Electronic Supplementary Material (ESI) for RSC Advances.

Facile preparation of high-quality perovskites for efficient solar cells via a fast conversion of wet PbI_2 precursor films

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Supplementary Figures

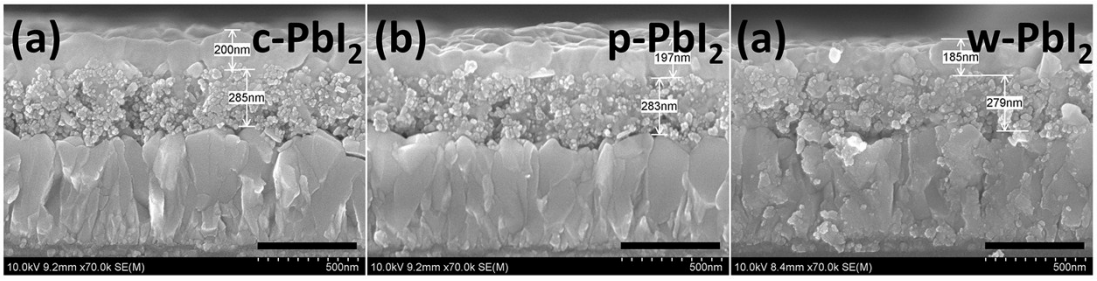

Fig. S1. Cross-sectional SEM images of the annealed $MAPbI_3$ films on mesoporous TiO_2 substrate prepared from their respective PbI_2 precursor films. The scale bars are 500 nm. The thickness of a $MAPbI_3$ capping layer on the top of the mp-TiO_2 prepared from c-PbI_2, p-PbI_2, and w-PbI_2 precursor film were determined to be 200, 197 and 185 nm, respectively. The increased thickness of capping layer of c-PbI_2-based perovskite film can be attributed to volume expansion occurred in the two-step sequential deposition process based on well-crystallized and compact PbI_2, which is because of the growth of perovskite crystals with the intercalation of organic iodides into PbI_2 skeleton.[1, 2]

