

Supporting information

Highly Efficient bifacial CsPbIBr₂ solar cells with TeO₂/Ag transparent electrode and unsymmetrical carrier transport behavior

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1. Experimental details

1.1 Materials and Reagents

Ultradry PbBr_2 (99.999%) and CsI (99.998%) and anhydrous DMSO (99.9+%, ACS reagent) are purchased from Alfa Aesar. FTO glasses (Pilkington, TEC-8, $8\Omega/\text{sq}$) are received from Yingkou OPV Tech New Energy Co., Ltd., China. All of them are used as received without further purification and modifications.

1.2 Preparation of c-TiO₂/FTO Substrate

FTO substrate was cleaned by sequential sonication in detergent, deionized water, acetone, and alcohol for 1 h. After being dried by N_2 flow, it was further treated by ultraviolet ozone cleaner for 15 min. After that, a ≈ 60 nm c-TiO₂ ETL can be coated on the cleaned FTO substrate by spin-coating a TiO₂ sol at 3500 rpm for 30 s with immediate thermal annealing at 480°C for 60 min in air.

1.3 Preparation of CsPbIBr₂ Film

1.0M CsPbIBr_2 precursor solution was prepared by fully dissolving 260 mg CsI and 367 mg PbBr_2 in 1 mL DMSO at room temperature. CsPbIBr_2 precursor film can be obtained by spin-coating precursor solution on c-TiO₂/FTO substrate at 1500 rpm for 20 s and 5000 rpm for 60 s. As an optimized device, 50 μL methanol solution containing 30 mg/mL CsI is spin-coated on CsPbIBr_2 precursor film at 5000 rpm for 20 s. After being washed with IPA, it was annealed at 280°C for 10min.

Thus, the CsPbIBr₂ film was obtained and labeled as intermolecular exchange [1]. For comparison, the one as control device without CsI treatment. The film was obtained by directly annealing the CsPbIBr₂ precursor film under 280°C for 10 min.

1.4 Fabrication of Solar Cells

The 75uL 90 mg/ml spiro-OMeTAD CB solution prepared in advance was spin-coated on the perovskite film at a speed of 1000 rpm for 5 s and 3000 rpm for 45 s, and allowed to stand for two or three minutes. Place in a dry environment and wait for spiro-OMeTAD to be oxidized for one day, then deposit an 11 nm ultra-thin Ag electrode in the thermal evaporation system, next a 40 nm TeO₂ film was coated as an optical modifying layer, and finally evaporate a 100 nm Ag film to act as the contact electrodes. The thicknesses of Ag and TeO₂ were estimated in situ with a calibrated quartz crystal monitor. And this gives the bifacial semitransparent CsPbIBr₂ perovskite solar cells with an active area of 7 mm². At the same time, the cells of Glass/FTO/TiO₂/CsPbIBr₂/ spiro-OMeTAD/Ag(100 nm) and Glass/FTO/TiO₂/CsPbIBr₂/Carbon were prepared as the reference devices. Here, the carbon electrode was deposited by silkscreen-printing conductive carbon paste, followed by heating at 100 °C for 10 min in ambient air to promote the evaporation of residual solvent in printed Carbon layer.

1.5 Device characterization

The photovoltaic performances of bifacial PSCs were measured by the Keithley 2400 source meter under the simulated AM 1.5G (One sun) illumination of 1000 W/m^2 from a sunlight simulator (Sanei Electric, XES-300T1). The illumination intensity was calibrated by a reference silicon cell certificated by the National Renewable Energy Laboratory. The external quantum efficiency of PSCs was measured by the quantum efficiency measurement system (SCS10-X150, Zolix instrument. Co. Ltd). The transmittance spectra were measured by the UV-visible spectrophotometer (Perkin-Elmer Lambda 950). The film morphology were characterized by a JSM-7800F extreme-resolution analytical field emission scanning electron microscope (SEM), the crystal quality were assessed by the X-ray diffractometer (D8 Advance, Bruker, Germany) and steady photoluminescence (PL) spectra recorded by a Fluorescence Spectrophotometer (Hitachi F-7000) with a 460 nm excitation. Transient photovoltage (TPV) measurement was performed with the same system excited by a 405 nm (50 Hz, 20ms) pulse laser. A digital oscilloscope (Tektronix, D4105, Beaverton, OR, USA) was used to record the photocurrent or photo-voltage decay process with a sampling resistor of 50Ω or $1\text{M}\Omega$, respectively. EIS measurements were performed on an electrochemical workstation (CHI600E, Shanghai Chenhua) with a 10 mV amplitude perturbation and frequencies between 100Hz and 1MHz. Nyquist plots were recorded on the same system under AC excitation

