Supporting Information

"Double-Sided Tape" Modifier Bridging TiO_2/Perovskite Buried Interface for Efficient and Stable All-Inorganic Perovskite Solar Cells

Junjie Ti,[a,1] Jingwei Zhu,[a,1] Benlin He,*[a] Zhihao Zong,[a] Xinpeng Yao,[a] Rui Tui,[a] Hao Huang,[d] Cong Chen,[e] Haiyan Chen,[a] Yanyan Duan[c] and Qunwei Tang*[b]

[a] *School of Materials Science and Engineering, Ocean University of China, 238 Songling Road, Qingdao 266100, P.R. China*

[b] *College of Information Science and Technology, Jinan University, 601 Huangpu Avenue West, Guangzhou 510632, P.R. China*

[c] *State Centre for International Cooperation on Designer Low-Carbon and Environmental Material (SCICDLCEM), School of Materials Science and Engineering, Zhengzhou University, Zhengzhou 450001, P.R. China*

[d] *Guangxi Key Laboratory of Processing for Non-ferrous Metals and Featured Materials, School of Resources, Environment and Materials, Guangxi University, Nanning 530004, China*

[e] *College of Materials Science and Engineering & Institute of New Energy and Low-Carbon Technology, Sichuan University, Chengdu 610065, China*

[1] *The authors contributed equally to this work.*

*** Corresponding Authors**. E-mail: blhe@ouc.edu.cn, tangqunwei@jnu.edu.cn

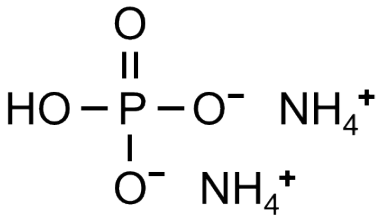

Fig. S1. The chemical structural formula of DAP.

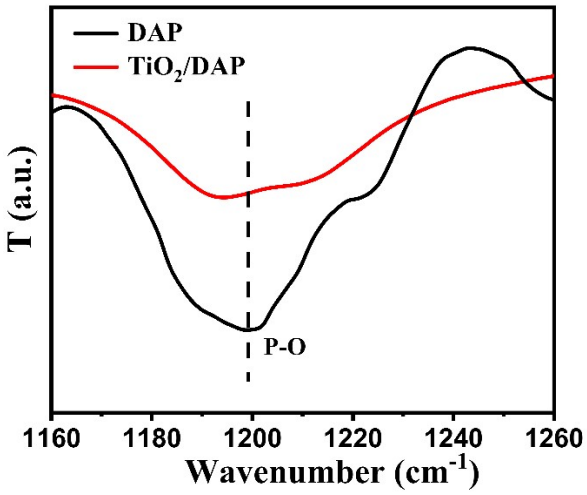

Fig. S2. FTIR spectra of P-O bond for DAP and TiO_2/DAP samples.

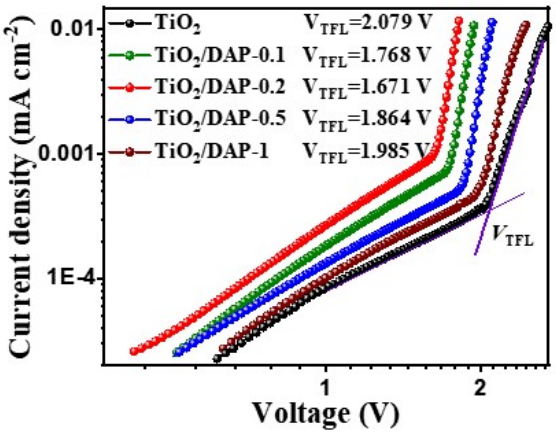

Fig. S3. The dark *J-V* curves of ETL-only devices with a structure of FTO/TiO_2/with and without DAP/Carbon.

