

Adjusting energy level alignment between HTL and CsPbI₂Br to improve solar cell efficiency

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SUPPORTING INFORMATION

Experimental section

Device fabrication

The ITO substrates (15 Ω sq⁻¹) were washed by detergent, and then cleaned with deionized water, acetone and isopropanol for 15 min. each, followed by a UV-ozone treatment for 15 min. The SnO₂ nanoparticle solution (Alfa Aesar) was diluted to 2.5% with deionized water, and then spin-coated onto the ITO glasses at a speed of 3000 rpm for 30 s. The SnO₂/ITO samples were annealed at 150 °C for 30 min. After cooling down to room temperature, these samples were further treated with UV-ozone for 10 min, then transferred to a nitrogen-filled glovebox. The CsPbI₂Br solution was prepared by dissolving CsI (0.8 mmol), PbI₂ (0.4 mmol), and PbBr₂ (0.4 mmol) in 1 mL DMF/DMSO mixed solvent (4 : 1, v/v). The solution was heated at 60 °C for 4 h. After filtering with a 0.22 μm PTFE filter, the solution was spin-coated onto the SnO₂/ITO substrates at 2700 rpm for 30 s, and annealed at 250 °C for 10 min. PVK and PTAA were dissolved in chlorobenzene with the same concentration of 10 mg mL⁻¹. A certain amount of PVK solution was added into the PTAA solution to prepare PTAA films with 0%, 5%, 7.5%, and 10% of PVK. After stirring overnight, the PTAA: x% PVK mixture solution was spin-coated onto perovskite layer and annealed at 100 °C for 10 min. Finally, MoO₃ (~6 nm) and Ag (~80 nm) were sequentially evaporated onto the HTL layer in a thermal evaporator (Pudi Vacuum). The effective area is 8 mm².

Characterization

For the current density–voltage (*J*–*V*) characterizations, a Newport xenon 300 W lamp was used to provide a light intensity of 100 mW cm⁻², which was calibrated with a certified silicon photodiode (Hamamatsu). *J*–*V* curves were characterized by using a Keithley 2450 source meter. A QE-R solar cell quantum efficiency measurement system developed by Enli Technology was employed to obtain external quantum effi-

ciency spectra at a given frequency of 210 Hz. A Bruker D8 ADVANCE X-ray diffractometer was used to acquire X-ray diffraction (XRD) patterns with a Cu-Kα radiation source (λ = 1.5418 Å). A double beam electron microscope (FEI Helios NanoLab G3 UC) was used to conduct morphological imaging characterization at 2 kV. Thermo scientific Evolution 201 UV–visible spectrophotometer was employed to measure UV–vis absorption spectra. The electrochemical workstation (Ivium Vertex, Netherland) was used to obtain Nyquist plots at 0.7 V in dark. Helium Iα excitation source of 21.22 eV at 5 V was applied to obtain ultraviolet photoelectron spectroscopy with spectra Photoelectron Spectrometer (SPCES-Lab). The transient photocurrent (TPC) spectrum excited by nitrogen laser (337 nm, n100, Stanford) was recorded by digital oscilloscope (dso-x3104a, Agilent). The impedance is 50 Ω.

To further verify the charge recombination loss in our devices, the electrochemical impedance spectroscopy (EIS) measurements were conducted to unveil the charge recombination loss in the bulk of CsPbI₂Br layer and the charge transfer properties at the PTAA/CsPbI₂Br interface. Fig. S9 shows Nyquist plots acquired from the devices based on the PTAA-only and PTAA: 7.5% PVK layers. The equivalent circuit mod-

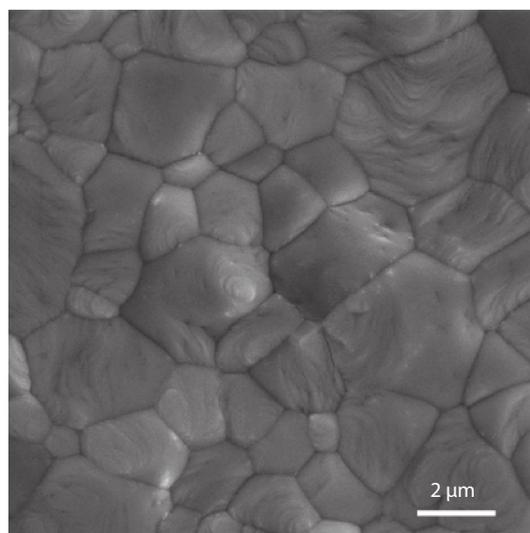


Fig. S1. SEM image of CsPbI₂Br film.

