Supporting Information

Low-temperature Interfacial Engineering for Flexible $CsPbI_2Br$ Perovskite Solar Cells with High Performance Beyond 15%

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Experimental Sections

Materials: Zinc oxide (ZnO) and the sol-gel-derived Aluminum-doped zinc oxide (AZO) precursor solution were prepared according to the literature. [1, 2] High-conductivity PEDOT:PSS (hc-PEDOTPSS) was obtained by following previously reported process. [3] Ethanol (EtOH, anhydrous, ≥99.9%), dimethyl sulfoxide (DMSO, anhydrous, ≥99.9%), chlorobenzene (CB) were purchased from Sigma-Aldrich and used as received. Tin (IV) oxide (SnO_2, 15% in H_2O colloidal dispersion), cesium iodide (CsI, 99.999%), tert-butyl cyanoacetate (t-BCA, 98%), lead bromide ($PbBr_2$, 99.999%) were obtained from Alfa Aesar and used as received. Poly[bis(4-phenyl) (2,4,6-trimethylphenyl) amine] (PTAA, ≥99.5%) was purchased from Xi'an Polymer Light Technology Corp and used as received. Lead iodide (PbI_2, 99.999%) was obtained from TCI and used as received.

Perovskite Solar Cell fabrication and characterization: PET substrates were cleaned by sequential ultrasonic treatment in detergent, deionized water, acetone and isopropyl alcohol, and then dried with a nitrogen stream. After 3 minutes of UV-ozone treatments, hc-PEDOT:PSS was spun coated on PET with 2000 rpm for 40 s, followed by thermal annealing at 120 ^oC for 15 min. The AZO solution was spun onto hc-PEDOT:PSS substrates for one time (2000 rpm, 60 s), after thermal annealing at 120 ^oC for 20 min in air, the substrates were transferred into glove box filled with N_2. 1.2 M CsI, 0.6 M PbI_2 and 0.6 M $PbBr_2$ were dissolved in DMSO, and stirred overnight at 60 ^oC in a glovebox. The fully dissolved perovskite precursor solution was spun coated onto the AZO layer at 2800 rpm for 45 s and then heated at 120 ^oC for 10 min to form perovskite films. After the substrate cooling down to room temperature, t-BCA (1 mg/ml, CB) was spun on the perovskite film at 4500 rpm for 30 s, and thermally annealed by 100 ^oC for 5 min. PTAA (10 mg/mL, CB) was spun coated on passivation layer at

3000 rpm for 30 s. Finally, 6 nm MoO_3 and 80 nm Ag anode were deposited by thermal evaporation using a metal shadow mask. The device area was 0.1 cm^2. All devices' measurements were performed under an ambient atmosphere at room temperature.

Fabrication of Electron-only device: A thin layer of electron transport layer AZO was spin-coated on a flexible electrode at 2000 rpm for 1 min, and subsequently annealed at 120 ^oC for 20 min. 1.2 M CsI, 0.6 M PbI_2 and 0.6 M $PbBr_2$ were dissolved in DMSO, and stirred overnight at 60 ^oC in a glovebox. The fully dissolved perovskite precursor solution was spun coated onto the AZO layer at 2800 rpm for 45 s and then heated at 120 ^oC for 10 min to form perovskite films. After the substrate cooling down to room temperature, t-BCA (1 mg/ml, CB) was spun on the perovskite film at 4500 rpm for 30 s, and thermally annealed by 100 ^oC for 5 min. Subsequently, the $PC_{61}BM$ (20 mg/mL in anhydrous chlorobenzene) and BCP (0.5 mg/mL in anhydrous ethanol) were then sequentially deposited by spin coating at 2000 rpm for 30 s and 4500 rpm for 60 s, respectively. Finally, 80 nm Ag anode were deposited by thermal evaporation through shadow masks.

Fabrication of Hole-only device: A thin layer of hole transport layer PEDOT:PSS CLEVIOSTM Al4083 was spin-coated on a flexible electrode at 4,000 rpm for 1 min, and subsequently annealed at 120 ^oC for 15 min. 1.2 M CsI, 0.6 M PbI_2 and 0.6 M $PbBr_2$ were dissolved in DMSO, and stirred overnight at 60 ^oC in a glovebox. The fully dissolved perovskite precursor solution was spun coated onto the PEDOT:PSS layer at 2800 rpm for 45 s and then heated at 120 ^oC for 10 min to form perovskite films. After the substrate cooling down to room temperature, t-BCA (1 mg/ml, CB) was spun on the perovskite film at 4500 rpm for 30 s, and thermally annealed by 100 ^oC for 5 min. PTAA (10 mg/mL, CB) was spun coated on passivation layer at 3000 rpm for 30 s. Finally, 6 nm MoO_3 and 80 nm Ag anode were deposited by thermal evaporation through shadow masks.