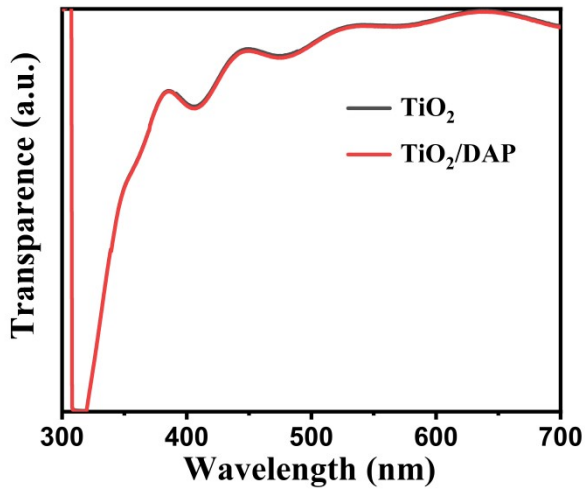

Fig. S4. The transmission spectra of TiO_2 and TiO_2/DAP films.

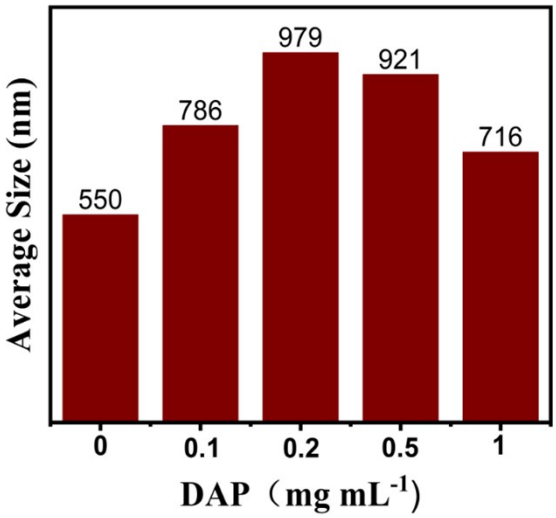

Fig. S5. The average grain size distribution statistical histogram of perovskite films deposited on various TiO_2 layers.

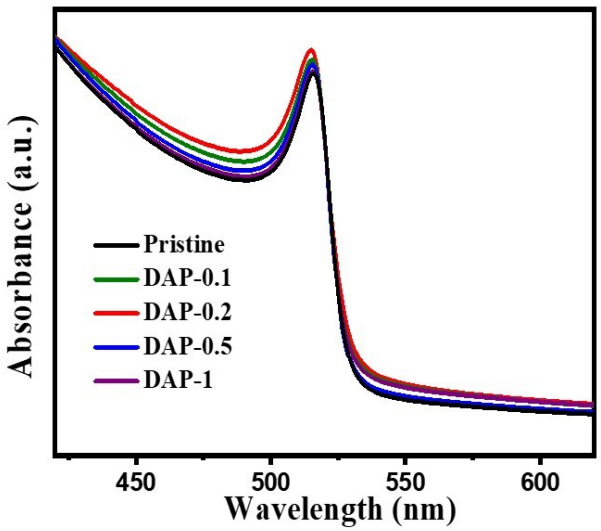

Fig. S6. UV-vis absorption spectrums of perovskite films deposited on various TiO_2 ETLs.

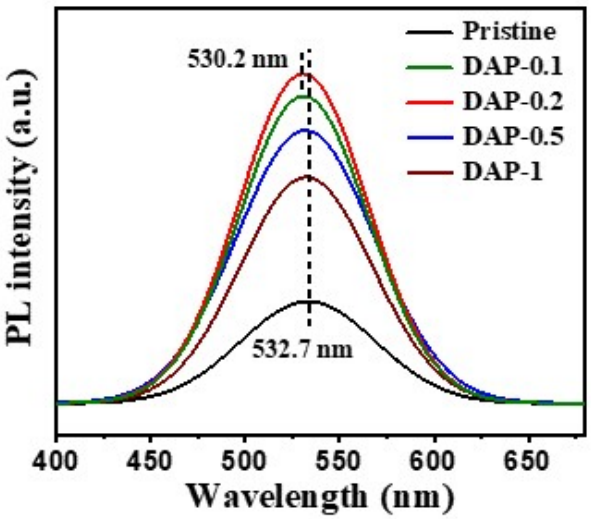

Fig. S7. The steady-state PL spectra excited from air side of perovskite films directly deposited on various glass substrates.

