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Design of Surface Termination for High-Performance Perovskite Solar Cells

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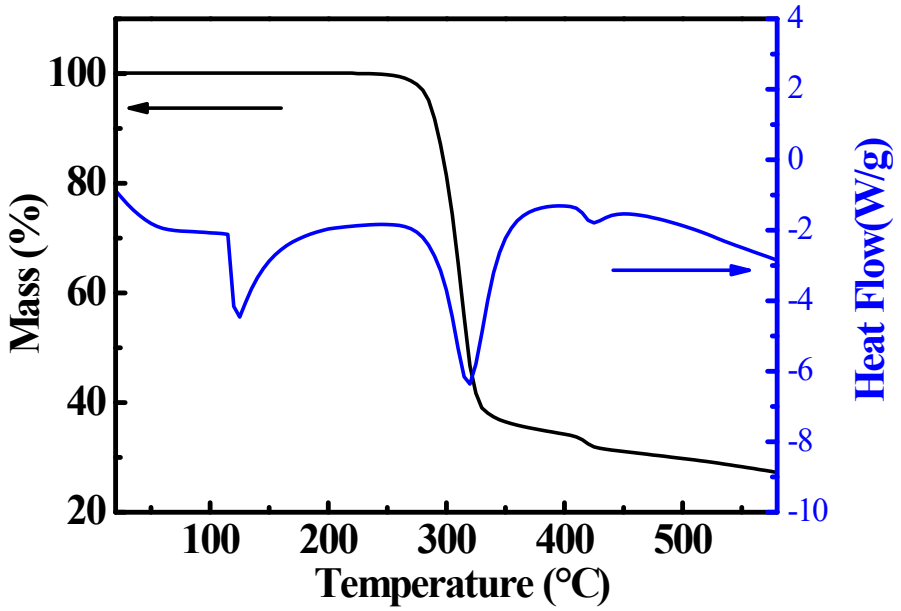

Fig. 1. Thermogravimetric analysis (TGA) of GASCN.

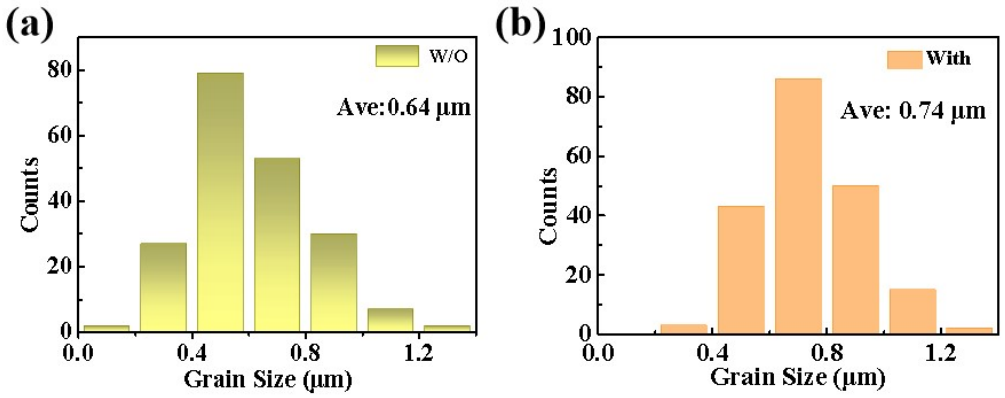

Fig. 2. Corresponding statistical size distributions for the $MAPbI_3$ films with GASCN contents, respectively. All films were deposited on glass/FTO/c-TiO_2 substrates.

