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Supplementary Information

Dramatically promoted crystallization control of organolead triiodide perovskite film by a homogeneous cap for high efficiency planar-heterojunction solar cells

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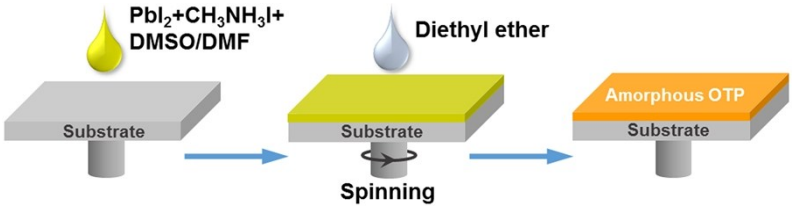

Fig. S1 Schematically illustrating the experimental processes for preparing precursor OTP film via anti-solvent assisted one-step spin-coating route reported by Park et al.[1].

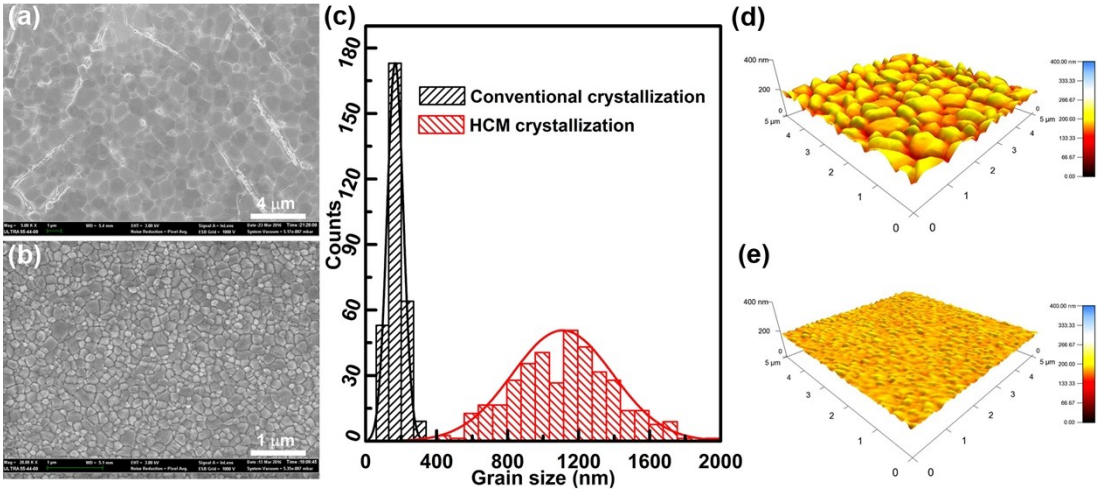

Fig. S2 Top-view SEM images used for the statistics of the grain size distribution of (a) HCM crystallized OTP film and (b) control sample obtained by conventional crystallization, respectively. (c) The corresponding histograms of statistic grain size distribution. (d) and (e) The corresponding AFM topographical images with the scanning area of 5×5 μm^2.

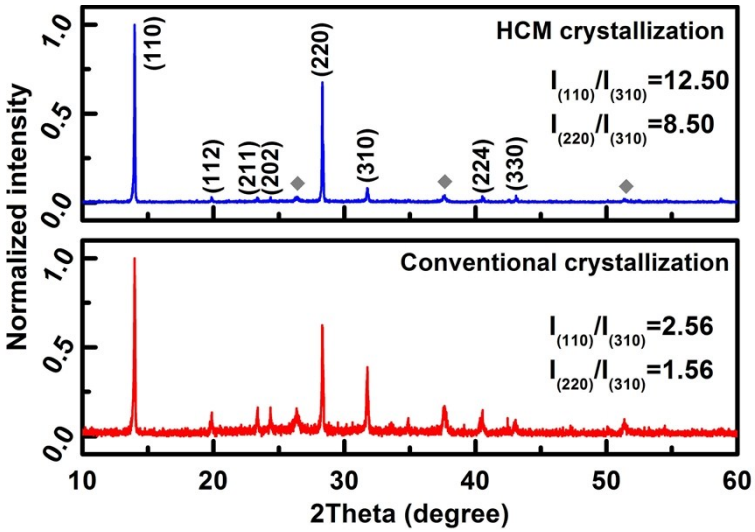

Fig. S3 Normalized intensity XRD spectra of HCM crystallized OTP film and control sample obtained by conventional crystallization. The solid diamond symbols correspond to the signals from TiO_2/FTO substrate. The insert gives the peak intensity ratios for (110) to (310) planes and (220) to (310) planes.