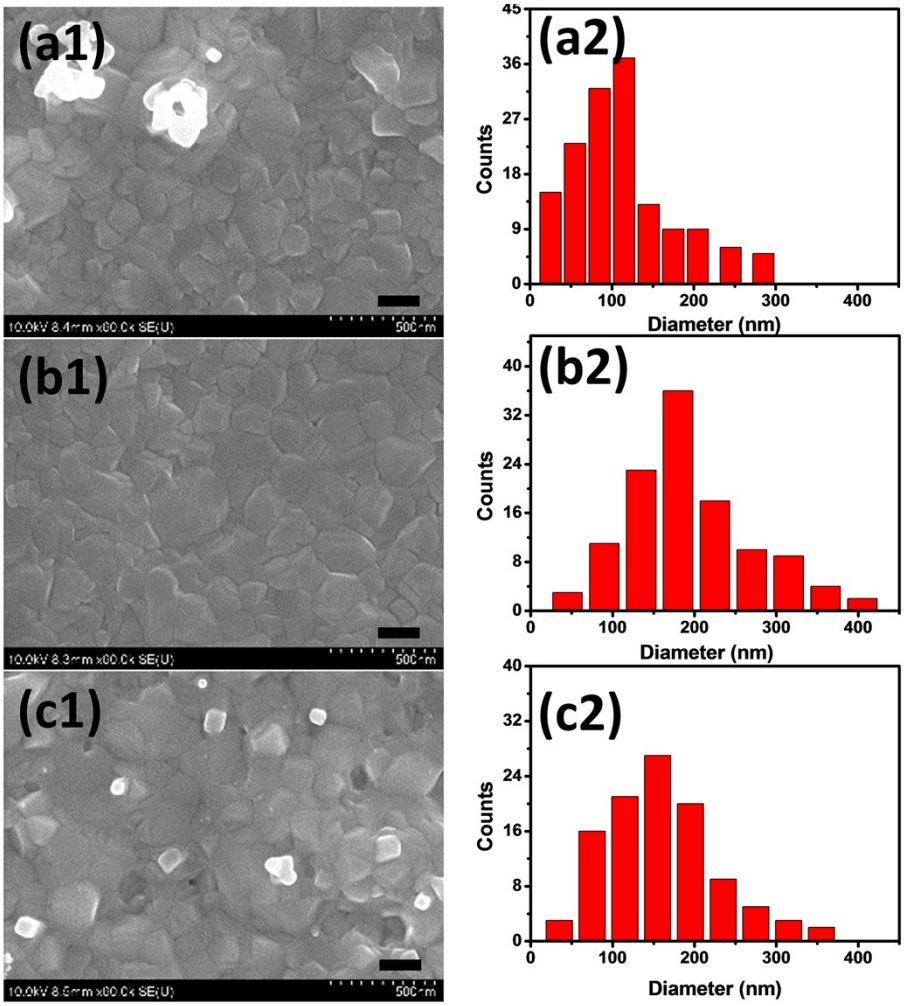

Fig. S2. Grain size distribution of the SEM images to the left. The perovskite films were prepared from c-PbI_2 (a1), w-PbI_2 (b1) and p-PbI_2 precursor films (c1), respectively. The scale bars in the SEM images are 200 nm. The average grain size of perovskites prepared with c-PbI_2, w-PbI_2 and w-PbI_2 precursor films are determined to be 127.1±64.7, 172.7±74.7 and 148.6±71.7 nm, respectively.

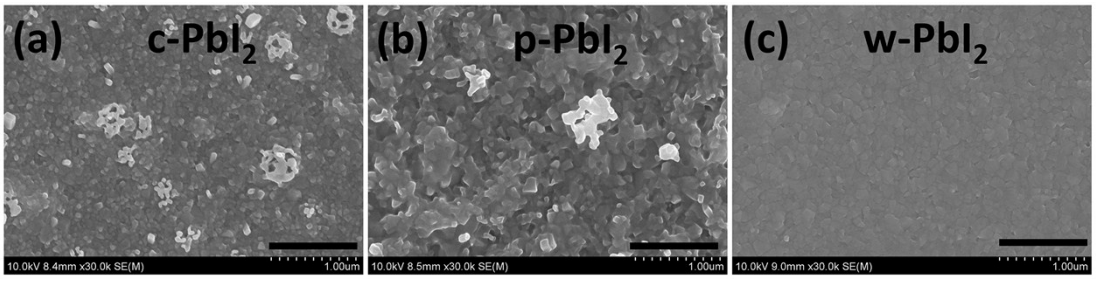

Fig. S3. Top-view SEM images of the as-prepared annealing-free perovskite films prepared from their respective PbI_2 precursor films. The scale bars are 1μm.

