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Supporting information

Low-temperature sprayed carbon electrode in modular HTL-free perovskite solar cells: comparative study on the choice of carbon sources

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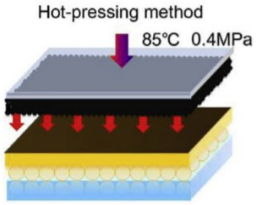
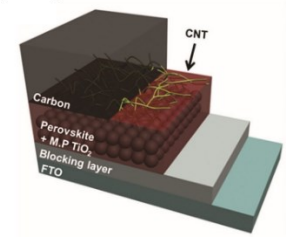
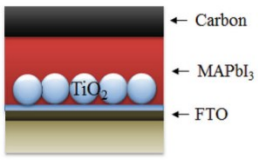
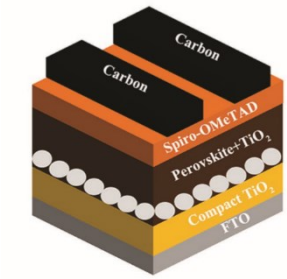
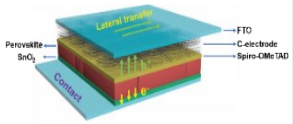
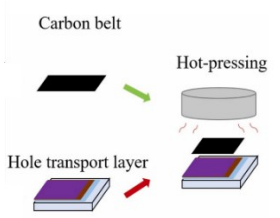
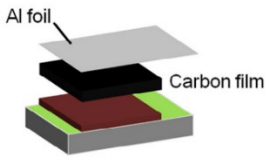
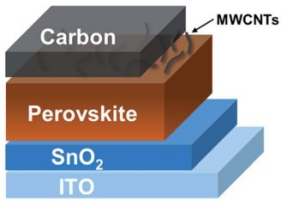
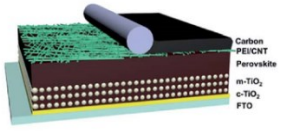
Table S1. A summary of device structure and photovoltaic performance of C-PSCs.

Carbon-electrode PSCs Structure	Perovskite	Cell Area (cm^2)	V_{oc} (V)	J_{sc} (mA cm^{-2})	PCE (%)	Ref.
	$Cs_{0.05}(MA_{0.17}FA_{0.83})_{0.95}Pb(I_{0.83}Br_{0.17})_3$	0.30 1.00	0.89 0.84	22.29 22.39	10.84 8.72	11
	$Cs_{0.06}(MA_{0.17}FA_{0.83})_{0.94}Pb(I_{0.84}Br_{0.16})_3$	0.12 1.00	0.96 0.99	18.66 17.57	13.57 8.18	48

	$MAPbI_3$	0.07 1.00	1.04 0.99	21.27 19.63	14.38 9.72	40
	$FA_xMA_{1-x}PbI_yBr_{3-y}$	0.1	1.08	23.33	19.2	16
	$Cs_{0.05}(FA_{0.85}MA_{0.15})_{0.95}Pb(I_{0.85}Br_{0.15})_3$	1.30	1.05	22.78	18.65	35
	$CsFA_{0.83}MA_{0.17}PbI_{2.53}Br_{0.47}$	0.115	1.09	21.1	15.3	32
	$CH_3NH_3PbI_3$	0.08	1.002	21.30	13.53	25
	$FA_xMA_{1-x}PbI_yBr_{3-y}$	1.0	1.01	20.48	12.34	34
	$CsPbI_3$	0.08	0.80	18.58	10.55	50

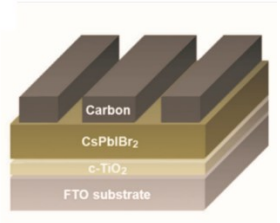
	$CsPbIBr_2$	0.09	1.245	10.66	9.16	33

Table S2. Parameters of the TRPL spectroscopy based on the pristine perovskite films and perovskite/carbon sample with various carbon sources.