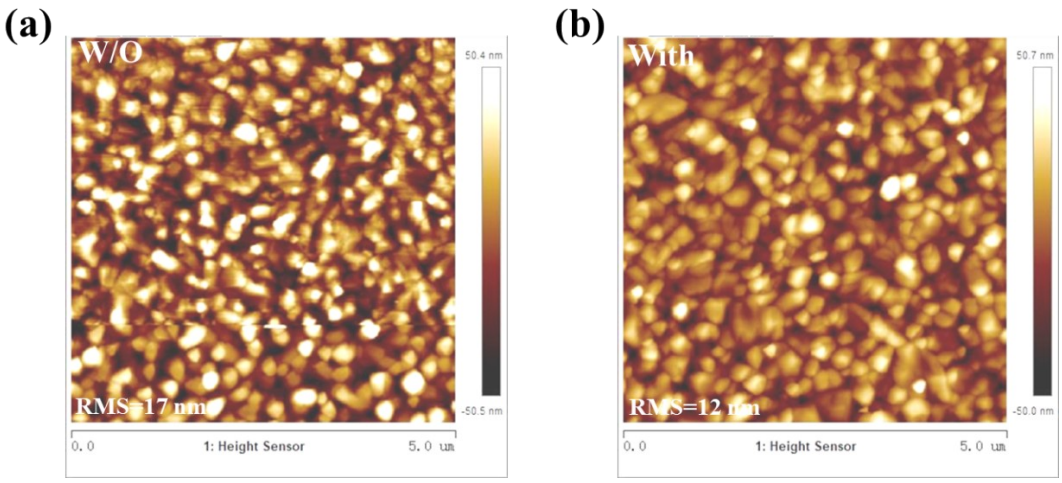

Fig. 3. Atomic force microscopy (AFM) images of the pristine and 0.05mg/ml GASCN treated film. The region of the AFM images is 5 × 5 μm.

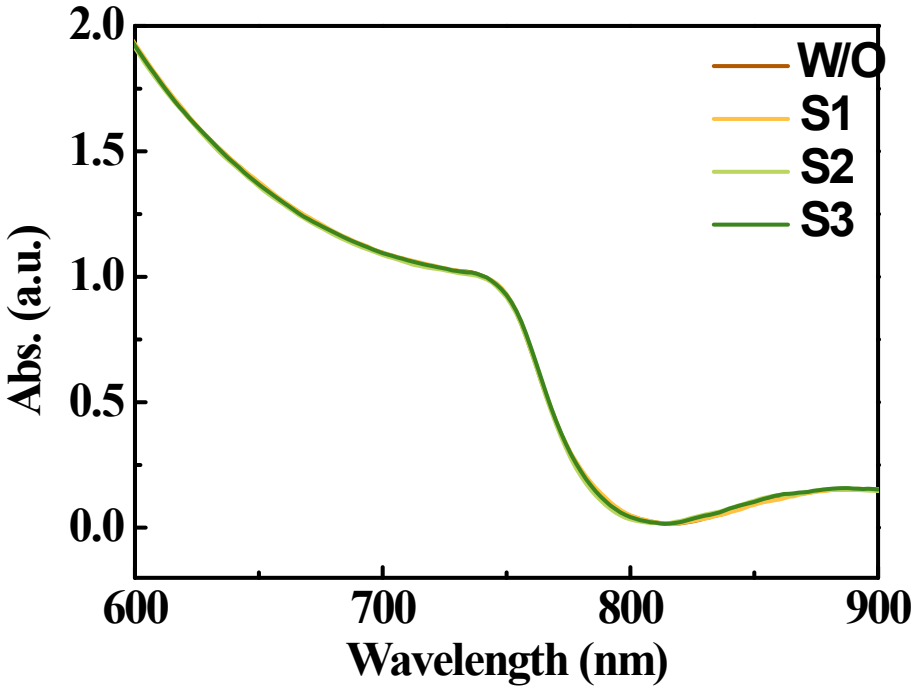

Fig. 4. UV-vis absorbance spectrum of the perovskite films with the different amounts of GASCN modification.

