

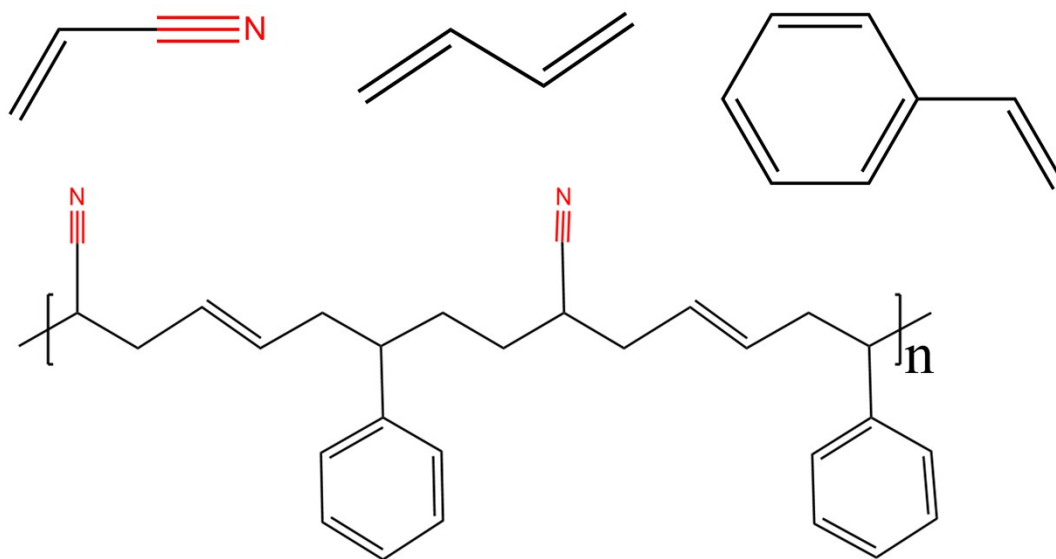
## Supporting Information

### **Enhanced comprehensive performance of carbon-based hole-transport-layer-free CsPbI<sub>2</sub>Br solar cells by a low-cost and stable long chain polymer**