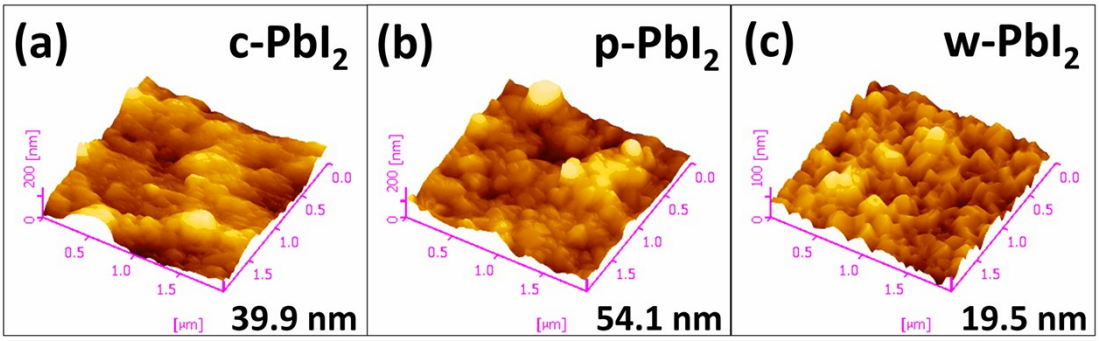

Fig. S4. AFM topographical 3D images (2 μm ×2 μm) of our perovskite films fabricated from their respective PbI_2 precursor films. The perovskite film prepared from annealing-free, wet PbI_2 (w-PbI_2) is uniform and ultra-smooth surface (RMS is 19.5 nm) due to the direct substitution of DMF molecules by MAI molecules in wet PbI_2 precursor film.

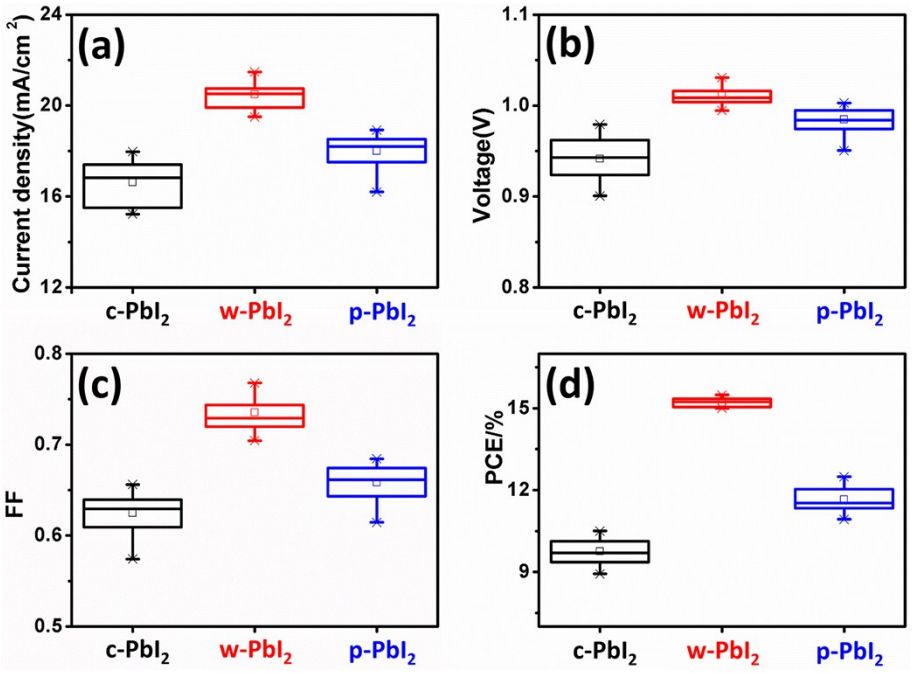

Fig. S5. Box charts of photovoltaic parameters for the perovskite solar cells fabricated with different $MAPbI_3$ films prepared from the various PbI_2 precursor films. The data were obtained from 20 individual cells.

