Supporting Information

Effective Charge Separation of Inverted Polymer Solar Cells using Versatile MoS₂ Nanosheets as Electron Transport Layer

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† Electronic supplementary information (ESI) available: additional topographic images, and additional ADF and ABF images, and additional UPS data of PEIE/ITO/glass, and additional energy band diagram.
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1. Experimental.

2. The topographic images of MoS$_2$ NSs and line profile.

3. The topographic images of SiO$_2$ on Si substrate and PEIE on SiO$_2$ on Si substrate.

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1. Experimental

1.1. Exfoliation of MoS$_2$ Nanosheets: 2 g of the commercial MoS$_2$ sheets (2D semiconductor) was dispersed in 1 L of ethanol. The MoS$_2$ dispersion was treated by tip-sonication for 5 hours. Then, the MoS$_2$ dispersion was centrifuged for 30 min at 8000 rpm to collect a supernatant and remove precipitation. The initial concentration of the MoS$_2$ NS supernatant was 100 ± 2.7 mg/L. Then, the supernatant was diluted in 1mL of ethanol and the concentration was estimated to be about 16.67 mg/L.

1.2. Characterization of MoS$_2$ Nanosheets: The MoS$_2$ NSs were characterized through TEM, AFM, Raman spectroscopy, UV-vis absorption spectroscopy, and PL spectroscopy. The TEM images were obtained using a Philips Tecnai G2 F20 TEM microscope with an accelerating voltage of 200 kV. The AFM surface images and height distribution were measured by Park system NX10. The MoS$_2$ NSs solution in DMF was deposited on a silicon wafer by spin coating and drying under ambient conditions. The AFM image was obtained in a tapping mode. Different concentrations of MoS$_2$ NSs solution were used to achieve layers deposited on SiO$_2$/Si substrate. Raman spectra of MoS$_2$ NSs were obtained using a Horiba Jobin Yvon-Labram HR UV visible NIR Raman microscope spectrometer with a 514 nm Ar laser. Ultraviolet-visible (UV-vis) absorption spectra of the MoS$_2$ NSs measured with a Shimadzu, UV-2600 UV-vis spectrophotometer. PL spectra were obtained using a Horiba NanoLog-C. To demonstrated the MoS$_2$ NSs of energy levels using electrochemical cyclic voltammetry. MoS$_2$ NSs were dispersed in a 0.5% Nafion solution (isopropanol and dimethylformamide) and dropped on glassy carbon electrode to serve as the working electrode. The counter electrode and the reference electrode were platinum wire and Ag/AgNO$_3$ (in 0.01 M MeCN), respectively, and the electrolyte was a
solution of 0.1 M tetrabutylammonium phosphorus hexa fluoride (TBAPF6) in anhydrous acetonitrile. Measurements were performed at room temperature with a scan rate of 100 mV/s.

1. 3. Device fabrication process of iPSCs: 1.5 × 1.5 cm² patterned ITO-coated glass substrates were cleaned with acetone, ethanol, and 2-propanol, respectively. The ITO-coated glass was treated with O₂ plasma for 40 sec. PEIE layer was spin-coated on the ITO-coated glass at 4000 rpm for 40 sec, and the substrate was annealed at 110 °C for 10 min. After annealing, 0.1 ml of MoS₂ NSs in 1 ml of ethanol solution was spin-coated on the PEIE surface at 4000 rpm for 40 sec, followed by annealing at 140 °C for 10 min under ambient conditions. The substrates were transferred into a nitrogen-filled glove box. The blend of P3HT:PC₆₀BM (1.5:1 (by weight)), PTB7:PC₇₁BM (1:1.5 (by weight)), PTB7-Th:PC₇₁BM (1:1.5 (by weight)) solution were spin-coated onto the MoS₂ NSs interlayer at 2000 rpm for 10 sec and 1400 rpm for 10 sec, respectively. Finally, MoO₃ (10 nm) and Ag (80 nm) were deposited by thermal evaporation on the active layer under a pressure of 1x10⁻⁶ Torr.