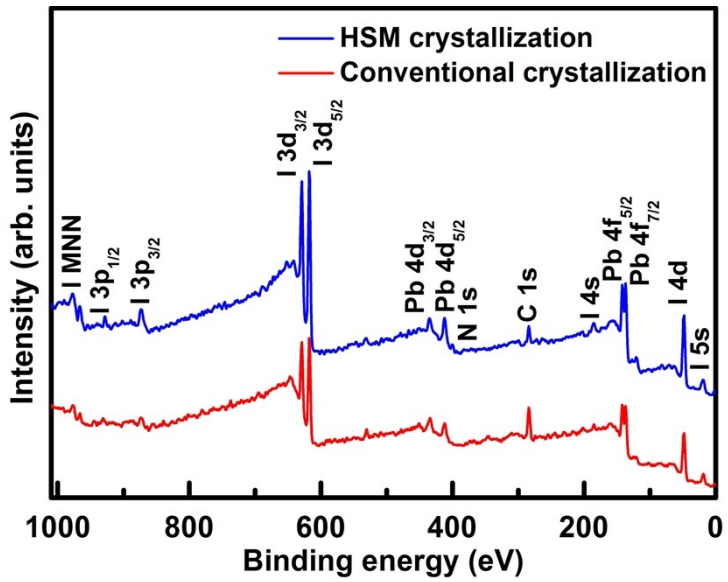

Fig. S4 Low-resolution XPS survey spectra of HCM crystallized OTP film and control sample obtained by conventional crystallization, respectively.

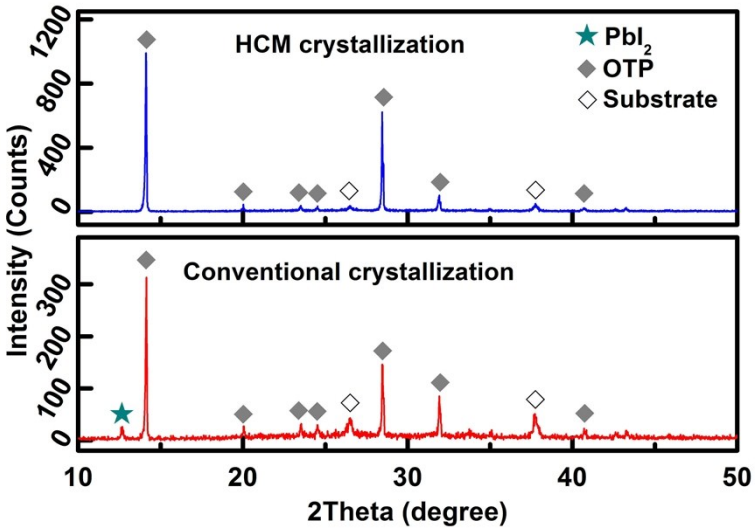

Fig. S5 XRD spectra of HCM crystallized OTP film and control sample obtained by conventional crystallization with the thermal annealing temperature and time to be 150 °C and 40 min. The solid diamond symbols signify the diffraction peaks of OTP with the solid star symbol denoting the diffraction peak of PbI_2 and the hollow diamond symbol denoting the signals of TiO_2/FTO substrate.

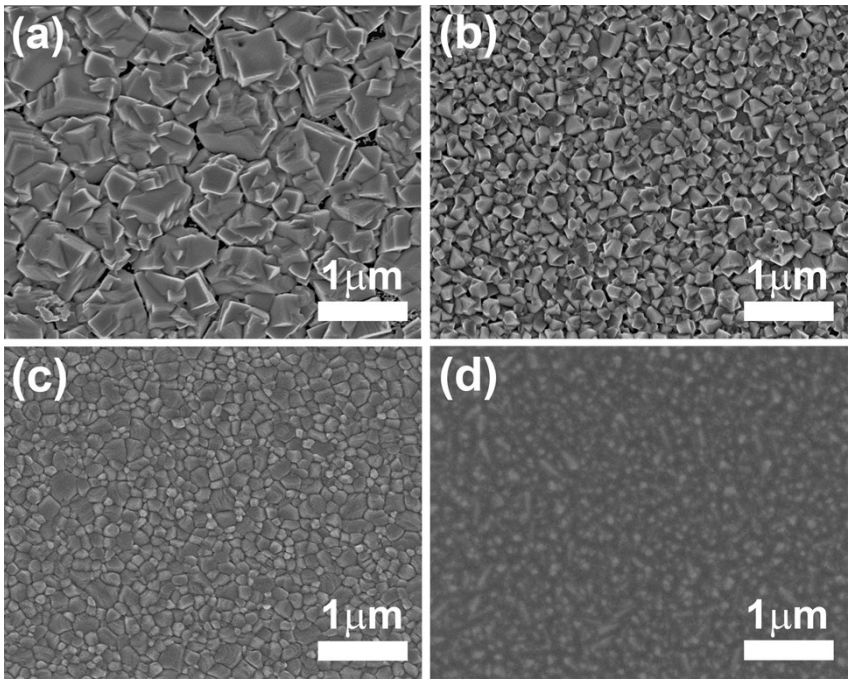

Fig. S6 (a-d) Top-view SEM images of the different caps of (a) two-step spin-coating deposited OTP film with CH_3NH_3I concentration of 7.5 mg mL^{-1}, (b) two-step spin-coating deposited OTP film with CH_3NH_3I concentration of 10.0 mg mL^{-1}, (c) one-step deposited OTP film prepared by conventional crystallization, and (d) compact TiO_2.