$$Y(t)=A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2} + y_0 \ * \text{MERGEFORMAT (1)}$$

$$\tau_{ave} = \frac{A_1\tau_1}{A_1\tau_1 + A_2\tau_2}\tau_1 + \frac{A_2\tau_2}{A_1\tau_1 + A_2\tau_2}\tau_2 \ * \text{MERGEFORMAT (2)}$$

Sample	τ_1 (ns)	Standard error	τ_2 (ns)	Standard error	τ_{ave} (ns)
Perovskite	25.94	0.89	437.97	11.55	254.91
MWCNT	4.94	0.17	43.65	0.83	21.65
Graphene	3.64	0.12	68.98	1.36	29.28
CNC	15.35	0.83	133.66	9.56	51.77

Table S3. Parameters of the ultraviolet photoelectron spectra based on different carbon sources.

$$\varphi = 21.2 - E_{cutoff} + E_{Fermi} \ * \text{MERGEFORMAT (3)}$$

Carbon Sources	E_{cutoff} (eV)	E_{Fermi} (eV)	Work function (eV)
CNT	16.67	0.49	5.02
CNC	16.72	1.65	6.13
Graphene	16.75	0.69	5.14

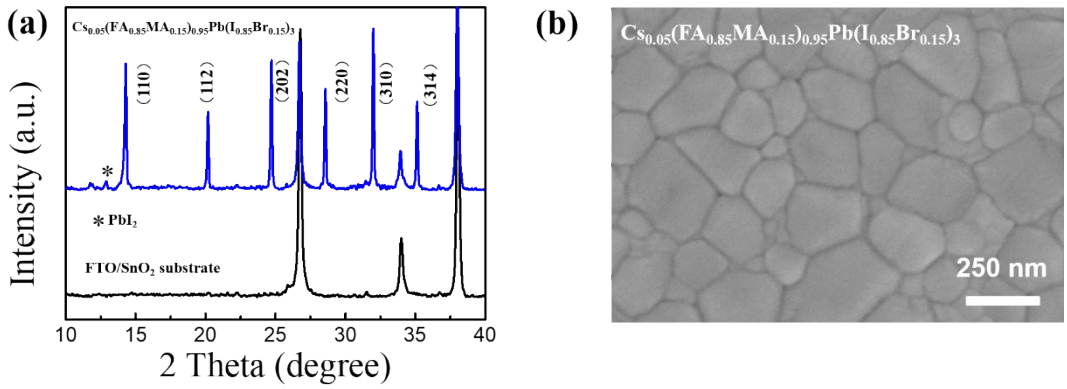


Fig. S1. (a) XRD patterns of the as-grown perovskite films. (b) top-view SEM images of perovskite films.

