

Supporting Information

Scalable Post-Treatment for Improved Self-Assembled Monolayer Coverage in Perovskite Solar Cells

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Keywords: Perovskite, Self-Assembled Monolayer, Buried Interface

Methods

Precursor preparation:

The perovskite precursor solution was prepared by mixing 1.8M FAPbI₃, 1.8M CsI, and 1.8M MAPbBr₃ in a volume ratio of 100:10:3. The 1.8M FAPbI₃ precursor solution was made by dissolving equimolar amounts of 1.8M formamidinium iodide (FAI, >99.99%, GreatCell Solar) and lead iodide (PbI₂, 99.99%, TCI), with the addition of 15 mol% methylammonium chloride (MACl), in a solvent mixture of DMF and NMP (in an 8:2 volume ratio). The 1.8M CsI solution was prepared by dissolving equimolar cesium iodide (CsI, >99%, Genestarbio) in DMSO. The 1.8M MAPbBr₃ precursor solution was made by dissolving equimolar amounts of 1.8M methylammonium bromide (MABr, >99.99%, GreatCell Solar) and lead bromide (PbBr₂, 99.99%, TCI) in a DMF and NMP solvent mixture (in an 8:2 volume ratio). All precursor solutions (FAPbI₃, CsI, and MAPbBr₃) were stirred at room temperature for two hours to ensure complete dissolution. MeO-2PACz and MACl were purchased from TCI, while PFN-Br was obtained from Ways.

Device fabrication

FTO glass substrates were cleaned by sonication in deionized water, acetone, and isopropanol for 10 minutes each, followed by drying under a nitrogen stream. The cleaned substrates were treated with UV ozone for one hour. For cooled moisture condensation (CMC) method, the substrates were cooled in a 70% RH humidity environment for 15 seconds. The cooled substrates were then coated with 1 mM MeO-2PACz for 10 seconds and spin-coated at 3000 rpm for 20 seconds to deposit the layer. Afterward, the FTO/MeO-2PACz substrates were heated on a hot plate at 100°C for 10 minutes. A PFN-Br solution (0.05 wt% in methanol) was spin-coated at 4000 rpm for 20 seconds.

The perovskite layer was fabricated using a vacuum method. Specifically, the perovskite precursor solution was spin-coated at 8000 rpm for 10 seconds, placed in a vacuum chamber, and evacuated to a pressure of 1.4×10^{-1} torr. The substrates were then removed and annealed on a hot plate at 60°C for five minutes and at 100°C for 60 minutes to form the perovskite layer. The final device was completed by thermal evaporation of C60 (40 nm), BCP (5 nm), and Ag (100 nm).

Characterizations:

The current density-voltage (J-V) characteristics were obtained using an AM 1.5G solar simulator (Enlitech), calibrated with a silicon reference cell. Measurements were conducted in both forward and reverse directions. For the reverse scan, measurements started from the open-circuit voltage (V_{oc}) and proceeded to the short-circuit current (J_{sc}), covering a voltage range of 1.2 V to -0.2 V. The forward scan was performed in the opposite direction, from J_{sc} to V_{oc} , spanning -0.2 V to 1.2 V. Both scans were performed at a rate of 20 mV/s. Surface morphology of the perovskite films was assessed using scanning electron microscopy (SEM) on a Zeiss ULTRA PLUS instrument. For detailed electrical surface mapping, contact atomic force microscopy (C-AFM) was employed using the Bruker Dimension ICON system in Scan Asyst mode. Electrical surface irregularities were examined with a bias voltage of 200 mV applied to the films. Steady-state photoluminescence (PL) data were collected using an FS5 spectrofluorometer (Edinburgh Instruments Ltd.), with an excitation wavelength of 600 nm. The signal was acquired at a scanning rate of 1 nm/s and averaged over two sequential scans. Mott-Schottky analysis, open-circuit voltage decay (OCVD) and transient photocurrent decay (TPC) are conducted by Piasos. X-ray Photoelectron Spectroscopy (XPS) was performed using a PHI 5000 VersaProbe III system (ULVAC-PHI Ltd.) to investigate the chemical composition and states of the elements within the perovskite films. The samples were prepared and maintained in dark, inert conditions prior to analysis to preserve their integrity.

