

Electronic Supplementary Information

Stable semi-transparent $\text{CH}_3\text{NH}_3\text{PbI}_3$ planar sandwich solar cells

**Jin Hyuck Heo^a, Hye Ji Han^a, Minho Lee^a, Myungkwan Song^b, Dong Ho Kim^b,
Sang Hyuk Im^{a,*}**

^a Functional Crystallization Center (FCC), Department of Chemical Engineering, Kyung Hee University, 1732 Deogyong-daero, Giheung-gu, Yongin-si, Gyeonggi-do 446-701, Republic of Korea

^b Advanced Functional Thin Film Department, 797 Changwondaero, Seongsangu, Changwon, Gyeongnam, 642-831, Republic of Korea

* To whom correspondence should be addressed. E-mail: imromy@khu.ac.kr

Materials and method

MAI (methylammonium iodide) was synthesized by reacting 50 mL hydriodic acid (57% in water, Aldrich) and 50 mL methylamine (40 % in methanol, Junsei Chemical Co. Ltd.) in a 250-mL round-bottom flask at 0 °C for 2 h with stirring. The solution mixture was dried by evaporation at 50 °C for 1h to yield white MAI powder. To purify MAI powder, the MAI powder was dissolved in ethanol, recrystallized from diethyl ether, and finally dried at room temperature in a vacuum oven for 24 h. And then 40 wt% of MAPbI_3 solution was prepared by mixing the MAI powder and PbI_2 (Aldrich) (1:1 mole ratio) in N,N-dimethylformamide (DMF, Aldrich) at 60 °C for 30 min. Then 0.1 mL hydriodic acid (57% in water, Aldrich) was added in 1 mL MAPbI_3 /DMF solution for further process.

To fabricate the MAPbI_3 sandwich solar cells, a 50-nm-thick TiO_2 electron conductor was deposited on a cleaned F-doped SnO_2 (FTO, Pilkington, TEC8) substrate by spray pyrolysis deposition (SPD) method with 20 mM of titanium diisopropoxide bis(acetylacetonate) (Aldrich) solution at 450 °C. A pin-hole free MAPbI_3 thin-film was

deposited on the TiO_2/FTO substrate by spin-coating of 40 wt% MAPbI_3 solution with hydriodic acid additive at 3000 rpm for 200 s and was dried on a hot plate at 100 °C for 2 min under ambient atmosphere. A counter electrode was prepared by spin-coating of a filtered poly(3,4-ethylenedioxythiophene):poly(styrenesulfonic acid) (PEDOT:PSS, Clevios, Al4083)/methanol (1:2 vol:vol) solution on a cleaned ITO (Indium tin oxide) glass substrate at 3000 rpm for 60s and subsequent drying at 150 °C for 20 min. 0.2 mL poly-3-hexylthiophene (P3HT, Lumtec) or poly-triarylamine (PTAA, EM index) hole transporting material (HTM)/toluene solution with Li-bis(trifluoromethanesulfonyl) imide (Li-TFSI, Aldrich):acetonitrile (ACN) solution and tert-butylpyrrolidine (t-BP, Aldrich):ACN solution additives (HTM/toluene/Li-TFSI solution/t-BP solution = 20 mg/1 mL/15 μL (170 mg:1 mL)/30 μL (1 mL:1 mL) was drop-casted on the $\text{MAPbI}_3/\text{TiO}_2/\text{FTO}$ substrate and the counter electrode of PEDOT:PSS/ITO substrate was covered on the wet P3HT or PTAA/ $\text{MAPbI}_3/\text{TiO}_2/\text{FTO}$ substrate. Then the two substrates were pressurized by double clip (Whasin) as shown in Fig. S1 and were dried for 4 h under ambient atmosphere. After completion of drying process, the double clips were removed. These sandwich type devices without edge sealing were used for further characterization. All device fabrications were conducted below 50 % of relative humidity.

