Supporting Information

Benefits of Very Thin PCBM and LiF Layer for Solution-Processed P-I-N Perovskite Solar Cells

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Experimental Section

Device fabrication: The patterned ITO substrates were cleaned with ultrasonication in acetone and 2% Helmanex soap in water, followed by washing with de-ionized water and then, in isopropyl alcohol. Finally, the substrates were treated under UV-ozone for 15 min to remove the last traces of the organic residues. A filtered dispersion of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonic acid) (PEDOT:PSS, levios, Al4083) was deposited on the top of the ITO by spin-coating at 3,000 r.p.m. for 60 s and subsequently dried at 150 ^{o}C for 20 min. CH_3NH_3I was prepared by reaction with hydroiodic acid (30 mL, 57% in water, Aldrich) and methylamine (27.9 mL, 40% in methanol, Junsei Chemical Co., Ltd.) in a 250 mL round-bottomed flask at 0˚C for 2 h with stirring. The precipitate was recovered by evaporating the solution at 50˚C for 1 h. The product was dissolved in ethanol, recrystallized using diethyl ether, and dried at 60˚C under vacuum for 24 h. The CH_3NH_3I and PbI_2 (Aldrich) were stirred in a mixture of dimethyl sulfoxide (DMSO):γ-butyrolactone (GBL) (3:7, v/v) at 60˚C for 12 h. The perovskite precursor solution of CH_3NH_3I and PbI_2 (1:1 molar ratio) was deposited onto PEDOT:PSS/ITO substrate by a consecutive two-step spin-coating process at 1000 r.p.m. and at 4,000 r.p.m. for 20 and 60 s, respectively, and the toluene in final spin-stage was dripped onto the substrate during spin coating. The perovskite-precursor

coated substrate was dried onto a hot plate at 100 ^{o}C. Afterwards, the phenyl-C61-butyric acid methyl ester (PCBM) layer was deposited by spin-coating of the different concentration solution (8, 12, 15, 20, and 25 mg ml^{-1}) of PCBM (nano-C) at 1200 r.p.m. for 60 s. Finally, the device was completed with evaporation in a high vacuum of Al contact electrodes after evaporation of LiF (~0.5 nm) layer through shadow mask. The active area of Al electrodes in the fabricated device was 0.09 cm^2. All the devices were encapsulated with glass cap to avoid the oxygen and the moisture.

For a large area photovoltaic module, the patterned ITO-glass substrate has ten ITO strips (0.8 cm × 10 cm) each separated by 2 mm wide etched areas, where are interconnected in series. (See Figure 4b) The active area of one cell is adjusted to be 0.6 cm × 10 cm. All the layers are prepared according to the same process to the fabrication of the unit cell.

Measurements: The J–V curves were measured using a solar simulator (Newport, Oriel Class A, 91195A) with a source meter (Keithley 2420) at 100 mA cm^{-2} illumination AM 1.5G and a calibrated Si-reference cell certificated by NREL. The *J–V* curves of all devices were measured by masking the active area with a metal mask of area 0.04 cm^2. The External quantum efficiency (EQE) was measured using a power source (Newport 300 W Xenon lamp, 66920) with a monochromator (Newport Cornerstone 260) and a multimeter (Keithley 2001).

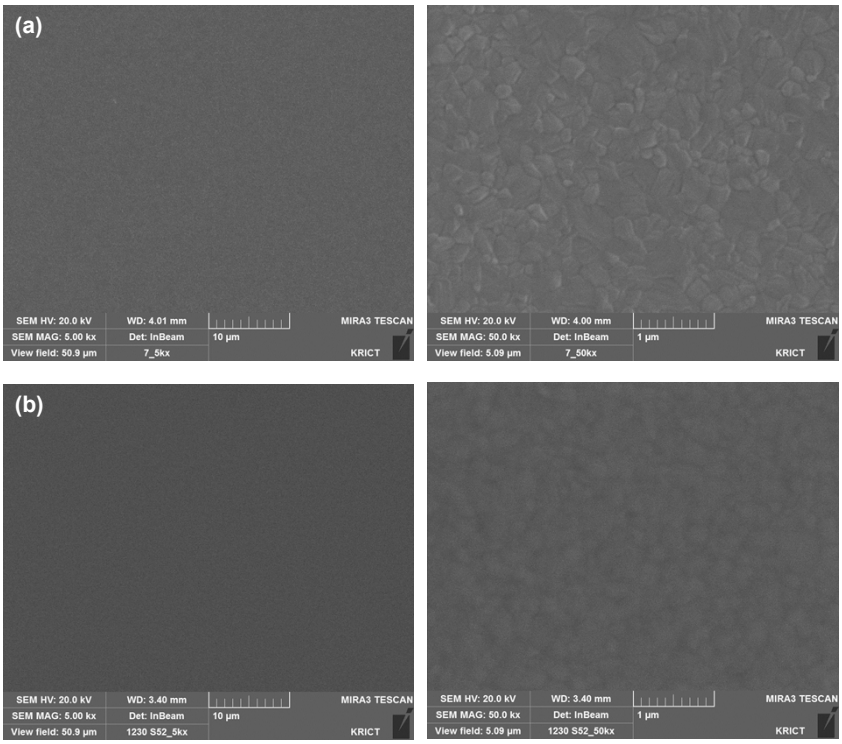


Figure S1. (a) SEM image of the top surface of a $CH_3NH_3PbI_3$ film on an ITO/PEDOT:PSS surface. (b) SEM image of the top surface of a PCBM film on an ITO/PEDOT:PSS/$CH_3NH_3PbI_3$ surface. The thickness of PCBM layer is 55 nm.

