

Functionalized Polymer Modified Buried Interface for Enhanced Efficiency and Stability of Perovskite Solar Cells

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Experimental Section

Materials and solvents

SnO₂ colloidal solution (15 wt% in water) was brought from Alfa Aesar. Lead (II) bromide (PbBr₂, 99.9%), Formamidinium hydroiodide (FAI, 99.9%), bis(trifluoromethane) sulfonimide lithium salt (Li-TFSI, 99%), 2,2',7,7'-Tetrakis[N,N-di(4-methoxyphenyl)amino]-9,9'-spirobifluorene (Spiro-OMeTAD, 99.86%) and 4-*tert*-butyl pyridine (*t*BP, 99%) were purchased from Advanced Election Technology CO., Ltd. Lead (II) iodide (PbI₂, 99.99%), Methylammonium bromide (MABr, 99.9%), lead (II) chloride (PbCl₂, 99.99%) and methylamine hydrochloride (MACl, 99.5%) were purchased from Xi'an Polymer Light Technology Corp. Cesium iodide (CsI, 99.99%) and Rubidium iodide (RbI) were obtained from Aladdin. Povidone-iodine (PV-I) was got from Macklin. Chlorobenzene (CB, >99.9% purity), N, N-dimethylformamide (DMF, 99.8%) and dimethyl sulfoxide (DMSO, 99.8%) were purchased from Sigma-Aldrich. All the chemicals were used without further purification.

Device Fabrication:

Laser patterned ITO glasses (7~9 Ω per square) were ultrasonically cleaned with detergent water and ethanol for 20 min in sequence. After being blown dry by nitrogen (99.99%), the ITO was treated by ultraviolet ozone (UV-O₃) for 20 min. Then, the SnO₂ colloidal solution was prepared by mixing the SnO₂ solution and deionized water at a volume rate of 1/3. Diluted SnO₂ colloidal solution was spin-coated on the ITO substrates at 3000 rpm for 30 s and then the SnO₂ film was annealed at 150 °C for 30 min. After cooling down to room temperature and then treated by UV-O₃ for 20 min. For PV-I treated, different concentrations of PV-I (0, 0.2, 0.5, and 1 mg/mL) were dissolving in DMSO, and then 30 μL of PV-I was spin-coated onto the SnO₂ film at a speed of 5000 rpm for 30s without further annealing. Then, all substrates were transferred to an argon-filled glovebox for perovskite deposition. The 1.55 M perovskite (Rb_{0.02}(FA_{0.95}Cs_{0.05})_{0.98}PbI_{2.91}Br_{0.03}Cl_{0.06}) precursor solution was prepared by dissolving FAI of 248.16 mg, CsI of 19.73 mg, RbI of 6.58 mg, PbI₂ of 682.73 mg, and PbBr₂ of 8.53 mg PbCl₂ of 12.74 mg, and MACl (additive) of 35 mg in the mixed solvents of DMF and DMSO ($V_{\text{DMF}} : V_{\text{DMSO}} = 4 : 1$). The as-prepared perovskite precursor solution was filter by the 0.22 μm PTFE filter before use. The perovskite film was deposited by the consecutively spin-coating process at 4000 rpm for 30 s where 80 μL CB antisolvent was dripped on perovskite films at 16 s before

ending the program, and the film was then annealed at 130 °C for 30 min. The Spiro-OMeTAD solution was prepared by mixing 72.3 mg Spiro-OMeTAD, 28.8 μL of 4-*tert*-butyl pyridine (*t*BP) and 17.5 μL of lithium bis(trifluoromethane sulfonyl)imide (Li-TFSI) stock solution (520 mg Li-TSFI in 1 mL acetonitrile) in 1 mL CB. Subsequently, 20 μL Spiro-OMeTAD solution was spin-coated onto the perovskite films at 4000 rpm for 33 s to form the hole transport layer. Finally, about 100 nm metal counter electrode was thermally evaporated on the top of Spiro-OMeTAD film under a vacuum of 3×10^{-5} Pa through using a shadow mask.

Characterization:

J-V curves were obtained using a solar simulator equipped and a Keithley 2400 source meter, and the black metal mask was employed to define the effective active area of the device to be 0.1 cm². *J-V* curves were measured from -0.01 V to 1.15 V (forward scan) or from 1.15 V to -0.01 V (reverse scan) with a scan rate of 110 mV/s. The PCE of the PSCs was tested under air and dark conditions. XRD, Polar diagram and grazing incidence X-ray diffraction (GIXRD) patterns were collected using a PANalytical Empyrean diffractometer equipped with a Cu K α radiation ($\lambda = 1.54056 \text{ \AA}$). XPS measurements were performed on a Thermo-Fisher ESCALab 250Xi system with a monochromatized Al K α under the pressure of 5.0×10^{-7} Pa. Fourier transformed infrared (FTIR) spectra were obtained on an FTIR spectrometer (Thermo Fisher Scientific). UV-Vis spectra were measured on an Agilent 8453 UV-Vis G1103A spectrometer. PL and TRPL were recorded by Edinburgh FLS1000, where the excitation wavelength of TRPL was provided by a 450 nm laser. SEM images were performed on SEM (JXA-8530F Plus).