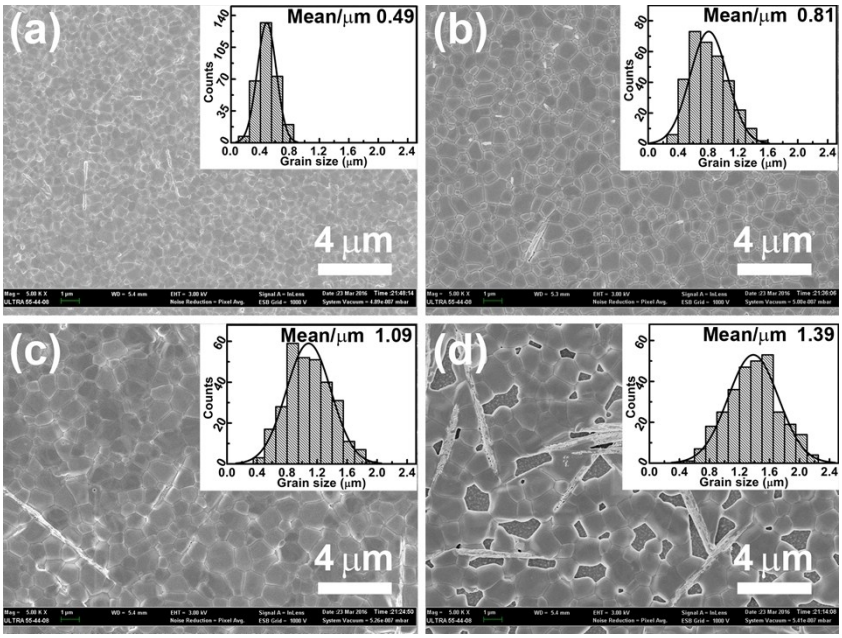

Fig. S7 Top-view SEM images used for the statistic of grain size distribution of HCM crystallized OTP films with different caps of (a) two-step deposited OTP film with CH_3NH_3I concentration of 7.5 mg/mL, (b) two-step deposited OTP film with CH_3NH_3I concentration of 10.0 mg/mL, (c) one-step deposited OTP film obtained by conventional crystallization, and (d) compact TiO_2, respectively. The insert gives the corresponding histogram of statistic grain size distribution.

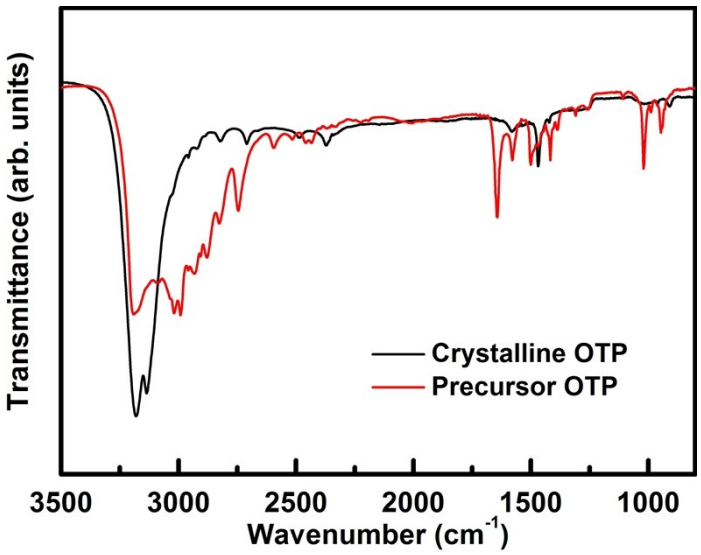

Fig. S8 FTIR spectra of the precursor film and HCM crystallized OTP film, respectively.