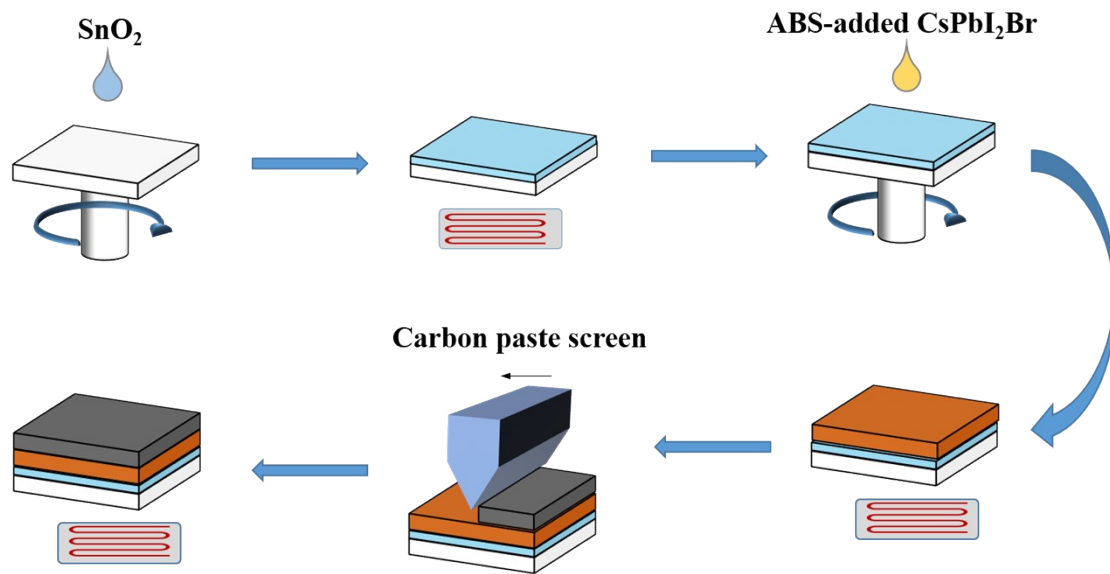
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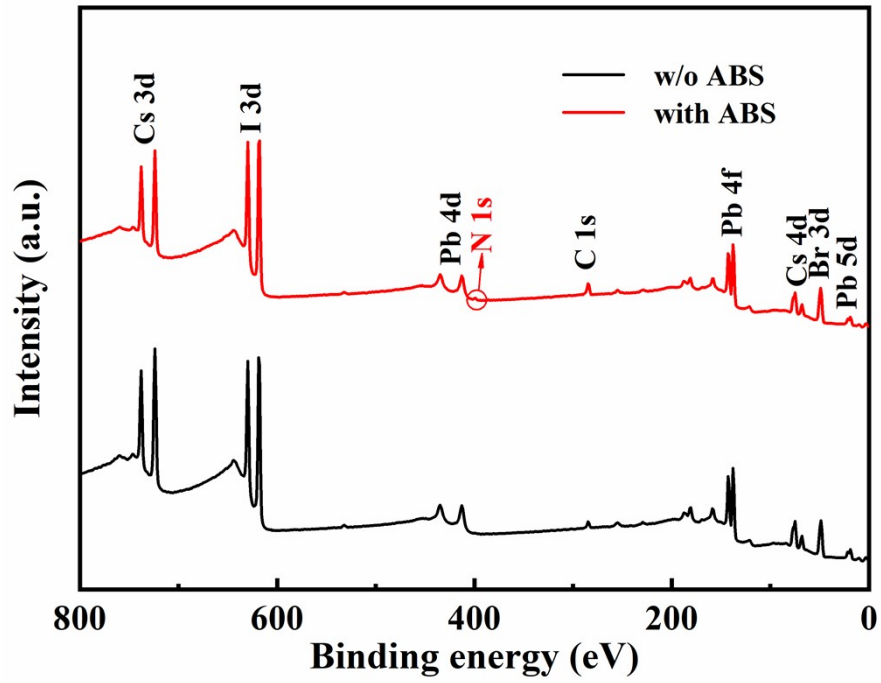
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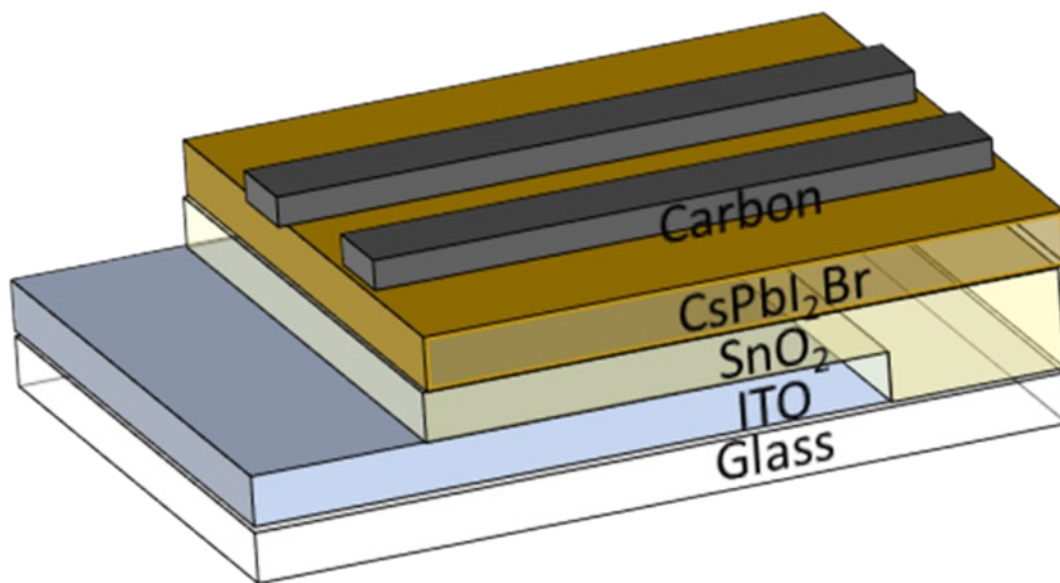
**Figure S1.** Molecular structures of the precursors (Top: acrylonitrile, butadiene and styrene from left to right) for synthesizing ABS (bottom).



