

# Supporting Information for

## Progress in Air-Processed Perovskite Solar Cells: From Crystallization to Photovoltaic Performance

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Table S1 The details device structure, fabrication conditions, device area, and photovoltaic parameters of OHP solar cells listed in Figure 9.

Lab-scale OHP solar cells								
Device Structure	Method for perovskite	Processing Environment	Device Area (cm <sup>2</sup> )	J <sub>sc</sub> (mA/cm <sup>2</sup> )	V <sub>oc</sub> (V)	FF (%)	PCE (%)	Ref
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> (Dye-sensitized solar cell)	Spin-coating	/	0.24	11.0	0.61	0.57	3.81	[3]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> (Dye-sensitized solar cell)	Spin-coating	/	0.309	15.82	0.706	0.59	6.54	[97]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Spiro-MeTAD/Au	Spin-coating & dip coating	Glovebox	0.209	21.3	1.00	0.66	14.1	[10]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PTAA /Au	Spin-coating	Glovebox	0.16	21.3	1.04	0.73	16.2	[98]
FTO/TiO <sub>2</sub> /(FAPbI <sub>3</sub> ) <sub>1-x</sub> (MAPbBr <sub>3</sub> ) <sub>x</sub> /PTAA/Au	Spin-coating	Glovebox	0.16	24.6	1.06	0.77	20.1	[99]
FTO/TiO <sub>2</sub> /FAPbI <sub>3</sub> /PTAA/Au	Spin-coating	Glovebox	0.0946	25.0	1.1	0.80	22.1	[10]
ITO/SnO <sub>2</sub> /FA <sub>1-x</sub> MA <sub>x</sub> PbI <sub>3</sub> /PEAI/Spiro-OMeTAD/Au	Spin-coating	Glovebox	0.108	25.2	1.18	0.78	23.3	[100]
/	/	/	/	/	/	/	24.2*	[10]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /P3HT/Ag	Spin-coating	In air with RH 50%	0.09	18.85	0.64	0.41	5.67	[90]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Carbon/PANI	Spin-coating	In air	/	13.6	0.65	0.45	4.00	[92]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Spiro-OMeTAD/Ag	Low-pressure chemical vapor deposition	In air with RH 60%	0.12	21.7	0.91	0.65	12.73	[91]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> (SCN) <sub>x</sub> /Spiro-OMeTAD/Au	Spin-coating	In air with RH 70%	/	21.1	0.96	0.75	15.12	[54]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Spiro-OMeTAD/Au	Spin-coating	In air with RH 35%	0.1256	19.01	1.04	0.73	14.55	[51]
FTO/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Spiro-OMeTAD/Au	Multi-flow air knife	In air with RH 40%	0.1	23.50	1.09	0.69	17.71	[93]
ITO/PolyTPD/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /C <sub>60</sub> /BCP/Ag	Spin-coating	In air with RH 70%	0.1	23.03	1.05	0.75	18.1	[33]
ITO/m-PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Ca/Al	Blowing assisted drop-casting	In air	0.1	22.64	1.11	0.77	19.48	[59]
ITO/ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Spiro-OMeTAD/Ag	Spin-coating	In air with RH 55-65%	0.04	22.17	1.07	0.77	18.34	[52]
ITO/PolyTPD/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /C <sub>60</sub> /BCP/Ag	Spin-coating	In air with RH 70%	0.1	23.36	1.00	0.67	15.56	[50]
FTO/TiO <sub>2</sub> /meso-TiO <sub>2</sub> /Cs <sub>0.05</sub> (FA <sub>0.83</sub> MA <sub>0.17</sub> ) <sub>0.95</sub> Pb(I <sub>0.83</sub> Br <sub>0.17</sub> ) <sub>3</sub> /Spiro-OMeTAD/Au	Spin-coating	In air with RH 20-35%	0.25	23.6	1.14	0.77	20.8	[44]
ITO/SnO <sub>2</sub> /MAPbI <sub>3</sub> /Spiro-OMeTAD/Au	Air blading	In air	0.09	23.46	1.09	0.79	20.08	[94]
ITO/SnO <sub>2</sub> /MAPbI <sub>3</sub> /Spiro-OMeTAD/Au	Air blading	In air with RH 60%	0.1	22.97	1.06	0.80	19.39	[95]
ITO/SnO <sub>2</sub> /Cs <sub>0.21</sub> FA <sub>0.56</sub> MA <sub>0.23</sub> (I <sub>0.98</sub> Br <sub>0.02</sub> ) <sub>3</sub>	Spin-coating	In air with RH 40%	/	23.37	1.11	0.71	18.38	[96]