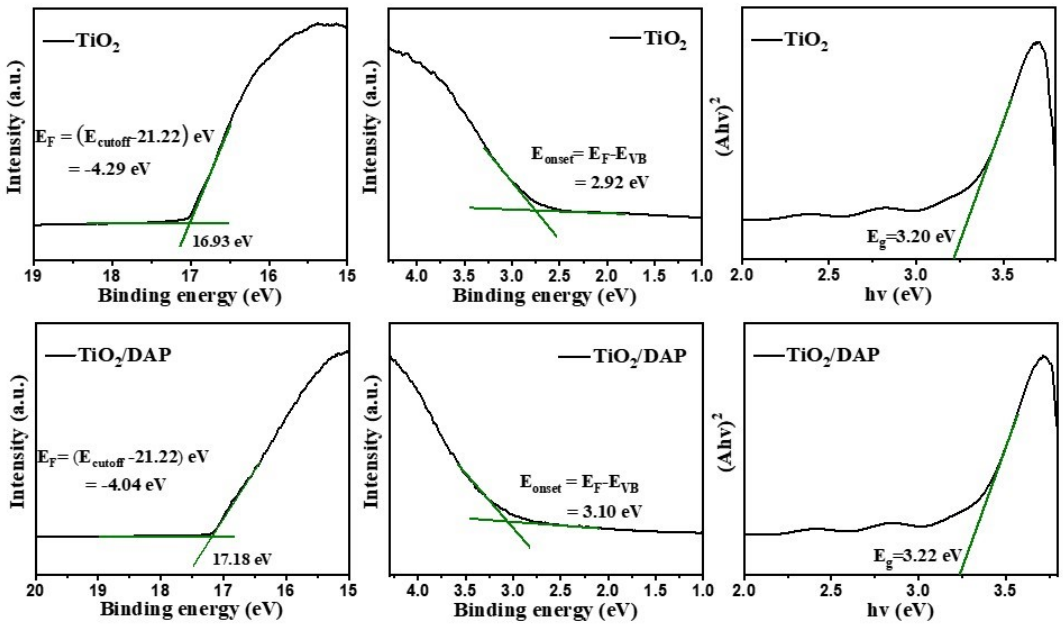

Fig. S8. Ultraviolet photoelectron spectra (UPS) and the curves of $(Ahv)^2$ as a function of hv for various TiO_2 films.

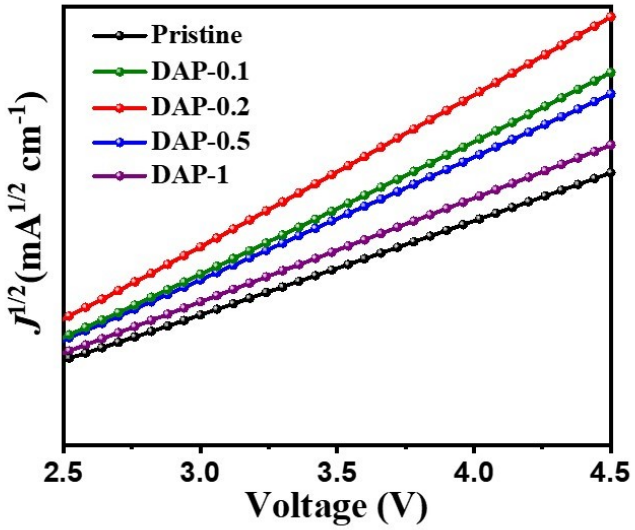

Fig. S9. The $J^{1/2}$-V curves of electron-only devices with a structure of FTO/TiO_2/without or with DAP/perovskite/PCBM/carbon.