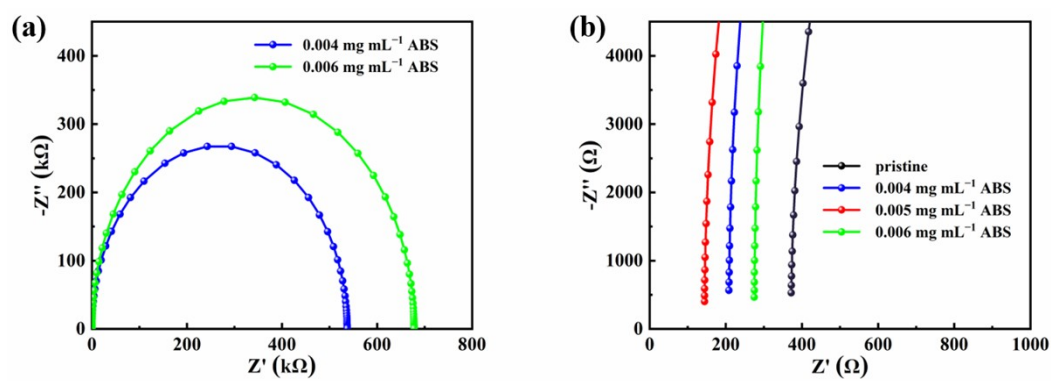
**Figure S2.** Main procedures of fabricating an HTL-free CsPbI<sub>2</sub>Br solar cell with a carbon electrode.



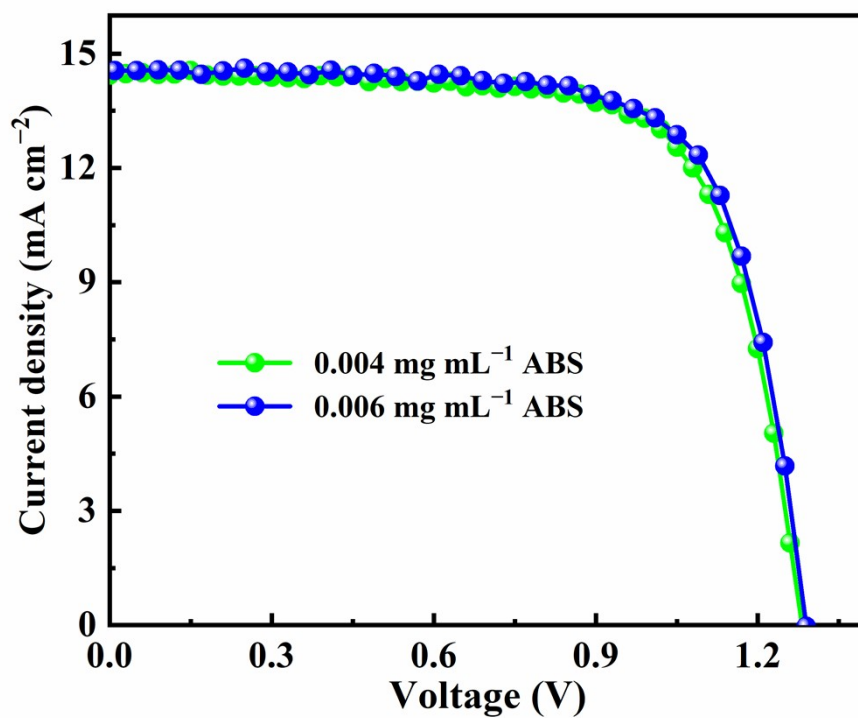
**Figure S3.** XPS full spectra of the pristine and ABS-added CsPbI<sub>2</sub>Br layers.