To measure the photovoltaic performance of sandwich solar cells, we measured the current density-voltage (J-V) curves by using a solar simulator (Peccell, PEC-L01) with a potentiostat (IVIUM, IviumStat) under illumination of 1 sun (100 mW/cm^2 AM 1.5G) which is calibrated by Si-reference cell certificated by JIS (Japanese Industrial Standards). For the measurement J-V hysteresis with respect to the scan direction, we set the forward and reverse scan rate to 10 $\text{mV}/200$ ms. The J-V curves of all devices

were measured by masking the active area with metal mask of 0.096 cm² or 1 cm². The external quantum efficiency (EQE) was measured by a power source (ABET 150W Xenon lamp, 13014) with a monochromator (DONGWOO OPTORN Co., Ltd., MonoRa-500i) and a potentiostat (IVIUM, IviumStat). All measurements were conducted at air conditions. For checking the air and humidity stability, we stored the sandwich type solar cells under ambient condition and measured their efficiency and relative humidity in every day during 20 days. We also checked the current density variation with light soaking time under applied voltage at maximum power point. For this the unsealed sandwich type solar cell was exposed by continuous 1 Sun light of solar simulator (100 mW/cm²) for 3600 s at air condition. During the continuous light illumination, the temperature of device was gradually increased to ~ 60 °C.

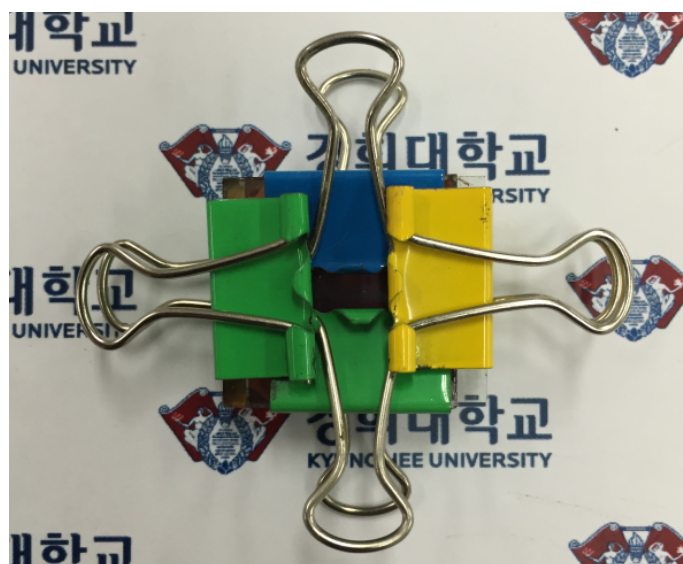


Fig. S1. Photograph of sandwich solar cell pressurized by double clip for laminating and drying process.