1. 4. Characterization of iPSCs structures: The structures of iPSCs with the MoS₂ NSs/PEIE/ITO/glass were characterized through Focused Ion Beam (FIB), TEM and STEM-EDS. In detail, The FIB was applied for cross-sectional sample preparation of iPSCs with both TEM and STEM-EDS. Thinly sliced TEM samples were prepared using a FIB (Nova nanolab 600 Dual Beam) operating at 5~20 kV with Ga ions. The TEM images were obtained using a Philips Tecnai G2 F20 TEM microscope with an accelerating voltage of 200 kV. The STEM-EDS images were obtained using a JEM-ARM200F Cs-corrected STEM.

1. 5. Photovoltaic device characterization of iPSCs with/without MoS₂ Nanosheets: The iPSCs were then tested in air under an AM 1.5 G illumination of 100 mW/cm² (Oriel 1 kW solar
simulator), which was calibrated with the International System of Units (SI) (SRC-1000-TC
KG5-N, VLSI Standards, Inc) for accurate measurement. The external quantum efficiency (EQE)
was measured using an Oriel IQE-200 (Newport), a calibrated Si UV detector and an SR8570
low noise current amplifier. In TCSPC, photo-excitation was carried out by the mode-locked
titanium: sapphire laser with the wavelength of 400 nm and luminescence signals were detected
by an InGaAs based photomultiplier detector.
2. The topographic images of MoS$_2$ NSs and line profile.

Figure S1. Topographic images and line profiles of the MoS$_2$ NSs before (a) and (c) after tip-sonication exfoliation and centrifugation process (b) and (d) corresponding to the white line in the topographic images.
3. The topographic images of SiO\textsubscript{2} on Si substrate and PEIE on SiO\textsubscript{2} on Si substrate.

**Figure S2.** Topographic images RMS values of the SiO\textsubscript{2} on Si (a) and PEIE on SiO\textsubscript{2} on Si substrate (b).
4. The annular dark field (ADF) and annular bright field (ABF) images of the MoS$_2$ NSs on the PEIE/ITO/glass.

**Figure S3.** The annular dark field (ADF) and annular bright field (ABF) images of the MoS$_2$ NSs.

We performed image analysis of MoS$_2$ NSs by a spherical aberration-corrected scanning TEM (Cs-TEM). As shown in Fig. S2(a) and S2(b), we show the annular dark field (ADF) and annular bright field (ABF) images of the MoS$_2$ NSs and the ITO/glass substrate. Through the ADF and ABF images, the contrast of the MoS$_2$ NSs layer was improved by confirming the shape of the MoS$_2$ NSs. As a result, MoS$_2$ NSs applied as ETL could be clearly confirmed.
5. The work function (Φ) of the spin-coated PEIE onto ITO/glass and ITO/glass calculated by using an ultraviolet photoelectron spectroscopy (UPS).

![Graph showing intensity vs kinetic energy for PEIE/ITO and ITO](image)

**Figure S4.** The work function of the PEIE/ITO/Glass and ITO/Glass.

The work function value of the ITO, PEIE/ITO structures obtained 4.29 eV, 3.36 eV, respectively. In this structure, the PEIE layer plays a role as a surface modifying layer on the ITO layer for good electron selectivity and efficiently promotes electron transportation to decrease the work function of ITO due to the formation of dipole layer.¹
6. Energy level diagram of the iPSCs with the MoS$_2$ interlayer highlighting pathways for charge generation and transport.

**Figure S5.** Energy level diagrams of the inverted PSCs with the MoS$_2$ interlayer.

The energy level diagrams of iPSCs demonstrate the possible pathways of current generation and transportation after the excitation of the photoactive materials P3HT:PC$_{60}$BM (a) and PTB7-th:PC$_{71}$BM (b)$^2$. 
7. The structure of optical absorption simulation.

![Diagram of the solar cell structure](image)

**Figure S6. The structure of optical absorption simulation.**

The solar cell structure used for the simulation is the Ag (80 nm)/MoO₃ (10 nm)/PTB7:PC₇₁BM (120 nm)/MoS₂ (8.31 nm)/PEIE (3 nm)/ITO (150 nm)/glass (1.1 mm) structures. MoS₂ NS diameter: 25 nm and spacing between the edges of MoS₂ NS: 104 nm.
Reference
