

Supporting Information

Efficient all-air processed mixed cation carbon-based perovskite solar cells with ultra-high stability

Junshuai Zhou^a, Jiaojiao Wu^a, Nan Li^a, Xitao Li^a, Yan-Zhen Zheng^{*a}, Xia Tao^{*a}

^a State Key Laboratory of Organic-Inorganic Composites, Beijing University of Chemical Technology, Beijing 100029, China

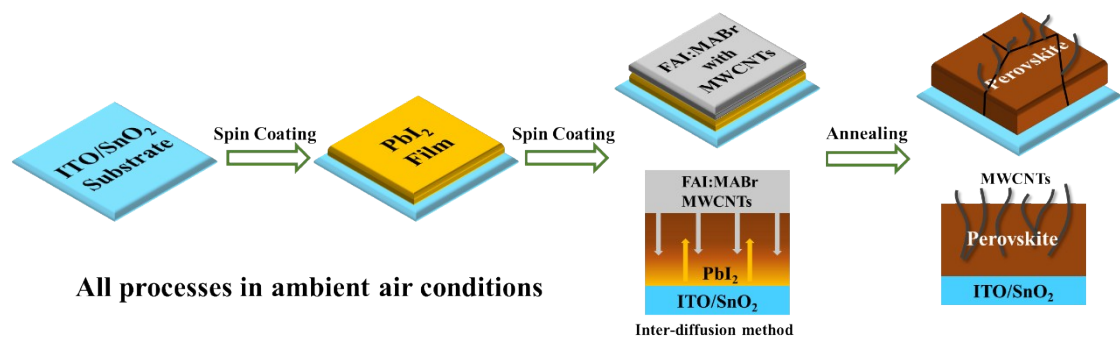


Fig. S1 Schematic illustration of inter-diffusion two-step method of fabricating perovskite films.

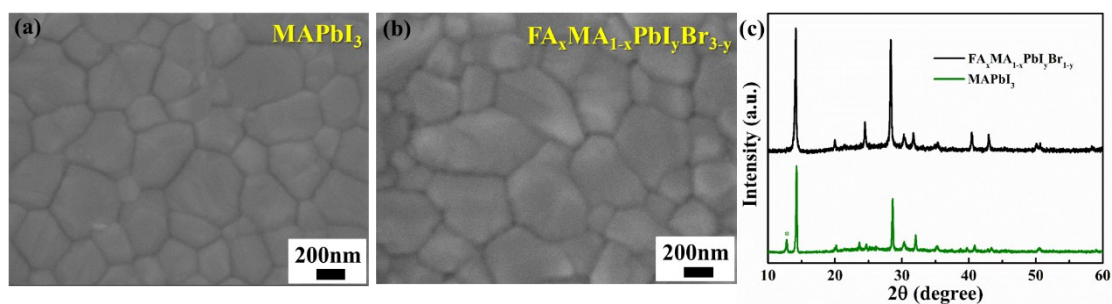


Fig. S2 (a,b) Top view SEM image of MAPbI_3 and $\text{FA}_x\text{MA}_{1-x}\text{PbI}_y\text{Br}_{3-y}$ perovskite films and (c) XRD patterns of MAPbI_3 and $\text{FA}_x\text{MA}_{1-x}\text{PbI}_y\text{Br}_{3-y}$ perovskite films. Asterisks denote the major reflections of PbI_2 .

