

## Supporting Information

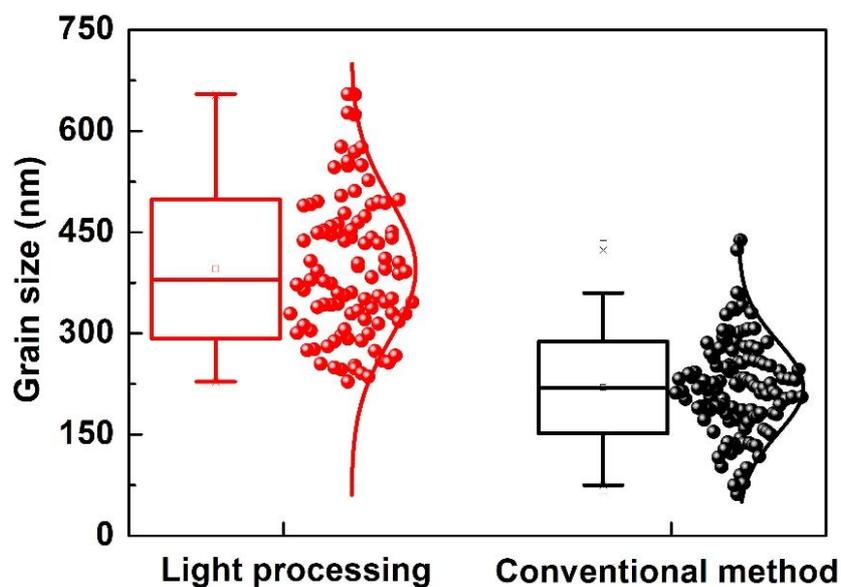
### **Light Processing Enables Efficient Carbon-Based, All-Inorganic Planar CsPbIBr<sub>2</sub> Solar Cells with High Photovoltages**