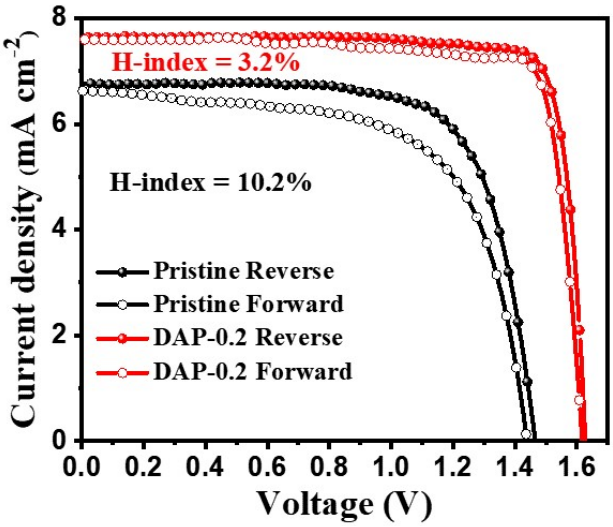

Fig. S10. The *J-V* curves under forward and reverse scan directions of the control and optimized PSCs.

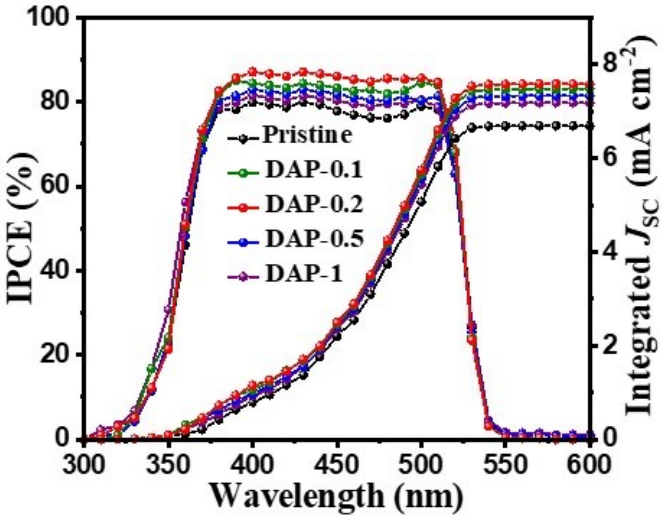

Fig. S11. IPCE spectra and integrated current density of various devices.