Device Characterization

Scanning electron microscopy (SEM) measurements were performed with an S 4800 and operated at an acceleration voltage of 5 kV. X-ray diffraction patterns (XRD) measured using D/MAX-TTRIII (CBO) with Cu Kα radiation (λ=1.542 Å). UV-vis absorption spectra were recorded on a SHIMADZU, UV-2600 spectrophotometer in the 250–800 nm wavelength range at room temperature. Current density–voltage (*J*–*V*) characteristics were measured using a source meter (Keithley 2400) under 100 mW cm^{-2} simulated AM 1.5 G irradiation from Abet solar simulator, the scan range is from –0.2 V to 1.3 V or from 1.3 V to –0.2 V. The steady-state photoluminescence and time-resolved photoluminescence (TRPL) were obtained using an Edinburgh instrument FLS920 spectrometer. X-ray photoelectron spectroscopy (XPS) measurements were carried out using a thermo-VG scientific ESCALAB 250 photoelectron spectrometer. The EQE was characterized by the QE-R systems (Enli Tech.). Impedance spectroscopy measurements were carried out using an LCR impedance analyzer (Keysight E4990A) under an alternating electric field of 30 mV peak-to-peak value. The repeated bending and stretching cycles were performed by a custom-made stretching machine which was actuated by a stepper motor. The thickness of the AZO was measure using a Dektak XT profilometer (Bruker).

Supporting Figures and Tables

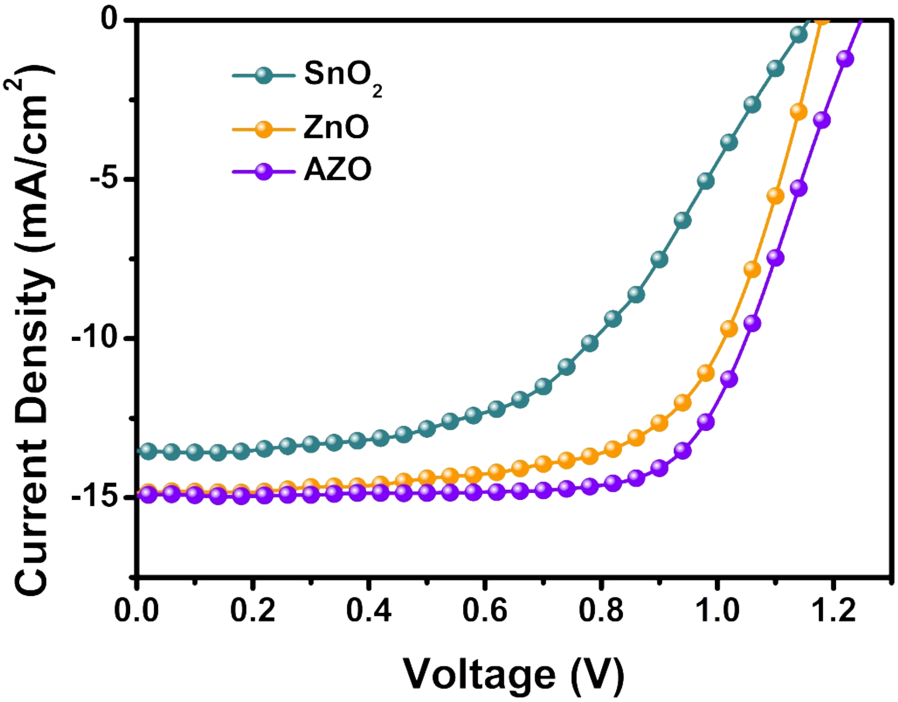

Figure S1. Current density–voltage (*J*–*V*) curves of flexible PSCs with different ETLs under standard AM 1.5 illuminations (100 mW/cm^2).

