Supplementary Information

Aging of Transition Metal Dichalcogenide Monolayers

Jian Gao[1], Baichang Li[1], Jiawei Tan[1], Phil Chow[1], Toh-Ming Lu[2*] and Nikhil Koratkar[1,3*]

[1]Materials Science and Engineering, Rensselaer Polytechnic Institute, Troy, NY 12180, USA

[2]Physics, Applied Physics, and Astronomy, Rensselaer Polytechnic Institute, Troy, NY 12180, USA

[3]Mechanical, Aerospace and Nuclear Engineering, Rensselaer Polytechnic Institute, Troy, NY 12180, USA

* Address Correspondence to T.M.L. (lut@rpi.edu) and N.K. (koratn@rpi.edu)

TABLE OF CONTENTS

Properties of polymer films:

The permeability to water moisture and O_2 of PMMA is 1.10×10^5 and 21.9 $cm^3 \cdot mil/(100\ in^2 \cdot 24\ hr \cdot atm)$,[1] respectively at 25 °C. The permeability to water moisture and O_2 of parylene C is 0.14 and 7.1 $cm^3 \cdot mil/(100\ in^2 \cdot 24\ hr \cdot atm)$ at 23 °C.[2]

Stoichiometric calculation from XPS:

The stoichiometric ratio (C_A:C_B) is calculated by dividing the core level areas (S_A, S_B) after adjusting for their residual sensitivity factors (RSF),

$$\frac{C_A}{C_B} = \frac{\frac{S_A}{RSF_A}}{\frac{S_B}{RSF_B}}$$

The corrected RSF of Mo3d, W4f, S2p, O1s, C1s are 88.654, 96.748, 17.899, 16.557, 6.948, respectively, from the PHI Multipak software database.