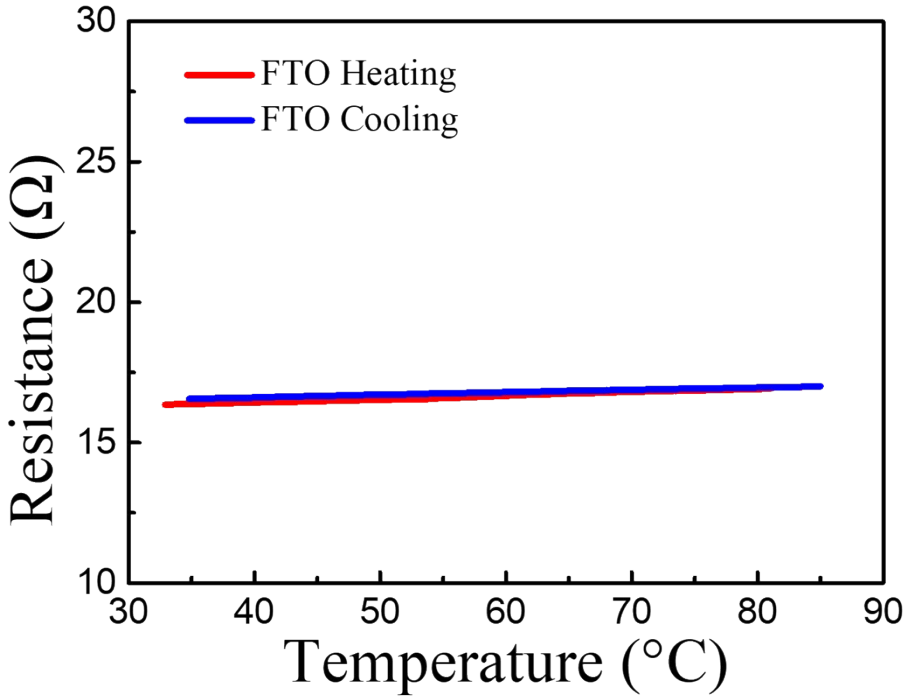
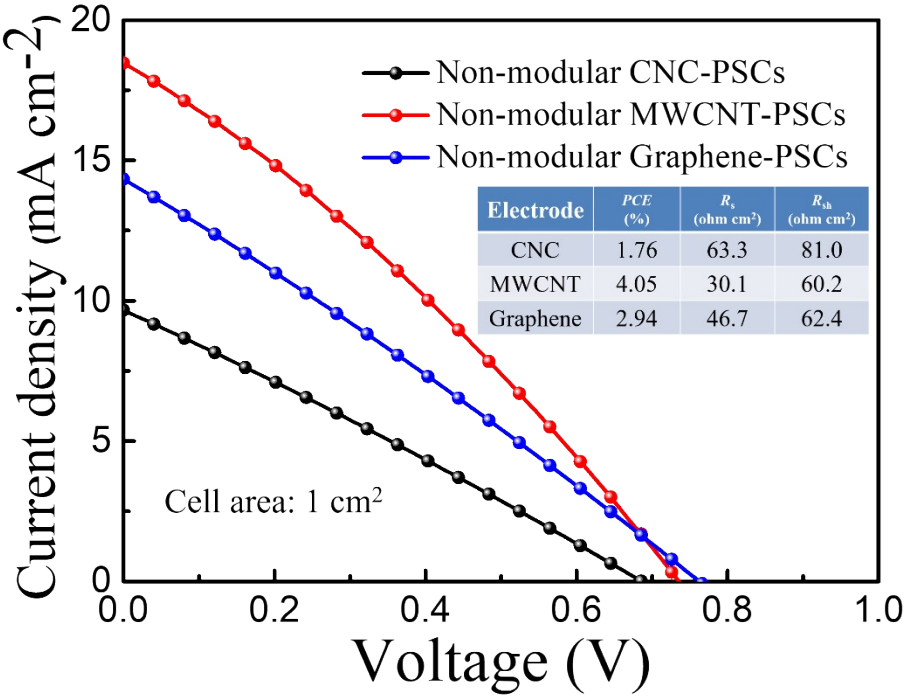


Fig. S2. The resistance- temperature curves of FTO film under heating and cooling process.



Electrode	*PCE* (%)	R_s (ohm cm²)	R_{sh} (ohm cm²)
CNC	1.76	63.3	81.0
MWCNT	4.05	30.1	60.2
Graphene	2.94	46.7	62.4

Fig. S3. J-V curves (cell area: 1 cm^2) under solar simulator AM 1.5 of different carbon-based non-modular PSCs (CNC, MWCNT, Graphene).