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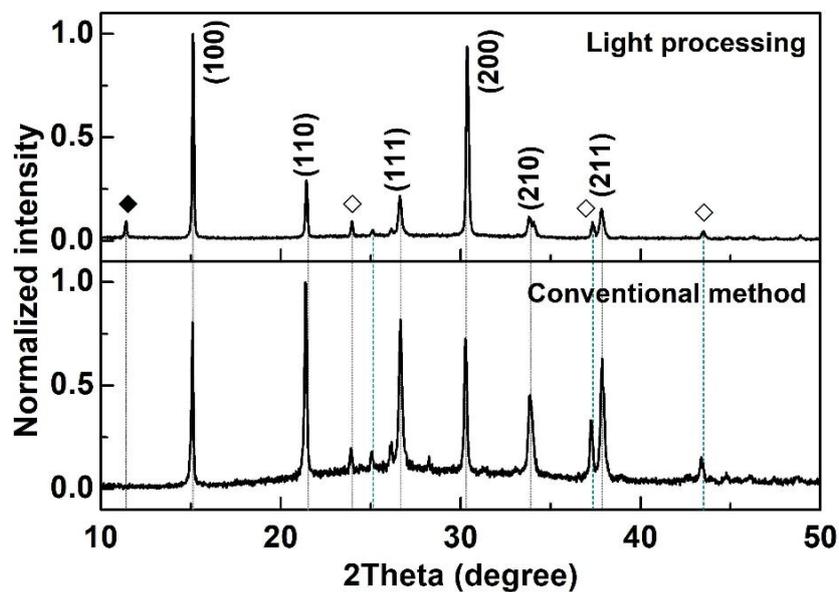
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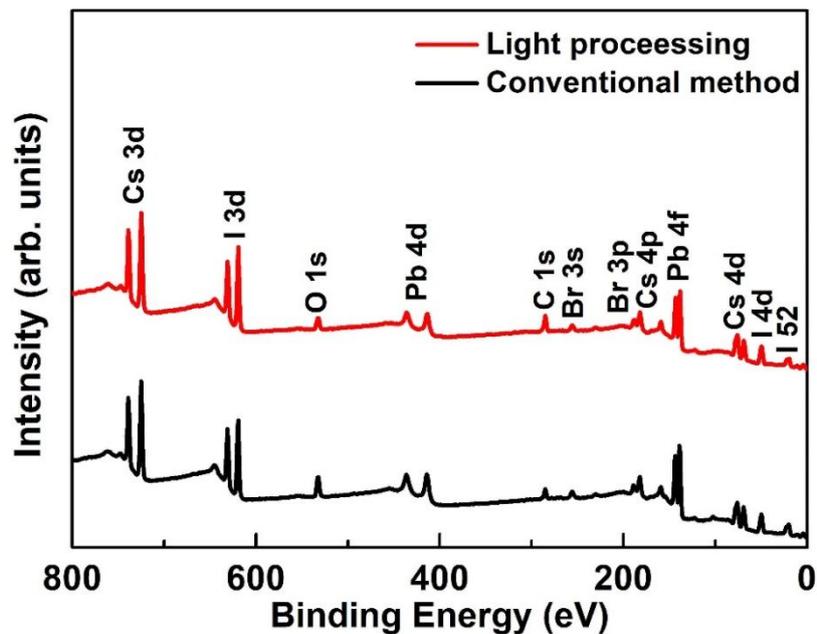
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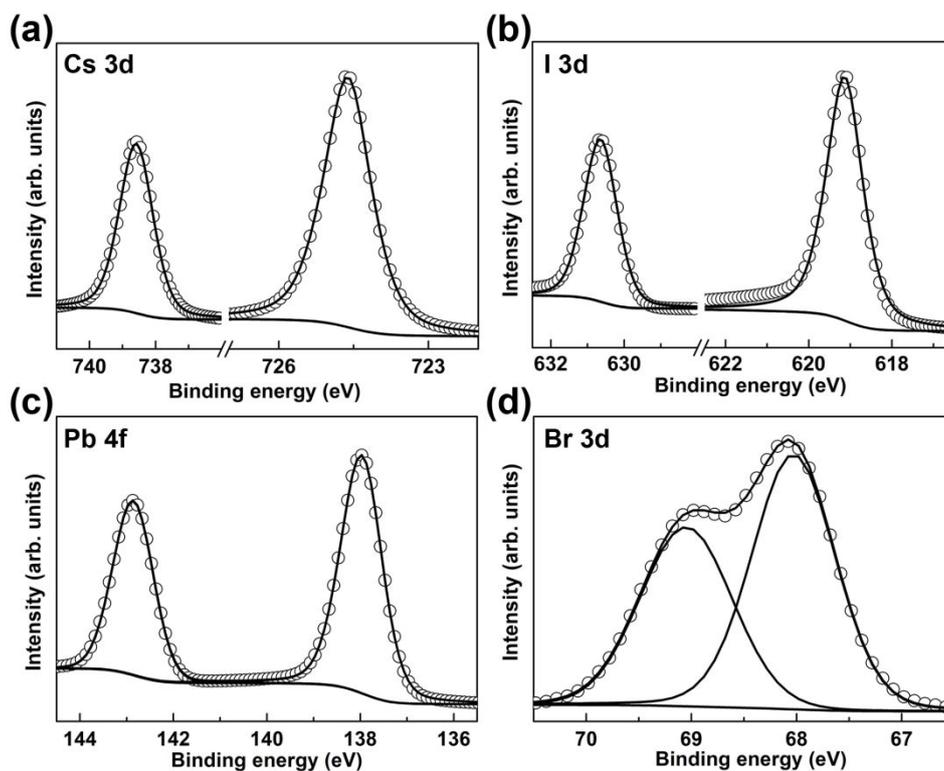
**Figure S1.** Statistic grain sizes distributions in the CsPbIBr<sub>2</sub> films prepared by light processing and conventional method, respectively.