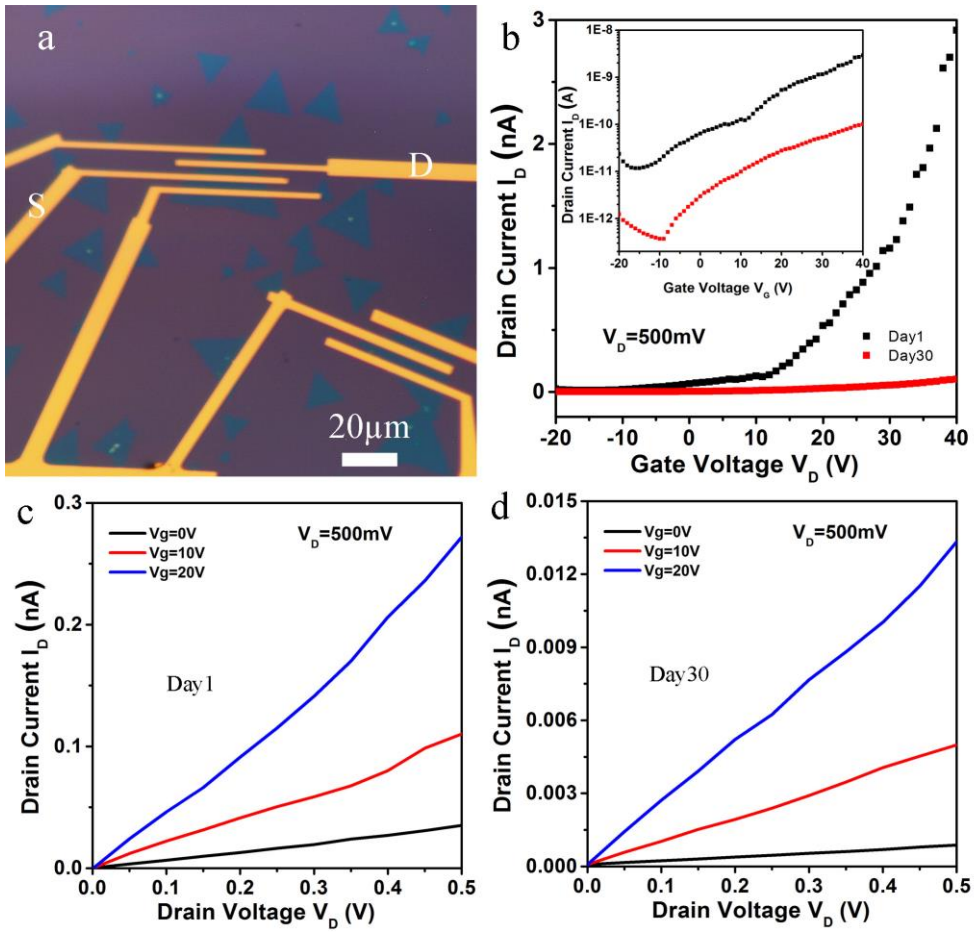

Figure S1: (a) Transistor device using CVD grown monolayer MoS_2 sheet on SiO_2/Si substrate. (b-d) The device was stored in vacuum (~ 1 Torr) for 3 months. The Day 1 data is when the sample was removed from the vacuum and tested. The Day 30 data is taken after the same device was continuously exposed to the ambient (room temperature and atmospheric pressure) for 30 days and then re-tested. We find up to two orders of magnitude reduction in the drain current in response to the applied gate and drain voltages, which confirms severe reduction in device performance due to aging. Note that the initial current (the Day 1 data) is already lower than the typical values reported in the literature, probably due to storage for 3 months in marginal vacuum.

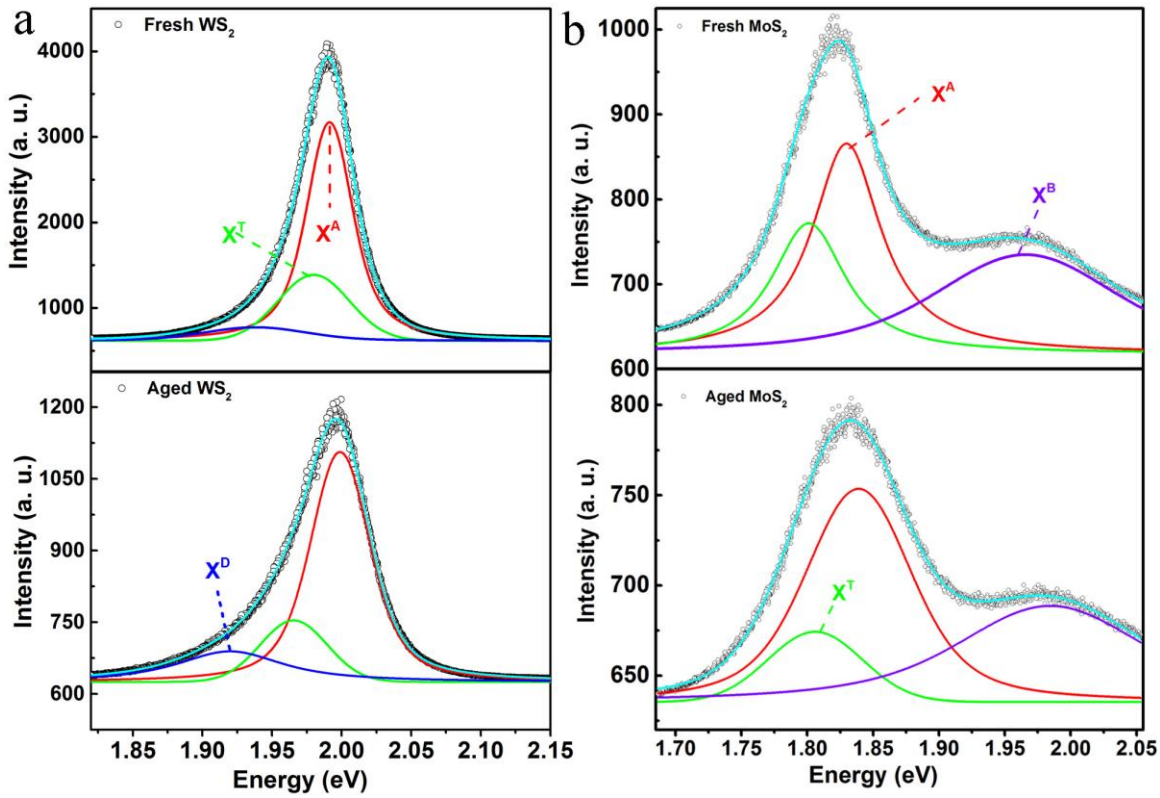

Figure S2: (a) PL spectra of fresh (top) and 1-year aged (bottom) CVD-grown monolayer WS_2. The A-exciton peak (red), trion peak (green), defect peak (blue) are marked after curve fitting of the PL spectral lineshape. The defect peak intensity increased significantly in the aged WS_2 samples. (b) PL spectra of fresh (top) and 1-year aged (bottom) CVD-grown monolayer MoS_2. The A-exciton (red), B-exciton (violet), and trion (green) contributions are marked on the plot.