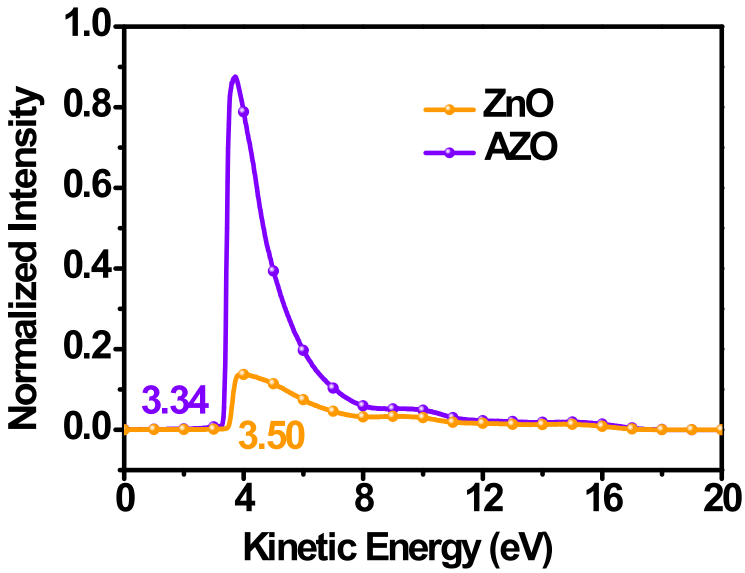

Figure S2. Ultraviolet photoelectron spectroscopy (UPS) spectra of ZnO and AZO.

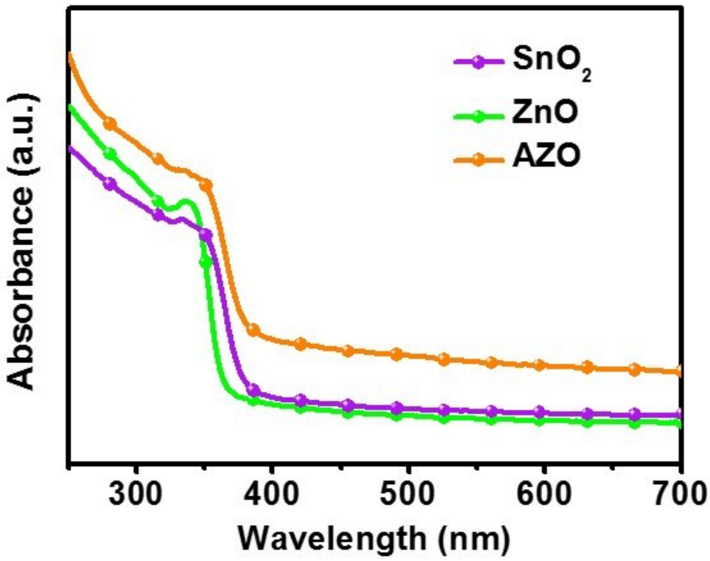

Figure S3. UV-vis absorption spectra of SnO_2, ZnO and AZO films.

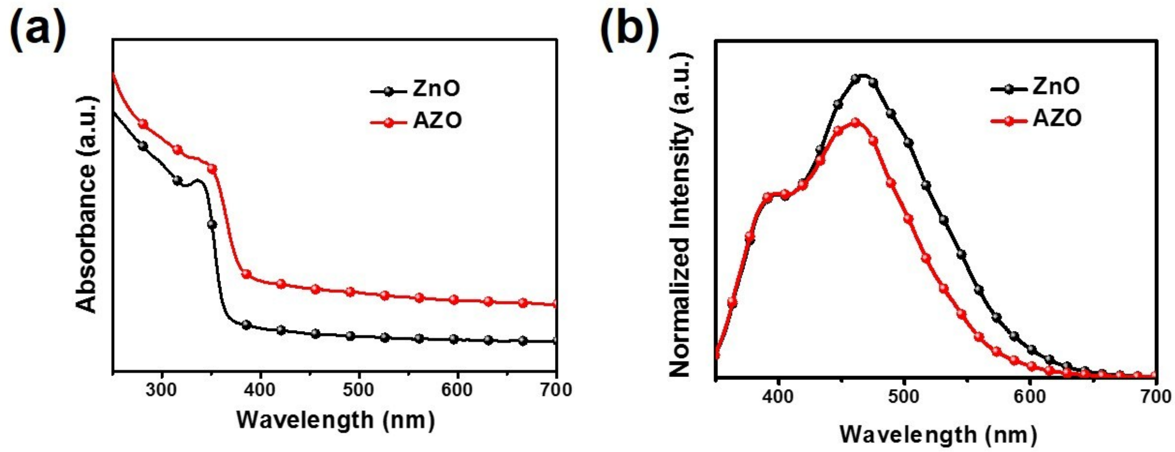

Figure S4. a) UV-vis absorption spectra and b) PL spectra of ZnO and AZO films.