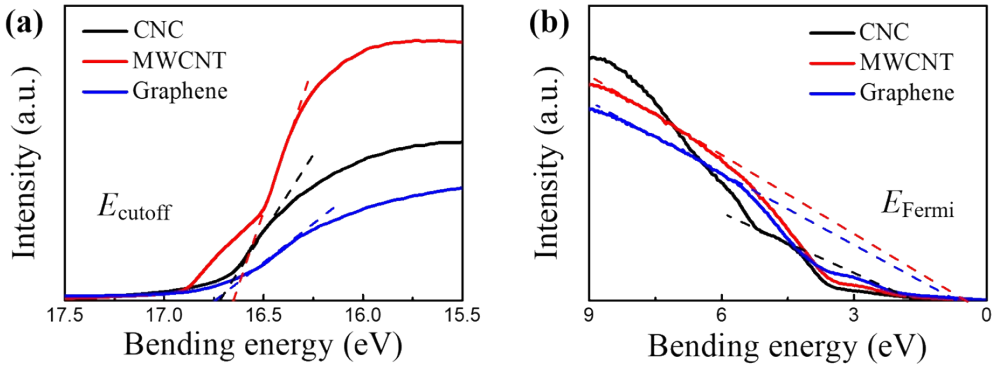


Fig. S4. (a) the cut-off energy (left part of Fig.4c) and (b) Fermi edge (right part of Fig.4c)

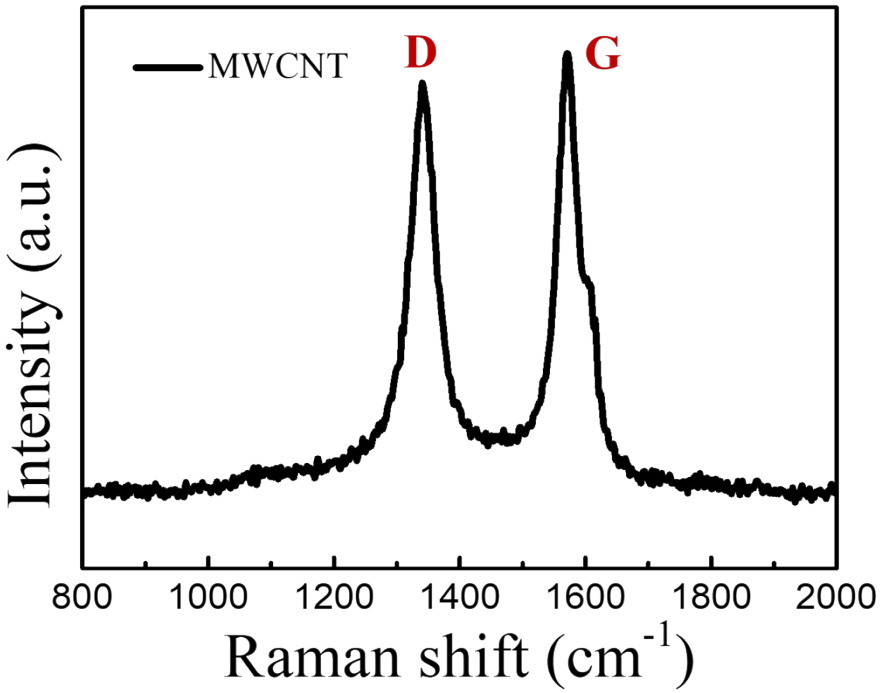


Fig. S5. Raman spectra of MWCNT, where the D-band originated from structural defects in carbon materials; the other G-band originated from graphite structure.