amplitude of 30 mV at a frequency of 5kHz. All the measurements were performed at room temperature under ambient atmosphere.

2. Experimental results

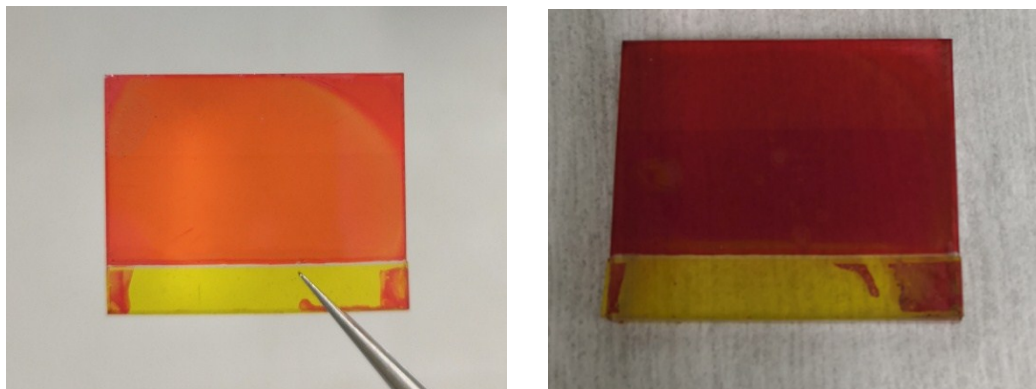


Figure S1 Photos of control (left) and CsI treated (right) CsPbIBr₂ films.

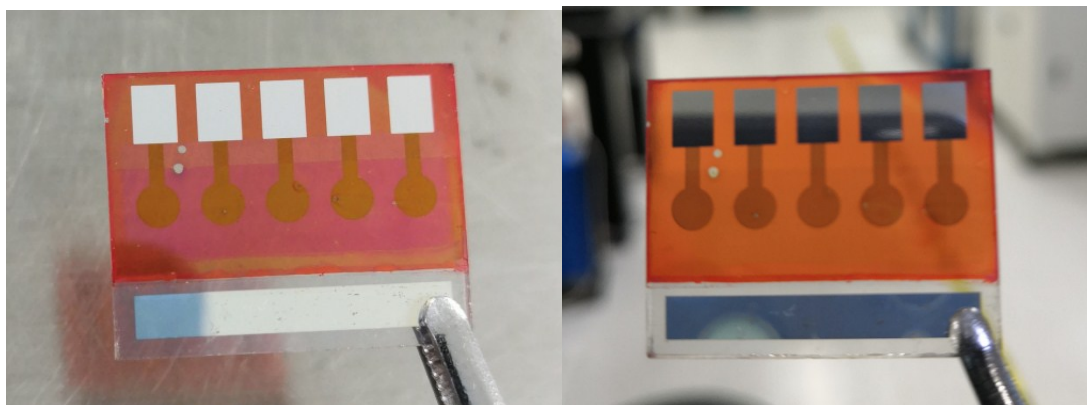


Figure S2 Photos of bifacial CsPbIBr₂ perovskite solar cells taken from the TeO₂/Ag side (left) and FTO side (right).