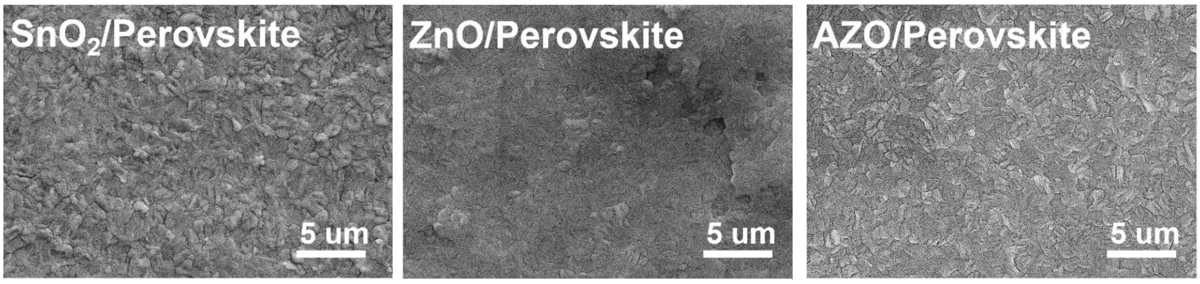

Figure S5. Top-viewed SEM images of Perovskite deposited on SnO, ZnO and AZO films.

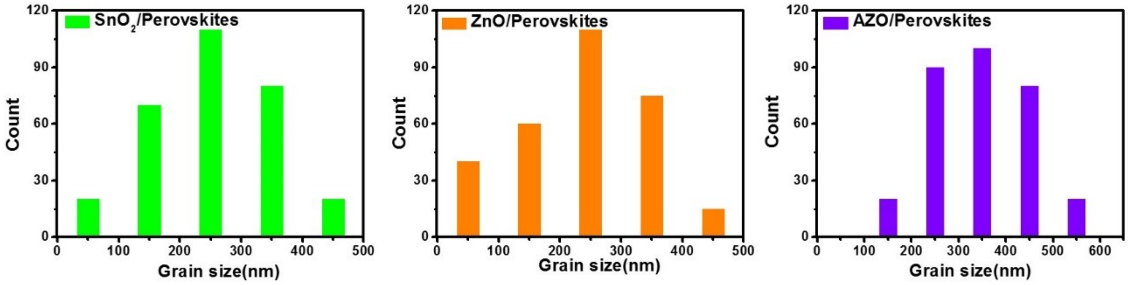

Figure S6. Grain size distributions of $CsPbI_2Br$ flms with different ETLs (at low magnification).

Table S1. Device photovoltaic performance with different ETLs under standard AM 1.5 illuminations (100 mW/cm^2).

ETL	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
SnO_2	1.16	13.53	51.77	8.11
ZnO	1.18	14.85	65.09	11.39
AZO	1.25	14.94	68.37	12.74