/Spiro-OMeTAD/Ag								
<b>Air processed large-scale OHP solar cells</b>								
ITO/PEDOT:PSS/ CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /PCBM/Ca/Al	Spray coating	In air	/	16.8	0.92	0.72	11.1	[63]
FTO/TiO <sub>2</sub> / CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> / Spiro-OMeTAD/Ag	Spray coating	In air	0.065	20.6	1.03	0.62	13	[58]
FTO/TiO <sub>2</sub> / CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> / Spiro-OMeTAD/Au	Spray coating	In air	1	18.59	1.03	0.68	13.09	[66]
FTO/TiO <sub>2</sub> /ZrO <sub>2</sub> / (5-AVA),(MA) <sub>1-x</sub> PbI <sub>3</sub> /Carbon	Printing	In air	0.5	22.8	0.86	0.66	12.8	[101]
FTO/TiO <sub>2</sub> /ZrO <sub>2</sub> /Carbon/MAPbI <sub>3</sub> (module)	Printing	In air	70	1.77	9.63	0.63	10.74	[103]
FTO/compact-TiO <sub>2</sub> /(meso-TiO <sub>2</sub> /meso-ZrO <sub>2</sub> /meso-carbon)/(5-AVA) <sub>x</sub> (MA) <sub>1-x</sub> PbI <sub>3</sub> (module)	Printing	In air	49	2.0	9.3	0.56	10.4	[56]
FTO/TiO <sub>2</sub> /Cs <sub>0.1</sub> (FA <sub>0.83</sub> MA <sub>0.17</sub> ) <sub>0.9</sub> Pb(I <sub>0.83</sub> Br <sub>0.17</sub> ) <sub>3</sub> /Spiro-OMeTAD/Au	Printing	In air with RH 45%	0.09	21.5	1.06	0.67	15.3	[104]
ITO/ZnO/MAPbI <sub>3</sub> /P3HT/Ag	Slot-die	In air with RH 30-40%	0.1	20.38	0.98	0.60	11.96	[71]
ITO/ZnO/MAPbI <sub>3</sub> / Bifluo-OMeTAD/MoO <sub>3</sub> /Ag	Slot-die	In air with RH 30-40%	0.1	19.7	1.1	0.68	14.7	[105]
ITO/ZnO/MAPbI <sub>3</sub> /P3HT/MoO <sub>3</sub> /Ag	Slot-die	In air	0.1	17.21	1.1	0.67	12.7	[106]
FTO/TiO <sub>2</sub> / CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> / Spiro-OMeTAD/Au	Doctor blading	In air	10.1	4.3	4.11	0.58	10.3	[107]
FTO/Graphene-TiO <sub>2</sub> / CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> / Spiro-OMeTAD/Au (module)	Doctor blading	In air	50.6	2.26	8.57	0.65	12.6	[108]
FTO/TiO <sub>2</sub> -PCBM/ CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> / Spiro-OMeTAD/Au	Doctor blading	In air with RH 10-20%	1.2	21.38	1.11	0.73	17.33	[109]
ITO/NiO <sub>x</sub> /MAPbI <sub>3</sub> /PC <sub>61</sub> BM/Ag	Doctor blading	In air with RH 40%	0.09	17.6	1.02	0.61	10.92	[110]
ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /PCBM/ZnO/Ag	R2R	In air	0.5	13.7	0.98	0.33	5.1	[111]
PET/TCO/ZnO/FA <sub>0.4</sub> MA <sub>0.6</sub> PbI <sub>3</sub> /PEDOT/MoO <sub>3</sub> /Ag	R2R	In air with RH 30-40%	0.1	19.6	1.04	0.54	11.0	[112]
ITO/m-PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Ca/Al	R2R	In air with RH 45%	0.1	17.39	0.99	0.65	11.16	[59]