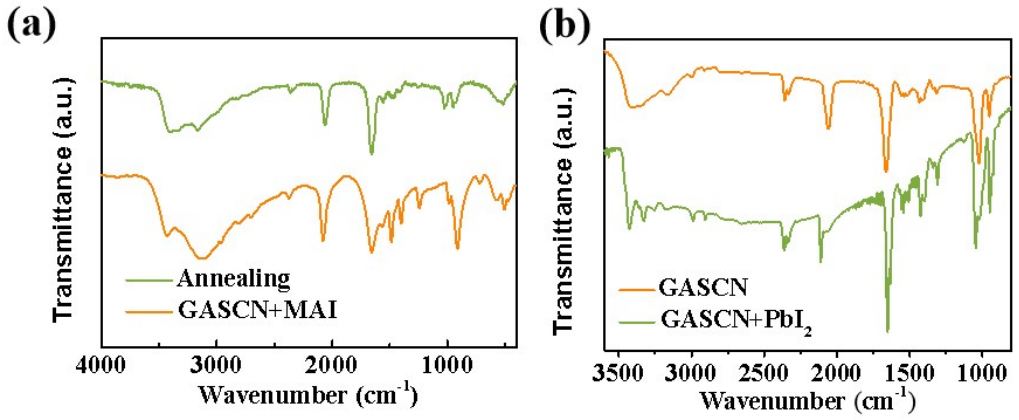

Fig. 5. Fourier transform infrared (FTIR) spectra of (a) GASCN and MAI before and after annealing and (b) GASCN with and without PbI_2 in DMSO solutions.

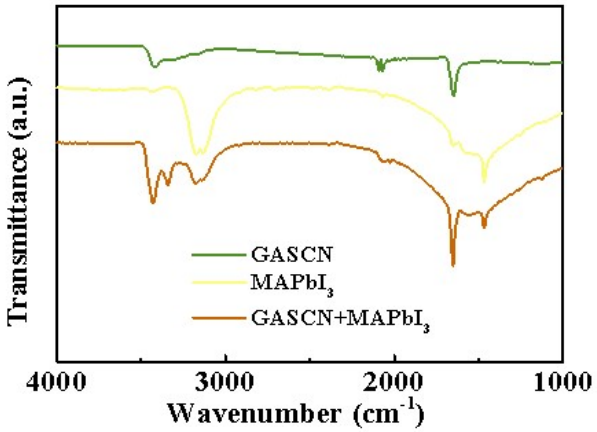

Fig. 6. Fourier transform infrared (FTIR) spectra of different films. All films were deposited on glass/FTO/c-TiO_2 substrates.