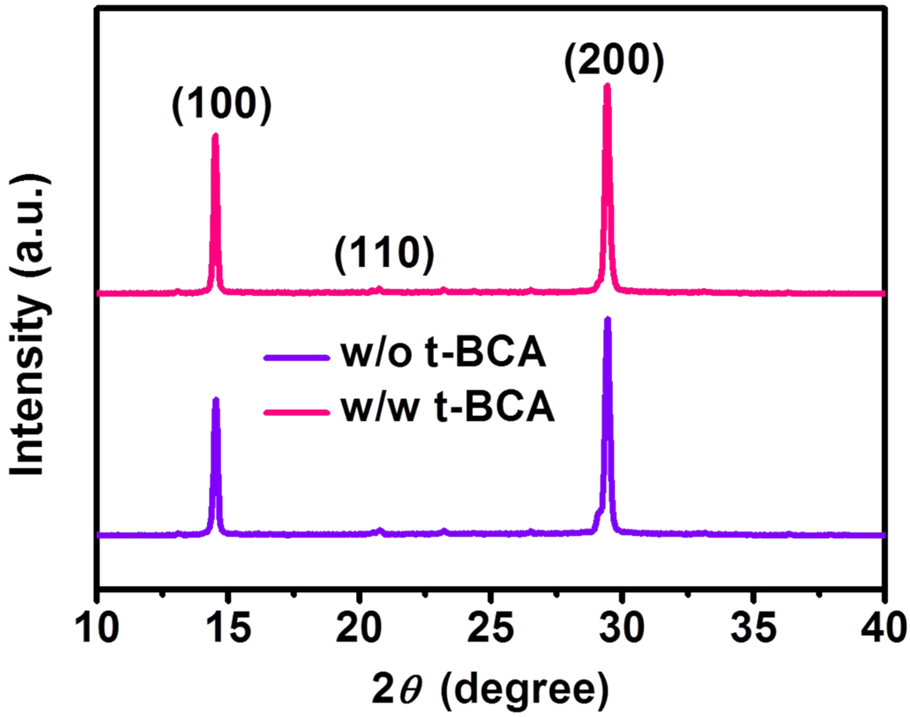

Figure S7. XRD patterns of perovskite films with and without t-BCA.

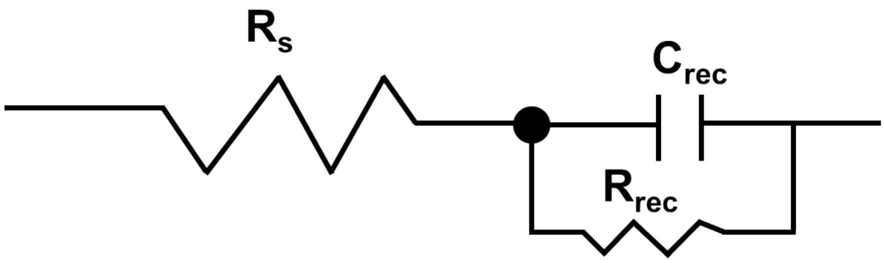

Figure S8. The corresponding circuit model for the solar cells with and without t-BCA layer.

Table S2. Fitting parameters of the EIS measurment with or without t-BCA layer.

	R_s (Ω)	**R_{rec} (Ω)**
w/o t-BCA layer	**32.78**	**101.2**
w/w t-BCA layer	**32.66**	**161.4**

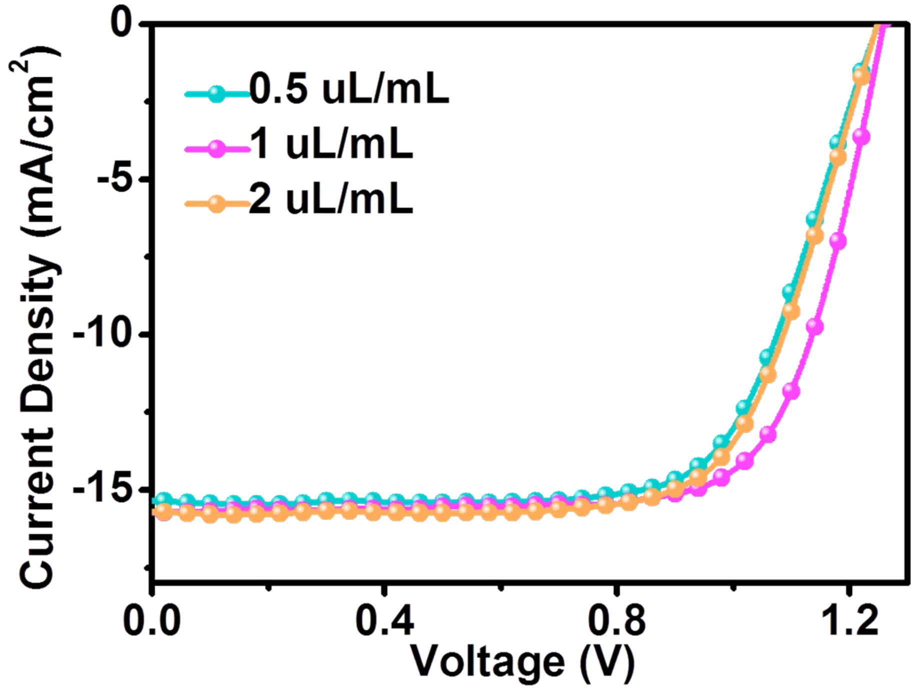

Figure S9. *J*–*V* curves of PSCs with different concentrations of t-BCA coated in perovskite films under standard AM 1.5 illuminations (100 mW/cm^2).