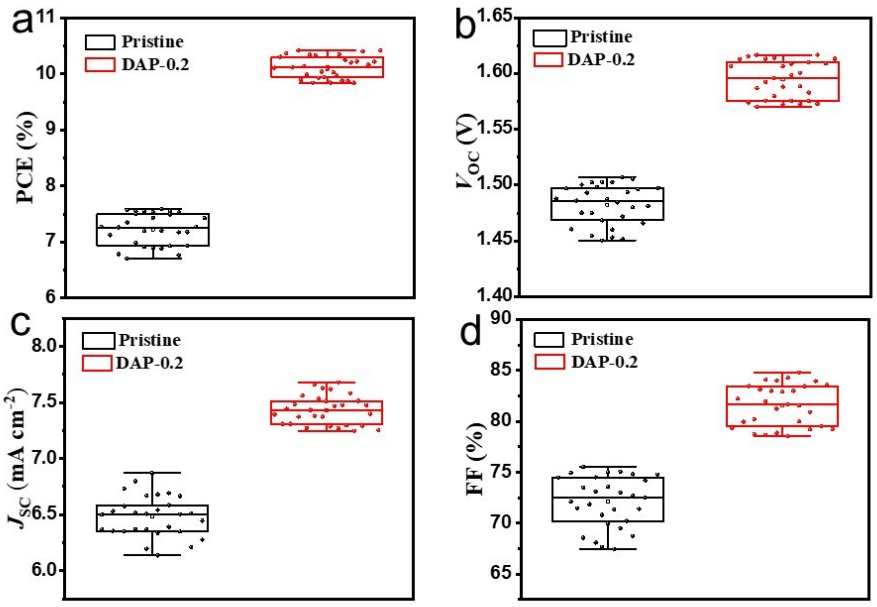

Fig. S12. Statistical distribution of (a) PCE, (b) V_{OC}, (c) J_{SC} and (d) FF for thirty random control and optimized $CsPbBr_3$ PSCs.

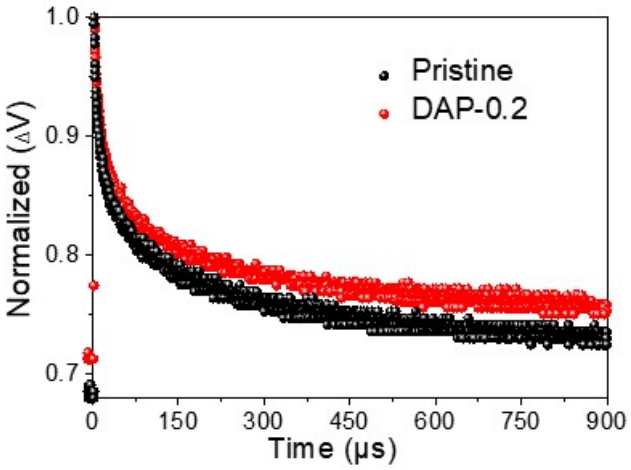

Fig. S13. TPV curves of various $CsPbBr_3$ PSCs under 0.3 sun illumination.

Table S1. The electron trap state density of TiO_2 films with and without DAP modification.

Samples	V_{TFL} (V)	N_t (10^{17} cm^{-3})
Pristine	2.079	5.69
DAP-0.1	1.768	4.76
DAP-0.2	1.671	4.52
DAP-0.5	1.864	5.01
DAP-1	1.985	5.37

Table S2. The electron trap state density and electron mobility of $CsPbBr_3$ films deposited on various TiO_2 ETLs.

Samples	V_{TFL} (V)	N_t (10^{16} cm^{-3})	μ_e (10^{-4} cm^2 V^{-1} s^{-2})
Pristine	1.635	2.06	4.52
DAP-0.1	1.356	1.71	8.41
DAP-0.2	1.291	1.62	11.7
DAP-0.5	1.444	1.83	7.12
DAP-1	1.541	1.95	5.48

Table S3. Summary of the parameters of $CsPbBr_3$ PSCs with champion PCE.

Devices	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	PCE (%)	Ref.
FTO/c-TiO_2/m-TiO_2/DAP/$CsPbBr_3$/Carbon	**7.57**	**1.621**	**84.05**	**10.31**	This work
FTO/c-TiO_2/m-TiO_2/$CsPbBr_3$/ Carbon	7.40	1.220	84.10	7.37	1
FTO/c-TiO_2/m-TiO_2/GQDs/$CsPbBr_3$/ Carbon	8.12	1.458	82.1	9.72	2
FTO/TiO_2/$CsPbBr_3$/SiQDs/spiro-OMeTAD/Ag	7.80	1.420	75.00	8.31	3
TiO_2/$CsPbBr_3$/Cu(Cr,Ba)O_2NCs/ Carbon	7.81	1.620	85.50	10.79	4
FTO/TiO_2/PTI-$CsPbBr_3$/spiro-OMeTAD/Ag	9.78	1.490	74.47	10.91	5
FTO/c-TiO_2/$CsPbBr_3$/$CsPbBr_3$-$CsPb_2Br_5$/$CsPbBr_3$-Cs_4PbBr_6/Carbon	9.24	1.461	75.39	10.17	6
FTO/Sb-TiO_2/$CsPbBr_3$/Carbon	6.70	1.654	80.40	8.91	7
FTO/TiO_2-AC/$CsPbBr_3$/ZnPc/Carbon	7.64	1.606	82.47	10.12	8
FTO/L-TiO_2:$MoSe_2$/$CsPbBr_3$/Carbon	6.70	1.615	78.70	10.02	9
FTO/c-TiO_2/m-TiO_2/$CsPbBr_3$/Carbon	7.13	1.380	62.0	6.10	10
FTO/c-TiO_2/m-TiO_2/Sm^{3+}-$CsPbBr_3$/Carbon	7.48	1.594	85.10	10.14	11

