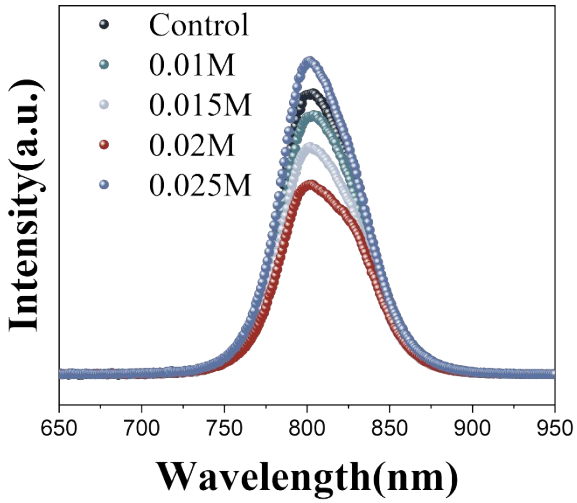

Figure S11. Steady-state PL spectra of perovskite films fabricated on SnO_2 substrates modified with different concentrations of PL-Glu.

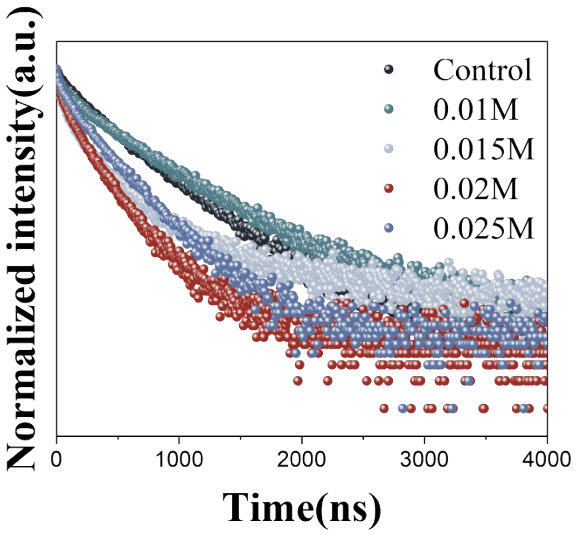

Figure S12. TRPL curves of perovskite films fabricated on SnO_2 substrates modified with different concentrations of PL-Glu.

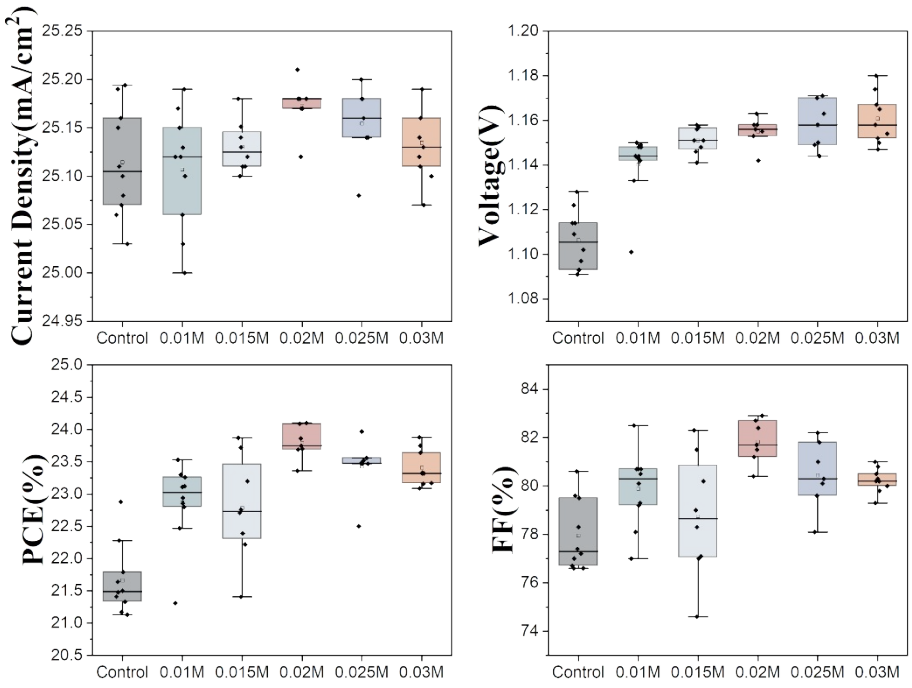

Figure S13. Statistic (a) Jsc, (b) Voc, (c) FF, and (d) PCE of PSCs based on SnO_2 modified by different concentrations of PL-Glu.

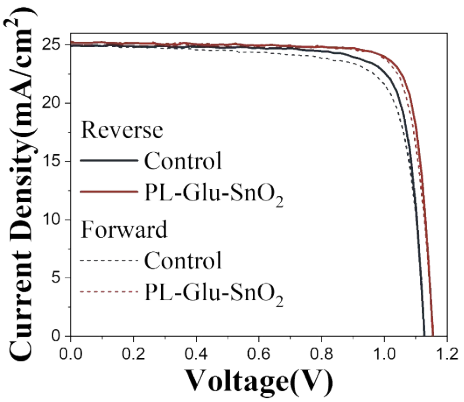

Figure S14. J-V curves under reverse and forward scanning directions.

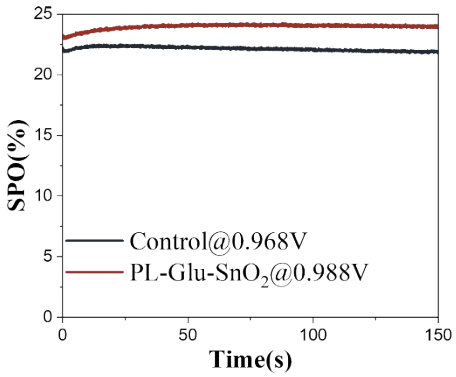

Figure S15. Steady-state photocurrent for control and PL-Glu-SnO_2 devices at the maximum power point under AM 1.5G one sun illumination, where the voltage is 0.968 and 0.988 V, respectively.