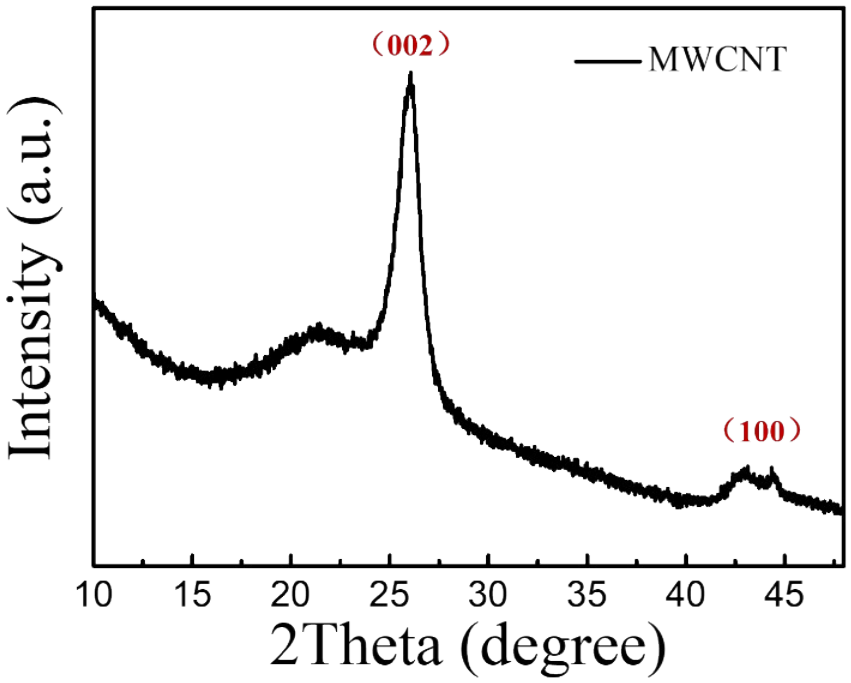


Fig. S6. XRD spectra of MWCNT.

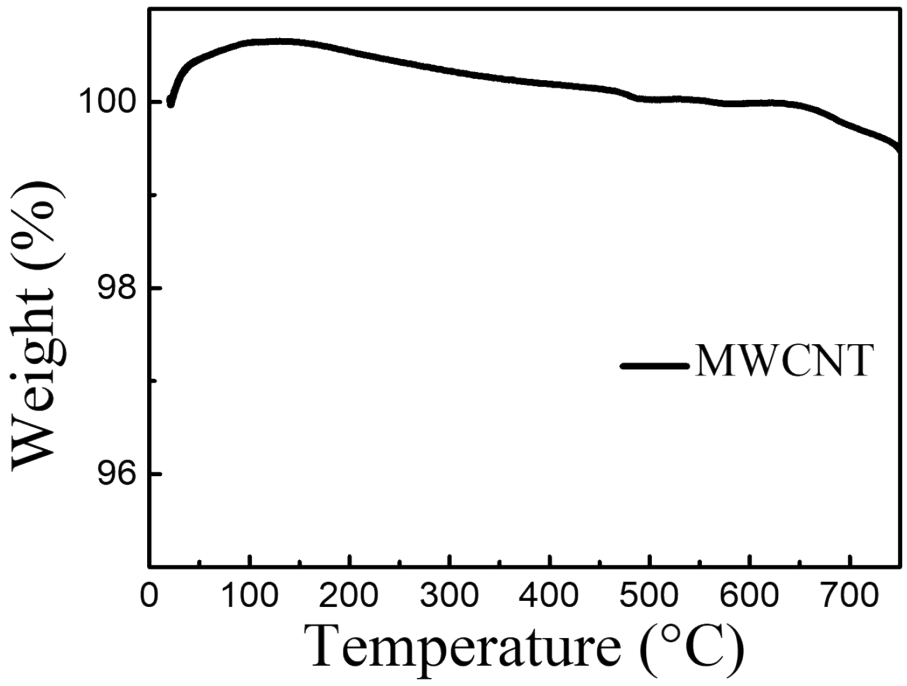


Fig. S7. Thermo gravimetric analysis (TGA) plot of MWCNT, which a convinced sign of thermal stability in the range of 25-800 ^{o}C.

