

Supporting Information

Ultra-Stable Two-Dimensional MoS₂ Solution for Highly Efficient Organic Solar Cells[†]

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Experimental Section

Synthesis of ce-MoS₂ sheets: The ce-MoS₂ nanosheets were synthesized through Li intercalation. Briefly, 0.3 g pristine MoS₂ crystals (Sigma-Aldrich) were immersed in 3 mL of 1.6 M butyllithium solution in hexane for 2 days inside a nitrogen glovebox. The Li_xMoS₂ was then filtrated, washed with 80ml of hexane to remove excess lithium and organic residues, and then removed from glove box. Intercalated MoS₂ compound was then suspended in water and sonicated for 2h. Exfoliated MoS₂ solution was then dialyzed against continuous water flow using MW 10,000 cut-off membranes for 5 days, followed by 4000 rpm centrifugation to remove any un-exfoliated aggregates. Material was then diluted with deionized water to 2 mg/ml for device fabrication.

Synthesis of m-MoS₂ sheets: The aqueous solution of CTAC is added to the DI water dispersion of ce-MoS₂ sheets by 1:2 weight ratio, and the mixture is shaken for 10min. The resulted mixture was then purified through high-speed centrifugation and re-dispersion.

Fabrication of OSCs: The devices were fabricated on commercially available indium tin oxide (ITO)-coated glass substrates. Glass/ITO substrates were pre-cleaned with a detergent, ultrasonicated in deionized water, acetone and isopropyl alcohol consecutively every 15 min, and then dried in an oven. After treatment in an ultraviolet ozone generator for 15 min on the substrates, the ce-MoS₂ or m-MoS₂ solution was spun onto the substrates at 2000 rpm for 40 s by 3 times to obtain a dense film. PTB7:PC₇₁BM (Both were purchased from 1-Material Inc) were dissolved in chlorobenzene with a 1:1.5 (10mg ml⁻¹:15mg ml⁻¹) weight ratio, and 3% volume ratio of 1,8-diiodoctane was added as a processing additive before use. The BHJ layer was spin-cast at 1300 rpm with a thickness of about 90 nm in a nitrogen glovebox. A conjugated polyelectrolyte (CPE), poly[(9,9-bis(3'-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN, obtained from 1-Material Inc) interlayer material was dissolved in methanol and deposited on top of the active layer. Subsequently, a 100 nm thick aluminum evaporated through a shadow mask to define the active layer area of the devices (~5.4 mm²) and form a top cathode.

Characterizations: TEM images were taken by a FEITECNAIG2F20-TWITEM and STEM images were taken by FEI Titan ChemiSTEM operated at 200 kV. X-Ray diffraction (XRD) analysis was carried out on a Rigaku D/max-2550PC X-ray diffractometer with Cu K α radiation ($\lambda=1.5406 \text{ \AA}$). The absorption spectra were recorded on a UV-Visible spectrophotometer (UV-2450, Shimadzu Corporation, Japan). The AFM images were taken by a Veeco Multimode atomic force microscopy in the tapping mode. X-Ray photoelectron spectroscopy (XPS) measurement was performed on a PHI Quantera SXM (ULVAC-PHI) with Al K α X-ray radiation as the X-ray source for excitation. The samples were deposited on the aluminum plate. The photoelectron take off angle was 45°, and the energy resolution was set to 1.4 eV. The XPS wide spectra and narrow spectra of C1s, and Au 4f were recorded. The binding energy corrections for the spectra were carried out using C1s peaks at 284.5 eV as sp² graphite. The J-V curves were measured with Keithley 236 measurement source units at room temperature in air. The photocurrent was measured under a calibrated solar simulator (Abet 300 W) at 100 mW cm⁻², and the light intensity was calibrated with a standard photovoltaic reference cell. External quantum efficiency (EQE) spectrum was measured with Stanford lock-in amplifier 8300 unit.

High angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) image (Figure S1a) shows individual Mo and S atoms and the honeycomb crystalline order of MoS₂. The inset in Figure S1a shows the schematic drawing of the monolayer flake of MoS₂ along the (001) plane. X-ray Diffraction (XRD) patterns showed the peak of (002) plane unchanged while the other peaks shrink dramatically for the ce-MoS₂ sheets (Figure 3e) when compared with the bulk MoS₂ powder (Figure S1b). These results reveal the successful formation of single-layer ce-MoS₂ sheets, which are ready to be dispersed in deionized (DI) water as a colloid solution.

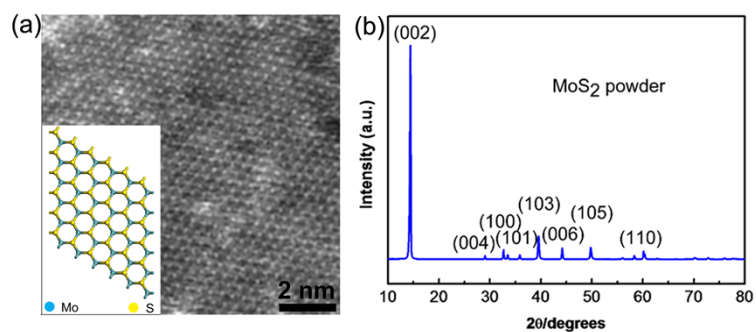


Figure S1. (a) High angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) image of ce-MoS₂ sheet. Inset: Structural models of MoS₂ (top view). (b) XRD patterns for MoS₂ powder.

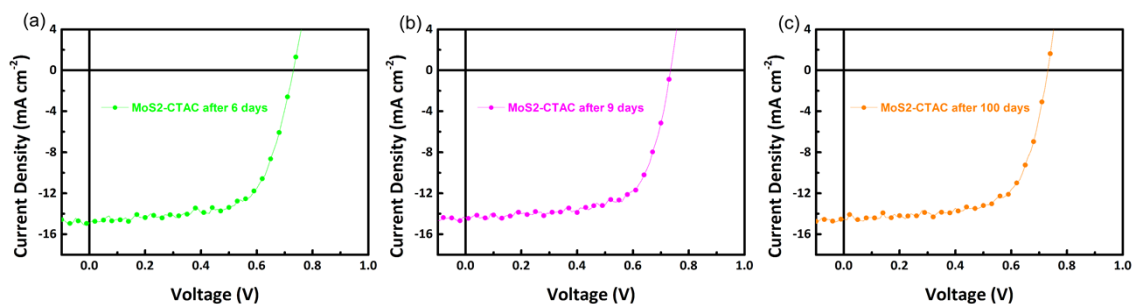


Figure S2. J-V curves of devices fabricated by using m-MoS₂ solution stored for 6 days (a), 9 days (b), 100 days (c).