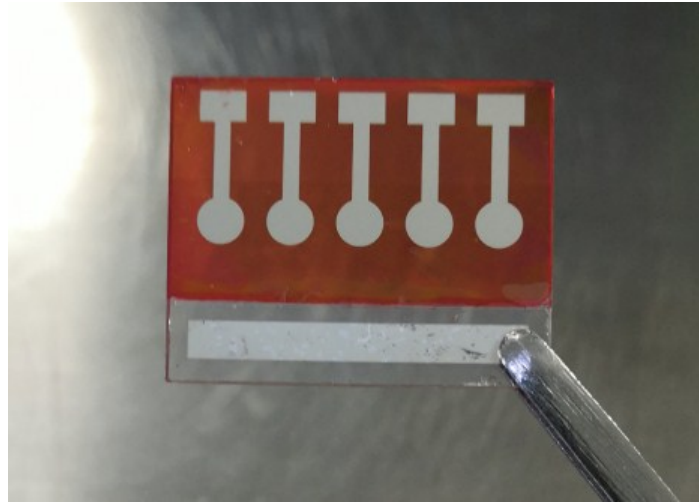


Figure S3 Photos of reference cell with 100 nm opaque Ag electrode, a structure of Glass/FTO/TiO₂/CsPbIBr₂/spiro-OMeTAD/Ag(100 nm).

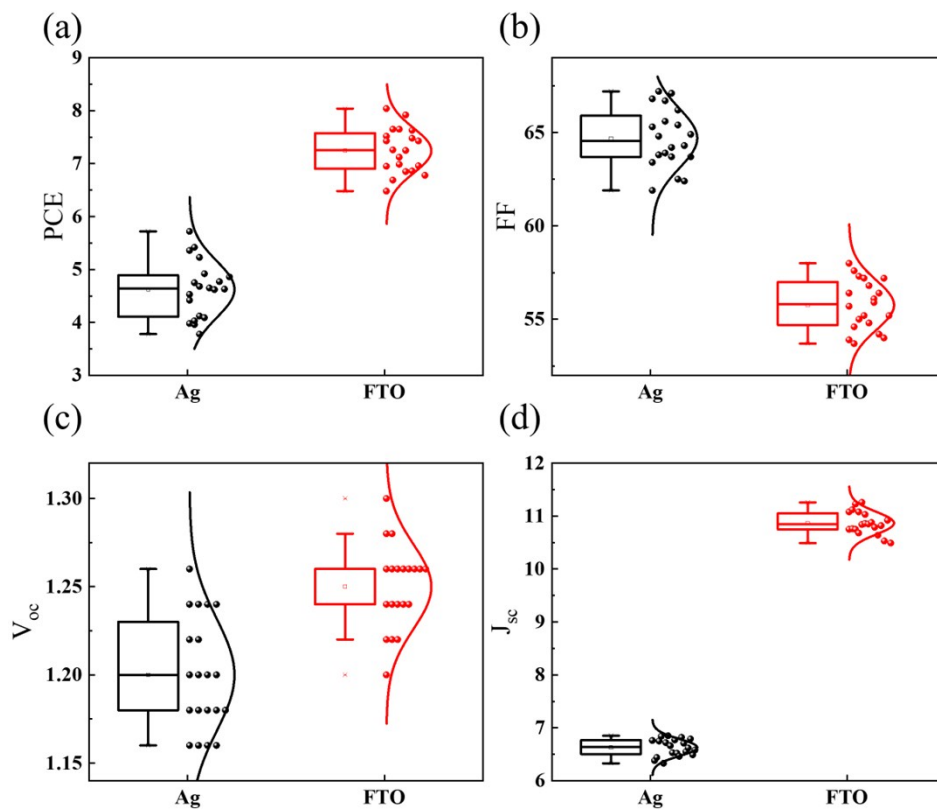


Figure S4 Statistical PCE, Voc, FF, Jsc of bifacial control CsPbIBr₂ PSCs with Ag/TeO₂ electrodes, which corresponding to the results of Fig. 3.