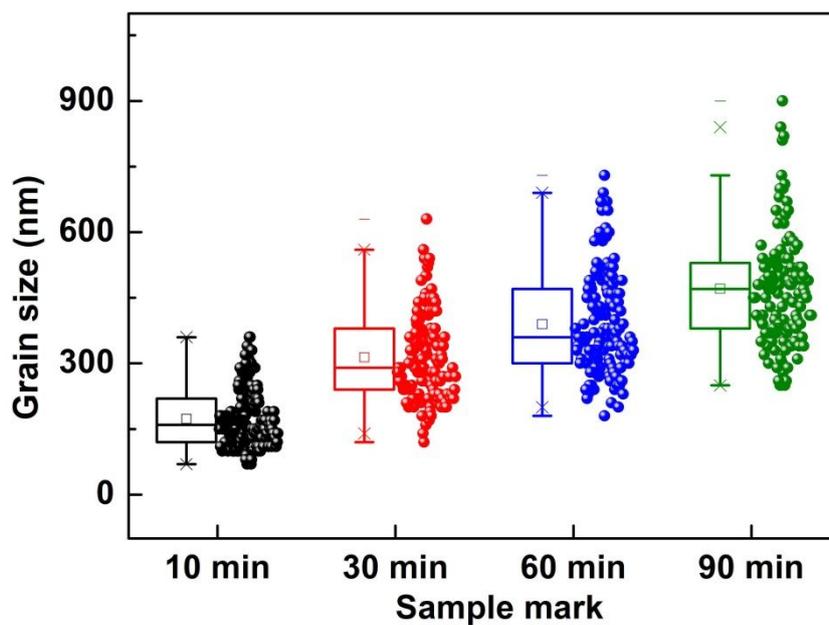
**Figure S2.** Normalized XRD patterns of CsPbIBr<sub>2</sub> films formed by light processing and conventional method, respectively. The symbols of solid diamond and open diamond represent the signals from PbBr<sub>2</sub> impurity phase and c-TiO<sub>2</sub>/FTO substrate, respectively.



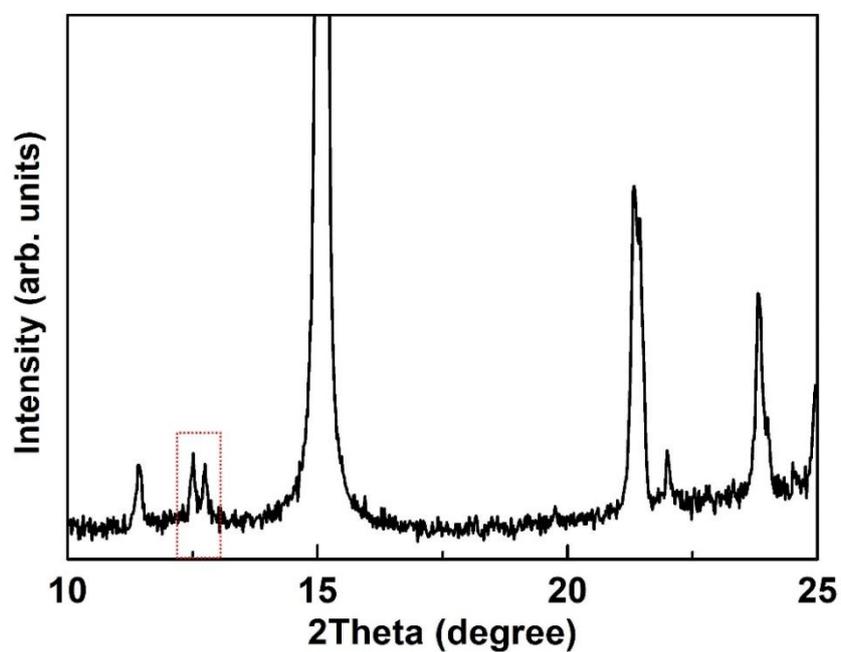
**Figure S3.** XPS survey spectra of CsPbIBr<sub>2</sub> films prepared by light processing and conventional method, respectively.