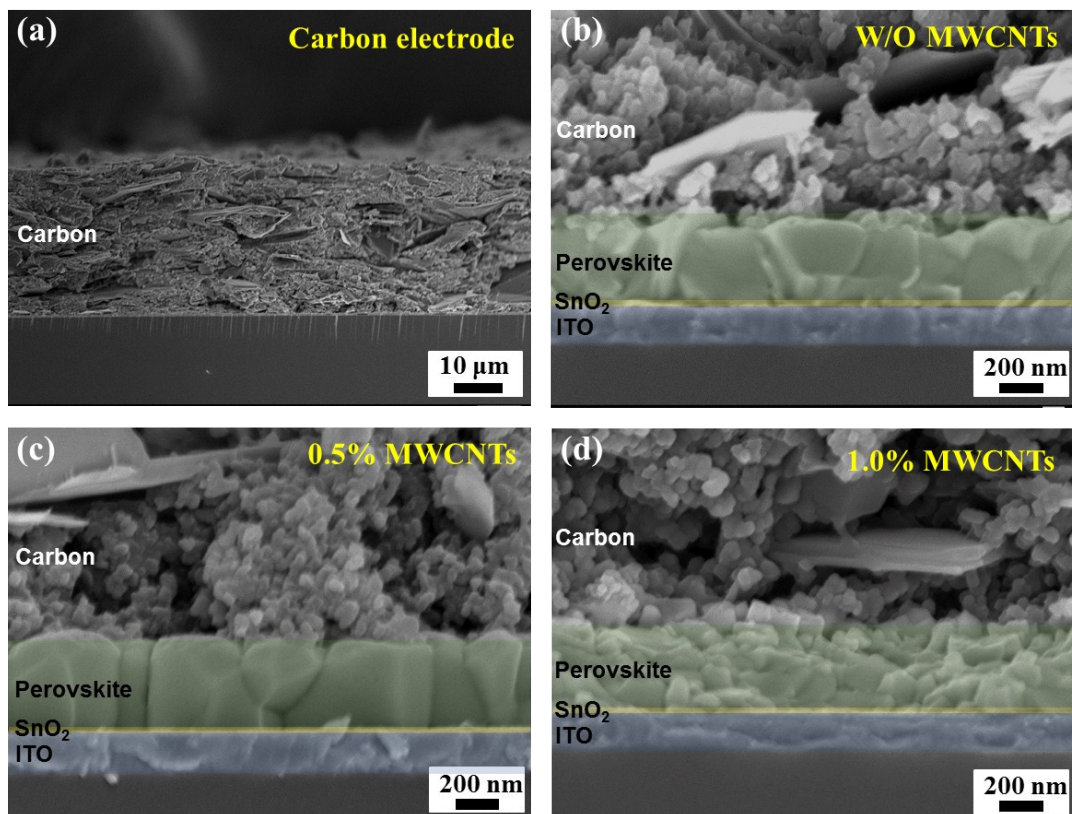


Fig. S3 (a) Cross-sectional SEM image of carbon electrode in ITO/SnO₂/FA_xMA_{1-x}PbI_yBr_{3-y} with MWCNTs/C device. (b-d) Cross-sectional SEM images of C-PSC devices with different MWCNTs doping contents.

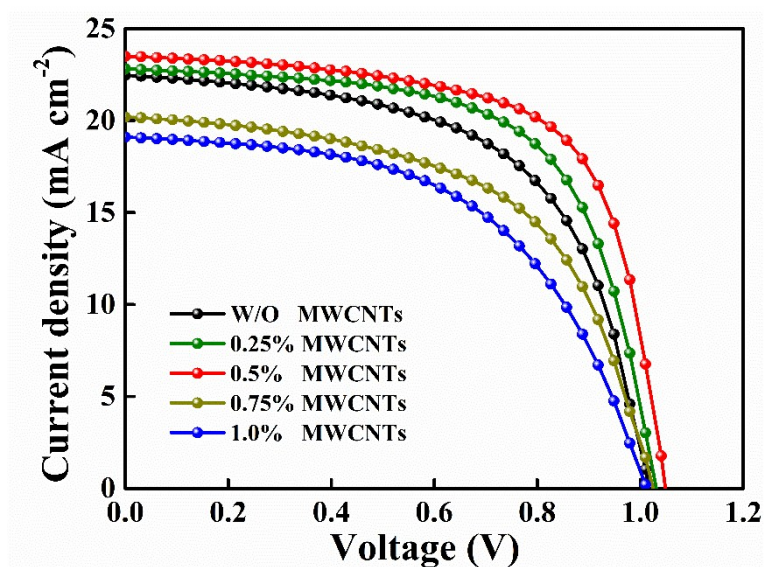


Fig. S4 J - V curves of the C-PSCs using the perovskite films with different MWCNTs (W/O, 0.25%, 0.5%, 0.75%, and 1.0%) under AM 1.5G 100 mW cm⁻² illumination.