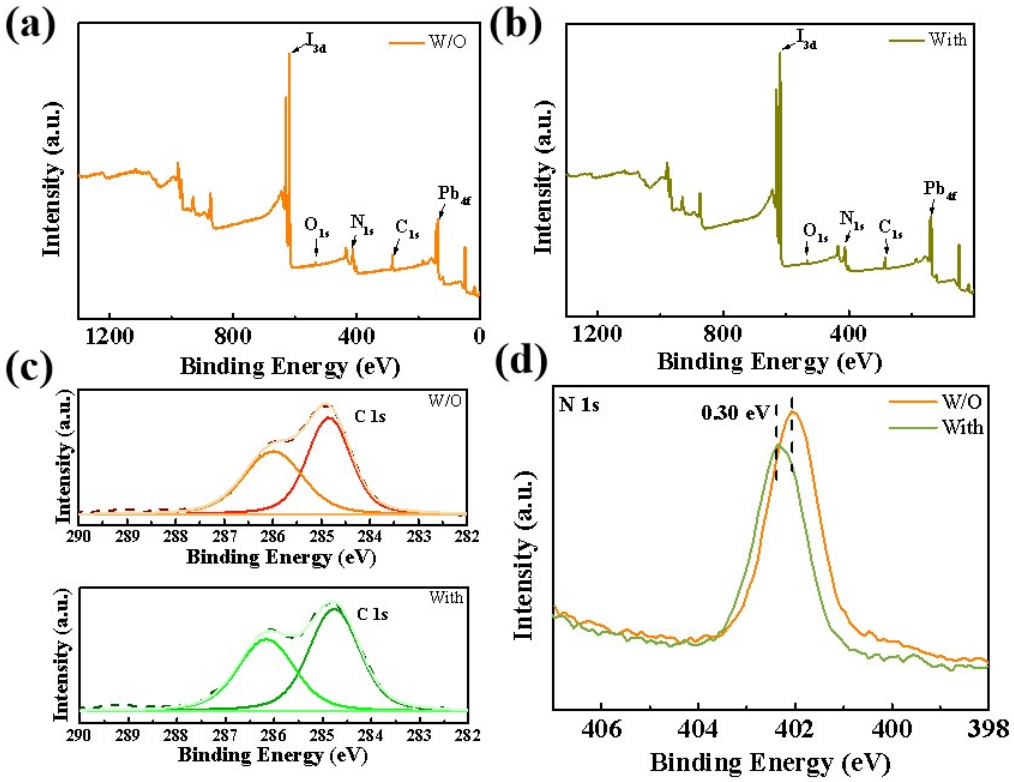

Fig. 6. XPS spectra of (a) pristine film and (b) GASCN treated film, and high resolution XPS of C 1s (c) and (d) N 1s of pristine and GASCN-$MAPbI_3$ films.

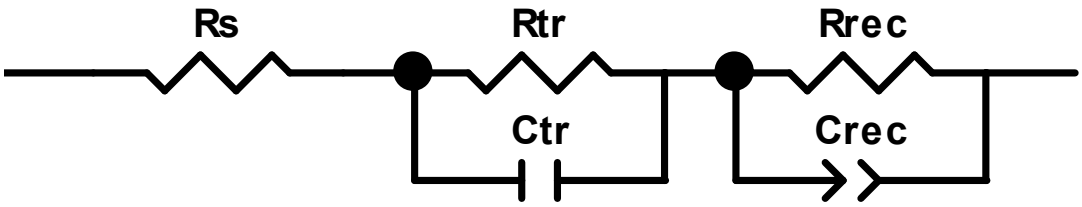

Fig. 7. Equivalent circuit model for the EIS spectra at bias voltage of 1.0 V. R_s, R_{tr}, R_{rec} and C_{rec} are series resistance, transport resistance, recombination resistance and the chemical capacitance, respectively.

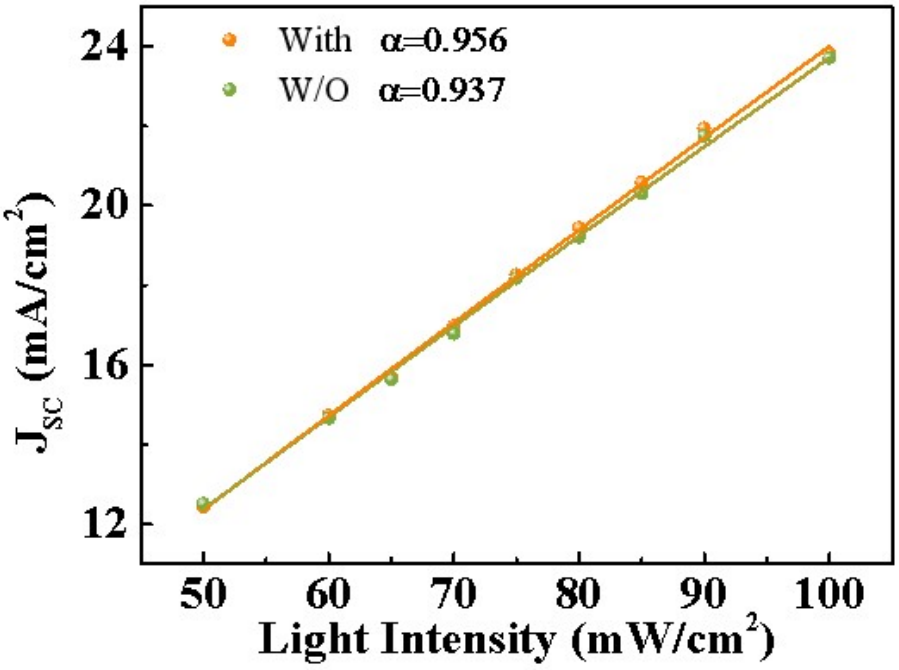

Fig. 8. The J_{sc} as a function of the light intensity for the corresponding devices.