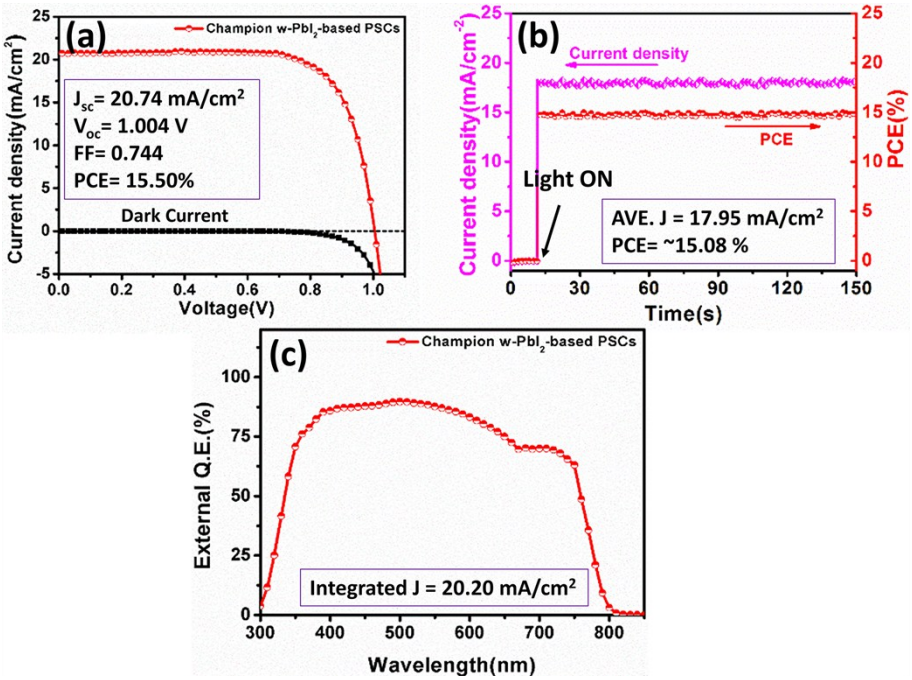

Fig. S6. (a) The J–V curves of a best $MAPbI_3$ solar cell prepared from w-PbI_2 precursor film with the highest efficiency of 15.49% (J_{sc}=20.74 mA/cm^2, V_{oc}=1.004 V, FF=0.744) under simulated 1 sun illumination. (b) Stability of power conversion efficiency as a function of time for the same cell measured at 0.84 V under simulated 1 sun illumination. (c) The external quantum efficiency (EQE) spectrum. The Jsc value is consistent with the EQE spectrum. It is worth to mention that there are reports showing that the integrated J_{sc} from EQE doesn't always match well with Jsc extracted from J-V measurement, which is presumably related to the mismatch (approximately 10%) between the EQE spectra and the photon flux AM 1.5 G spectrum,[3] and the difference in device characteristics (e.g., stability during measurement outside the glovebox) for solar cells.[2, 4]

Table S1. The full width at half maximum (FWHM) of $MAPbI_3$ (110) diffraction peak and PbI_2 (001) diffraction peak extracted from the XRD patterns (Fig. 3 in manuscript).

PbI_2 precursor films	PbI_2 (001) peak	$MAPbI_3$ (110) peak	
		Annealing-free	Annealed
c-PbI_2	0.414	0.525	0.369
w-PbI_2	0.472	0.470	0.270
p-PbI_2	0.382	0.495	0.362

It is well known that the smaller FWHM of diffraction peak generally means the higher crystallinity. For w-PbI_2 film, the largest FWHM (0.472) of PbI_2 (001) diffraction peak indicates the lowest crystallization, which is attributed to that the presence of high boiling DMF solvent in the as-prepared wet PbI_2 film retards the growth of PbI_2 crystals. After annealing at 70 ^oC for 30 min, the crystallinity of PbI_2 film increases, as the decreased FWHM (0.414) of c-PbI_2 film, which is due to the evaporation of solvent DMF molecules. The lowest FWHM (0.382) of PbI_2 (001) diffraction peak presents the highest crystallization of PbI_2 crystals in the p-PbI_2 film, which can be ascribed to an effective extraction of residual DMF by an anti-solvent of IPA molecules. For annealing-free $MAPbI_3$ films, tens of seconds at room temperature is effective and sufficient to transform PbI_2 into well-crystallized $MAPbI_3$ films with the lowest FWHM (0.382) of $MAPbI_3$ (110) diffraction peak, which can be attributed to the retarded crystallization of w-PbI_2 and a direct molecular exchange of DMF molecules intercalated in PbI_2 with MAI. Upon annealing, the FWHM of $MAPbI_3$ (110) diffraction peak for all samples significantly decreases, which indicates the increased crystallinity of annealed perovskites.

References

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2 T. Y. Zhang, M. J. Yang, Y. X. Zhao and K. Zhu, *Nano Lett.,* 2015, **15**, 3959-3963.

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