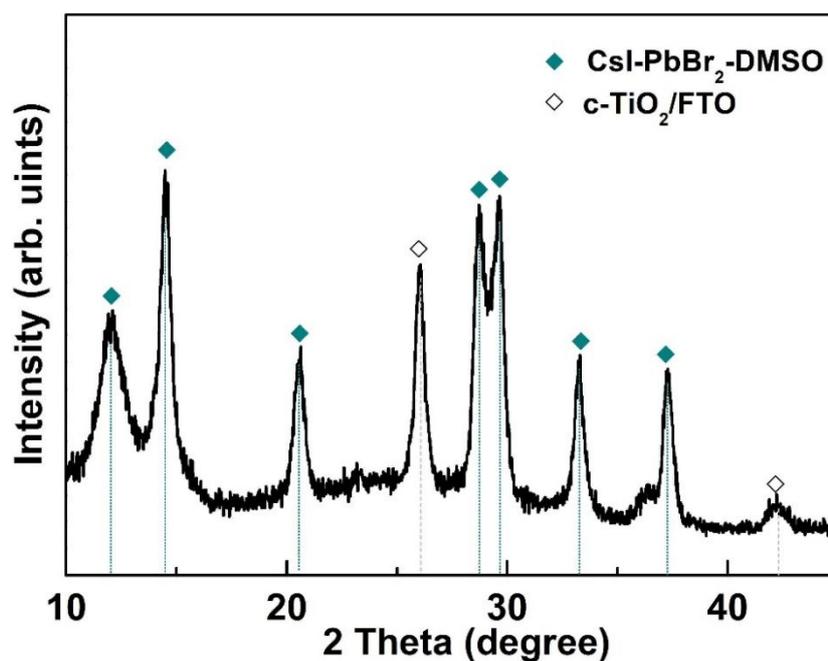
**Figure S4.** (a-d) Core-level XPS spectra of the CsPbIBr<sub>2</sub> film prepared by light processing after washing with absolute methanol.



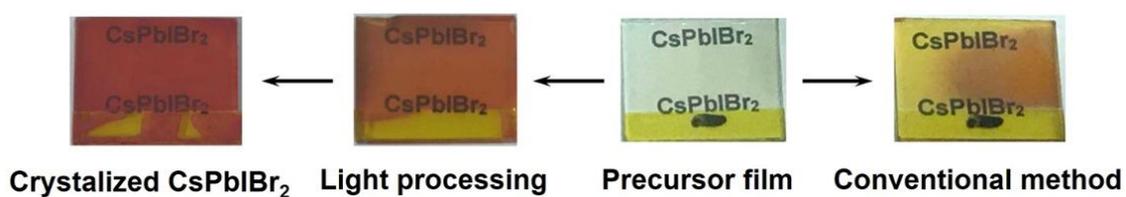
**Figure S5.** Statistic grain sizes distributions of the CsPbIBr<sub>2</sub> films with different light processing times of 10, 30, 60, and 90 min, respectively.



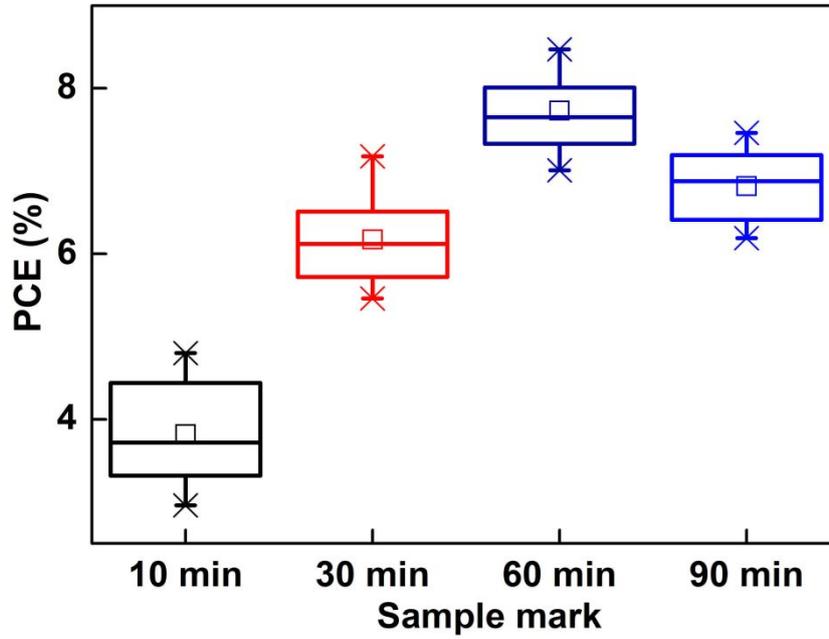
**Figure S6.** Enlarged XRD pattern of the CsPbIBr<sub>2</sub> film prepared by light processing with the time of 90 min. The red rectangle marks the signal from non-perovskite phase CsPbIBr<sub>2</sub>.