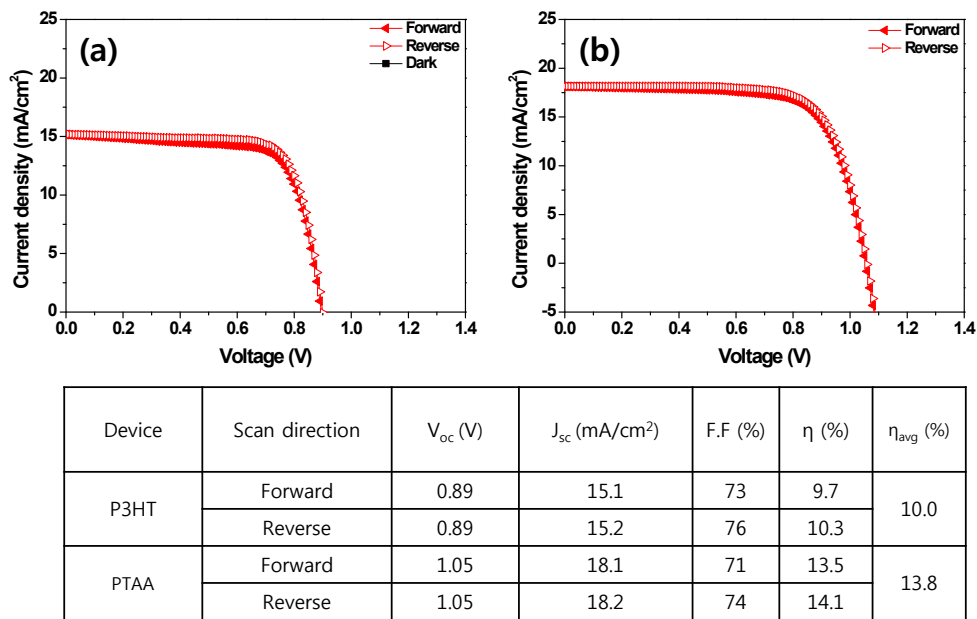


Fig. S2. J-V curves of sandwich solar cells with P3HT (a) and PTAA (b) HMT under back side illumination condition. Table is summary of photovoltaic properties of device (a) and (b).

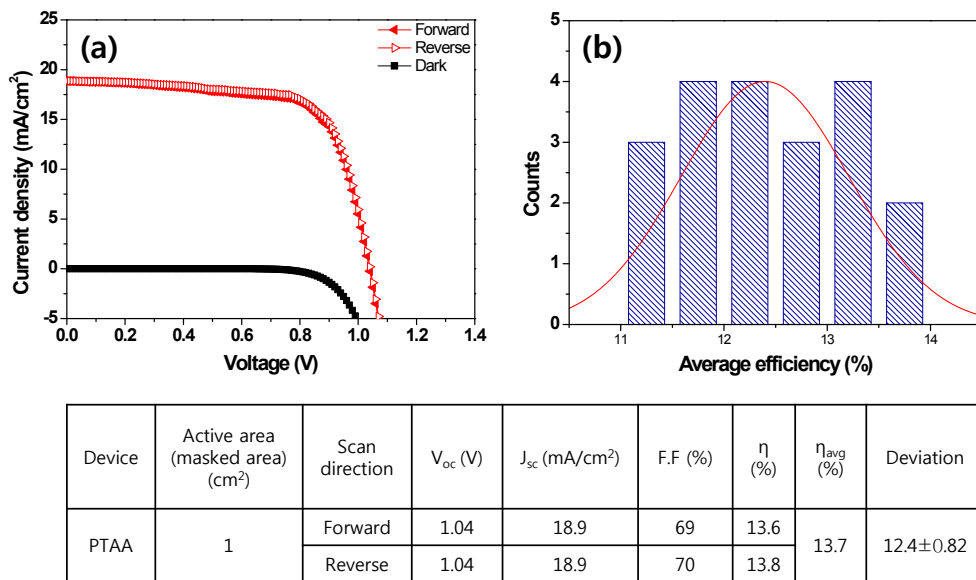


Fig. S3. (a) J-V curve of full FTO/TiO₂/MAPbI₃/PTAA with additives/PEDOT:PSS/ITO planar sandwich solar cell (best cell): scan rate = 10 mV/200 ms, delay time = 200 ms, front illumination to FTO side, masked area = 1 cm² and (b) deviation of average power conversion efficiency (η_{avg}). Table is summary of photovoltaic properties of device (a).

