Supplement Information

Solvent Annealing of PbI₂ for High-Quality Crystallization of Perovskite Films

for Solar Cells with Efficiency Exceeding 18%

Yafei Wang^a, Shibin Li^{a*}, Peng Zhang^a, Detao Liu^a, Xiangling Gu^a, Hojjatollah Sarvari^b, Zhongbiao Ye^{a,b}, Jiang Wu^c, Zhiming Wang^d, and Zhi David Chen^{a,b*}

^a State Key Laboratory of Electronic Thin Films and Integrated Devices, and School of

Optoelectronic Information, University of Electronic Science and Technology of China

(UESTC), Chengdu, Sichuan 610054, China

^b Department of Electrical & Computer Engineering, and Center for Nanoscale Science & Engineering, University of Kentucky, Lexington, Kentucky 40506, USA

^c Department of Electronic and Electrical Engineering, University College London, Torrington Place, London WC1E 7JE,United Kingdom

^d Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China, Chengdu, 610054, China

*Corresponding authors: shibinli@uestc.edu.cn, <u>zhichen@engr.uky.edu</u>

In this investigation, using the DMSO SA-TA process, a series of experiment parameters were studied to optimize the performance of solar cells. At first, we studied the TA-TA, DMSO SA-TA, DMSO SA-SA as shown in Figure S1, because the TA-SA process did not result in good results as described in the text. We found that the DMSO SA-TA annealing process is the best among TA-TA and DMSO SA-SA processes. This result is consistent with our previous conclusion using DMF in the text. It should be noted that we used the same petri dish. We studied the effect of different DMSO volumes on the device performance as shown in Figure S2. We found negligible difference in PCE by using different volumes of DMSO solution. We chose the volume of 50 μ l in our subsequent experiments. The diameter of m-TiO₂ and the spin speed of PbI₂ were also investigated (Figure S3 and Figure S4). It can be seen in Figure S3 that m-TiO2 with the diameter of 30 nm results in better PCE. The first spin speed of PbI₂ significantly affected the device performance. As shown in Figure S4, the first spin speeds of 2500 rpm, 3000 rpm, and 3500 rpm resulted in PCEs of 14%, 15.7%, AND 17.1% separately. In addition, we studied thermal annealing (TA) of PbI₂ and the DMSO annealing of PbI₂ at 100°C for 10 min. After thermal annealing at 100°C, PbI₂ exhibits smooth and dense surface, while after annealing PbI₂ in DMSO at 100°C we obtained a surface that looks like needles but are actual many PbI₂ nanowires as shown in Figure S5. This suggests that lower temperature DMSO annealing is better for porous PbI₂.

Figure.S1 J-V curves and device performance of devices fabricated using different annealing processes (TA-TA, DMSO SA-TA and DMSO SA-SA).



Figure.S2 J-V curves and device performance of solar cells fabricated using different volumes of DMSO solvent in the DMSO SA-TA process.



Figure.S3 J-V curves and device performance of devices fabricated using $m-TiO_2$ with different diameters.



Figure.S4 J-V curves and device performance of devices fabricated using different spin-coating speeds for PbI₂.



Figure.S5 SEM images of Surface morphology of PbI_2 films obtained using different annealing processes at 100 °C.