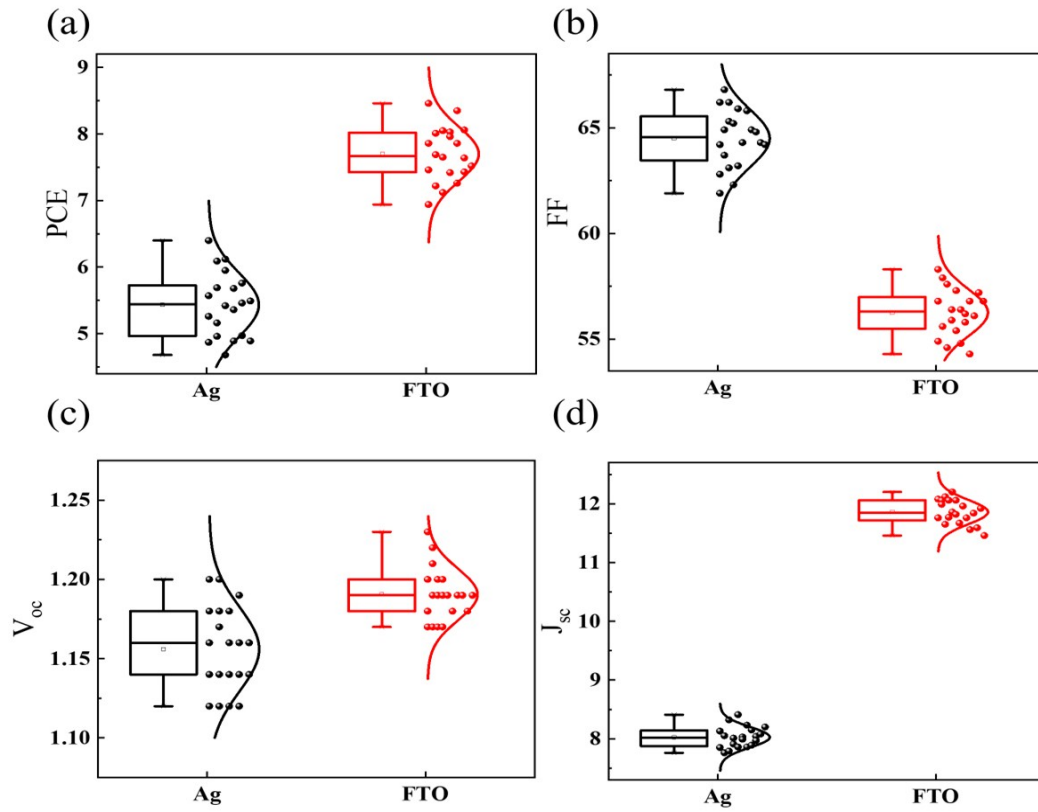
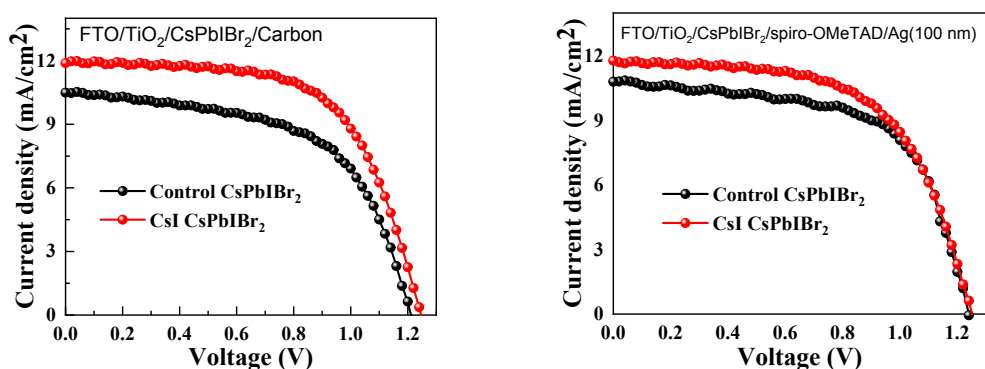


Figure S5 Statistical PCE, V_{oc} , FF, J_{sc} of bifacial CsI-CsPbIBr₂ PSCs with Ag/TeO₂ electrodes, which corresponding to the results of Fig. 3.