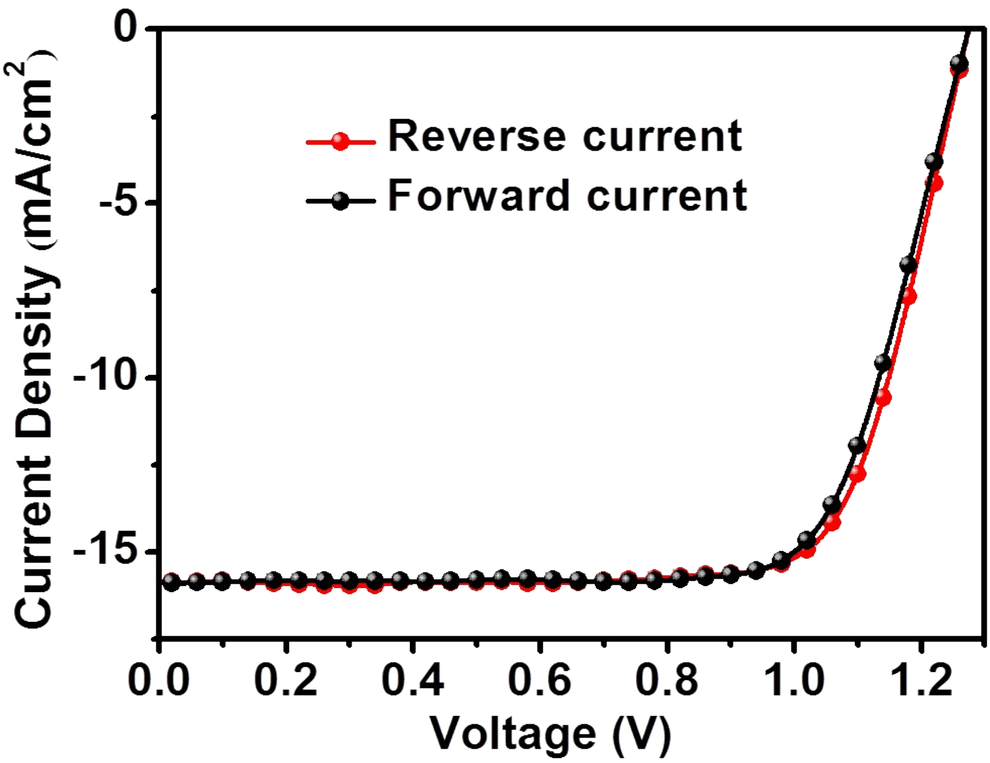

Figure S10. *J–V* curves of PSC measured in forward and reverse scans.

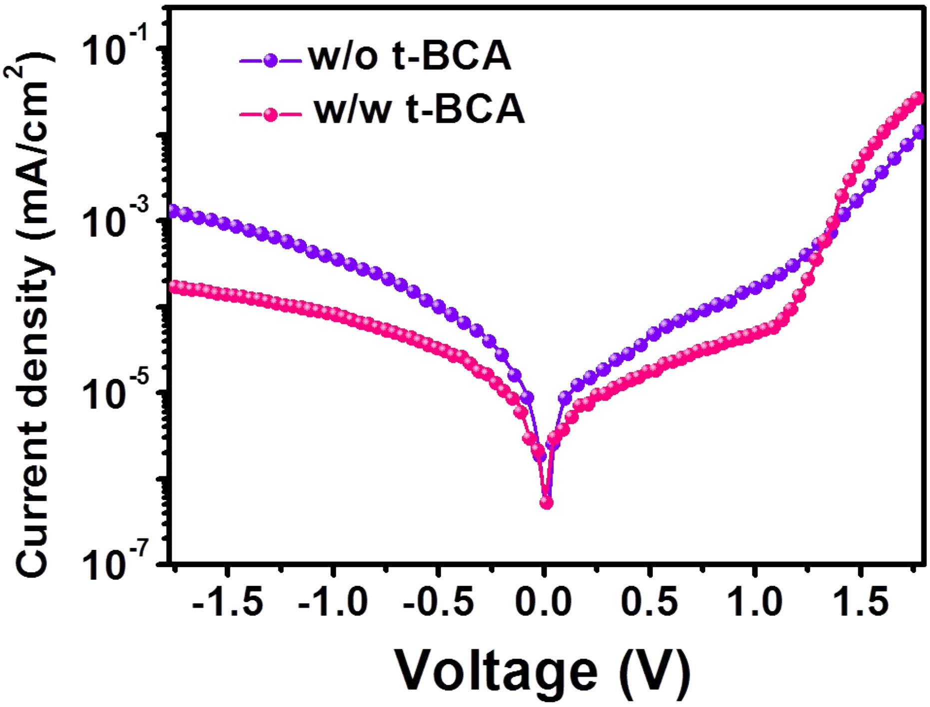

Figure S11. Dark *J*–*V* curves of PSCs with different conditions.