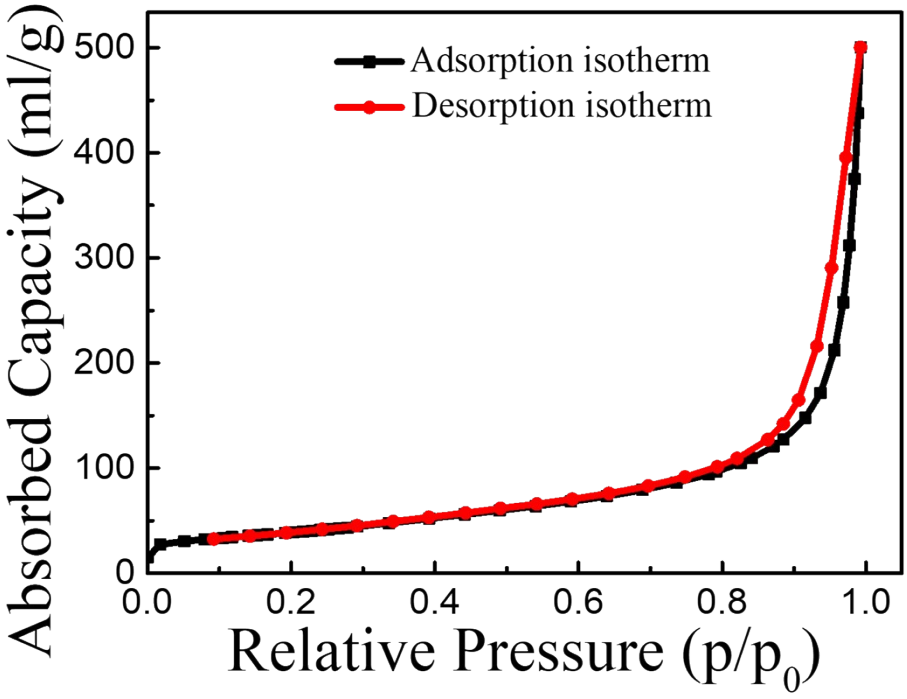


Fig. S8. Isotherm linear plot of MWCNT, and the specific surface area was 138.56 m^2/g.

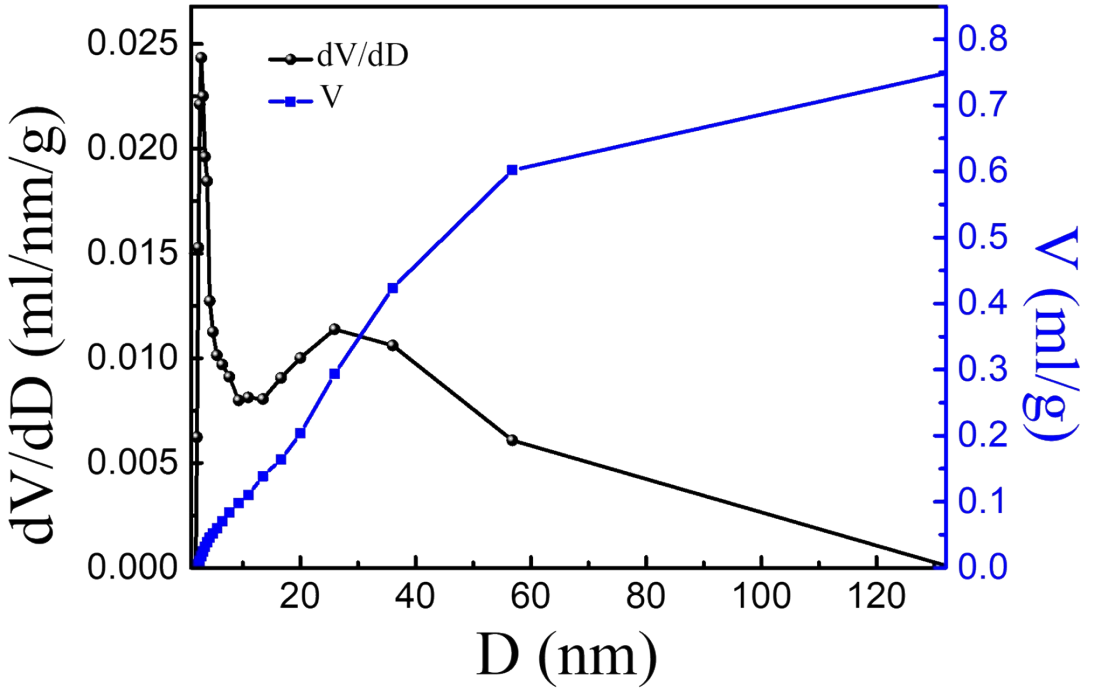


Fig. S9. Barrett-Joyner-Halenda (BJH desorption) pore volume & pore size curve of MWCNT, where the pore volume was 0.75 ml/g, the pore size was 16.31 nm.