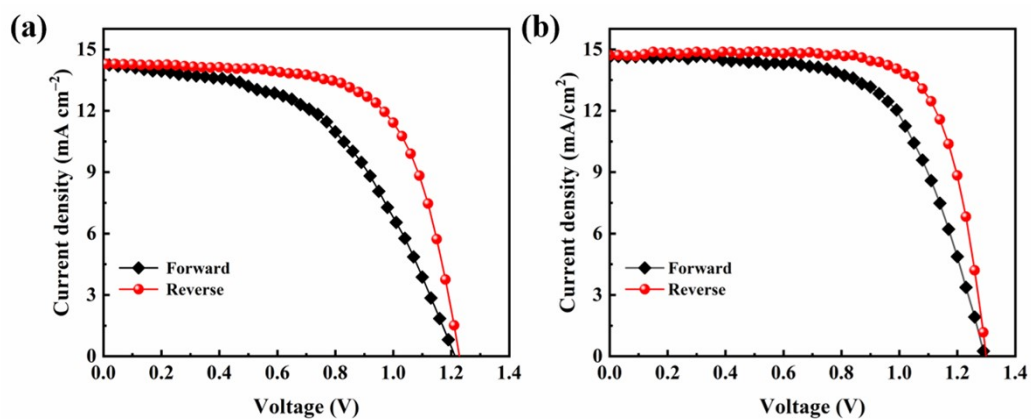
**Figure S4.** Device configuration of the HTM-free carbon-based CsPbI<sub>2</sub>Br solar cells.



**Figure S5.** (a) Nyquist plots of the EIS curves for the devices prepared with the ABS addition of 0.004 and 0.006 mg mL<sup>-1</sup>. (b) Enlarged plots of the EIS curves at high frequency for the devices prepared with different ABS additions.