\*The certified PCE obtained from NREL Efficiency chart, <https://www.nrel.gov/pv/cell-efficiency.html>, Access on April 17, 2019

## Reference

- [3] A. Kojima, K. Teshima, Y. Shirai and T. Miyasaka, *Journal of the American Chemical Society*, 2009, **131**, 6050-6051.
- [10] NREL Efficiency chart, <https://www.nrel.gov/pv/cell-efficiency.html>, Access on April 17, 2019.
- [33] Y. Cheng, X. Xu, Y. Xie, H.-W. Li, J. Qing, C. Ma, C.-S. Lee, F. So and S.-W. Tsang, *Solar RRL*, 2017, 1, 1700097.
- [44] T. Singh and T. Miyasaka, *Advanced Energy Materials*, 2018, 8, 1700677.

- [50] X. Xu, C. Ma, Y.-M. Xie, Y. Cheng, Y. Tian, M. Li, Y. Ma, C.-S. Lee and S.-W. Tsang, *Journal of Materials Chemistry A*, 2018, 6, 7731-7740.
- [51] J. Huang, X. Yu, J. Xie, D. Xu, Z. Tang, C. Cui and D. Yang, *ACS applied materials & interfaces*, 2016, 8, 21505-21511.
- [52] W.-T. Wang, J. Sharma, J.-W. Chen, C.-H. Kao, S.-Y. Chen, C.-H. Chen, Y.-C. Feng and Y. Tai, *Nano Energy*, 2018, 49, 109-116.
- [54] Q. Tai, P. You, H. Sang, Z. Liu, C. Hu, H. L. W. Chan and F. Yan, *Nature communications*, 2016, 7, 11105.
- [56] Y. Hu, S. Si, A. Mei, Y. Rong, H. Liu, X. Li and H. Han, *Solar RRL*, 2017, 1, 1600019.
- [58] S. Das, B. Yang, G. Gu, P. C. Joshi, I. N. Ivanov, C. M. Rouleau, T. Aytug, D. B. Geohegan and K. Xiao, *ACS Photonics*, 2015, 2, 680-686.
- [59] C. Zuo, D. Vak, D. Angmo, L. Ding and M. Gao, *Nano Energy*, 2018, 46, 185-192.
- [63] A. T. Barrows, A. J. Pearson, C. K. Kwak, A. D. F. Dunbar, A. R. Buckley and D. G. Lidzey, *Energy & Environmental Science*, 2014, 7, 2944-2950.
- [66] H. Huang, J. Shi, L. Zhu, D. Li, Y. Luo and Q. Meng, *Nano Energy*, 2016, 27, 352-358.
- [71] K. Hwang, Y.-S. Jung, Y.-J. Heo, F. H. Scholes, S. E. Watkins, J. Subbiah, D. J. Jones, D.-Y. Kim and D. Vak, *Advanced Materials*, 2015, **27**, 1241-1247.
- [90] M. Seetharaman S, P. Nagarjuna, P. N. Kumar, S. P. Singh, M. Deepa and M. A. G. Namboothiry, *Physical Chemistry Chemical Physics*, 2014, 16, 24691-24696.
- [91] P. Luo, Z. Liu, W. Xia, C. Yuan, J. Cheng and Y. Lu, *ACS applied materials & interfaces*, 2015, 7, 2708-2714.
- [92] P. Bhatt, K. Pandey, P. Yadav, B. Tripathi, C. K. P, M. K. Pandey and M. Kumar, *Solar Energy Materials and Solar Cells*, 2015, 140, 320-327.
- [93] L.-L. Gao, C.-X. Li, C.-J. Li and G.-J. Yang, *Journal of Materials Chemistry A*, 2017, 5, 1548-1557.
- [94] J. Ding, Q. Han, Q.-Q. Ge, D.-J. Xue, J.-Y. Ma, B.-Y. Zhao, Y.-X. Chen, J. Liu, D. B. Mitzi and J.-S. Hu, *Joule*, 2019, 3, 402-416.
- [95] R. Cheng, C.-C. Chung, H. Zhang, Z. Zhou, P. Zhai, Y.-T. Huang, H. Lee and S.-P. Feng, *Small*, 2019, 15, 1804465.
- [96] K. Huang, H. Li, C. Zhang, Y. Gao, T. Liu, J. Zhang, Y. Gao, Y. Peng, L. Ding and J. Yang, *Solar RRL*, 2019, 3, 1800318.
- [97] J.-H. Im, C.-R. Lee, J.-W. Lee, S.-W. Park and N.-G. Park, *Nanoscale*, 2011, **3**, 4088-4093.
- [98] S. Ryu, J. H. Noh, N. J. Jeon, Y. Chan Kim, W. S. Yang, J. Seo and S. I. Seok, *Energy & Environmental Science*, 2014, 7, 2614-2618.
- [99] W. S. Yang, J. H. Noh, N. J. Jeon, Y. C. Kim, S. Ryu, J. Seo and S. I. Seok, *Science*, 2015, 348, 1234-1237.
- [100] Q. Jiang, Y. Zhao, X. Zhang, X. Yang, Y. Chen, Z. Chu, Q. Ye, X. Li, Z. Yin and J. You, *Nature Photonics*, 2019, DOI: 10.1038/s41566-019-0398-2.
- [101] A. Mei, X. Li, L. Liu, Z. Ku, T. Liu, Y. Rong, M. Xu, M. Hu, J. Chen, Y. Yang, M. Grätzel and H. Han, *Science*, 2014, 345, 295-298.
- [103] A. Priyadarshi, L. J. Haur, P. Murray, D. Fu, S. Kulkarni, G. Xing, T. C. Sum, N. Mathews and S. G. Mhaisalkar, *Energy & Environmental Science*, 2016, 9, 3687-3692.
- [104] F. Mathies, H. Eggers, B. S. Richards, G. Hernandez-Sosa, U. Lemmer and U. W. Paetzold, *ACS Applied Energy Materials*, 2018, 1, 1834-1839.
- [105] T. Qin, W. Huang, J.-E. Kim, D. Vak, C. Forsyth, C. R. McNeill and Y.-B. Cheng, *Nano Energy*,

