

Article

# High Performance Planar Structure Perovskite Solar Cells Using a Solvent Dripping Treatment on Hole Transporting Layer

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## Experimental Section

PEDOT:PSS aqueous solution (Clevios P VP Al 4083) was purchased from Heraeus. Other chemicals, including PbI<sub>2</sub>, PbCl<sub>2</sub>, DMSO, DMF, and GBL, were supplied by Sigma-Aldrich. All materials were used as received. PEDOT:PSS films were prepared by spin coating its aqueous solution on pre-cleaned glass substrates. They were subsequently dried at 120 °C on a hot plate for 20 min. The treatment was performed by dropping 500 μL of a solvent onto each PEDOT:PSS film during the spin coating. After the PEDOT:PSS films were dried, they were then dried again at 120 °C.

### 2.2. Characterizations of Materials

The conductivities of the PEDOT:PSS films were measured by the four-probe technique with a Keithley 2400 source/meter. The electrical contacts were made by pressing indium on the four corners of each PEDOT:PSS film on glass substrate. X-ray photoelectron spectra (XPS) were collected with an Axis Ultra DLD X-ray photoelectron spectrometer equipped with an Al K $\alpha$  X-ray source (1486.6 eV). The atomic force microscopic (AFM) images were acquired with a Veeco NanoScope IV Multi-Mode AFM in tapping mode. The thicknesses of the PEDOT:PSS films were measured using an Alpha-Step IQ surface profiler. Photoluminescence spectra and time-resolved photoluminescence (TR-PL) spectra were measured using the Pico Quant Fluotime 300 by using a 510 nm picosecond pulsed laser.

### 2.3. Fabrication and Characterization of PSCs

ITO glass substrates were cleaned sequentially in detergent, deionized (DI) water, acetone, and isopropanol by sonication for 20 min. After drying under N<sub>2</sub> stream, the substrates were further treated with UV-ozone for 15 min. A PEDOT:PSS layer with a thickness of ~60 nm was prepared by spin coating Clevios P VP Al 4083 on ITO substrates at 2000 rpm for 1 min and subsequently annealed at 120 °C for 15 min in air. The substrates coated with PEDOT:PSS were then transferred into a glove box filled with highly pure N<sub>2</sub>. The moisture and oxygen levels in the glove box were less than 1 ppm. The perovskite layer was deposited by spin coating a solution consisting of 1.246 M PbI<sub>2</sub>, 0.154 M PbCl<sub>2</sub>, and 1.3 M MAI in cosolvent of DMSO:GBL (volume ratio = 3:7) at 1000 rpm for 20 s, and then at 3500 rpm for 60 s. After 50 s of the start of the spin coating, 1000 μL anhydrous toluene was dripped onto each spinning film. Then, the as-cast films were annealed at 100 °C for 20 min. The thickness of the perovskite thin films was around 320 nm. The phenyl-C61-butyric acid methyl ester

(PCBM) layer with a thickness of about 55 nm was deposited by spin coating a chlorobenzene solution of 20 mg/mL PCBM at 2000 rpm for 40 s. A sub-nanometer thick BCP layer was subsequently spin coated from its 0.05 wt.% isopropanol solution. The devices were completed by thermally depositing 100 nm-thick Ag in a vacuum of  $<1 \times 10^{-6}$  mbar. Each device had an area of 0.09 cm<sup>2</sup> and a shadow mask (0.075 cm<sup>2</sup>) was applied during the device measurement. The photovoltaic performance of the PSCs was tested in air with a computer-programmed Keithley 2400 source/meter and a Newport's Oriel class A solar simulator, which simulated the AM1.5 sunlight with energy density of 100 mW cm<sup>-2</sup> and was certified to the JIS C 8912 standard.