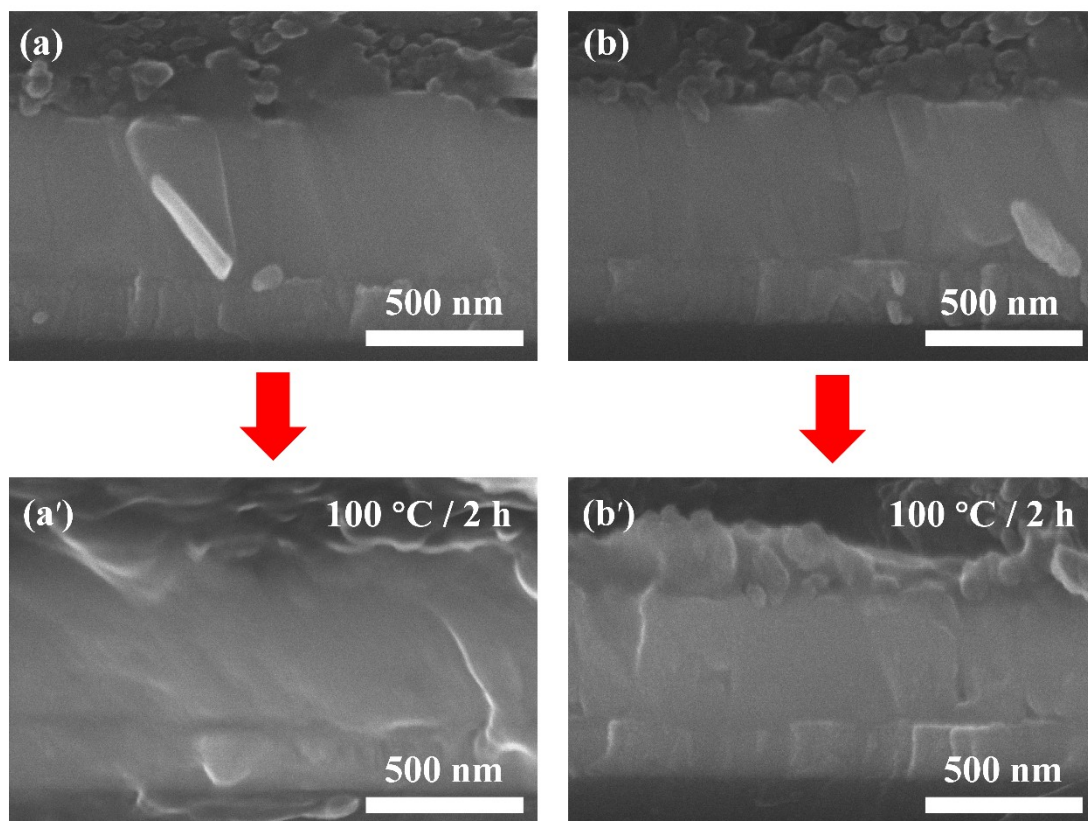


**Figure S6.** Illuminated  $J$ - $V$  curves at AM 1.5G for the devices prepared with the ABS addition of 0.004 and 0.006 mg mL<sup>-1</sup>

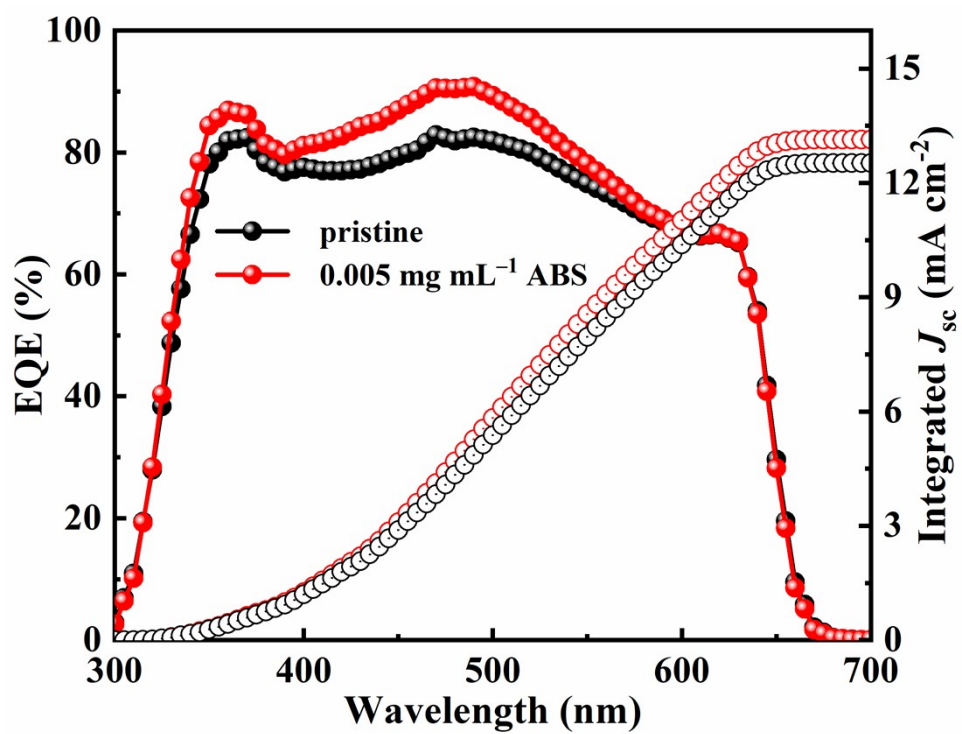


**Figure S7.**  $J$ - $V$  curves of the (a) pristine and (b) 0.005  $\text{mg mL}^{-1}$  ABS-added champion CsPbI<sub>2</sub>Br PSCs at the forward and reverse scans.