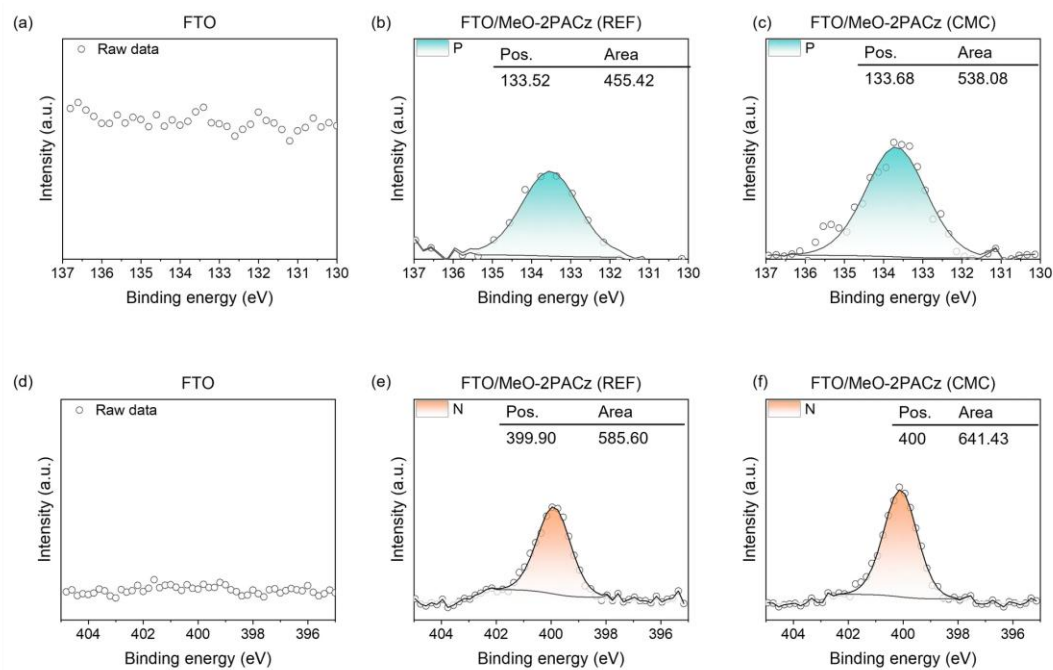


Figure S1. X-ray photoelectron spectroscopy (XPS) spectra of the phosphorus (P 2p) and nitrogen (N1s) regions for FTO, FTO/MeO-2PACz (REF), and FTO/MeO-2PACz (CMC) samples. (a) P 2p XPS spectrum of the bare FTO substrate (FTO/MeO-2PACz (REF) (c) FTO/MeO-2PACz (CMC), and N 1s XPS spectrum of the (d) bare FTO substrate, (e)FTO/MeO-2PACz (REF) (f) FTO/MeO-2PACz (CMC).

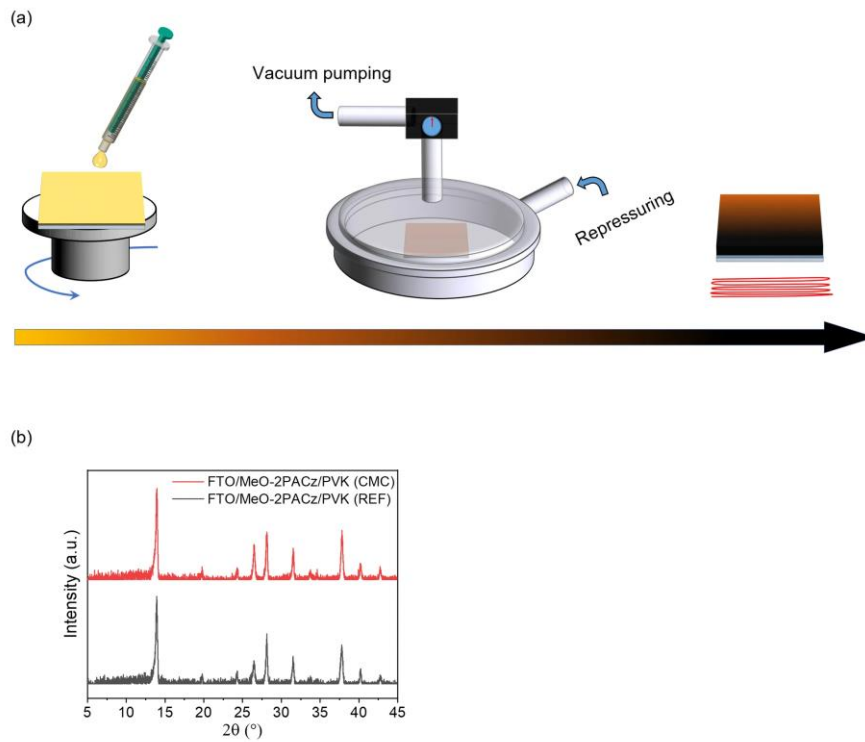


Figure S2. (a) Schematic illustration of the deposition process for the perovskite (PVK) layer on the FTO/MeO-2PACz substrate, involving spin-coating followed by vacuum-assisted processing. (b) X-ray diffraction (XRD) patterns of FTO/MeO-2PACz/PVK (CMC) and FTO/MeO-2PACz/PVK (Reference) samples.