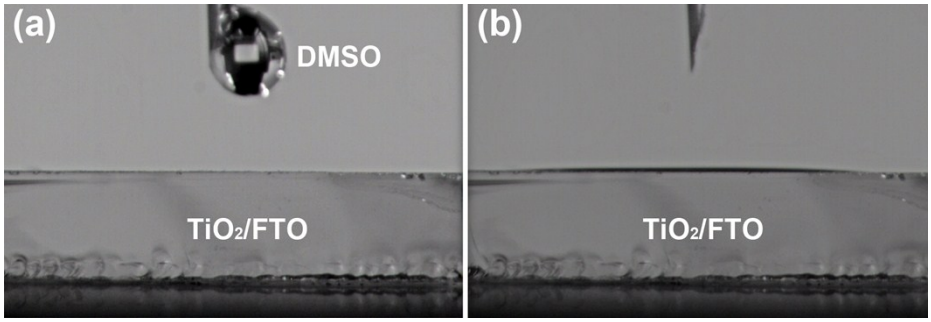

Fig. S9 Optical images of droplet of DMSO (a) before and (b) after dropping on the compact TiO_2/FTO substrate. The rapid spreading of DMSO droplet on compact TiO_2 indicates the super-wetting property of DMSO on compact TiO_2.

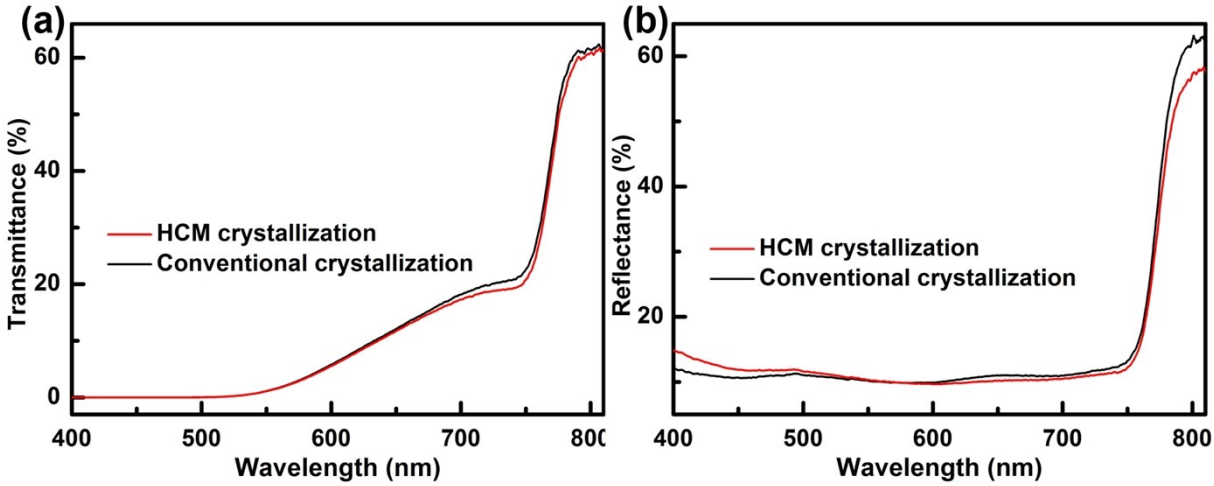

Fig. S10 (a) Transmission spectra and (b) reflection spectra of HCM crystallized OTP film and control sample obtained by conventional crystallization.

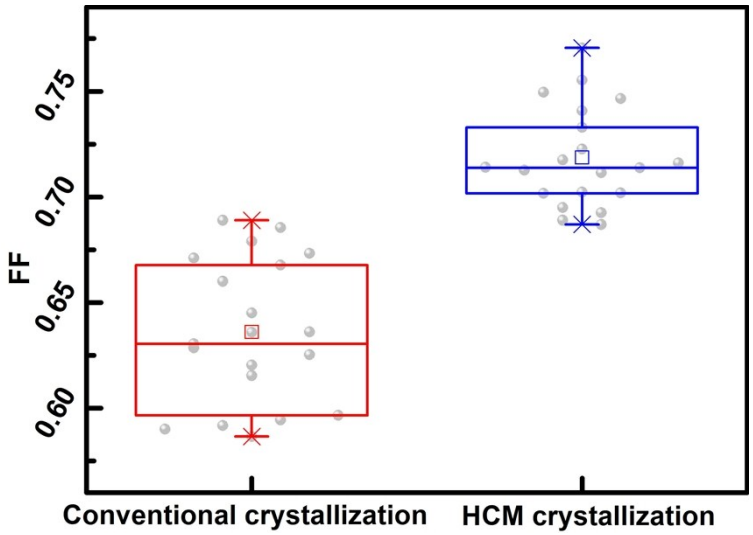

Fig. S11 Statistic FFs of 20 cells with control samples obtained by conventional crystallization (Conventional crystallization) and HCM crystallized OTP films (HCM crystallization), respectively.

References

1. N. Ahn, D.-Y. Son, I.-H. Jang, S. M. Kang, M. Choi and N.-G. Park, *J. Am. Chem. Soc.*, 2015, 137, 8696-8699.