Reference cells	PCE (%)	FF (%)	V _{OC} (V)	J _{SC} (mA/cm ²)
control- Ag(100 nm)	8.30	62.61	1.22	10.86
CsI- Ag(100 nm)	8.81	60.29	1.24	11.78
control-Carbon	8.35	63.16	1.20	11.03
CsI-Carbon	9.22	62.09	1.24	11.98

Figure S6 JV curves and photovoltaic parameters of reference CsPbIBr₂ PSCs based on opaque Ag(100 nm) and carbon electrodes.

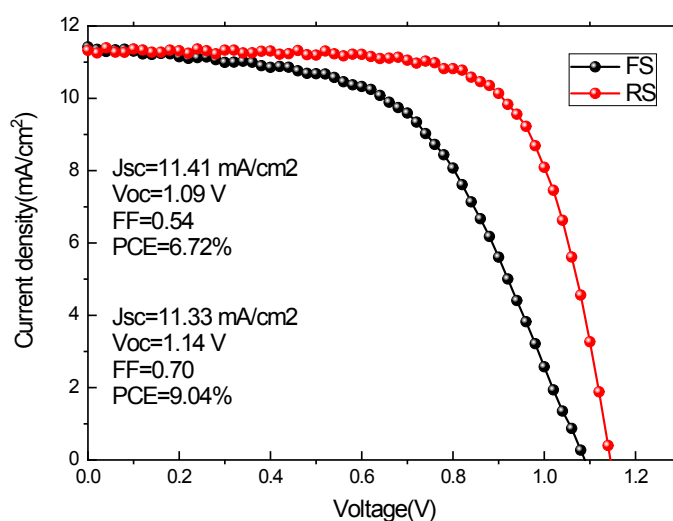


Fig. S7 (a) JV curves for control CsPbIBr₂ PSC under simulated AM 1.5 G illumination with forward scan (FS) and Reverse scan (RS).

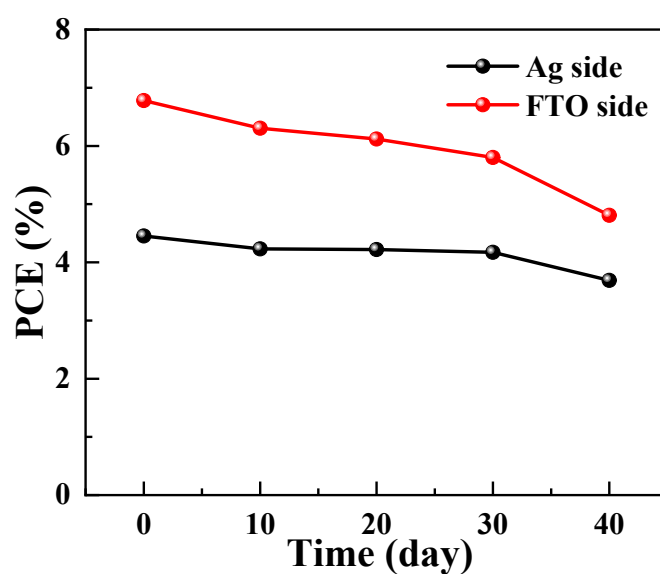


Fig. S8 Time-dependent PCEs for the un-encapsulated champion cells storing in a glovebox.

[1] Zhu, W.; Zhang, Q.; Chen, D.; Zhang, Z.; Lin, Z.; Chang, J.; Zhang, J.; Zhang, C.; Hao, Y. Intermolecular Exchange Boosts Efficiency of Air-Stable, Carbon-Based All-Inorganic Planar CsPbIBr₂ Perovskite Solar Cells to Over 9%. *Adv. Energy Mater.* 2018, 8, 1802080.