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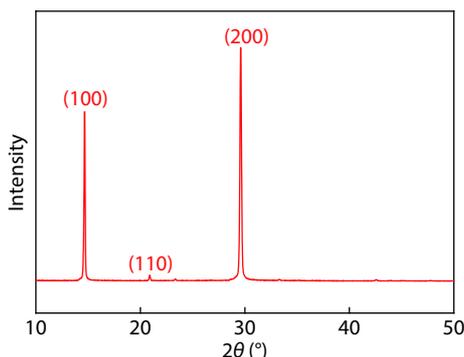
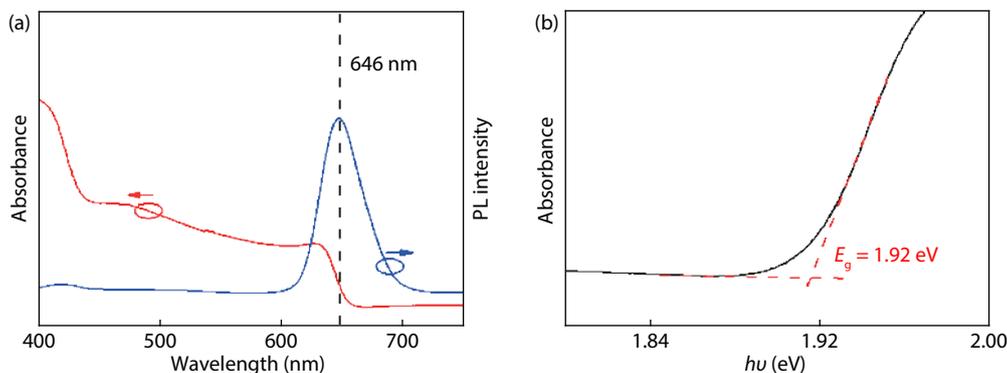
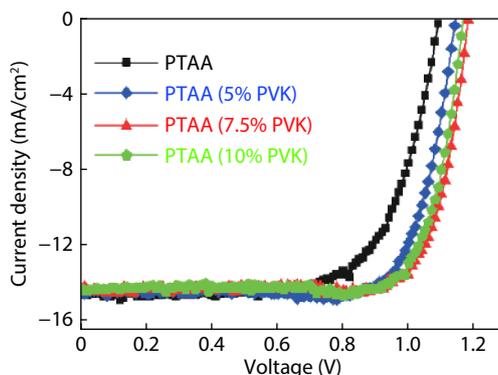
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Table 1. Device parameters from Fig. S4.

HTL	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
PTAA	14.4	1.10	70	11.0
PTAA (5% PVK)	14.1	1.18	77	12.8
PTAA (7.5% PVK)	14.5	1.19	79	13.6
PTAA (10% PVK)	14.4	1.18	78	13.2

Table 2. Fitting results of Nyquist plots.

HTL	R_s (Ω)	R_{sc} (Ω)	R_{rec} (k Ω)
PTAA	30	624	1.312
PTAA (7.5% PVK)	22	282	3.491

Fig. S2. XRD pattern for CsPbI₂Br film.Fig. S3. (a) UV-visible absorption and PL spectra for CsPbI₂Br film. (b) Estimation of optical bandgap.Fig. S4. J - V curves for CsPbI₂Br solar cells with PTAA HTL with different PVK content.

el as illustrated in the inset of Fig. 5(c) was employed to fit the Nyquist plots, which contains two primary interfaces with a selective contact resistance (R_{sc}) and a charge recombina-

tion resistance (R_{rec})^[1]. The fit results are listed in Table S2. It is shown that as compared to the PTAA-only device the R_{sc} was reduced from 624 to 282 Ω while the R_{rec} increased from