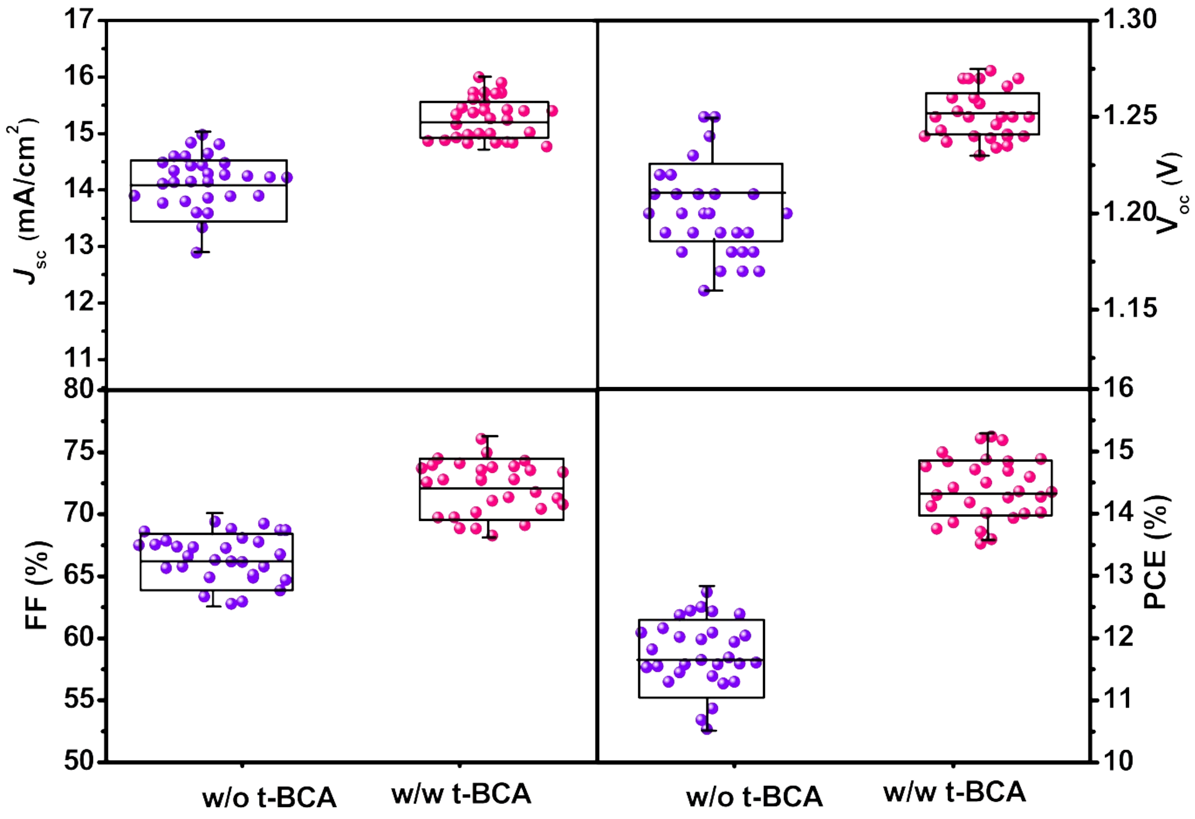

Figure S12. PV performance statistics of the $CsPbI_2Br$ and $CsPbI_2Br$/t-BCA layer based devices (30 devices).