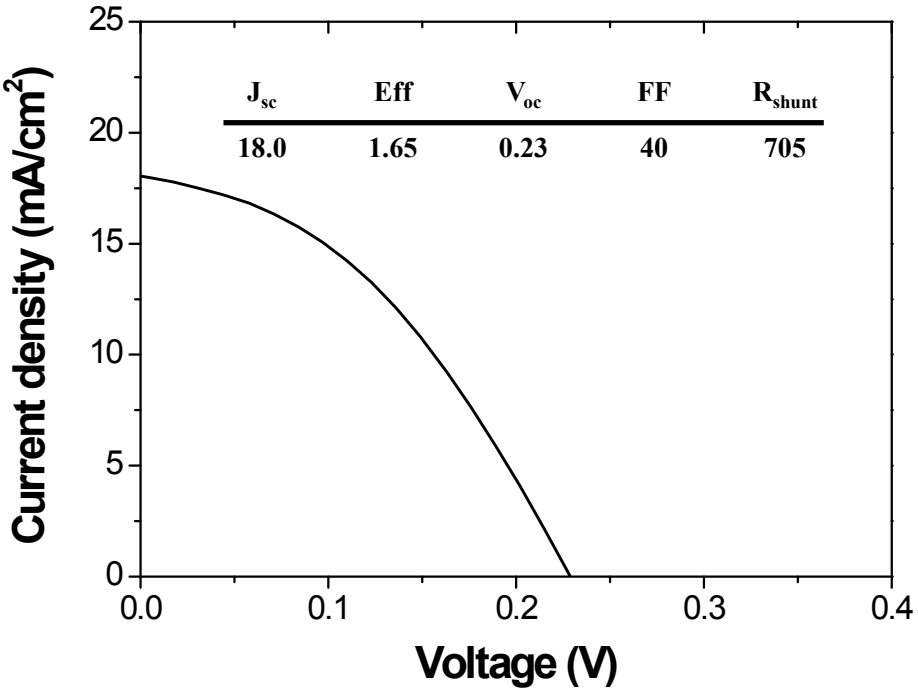
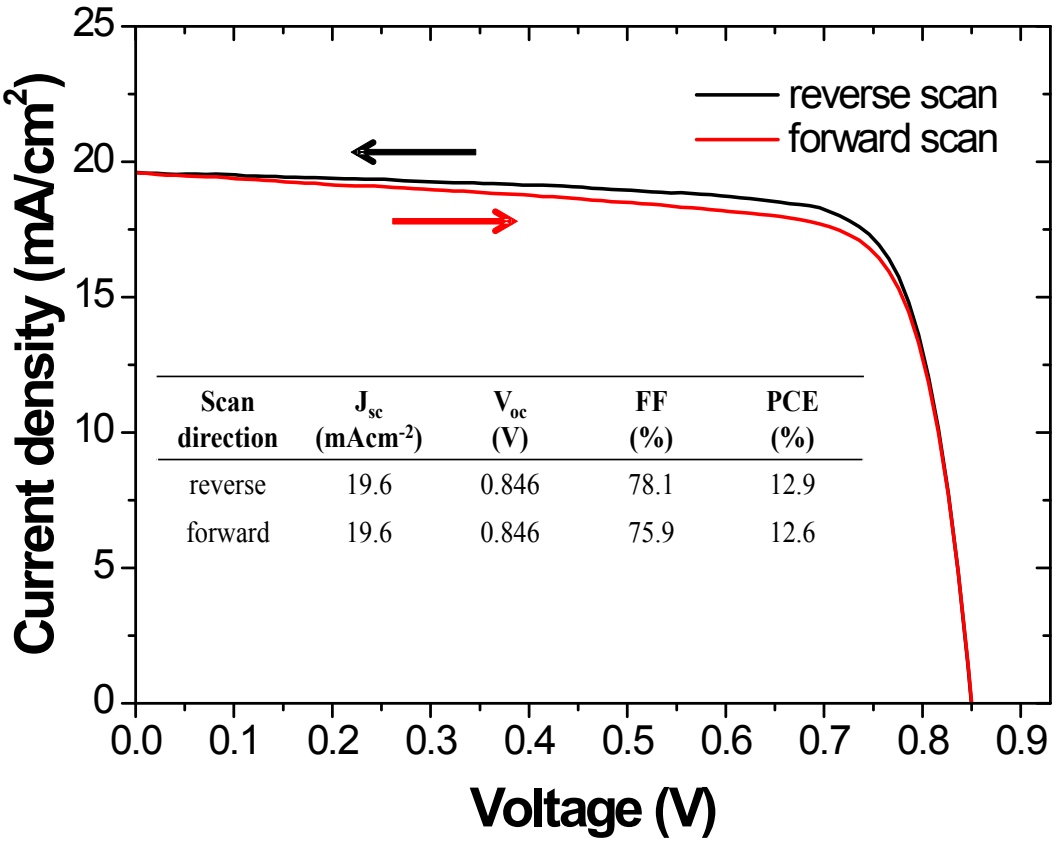