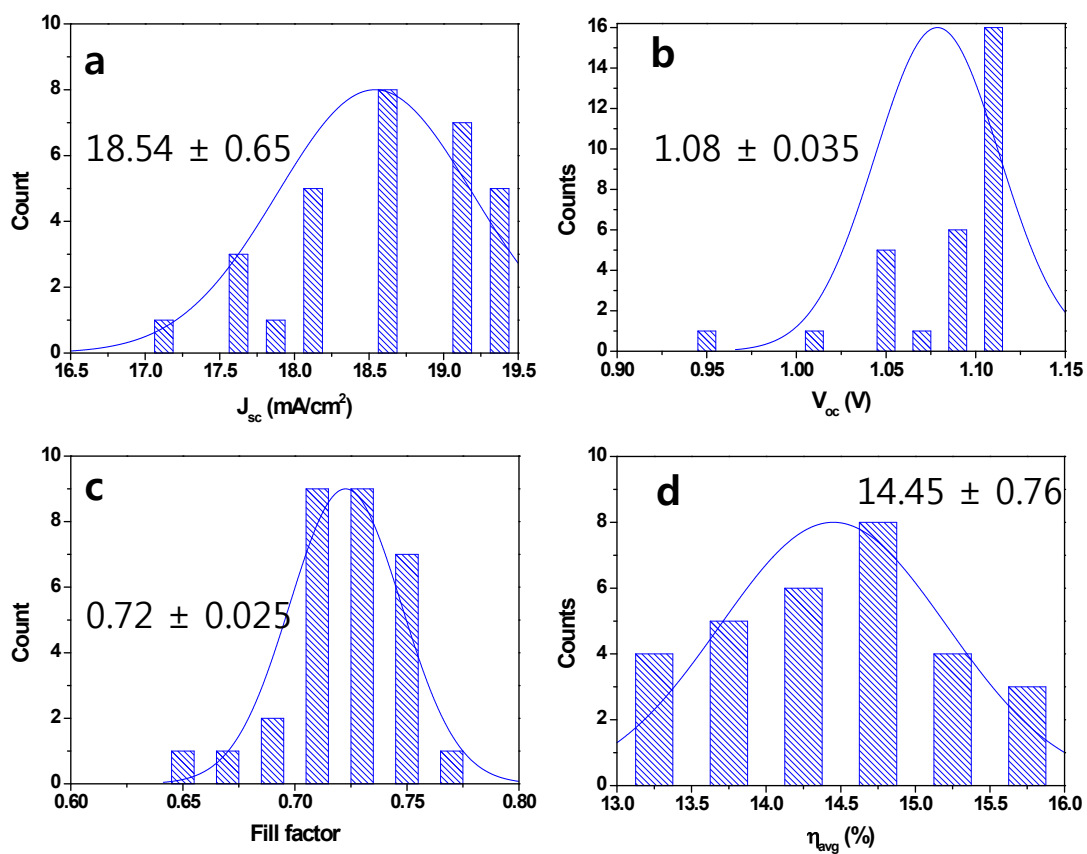


Fig. S4. Deviation of J_{sc} , V_{oc} , F.F. and η_{avg} of PTAA-2500 rpm sandwich solar cell.

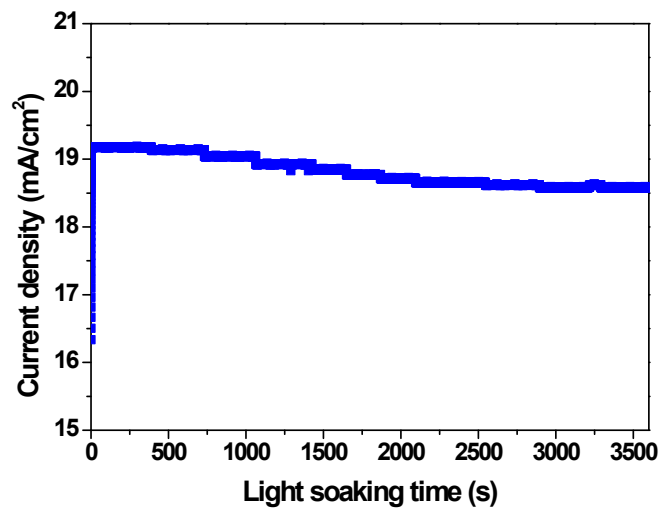


Fig. S5. The current density variation with light soaking time under applied voltage at maximum power point (1 Sun light illumination for 3600 s).