FTO/c-TiO_2/m-TiO_2/Sr^{2+}-$CsPbBr_3$/ Carbon	7.71	1.540	81.10	9.63	12
FTO/TiO_2/CQD-$CsPbBr_3$IO/Spiro-OMeTAD/Au	11.34	1.060	69.00	8.29	13
FTO/c-TiO_2/$CsPbBr_3$/Carbon	6.89	1.49	79.0	8.11	14
FTO/c-TiO_2/$CsPbBr_3$/Ti_3C_2-MXene/ Carbon	8.54	1.444	73.08	9.01	15
FTO/c-TiO_2/$CsPbBr_3$/spiro-OMeTAD/Au	5.60	1.500	62.00	5.40	16
FTO/c-TiO_2/m-TiO_2/$CsPbBr_3$+L-lysine/Carbon	7.64	1.565	81.0	9.68	17
FTO/c-TiO_2/m-TiO_2/Sn^{2+}-$CsPbBr_3$/ Carbon	7.66	1.370	82.22	8.63	18
FTO/TiO_2/$CsPbBr_3$/MnS/Carbon	8.28	1.520	83.00	10.45	19
FTO/c-TiO_2/m-TiO_2/$CsPbBr_3$/[BMMIm]Cl/Carbon	7.45	1.610	83.00	9.92	20

Table S4. The carrier lifetime parameters of perovskite films deposited on various TiO_2 ETLs.

Samples	τ_{ave} (ns)	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)
Pristine	0.78	0.69	54.33	13.95	45.67
DAP-0.1	0.26	0.37	62.85	7.63	37.15
DAP-0.2	0.15	0.22	61.92	5.31	38.08
DAP-0.5	0.34	0.45	63.36	8.74	36.64
DAP-1	0.52	0.61	64.18	10.96	35.82

The TRPL attenuation curve is fitted with a double exponential decay function: $I = A_1e^{-(\tau1-\tau0)/\tau1} + A_2e^{-(\tau1-\tau0)/\tau2}$ to obtain the carrier lifetime, where τ_1 represents the faster defect-related non-radiation recombination, τ_2 represents the slower radiation recombination part.[21] Through the following formula: $\tau_{ave} = (A_1\tau_1^2 + A_2\tau_2^2)/(A_1\tau_1 + A_2\tau_2)$, the average lifetime ($\tau_{ave}$) of photogenerated carriers can be obtained.

Table S5. EIS parameters of $CsPbBr_3$ PSCs with and without DAP modifier.