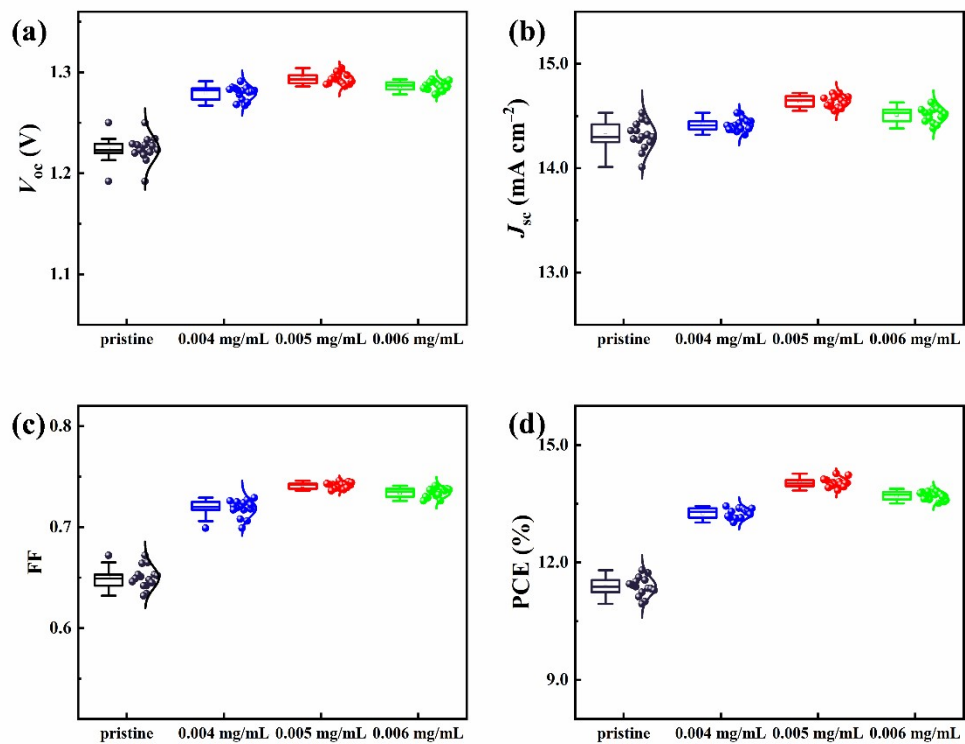




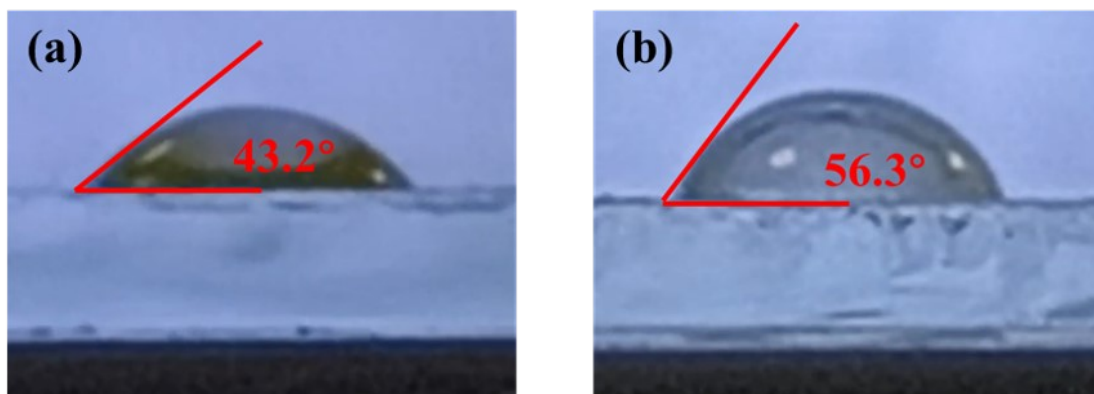
**Figure S8.** Cross-sectional SEM images of the (a, a') pristine device and (b, b') 0.005 mg mL<sup>-1</sup> ABS-added device before and after annealing at 100 °C for 2 h in air.