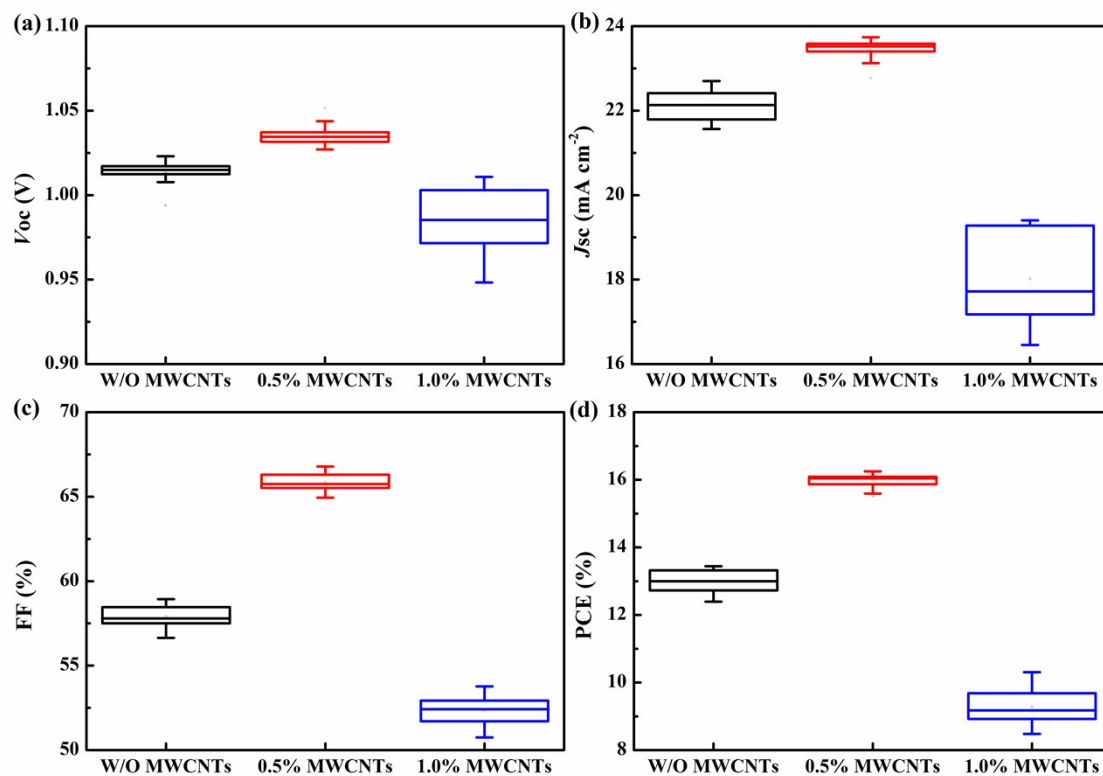


Fig. S5 Photovoltaic parameters statistics of 40 perovskite devices with different MWCNTs (W/O, 0.5% and 1.0%).

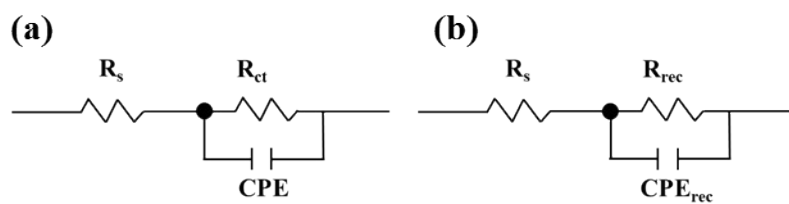


Fig. S6 The equivalent circuit model for C-PSCs in EIS under AM 1.5G illumination (a) and under dark condition (b).

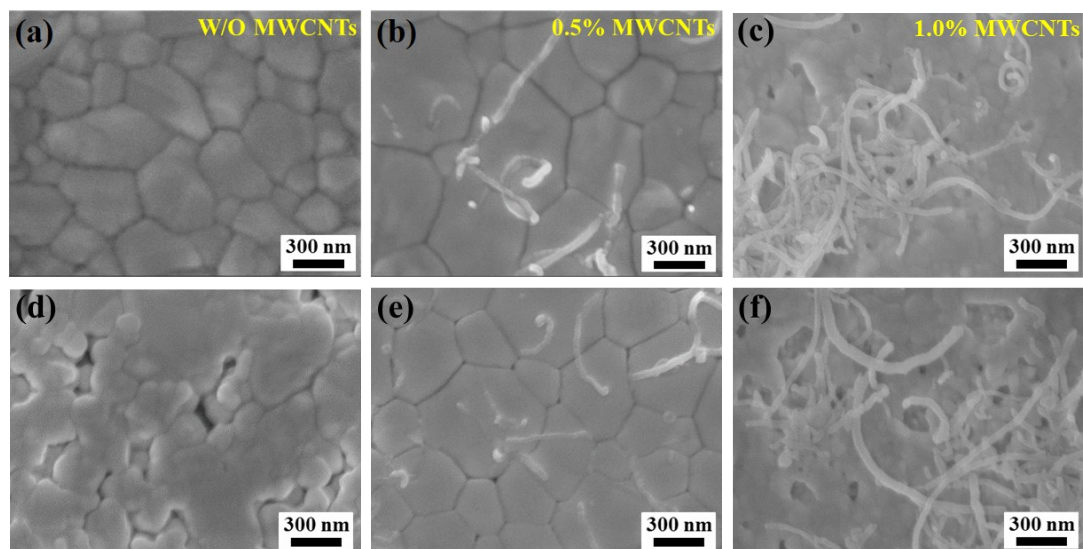


Fig. S7 SEM images of fresh (a-c) and air-aged (60 days, d-f) perovskite films with different MWCNTs (W/O, 0.5%, 1.0%) in air with a relative humidity of 30-80%.

Table S1 Parameters of the TRPL spectroscopy based on the perovskite films with different MWCNTs doping contents.