Devices	R_s (Ω cm^2)	R_{rec} (Ω cm^2)
Pristine	16.37	52.71
DAP-0.1	11.02	91.23
DAP-0.2	6.94	109.82
DAP-0.5	14.06	81.26
DAP-1	14.80	72.83

References

1. D. Huang, P. Xie, Z. Pan, H. Rao and X. Zhong, *J. Mater. Chem. A*, 2019, **7**, 22420-22428.
2. J. Duan, Y. Zhao, B. He and Q. Tang, *Angew. Chem., Int. Ed. Engl.*, 2018, **57**, 3787-3791.
3. S. Zhou, R. Tang, H. Li, L. Fu, B. Li and L. Yin, *J. Power Sources*, 2019, **439**, 227065.
4. J. Duan, Y. Zhao, Y. Wang, X. Yang and Q. Tang, *Angew. Chem., Int. Ed.*, 2019, **58**, 16147-16151.
5. G. Tong, T. Chen, H. Li, L. Qiu, Z. Liu, Y. Dang, W. Song, L. K. Ono, Y. Jiang and Y. Qi, *Nano Energy*, 2019, **65**, 104015.
6. G. Tong, T. Chen, H. Li, W. Song, Y. Chang, J. Liu, L. Yu, J. Xu, Y. Qi and Y. Jiang, *Sol. RRL*, 2019, **3**, 1900030.
7. Y. Xu, J. Duan, X. Yang, J. Du, Y. Wang, Y. Duan and Q. Tang, *J. Mater. Chem. A*, 2020, **8**, 11859-11866.
8. J. Zhu, M. Tang, B. He, W. Zhang, X. Li, Z. Gong, H. Chen, Y. Duan and Q. Tang, *J. Mater. Chem. A*, 2020, **8**, 20987-20997.
9. Q. Zhou, J. Du, J. Duan, Y. Wang, X. Yang, Y. Duan and Q. Tang, *J. Mater. Chem. A*, 2020, **8**, 7784-7791.
10. J. Liu, L. Zhu, S. Xiang, Y. Wei, M. Xie, H. Liu, W. Li and H. Chen, *Sustain. Energ. Fuels*, 2019, **3**, 184-194.
11. J. Duan, Y. Zhao, X. Yang, Y. Wang, B. He and Q. Tang, *Adv. Energy Mater.*,

2018, **8**, 1802346.

12. Y. Zhao, Y. Wang, J. Duan, X. Yang and Q. Tang, *J. Mater. Chem. A*, 2019, **7**, 6877-6882.
13. S. Zhou, R. Tang and L. Yin, *Adv. Mater.*, 2017, **29**, 1703682-1703690.
14. X. Wan, Z. Yu, W. Tian, F. Huang, S. Jin, X. Yang, Y. B. Cheng, A. Hagfeldt and L. Sun, *J. Energy Chem.*, 2020, **46**, 8-15.
15. T. Chen, G. Tong, E. Xu, H. Li, P. Li, Z. Zhu, J. Tang, Y. Qi and Y. Jiang, *J. Mater. Chem. A*, 2019, **7**, 20597-20603.
16. Q. A. Akkerman, M. Gandini, F. Di Stasio, P. Rastogi, F. Palazon, G. Bertoni, J. M. Ball, M. Prato, A. Petrozza and L. Manna, *Nat. Energy*, 2016, **2**, 16194.
17. W. Zhang, X. Liu, B. He, J. Zhu, X. Li, K. Shen, H. Chen, Y. Duan and Q. Tang, *ACS Appl*. *Mater*. *Interfaces*, 2020, **12**, 36092-36101.
18. H. Guo, Y. Pei, J. Zhang, C. Cai, K. Zhou and Y. Zhu, *J. Mater. Chem. C*, 2019, **7**, 11234-11243.
19. X. Li, Y. Tan, H. Lai, S. Li, Y. Chen, S. Li, P. Xu and J. Yang, *ACS Appl. Mater. Interfaces*, 2019, **11**, 29746-29752.
20. W. Zhang, X. Liu, B. He, Z. Gong, J. Zhu, Y. Ding, H. Chen and Q. Tang, *ACS Appl Mater Interfaces*, 2020, **12**, 4540-4548.
21. X. Cao, G. Zhang, Y. Cai, L. Jiang, W. Yang, W. Song, X. He, Q. Zeng, Y. Jia and J. Wei, *Green Chem.*, 2021, **23**, 470-478.