**Figure S9.** EQE spectra and the integrated  $J_{sc}$  of the devices prepared with the ABS addition of 0 and 0.005 mg mL<sup>-1</sup>.



**Figure S10.** Statistical charts of (a)  $V_{oc}$ , (b)  $J_{sc}$ , (c) FF and (d) PCE of the 15 independent devices for each ABS addition.



**Figure S11.** Water contact angle of the (a) pristine and (b)  $0.005 \text{ mg mL}^{-1}$  ABS-added CsPbI<sub>2</sub>Br layers.

**Table S1.** Molecular mass and density of ABS.

<b>Sample</b>	<b>Mw (g/mol)</b>	<b><math>\rho</math> (g/ml)</b>
ABS	633.91	1.05

**Table S2.** XRD data of the CsPbI<sub>2</sub>Br layers with different ABS additions.

Sample	(100)		(200)	
	$2\theta^\circ$	FWHM	$2\theta^\circ$	FWHM
pristine	14.63°	1.11	29.55°	0.58
0.004 mg ml <sup>-1</sup> ABS	14.62°	0.99	29.53°	0.47
0.005 mg ml <sup>-1</sup> ABS	14.62°	0.90	29.54°	0.46
0.006 mg ml <sup>-1</sup> ABS	14.63°	0.89	29.56°	0.48

**Table S3.** Carrier lifetimes estimated from the TRPL spectra of Figure 4d.

<b>Sample</b>	<b><math>\tau_1</math> (ns)</b>	<b><math>\tau_2</math> (ns)</b>	<b><math>A_1</math></b>	<b><math>A_2</math></b>	<b><math>\tau_{ave}</math> (ns)</b>
Pristine	1.11	8.86	227.25	2.18	1.66
0.005 mg mL <sup>-1</sup> ABS	0.92	12.47	292.37	2.41	2.08

**Table S4.** Summary of the reported photovoltaic parameters of the HTM-free carbon-based CsPbI<sub>2</sub>Br PSCs.

Strategy	$J_{sc}$ (mA cm <sup>-2</sup> )	$V_{oc}$ (V)	FF (%)	PCE (%)	Ref
ABS additive	14.69	1.30	74.7	14.27	This work
Cs <sub>2</sub> PtI <sub>6</sub> surface modification	14.85	1.28	72.0	13.69	[44]
PTU and PU interface modification	14.83	1.22	72.0	13.01	[45]
ATHPBr additive	14.28	1.30	78.1	14.50	[46]
non-hydrophilic Cl-containing SnO <sub>2</sub> ETL	14.70	1.26	76.2	14.11	[47]
CsPbI <sub>2</sub> Br/CsPb <sub>2</sub> I <sub>4</sub> Br heterostructure	14.60	1.32	79.1	15.25	[48]
HTAB surface modification	14.10	1.26	80.6	14.31	[49]
KTFA and CF <sub>3</sub> PMABr additive	14.98	1.27	73.7	14.05	[23]
P-F-PEAI/Br passivation layer	14.91	1.27	73.8	13.97	[50]
Ionic modified ZnO layer	14.95	1.27	75.1	14.25	[51]
EDTA interface modification	14.95	1.26	74.3	13.94	[52]



**Table S5.** Photovoltaic parameters of the champion devices fabricated with the pristine and 0.005 mg mL<sup>-1</sup> ABS-added CsPbI<sub>2</sub>Br layers at the forward and reverse scans.

Devices		$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF	PCE (%)	HI (%)
pristine	Forward	14.28	1.213	0.522	9.04	23.4%
	Reverse	14.30	1.228	0.672	11.80	
0.005 mg mL <sup>-1</sup> ABS	Forward	14.62	1.294	0.632	11.96	16.2%
	Reverse	14.69	1.300	0.747	14.27	