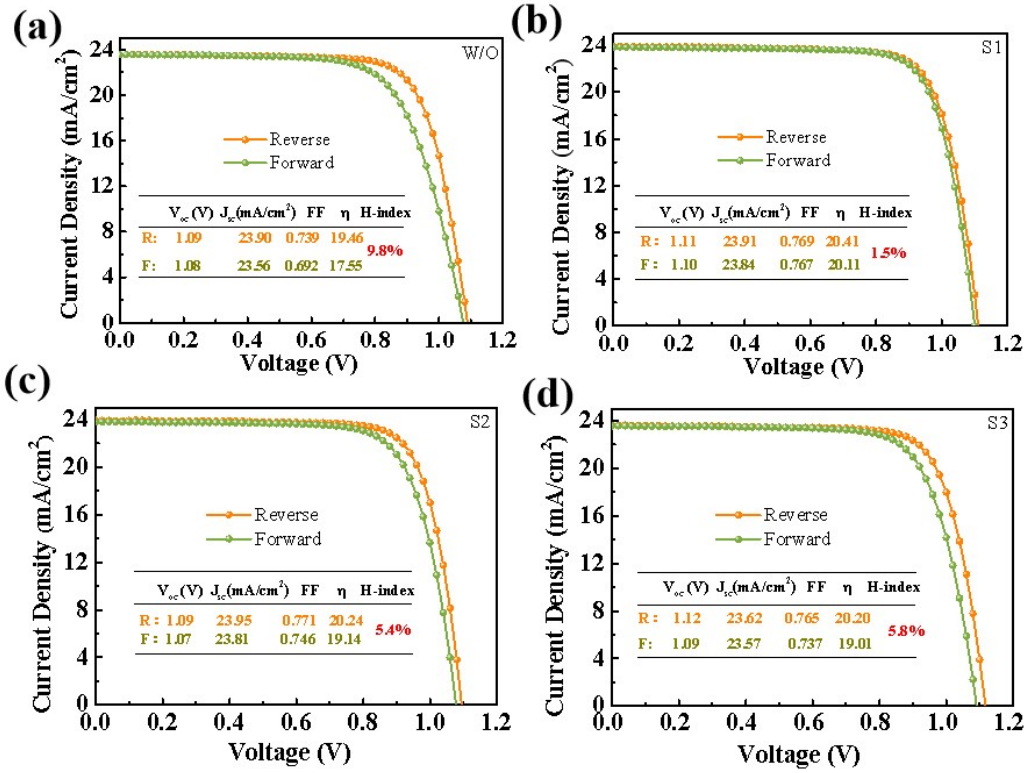

Fig. 9. The *J-V* curves of $MAPbI_3$ perovskite cells without and with GASCN treatment are recorded with forward and reverse voltage scanning directions under AM 1.5G irradiation (100 mW cm^{-2}).

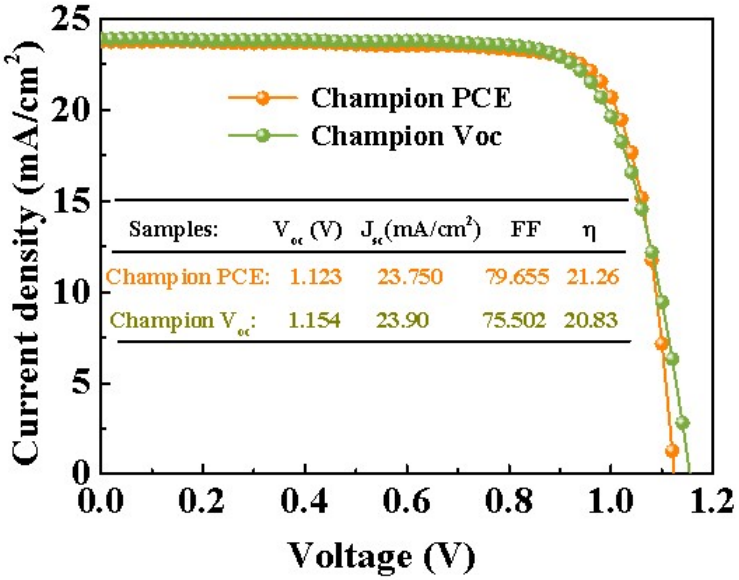

Fig. 11. *J-V* curve of device with the champion V_{OC} and champion PCE measured by reverse scan under standard AM 1.5G irradiation.

