Electronic Supplementary Information (ESI)

Efficiency enhancement of semi-transparent sandwich type CH₃NH₃PbI₃ perovskite solar cells with island morphology perovskite film by introduction of polystyrene passivation layer

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Experimental

Preparation of MAPbI₃ perovskite solution

MAI (CH₃NH₃I) was synthesized by reacting 50 mL methylamine (40 % in methanol, Junsei Chemical Co. Ltd.) with 50mL hydriodic acid (57% in water, Aldrich) in a 250 mL round-bottom flask at 0 °C for 2h with vigorous stirring. Solid precipitate was obtained by evaporating the solvent on a rotary evaporator at 50 °C for 1h. This white solid was then dissolved in ethanol, recrystallized from diethyl ether, and finally dried in a vacuum oven for 24h at room temperature to obtain pure MAI. 40 wt% MAPbI₃ perovskite solution was prepared by mixing 1:1 mole ratio of MAI powder and PbI₂ (Aldrich) in 1mL γ -butyrolactone (Aldrich) at 60 °C for 30 min under stirring with magnetic bar.

Device fabrication

To fabricate semi-transparent sandwich type MAPbI₃ perovskite solar cells with island morphology perovskite film, a ~50 nm-thick dense TiO₂ electron conducting layer was deposited on cleaned partially etched F-doped tin oxide (FTO, Pilkington, TEC8) glass substrate (FTO substrate = $2.5 \text{ cm} \times 2.5 \text{ cm}$, 2 line patterned electrode = $0.5 \text{ cm} \times 1 \text{ cm}$: see below Fig.) by spray pyrolysis deposition method using 20 mM of titanium diisopropoxidebis(acetylacetonate) (Aldrich) solution at 450 °C. The island morphology perovskite film was deposited on the TiO₂/FTO substrate by spin-coating of 40 wt % MAPbI₃/γ-butyrolactone perovskite solution at 5000 rpm for 120 s and was dried on a hot plate at 100 °C for 2 min. A polystyrene (Aldrich, average MW = 192,000) passivation interlayer was deposited by consecutive spin-coating and spin-washing process (see below Fig.). For this, we deposited PS passivation interlayer by spin-coating PS/toluene solution (0.1 g/1 mL) at 5000 rpm for 30 s (ramp time = 3s) and then the PS/ MAPbI₃/TiO₂/FTO substrate was accelerated/decelerated to specific spin speed (7000, 6000, 5000, and 4000 rpm). As soon as the spin speed reached to specific rpm (ramp time = 3 s), 0.5 mL toluene was dropped on the spinning PS/MAPbI₃ islands/TiO₂/FTO substrate. 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 on a hotplate at 150 °C for 20 min. A polytriarylamine (PTAA, EM index) hole transporting material (HTM)/toluene solution with Li-bis(trifluoromethanesulfonyl) imide (Li-TFSI, Aldrich):acetonitrile (ACN) solution and tert-butylpyrridine (t-BP, Aldrich):ACN solution additives (HTM/toluene/Li-TFSI solution/t-BP solution = 20 mg/1 mL/15 μ L (170 mg:1 mL)/30 μ L (1 mL:1 mL) was dropped on the MAPbI₃ islands/TiO₂/FTO substrate with and without PS passivation interlayer and the counter electrode of PEDOT:PSS/ITO substrate was covered on the wet PTAA/ MAPbI₃ islands/TiO₂/FTO substrate with and without PS passivation interlayer. Then the two substrates were pressurized by double clip and were fully dried. All device fabrications were conducted below 20 % of relative humidity.



Device characterization

The current density-voltage (J-V) curves were measured by a solar simulator (Peccell, PEC-L01) with a potentiostat (IVIUM, IviumStat) at under illumination of 1 Sun (100 mW/cm² AM 1.5G) and a calibrated Si-reference cell certificated by JIS (Japanese Industrial Standards). The J-V curves of all devices were measured by masking the active area with metal mask of 0.096 cm². To measure the hysteresis of J-V curves, the forward and reverse scan rate was set to 10 mV·200 ms⁻¹ (voltage delay time⁻¹). The external quantum efficiency (EQE) was measured by a power source (ABET, 150W Xenon lamp, 13014) with a monochromator (DONGWOO OPTRON Co., Ltd., MonoRa-500i) and potentiostat (IVIUM, IviumStat). Intensity-modulated photocurrent (IMPS) and photovoltage spectroscopy (IMVS) were measured by potentiostat (IVIUM, IviumStat) with light emitting diode (LED) (IVIUM, IM1225).



Fig. S1. The SEM surface image of MAPbI₃ islands/TiO₂/FTO substrate with PS passivation layer formed by spin-washing process at 4000 rpm (a, b) and at 7000 rpm (c, d).



Fig. S2. IMPS (a, b) and IMVS (c, d) of the semi-transparent sandwich type island MAPbI₃ perovskite solar cells (a, c) with and (b, d) without PS passivation interlayer.