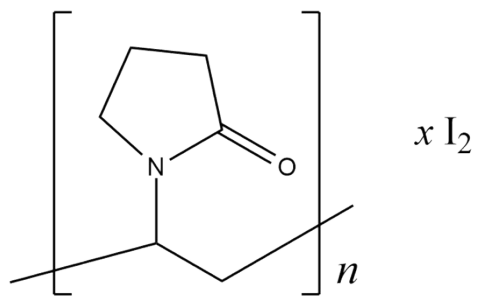


Figure S1 The structures of the PV-I.

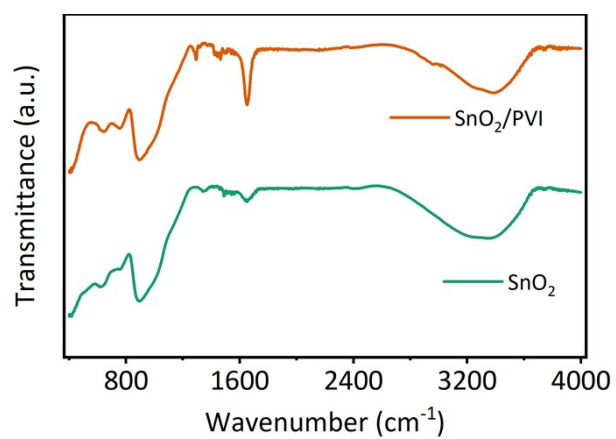


Figure S2 FTIR spectra of the SnO₂ without or with PV-I modification.

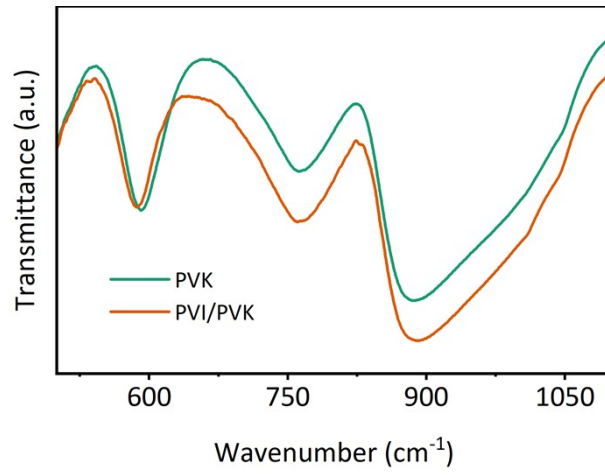


Figure S3 FTIR spectra of the pure perovskite and perovskite modified by PV-I.

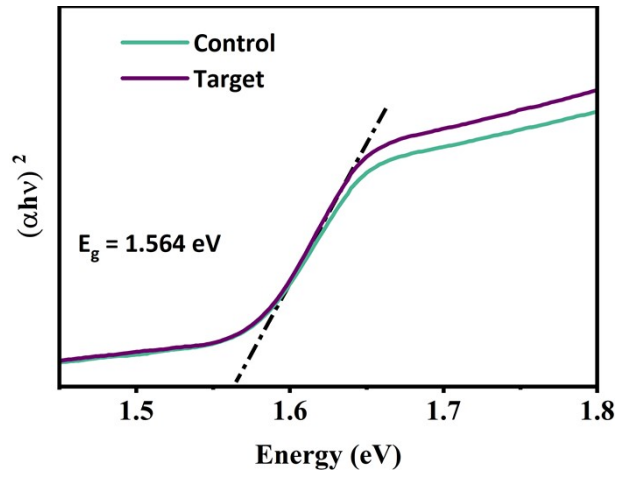


Figure S4 Tauc plots of the perovskite film with or without PV-I modification.

