

Electronic Supplementary Information

Pressure-assisted $\text{CH}_3\text{NH}_3\text{PbI}_3$ morphology reconstruction to improve the high performance of perovskite solar cells

Experimental section

Materials: PbI_2 was purchased from Sigma Aldrich, N, N-dimethylformamide (DMF) and chlorobenzene from Alfa Aesar. All the chemicals were directly used without further purification. $\text{CH}_3\text{NH}_3\text{I}$ was synthesized according to the literature.¹ Substrates are F-doped tin oxide conducting glass (FTO, Pilkington, thickness: 2.2 mm, sheet resistance 14 Ω /square). Before use, FTO glass was firstly washed with alcohol alkaline solution, rinsed with distilled water for several times and subsequently with ethanol in an ultrasonic bath, finally dried under air stream.

Device fabrication: 50 nm-thickness TiO_2 compact layer and 400 nm-thickness mesoporous TiO_2 anatase layer (Dyesol 18NRT, Dyesol) were deposited on FTO glass in sequence by spin-coating method and annealing at 500 °C. Then, $\text{CH}_3\text{NH}_3\text{PbI}_3$ was deposited on TiO_2 porous films according to sequential deposition method with a little modification.² That is, 1.2 M PbI_2 in DMF was spin-coated on the TiO_2 film at 3000 rpm for 60 s and heated at 90°C for 2 min, repeated once again, then soaked into $\text{CH}_3\text{NH}_3\text{I}$ isopropanol solution (10 mg/mL) for 30 min. The film color changes from yellow to dark brown. The obtained $\text{CH}_3\text{NH}_3\text{PbI}_3/\text{TiO}_2$ films were thoroughly rinsed with isopropanol, dried under air stream, heated at 100°C for 30 min in air on a hotplate, and finally transferred to a hot-press machine. The temperature of both fixed and moving hotplates were set at 100 °C, and pressure were set to 0.5 MPa. Unless otherwise specified, the hot-pressing operation time was 5 min for each device. For HTM-contained structure, spiro-OMeTAD solution was prepared and doped according to the literature,³ then spin-coated at 2000 rpm on the perovskite film to give a HTM layer. Finally, 80 nm of gold was thermally evaporated on top of the perovskite or HTM layer as counter electrode.

Characterization: The microscopic morphologies of the films were obtained with scanning electron microscopy (SEM, Hitachi, S-5200). UV-vis transmittance spectra were obtained on UV-2550 spectrophotometer, Shimadzu. The XRD patterns are recorded with a Japan Rigaku D/max-2500 rotation anode X-ray diffractometer equipped with graphite-monochromatized Cu $K\alpha$ radiation ($\lambda=1.54178\text{\AA}$). Time resolved photoluminescence (PL) spectra were recorded on PL spectrometer, Edinburgh Instruments, FLS 900, excited with a picosecond pulsed diode laser (EPL-445), which were measured at 775 nm after excitation at 445 nm.

Photovoltaic measurement: The cells were illuminated under AM 1.5 simulated sunlight (100 mW/cm^2) on Oriel Solar Simulator 91192. Photocurrent density-photovoltage (J - V) characteristics of the devices were recorded on a digital source meter (Keithley model 2602). A mask with a window of 0.1 cm^2 was clipped on the TiO_2 side to define the photoactive area of the cells.

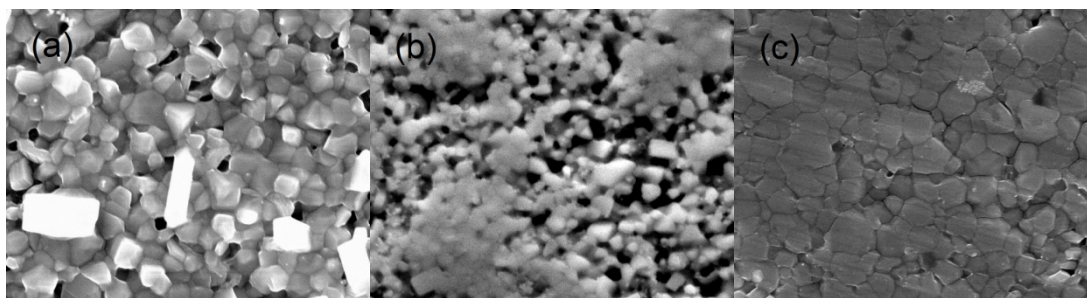


Fig. S1 SEM images of a) thermal annealed, b) pressed and c) hot-pressed perovskite films.