2017, 31, 210-217.

- [106] J.-E. Kim, Y.-S. Jung, Y.-J. Heo, K. Hwang, T. Qin, D.-Y. Kim and D. Vak, Solar Energy Materials and Solar Cells, 2018, 179, 80-86.
- [107] S. Razza, F. Di Giacomo, F. Matteocci, L. Cinà, A. L. Palma, S. Casaluci, P. Cameron, A. D'Epifanio, S. Licoccia, A. Reale, T. M. Brown and A. Di Carlo, Journal of Power Sources, 2015, 277, 286-291.
- [108] A. Agresti, S. Pescetelli, A. L. Palma, A. E. Del Rio Castillo, D. Konios, G. Kakavelakis, S. Razza, L. Cinà, E. Kymakis, F. Bonaccorso and A. Di Carlo, ACS Energy Letters, 2017, 2, 279-287.
- [109] M. Yang, Z. Li, M. O. Reese, O. G. Reid, D. H. Kim, S. Siol, T. R. Klein, Y. Yan, J. J. Berry, M. F. A. M. van Hest and K. Zhu, Nature Energy, 2017, 2, 17038.
- [110] Y. Peng, Y. Cheng, C. Wang, C. Zhang, H. Xia, K. Huang, S. Tong, X. Hao and J. Yang, Organic Electronics, 2018, 58, 153-158.
- [111] Z. Gu, L. Zuo, T. T. Larsen-Olsen, T. Ye, G. Wu, F. C. Krebs and H. Chen, Journal of Materials Chemistry A, 2015, 3, 24254-24260.
- [112] Y.-J. Heo, J.-E. Kim, H. Weerasinghe, D. Angmo, T. Qin, K. Sears, K. Hwang, Y.-S. Jung, J. Subbiah, D. J. Jones, M. Gao, D.-Y. Kim and D. Vak, Nano Energy, 2017, 41, 443-451.