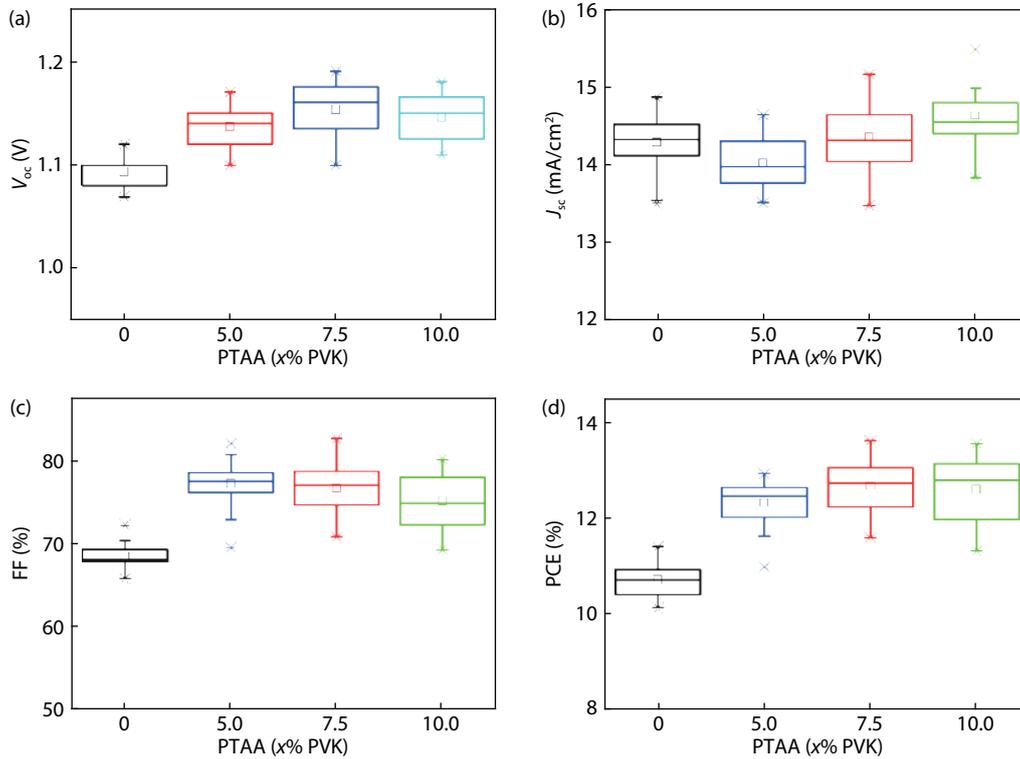


Fig. S5. Device performance change with PVK content.

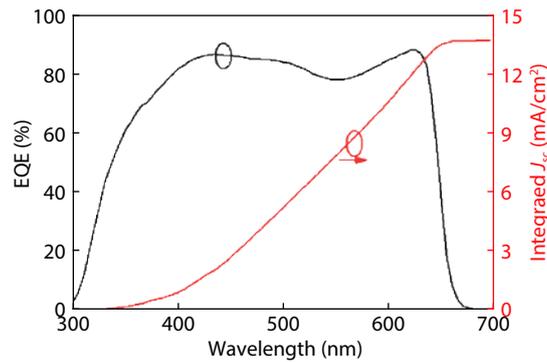
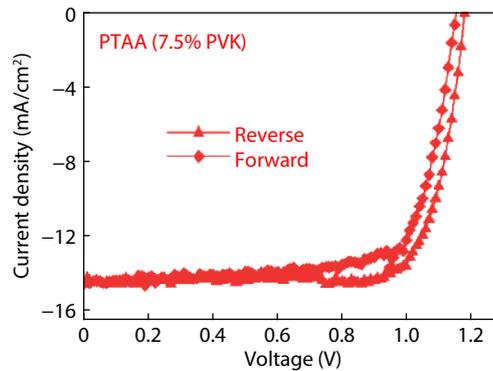


Fig. S6. The EQE spectrum and integrated current density for the solar cell with PTAA (7.5% PVK) HTL.

Fig. S7. J - V curves for the solar cell with PTAA (7.5% PVK) HTL.

1.312 k Ω to 3.491 k Ω with mixing 7.5% PVK into the PTAA layer. Such observation firmly proved that the electron-hole recombination loss was reduced in the CsPbI₂Br perovskite layer and the charge transfer efficiency was improved at the PTAA/CsPbI₂Br interface.