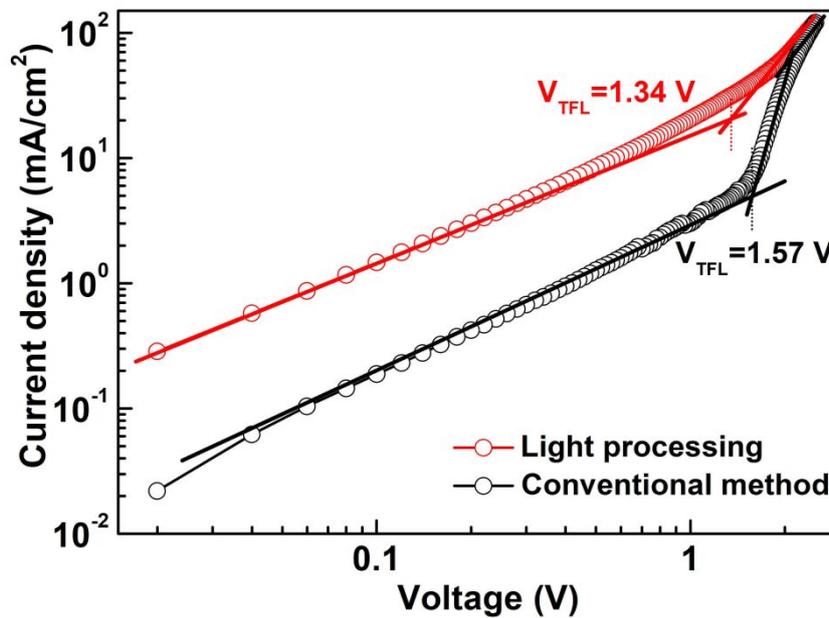
**Figure S7.** XRD pattern of the precursor CsPbIBr<sub>2</sub> film. It indicates that the film is composed of crystalline CsI-PbBr<sub>2</sub>-DMSO intermediate phase.



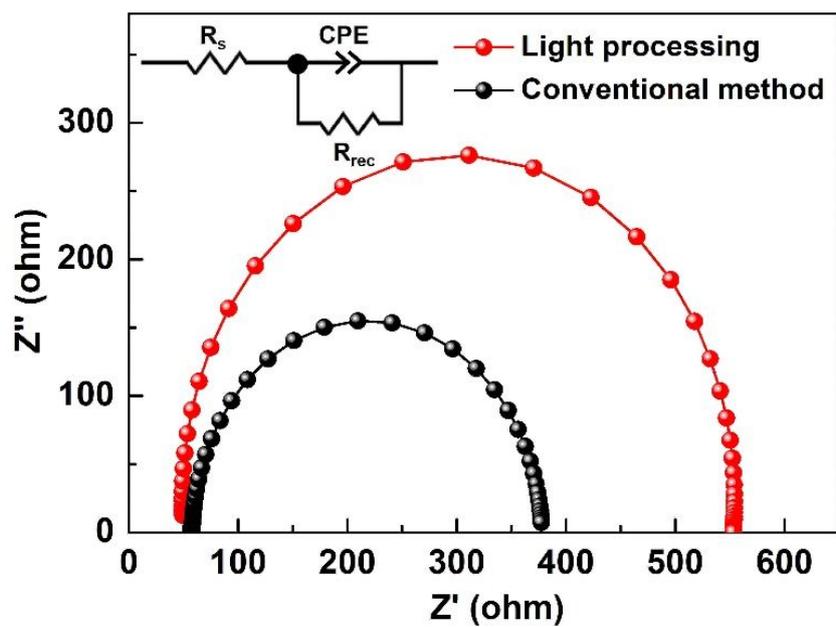
**Figure S8.** Optical images of the precursor CsPbIBr<sub>2</sub> film (Precursor film), the one after light processing (Light processing), and the one after being stored at 25 °C in dark for 60 min (Conventional method), as well as the ultimate CsPbIBr<sub>2</sub> film formed by light processing (Crystallized CsPbIBr<sub>2</sub>), respectively.