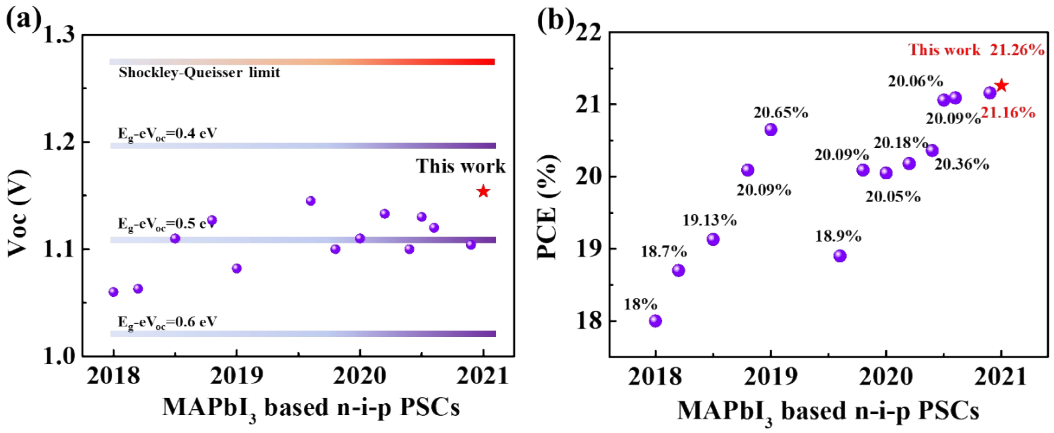

Fig. S12 Photovoltaic parameters statistics of $MAPbI_3$ based n-i-p PCSs in recent three years.

Table 1. Photovoltaic parameters for typical $MAPbI_3$ devices based on different solvent of GASCN with reverse scanning directions. All *J-V* curves were measured under simulated AM 1.5G illumination with a reverse scan rate of 0.15 V s^{-1}.

Sample	V_{oc} (V)	J_{sc} (mA/cm^2)	Fill Factor (%)	PCE (%)
CH	1.112 (1.103±0.007)	23.99 (23.69±0.18)	76.54 (74.92±1.46)	20.23 (19.58±0.48)
CB	1.154 (1.133±0.014)	23.95 (23.81±0.11)	79.65 (77.76±1.14)	21.26 (20.98±0.14)
EA	1.129 (1.100±0.018)	23.98 (23.76±0.17)	75.95 (74.01±1.29)	20.23 (19.35±0.58)
IPA	1.113 (1.083±0.012)	23.99 (23.67±0.16)	74.71 (72.63±1.40)	19.38 (18.61±0.41)

Table 2. Fitted parameters based on TR-PL spectra of the perovskite films.

Sample	τ_1 (ns)	% of τ_1	τ_2 (ns)	% of τ_2	τ_{ave} (ns)
W/O	22.095	37.20	12.808	62.80	22.095
S1	71.545	51.02	33.668	48.98	52.992
S2	65.687	57.86	27.374	42.14	49.540
S3	55.261	51.20	20.975	48.80	38.529

Table 3. Equivalent circuit fitting parameters for the EIS spectra.

Sample	R_s (Ω)	R_{tr} (Ω)	C_{tr} (nF)	R_{rec} (Ω)	C_{rec} (nF)
W/O	33.85	124	38.46	3130	12.18
With	33.36	385	71.08	7885	23.45

Table 4. Literature survey of recent works on polycrystalline $MAPbI_3$-based n-i-p PSCs.