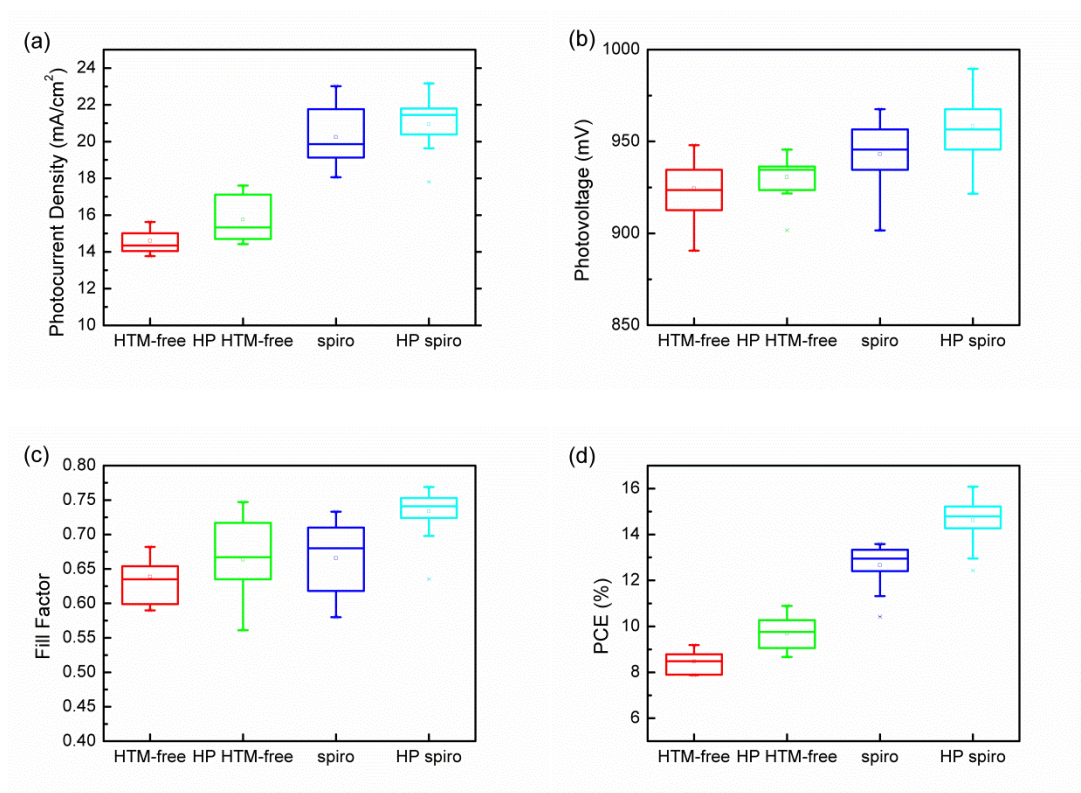


Fig. S2 Statistical data of six devices fabricated with P3HT and P3HT/GD, respectively: a) short-circuit photocurrent density (J_{sc}); b) open-circuit photovoltage (V_{oc}); c) fill factor (FF); d) power conversion efficiency (PCE).

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- 2 J.J. Shi, Y.H. Luo, H.Y. Wei, J.H. Luo, J. Dong, S.T. Lv, J.Y. Xiao, Y.Z. Xu, L.F. Zhu, X. Xu, H.J. Wu, D.M. Li and Q.B. Meng, *ACS Appl. Mater. Interfaces*, 2014, **6**, 9711.
- 3 J. Burschka, N. Pellet, S.J. Moon, R. Humphry-Baker, P. Gao, M.K. Nazeeruddin and M. Grätzel, *Nature*, 2013, **499**, 316.