Figure S2. (a) Photocurrent density-voltage (*J-V*) characteristics of $CH_3NH_3PbI_3$-PCBM heterojunction solar cell where the thickness of PCBM layer is ~40 nm. Measurement was carried out under simulated AM 1.5 100 mW cm^{-2} sunlight. The inset table shows the photovoltaic performance parameters, J_{sc} (mA/cm^2), efficiency (%), V_{oc} (V), FF and R_{shunt} (ohms).

Table S1. Photovoltaic parameters derived from *J-V* measurement for the devices based on $CH_3NH_3PbI_3$-PCBM heterojunction solar cells with different PCBM layer thickness.

Thickness (nm)	**J_{sc} ($mAcm^{-2}$)**	**V_{oc} (V)**	**FF (%)**	**PCE (%)**
55	19.5	0.844	77.5	12.8
100	17.9	0.816	74.4	10.8
120	16.7	0.803	74.7	10.0
140	16.0	0.789	67.9	8.6



Scan direction	J_{sc} ($mAcm^{-2}$)	V_{oc} (V)	FF (%)	PCE (%)
reverse	19.6	0.846	78.1	12.9
forward	19.6	0.846	75.9	12.6

Figure S3. Photocurrent density-voltage (*J-V*) characteristics of $CH_3NH_3PbI_3$-PCBM heterojunction device, measured with 10 mV voltage steps and a delay time of 40 ms with different scan directions. The inset table shows the photovoltaic performance parameters, J_{sc} (mA/cm^2), V_{oc} (V), FF (%) and PCE (%).

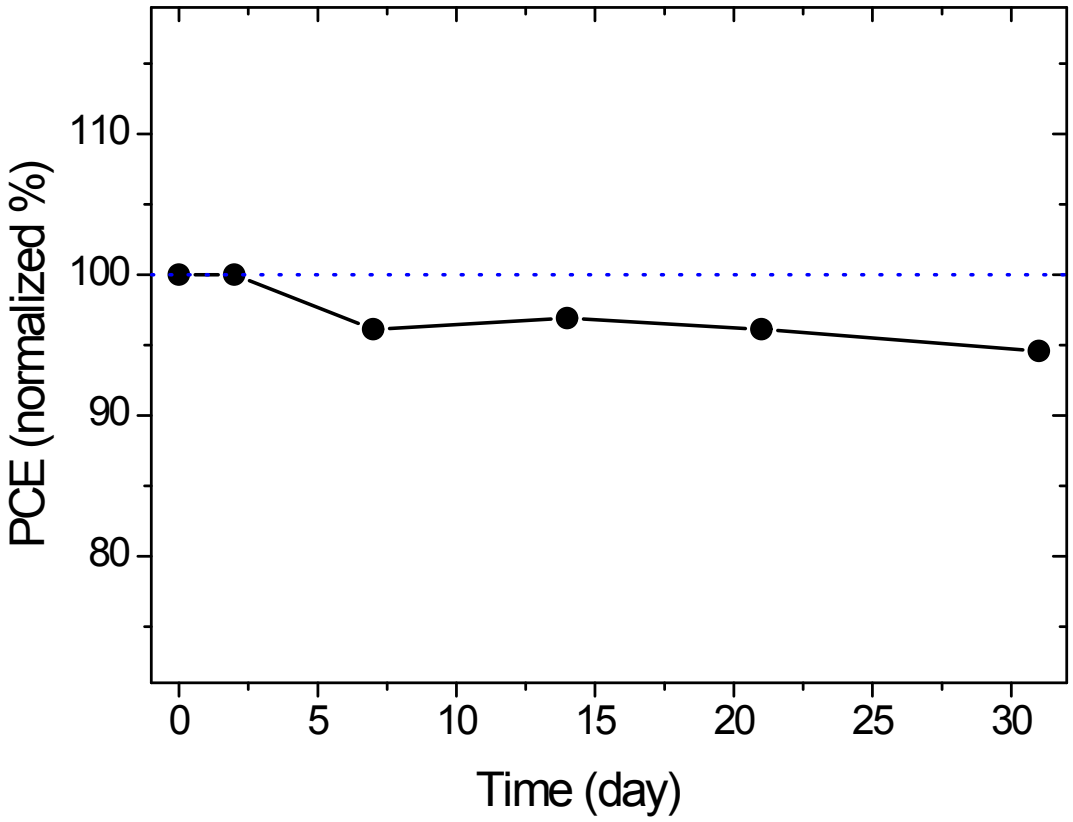


Figure S4. Normalized PCE of $CH_3NH_3PbI_3$-PCBM heterojunction device stored for different numbers of days. The encapsulated device is maintained in ambient air under dark.