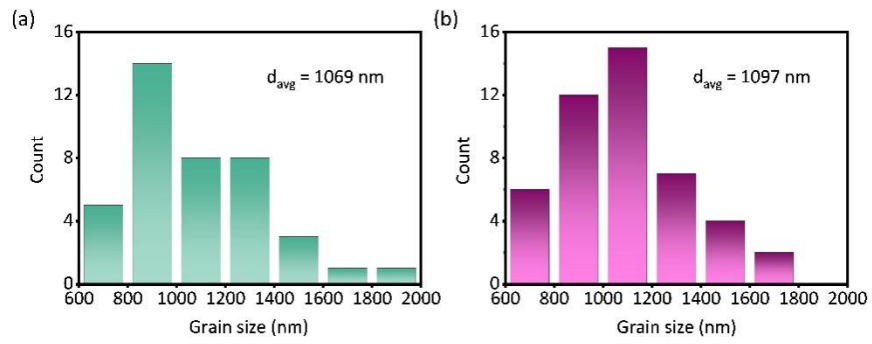


Figure S5 Grain size distribution statistics with the perovskite film (a) without and (b) with PV-I modification.

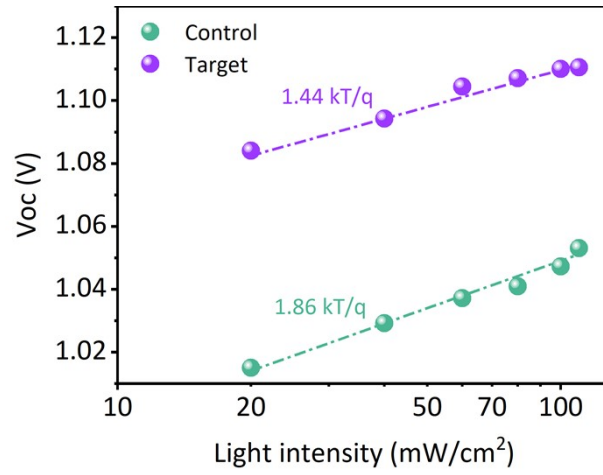


Figure S6 Light-intensity-dependent V_{OC} of the control and target devices.

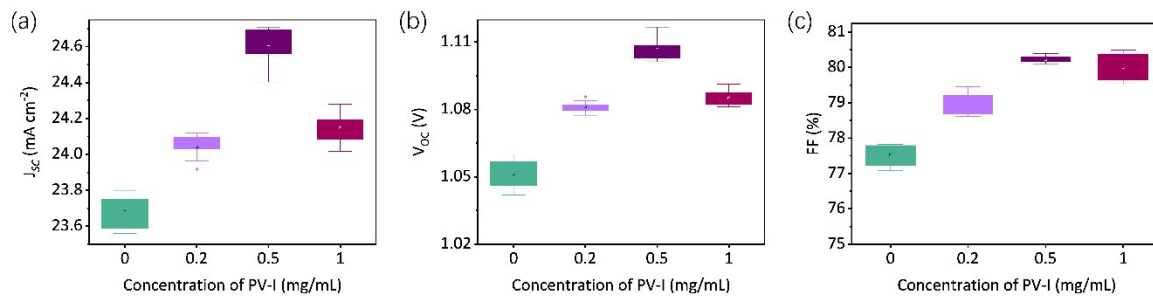


Figure S7. (a) J_{SC} , (b) V_{OC} , and (c) FF statistical diagrams of the devices modified by different concentrations of PV-I. The statistical data were collected from 20 cells for each concentration.

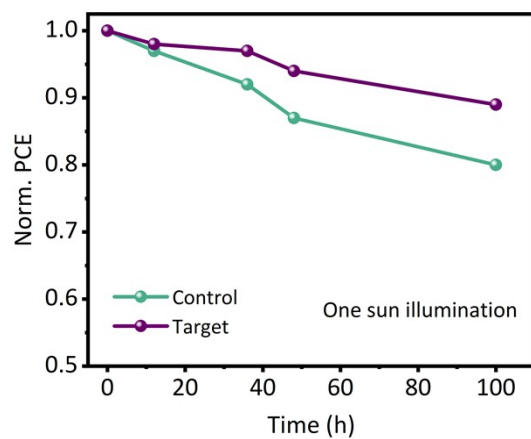


Figure S8 PCE evolution of the devices based on SnO₂ without and with PV-I modification aged under one sun illumination where the devices were located in the glovebox.

Table S1 Fitting results from TRPL kinetics in Figure 4b.

	Glass/PVK	Glass/PV-I/PVK
τ_1 (ns)	134.90	30.59
%	28.7	12.0
τ_2 (ns)	447.56	911.45
%	71.2	87.9
τ_{ave} (ns)	413.7	907.4

Table S2 Fitting results from TRPL dynamics in Figure 4d.

	ITO/SnO ₂ /PVK	ITO/SnO ₂ /PV-I/PVK
τ_1 (ns)	66.87	12.13
%	79.4	77.7
τ_2 (ns)	264.29	144.91
%	20.6	22.3
τ_{ave} (ns)	166.9	114.7