References

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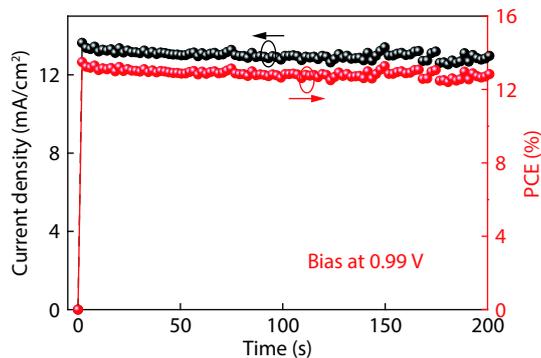


Fig. S8. Steady-state current and stabilized PCE at 0.99 V bias.

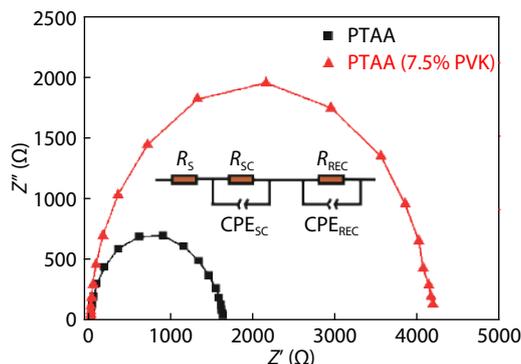


Fig. S9. Electrochemical impedance spectra (EIS) acquired from PTAA device and PTAA (7.5% PVK) device.

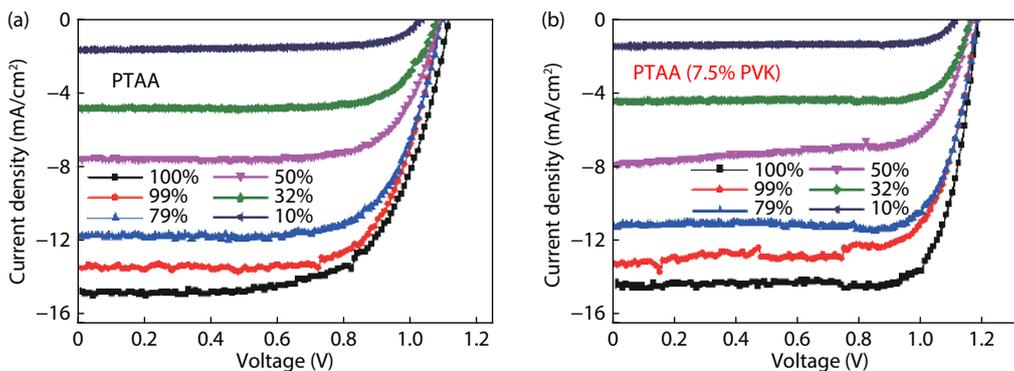


Fig. S10. J - V curves obtained under different light intensity for devices with (a) PTAA and (b) PTAA (7.5% PVK).

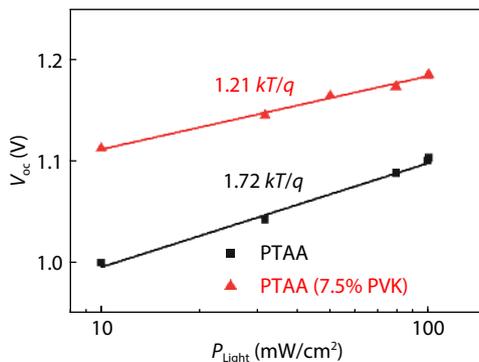


Fig. S11. V_{oc} vs light intensity plots for PTAA device and PTAA (7.5% PVK) device.

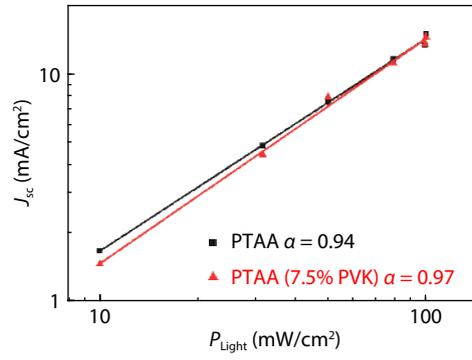


Fig. S12. J_{sc} vs light intensity plots for PTAA device and PTAA (7.5% PVK) device.