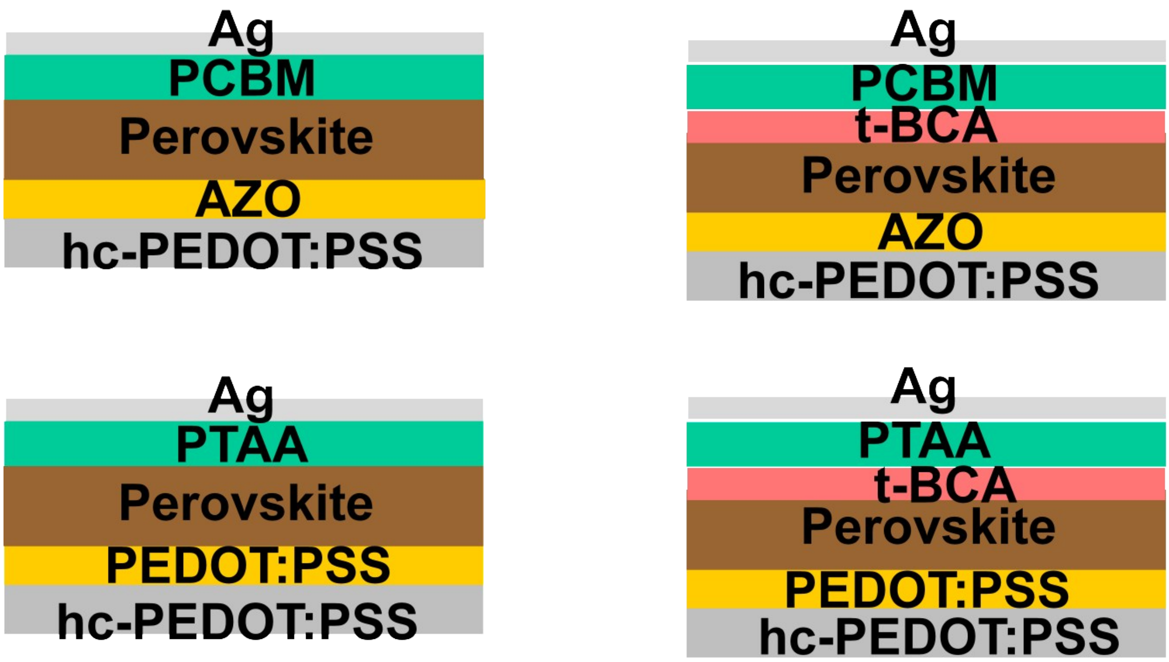

Figure S13. The device structures of the electron-only device and the hole-only device of flexible PSCs.

Table S3. A summary of the detail performance parameters of reported $CsPbX_3$ PSCs prepared at low temperature.

Configuration	Temperature (°C)	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	Ref.
FTO/TiO_2/$CsPbI_2Br$/Spiro-OMeTAD/Au (rigid)	120	1.18	14.9	77.2	13.5	11
PET/ITO/Nb_5O_2/$CsPbI_2Br$/Spiro-OMeTAD/Au (flexible)	130	1.19	14.6	67.3	11.7	11
ITO/NiO_x/$CsPbI_2Br$/C_{60}/BCP/Ag (rigid)	120	1.05	12.6	78.7	10.4	12
PET/ITO/NiO_x/$CsPbI_2Br$/C_{60}/BCP/Ag (flexible)	120	0.97	11.5	65.0	7.3	12
ITO/PEDOT/$CsPbI_2Br$/C_{60}/BCP/Ag (rigid)	Room-temperature	1.16	12.4	60.1	8.67	15
PET/ITO/PEDOT/$CsPbI_2Br$/C_{60}/BCP/Ag (flexible)	Room-temperature	1.05	12.0	51.4	6.50	15
PET/ITO/TiO_2/$CsPbI_{2.85}Br_{0.15}$/PTAA/Au (flexible)	-	0.96	18.73	72.8	13.14	10
FTO/c-TiO_2/$CsPbI_2Br$/Spiro-OMeTAD/Ag (rigid)	130	1.13	13.61	68.64	10.56	14
FTO/bl-TiO_2/m-TiO_2/$CsPbI_2Br$/PIF8-TAA /Au (rigid)	150	1.31	14.55	78.58	14.86	17
hc-PEDOT:PSS /AZO/$CsPbI_2Br$/t-BCA/PTAA/MoO_3/ Ag (flexible)	**120**	**1.26**	**15.87**	**75.41**	**15.08**	**This work**

References

1 P. Zhang, J. Wu, T. Zhang, Y. Wang, D. Liu, H. Chen, L. Ji, C. Liu, W. Ahmad, Z. D. Chen, S. Li, Adv. Mater., 2018, **30**, 1703737.

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