Samples	τ_1	% of τ_1	τ_2	% of τ_2	τ_{ave}
W/O MWCNTs	3.14	33.67	23.16	66.33	16.42
0.5% MWCNTs	2.73	35.86	19.85	64.14	13.71
1.0% MWCNTs	6.16	30.35	33.15	69.65	24.96

Table S2 Photovoltaic parameters of the best-performing C-PSCs with different MWCNTs doping content (W/O, 0.25%, 0.5%, 0.75% and 1.0%).

Samples	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
W/O MWCNTs	1.02	22.47	58.69	13.43
0.25% MWCNTs	1.03	22.81	62.82	14.76
0.5% MWCNTs	1.04	23.50	66.45	16.25
0.75% MWCNTs	1.02	20.19	57.15	11.66
1.0% MWCNTs	1.01	19.09	53.76	10.37

Table S3 Photovoltaic parameters statistics of 40 devices of the perovskite devices with different MWCNTs (W/O, 0.5% and 1.0%) measured under reverse scan (from V_{oc} to 0).

Samples	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
W/O MWCNTs	1.01±0.01	22.14±0.34	57.91±1.02	13.00±0.43
0.5% MWCNTs	1.04±0.01	23.45±0.26	65.84±0.61	15.98±0.27
1.0% MWCNTs	0.99±0.02	18.02±1.32	52.31±1.45	9.28±1.03

Table S4 Summary of photovoltaic performance and ambient stability for various PSCs.

Structure	PCE	Ambient Stability	RH	Ref.
FTO/bl-TiO ₂ /mp-TiO ₂ /mp-ZrO ₂ /Carbon(5-AVAI-CH ₃ NH ₃ PbI ₃)	9.53%	—	—	1
ITO/PEDOT:PSS/CH ₃ NH ₃ PbI _{3-x} Cl _x /PCBM/C ₆₀ /Al	12.98%	80% for 120 h	50%	2
FTO/n-TiO ₂ /CH ₃ NH ₃ PbI _{3-x} Cl _x /P3HT/Au	13.70%	—	35%	3
FTO/n-TiO ₂ /CH ₃ NH ₃ PbI _{3-x} (SCN) _x /spiro-OMeTAD/Au	15.12%	86.7% for 500 h	70%	4
FTO/bl-TiO ₂ /CH ₃ NH ₃ PbI _{3-x} (SCN) _x /spiro-OMeTAD/Au	16.59%	95% for 45 days	70%	5
FTO/nano-TiO ₂ /CH ₃ NH ₃ PbI _{3-x} Cl _x /spiro-OMeTAD/Au	17.56%	70% for 96 h	60%-80%	6
FTO/bl-TiO ₂ /mp-TiO ₂ /(FAPbI ₃) _{1-x} (MAPbBr ₃) _x /spiro-OMeTAD/Au	17.60%	—	—	7
FTO/bl-TiO ₂ /mp-TiO ₂ /Cs _x (FA _{0.83} MA _{0.17})(_{1-x})Pb(I _{0.83} Br _{0.17}) ₃ /Spiro-OMeTAD/Au	20.80%	93.75% for 18 weeks	20%-35%	8
ITO/SnO ₂ /PbI ₂ -FAI:MABr with 0.5% MWCNTs/Carbon	16.25%	93% for 22 weeks	30%-80%	This Work

Reference

1. S. G. Hashmi, D. Martineau, X. Li, M. Ozkan, A. Tiihonen, M. I. Dar, T. Sarikka, S. M. Zakeeruddin, J. Paltakari, P. D. Lund and M. Grätzel, *Adv. Mater. Technol.*, 2017, **2**, 1600183.
2. G. Wang, D. Liu, J. Xiang, D. Zhou, K. Alameh, B. Ding and Q. Song, *RSC Advances*, 2016, **6**, 43299-43303.
3. A. Rapsomanikis, D. Karageorgopoulos, P. Lianos and E. Stathatos, *Sol. Energy Mater. Sol. Cells*, 2016, **151**, 36-43.
4. Q. Tai, P. You, H. Sang, Z. Liu, C. Hu, H. L. Chan and F. Yan, *Nat. Commun.*, 2016, **7**, 11105.
5. Z. Zhang, Y. Zhou, Y. Cai, H. Liu, Q. Qin, X. Lu, X. Gao, L. Shui, S. Wu and J.-M. Liu, *J. Power Sources*, 2018, **377**, 52-58.
6. L. Zhang, X. Zhang, Y. Yu, X. Xu, J. Tang, X. He, J. Wu and Z. Lan, *Solar Energy*, 2017, **155**, 942-950.
7. K. Sveinbjörnsson, K. Aitola, J. Zhang, M. B. Johansson, X. Zhang, J.-P. Correa-Baena, A. Hagfeldt, G. Boschloo and E. M. J. Johansson, *J. Mater. Chem. A*, 2016, **4**, 16536-16545.

8. T. Singh and T. Miyasaka, *Adv. Energy Mater.*, 2018, **8**, 1700677.