**Figure S9.** Statistic PCEs of 20 independent cells based on the CsPbIBr<sub>2</sub> films with different light processing times of 10, 30, 60, and 90 min, respectively.



**Figure S10.** Hole-only dark J-V curves as well as the SCLC analyses for the CsPbIBr<sub>2</sub> films prepared by light processing and conventional method, respectively.



**Figure S11.** Fitted Nyquist plots for the cell fabricated by light processing and conventional method, respectively. The insert is the equivalent circuit for the fittings.

**Table S1.** Summary the  $E_{\text{loss}}$  values along with  $V_{\text{oc}}$  values and PCEs for pure CsPbIBr<sub>2</sub>-based solar cells reported so far.

Cell configuration	$E_g$ (eV)	$V_{\text{oc}}$ (V)	PCE (%)	$E_{\text{loss}}$ (eV)	Refs.
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	2.05	1.245	9.16	0.805	[25]
ITO/SnO <sub>2</sub> /CsPbIBr <sub>2</sub> /spiro-OMeTAD/Ag	2	1.15	8.54	0.85	[27]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	1.90	1.08	8.25	0.82	[22]
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /spiro-OMeTAD/Au	2.05	1.227	8.02	0.823	[32]
ITO/SnO <sub>2</sub> /C60/CsPbIBr <sub>2</sub> /spiro-OMeTAD/Au	2.04	1.18	7.34	0.86	[31]
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	2.05	1.142	6.55	0.908	[24]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /spiro-OMeTAD/Au	2.05	1.121	6.30	0.929	[28]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	1.89	0.96	6.14	0.93	[23]
FTO/NiO <sub>x</sub> /CsPbIBr <sub>2</sub> /ZnO/Al	2.1	1.01	5.57	1.09	[34]
FTO/NiO <sub>x</sub> /CsPbIBr <sub>2</sub> /MoO <sub>x</sub> /Au	2.08	0.85	5.52	1.23	[30]
ITO/ZnO/CsPbIBr <sub>2</sub> /spiro-OMeTAD/Ag	2.06	1.04	4.8	1.01	[33]
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Au	2.05	0.959	4.70	1.091	[29]

**Table S2.** Compositional atom ratios in CsPbIBr<sub>2</sub> films obtained by light processing and conventional method, respectively, as well as that for the former after washing with absolute methanol.

Samples	Cs (at.%)	Pb (at.%)	I (at.%)	Br (at.%)	(Cs+I)/(Pb+Br/2)
Light processing	10.28	5.92	8.93	13.04	1.54
Conventional method	10.01	8.19	9.02	18.25	0.97
Light processing after washing	9.85	7.87	9.23	17.61	1.14

**Table S3.** Summary of photovoltaic parameters of pure CsPbIBr<sub>2</sub>-based solar cells reported so far.

Cell configuration	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF	PCE (%)	Refs.
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	1.283	11.17	0.60	8.60	<b>This work</b>
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	1.245	10.66	0.69	9.16	[25]
ITO/SnO <sub>2</sub> /CsPbIBr <sub>2</sub> /spiro-OMeTAD/Ag	1.15	10.61	0.70	8.54	[27]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	1.08	12.32	0.62	8.25	[22]
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /spiro-OMeTAD/Au	1.227	9.69	0.674	8.02	[32]
ITO/SnO <sub>2</sub> /C60/CsPbIBr <sub>2</sub> /spiro-OMeTAD/Au	1.18	8.32	74.8	7.34	[31]
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	1.142	9.11	0.63	6.55	[24]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /spiro-OMeTAD/Au	1.121	7.9	0.70	6.30	[28]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Carbon	0.96	12.15	0.53	6.14	[23]
FTO/NiO <sub>x</sub> /CsPbIBr <sub>2</sub> /ZnO/Al	1.01	8.65	0.636	5.57	[34]
FTO/NiO <sub>x</sub> /CsPbIBr <sub>2</sub> /MoO <sub>x</sub> /Au	0.85	10.56	0.62	5.52	[30]
ITO/ZnO/CsPbIBr <sub>2</sub> /spiro-OMeTAD/Ag	1.04	8.78	0.525	4.8	[33]
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /Au	0.959	8.7	0.56	4.70	[29]