Device Structure	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)	PCE(%)	Ref.
SnO_2/$MAPbI_3$/FAI/HTL	22.99	1.127	77.50	20.09	1
c-TIO_2/m-TIO_2/$MAPbI_3$/AVA/HTL	22.30	1.060	76.00	18.00	2
SnO_2/$MAPbI_3$/$EDBEPbI_4$/HTL	23.17	1.110	73.90	19.13	3
SnO_2/$MAPbI_3$/QA/HTL	23.19	1.130	80.00	21.06	4
c-SnO_2/$MAPbI_3$/HTL	23.98	1.120	78.62	21.09	5
c-SnO_2/$MAPbI_3$/HTL	23.73	1.100	78.00	20.36	6
Li–SnO_2/$MAPbI_3$/HTL	23.24	1.133	76.66	20.18	7
c-TiO_2/SnO_2/$MAPbI_3$/HTL	23.39	1.104	81.90	21.16	8
c-TiO_2/SnO_2/$MAPbI_3$/HTL	22.92	1.110	79.00	20.05	9
c-TiO_2/$MAPbI_{3-x}Cl_x$/HTL	21.91	1.063	80.00	18.70	10
SnO_2/Chol-SnO_2/$MAPbI_3$/HTL	22.80	1.145	72.41	18.90	11
c-TiO_2/$MAPbI_3$/HTL	23.89	1.100	76.40	20.09	12
c-TiO_2/SnO_2/$MAPbI_3$/HTL	23.31	1.082	82.25	20.65	13
TiO_2/$MAPbI_3$/HTL	23.75	1.123 1.154 (highest)	79.66	21.26	This work

Respectively, all HTL in the table refer to Spiro-OMeTAD.

References:

1 Y. Zou, H.-Y. Wang, Y. Qin, C. Mu, Q. Li, D. Xu and J. Zhang, *Adv. Funct. Mater.*, 2019, **29**, 1805810.

2 T. Ye, A. Bruno, G. Han, T. M. Koh, J. Li, N. F. Jamaludin, C. Soci, S. G. Mhaisalkar and W. L. Leong, *Adv. Funct. Mater.*, 2018, **28**, 1801654.

3 P. Li, Y. Zhang, C. Liang, G. Xing, X. Liu, F. Li, X. Liu, X. Hu, G. Shao and Y. Song, *Adv. Mater.*, 2018, **30**, e1805323.

4 Q. He, M. Worku, H. Liu, E. Lochner, A. J. Robb, S. Lteif, J. S. R. Vellore-Winfred, K. Hanson, J. B. Schlenoff, B. J. Kim and B. Ma, *Angew. Chem. Int. Ed.*, 2021, **60**, 2485-2492.

5 Z. Liu, F. Cao, M. Wang, M. Wang and L. Li, *Angew. Chem. Int. Ed.*, 2020, **59**, 4161-4167.

6 W. Hou, G. Han, T. Ou, Y. Xiao and Q. Chen, *Angew. Chem. Int. Ed.*, 2020, **59**, 21409-21413.

7 K. Jung, W.-S. Chae, J. W. Choi, K. C. Kim and M.-J. Lee, *J. Energy Chem.*, 2020, **59**, 755-762.

8 X. Wang, Y. Wang, T. Zhang, X. Liu and Y. Zhao, *Angew. Chem. Int. Ed.*, 2020, **59**, 1469-1473.

9 N. Wei, Y. Chen, Y. Miao, T. Zhang, X. Wang, H. Wei and Y. Zhao, *J. Phys. Chem. Lett.*, 2020, **11**, 8170-8176.

10 F. Wu, X. Yue, Q. Song and L. Zhu, *Sol. RRL.*, 2018, **2**, 1700147.

11 J. Yan, Z. Lin, Q. Cai, X. Wen and C. Mu, *ACS Appl. Energy Mater.*, 2020, **3**, 3504-3511.

12 Z. Yao, W. Zhao, Z. Xu, J. Wen and S. Liu, *Small*, 2020, **16**, e2003582.

13 Y. Wang, H. Zhang, T. Zhang, W. Shi, M. Kan, J. Chen and Y. Zhao, *Sol. RRL*., 2019, **3**, 1900197.