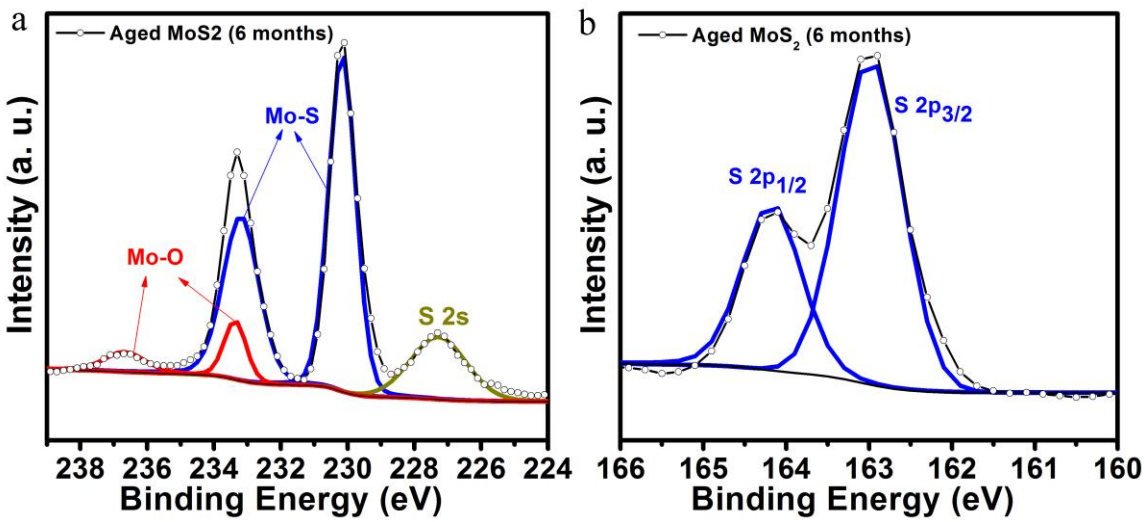

Figure S3: XPS spectra of Mo 3d (a) and S 2p (b) of MoS_2 stored in air for 6 months.

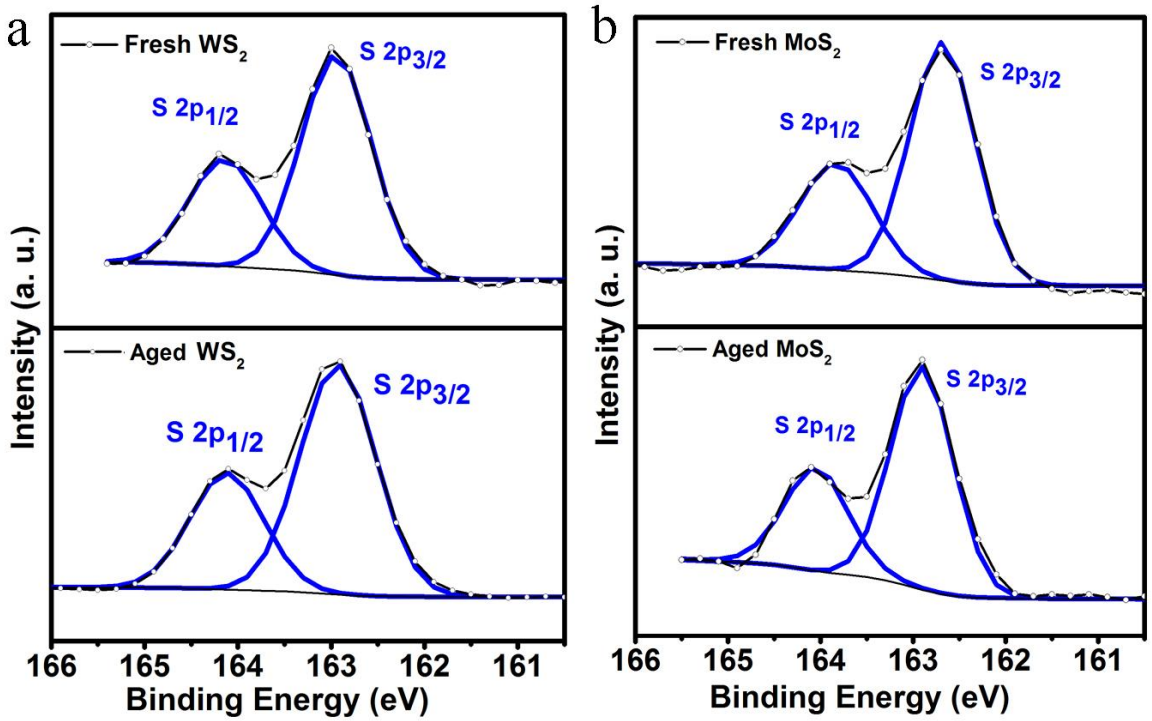

Figure S4: XPS spectra of the core-level binding energies of S 2p of WS_2 (a) and MoS_2 (b). There is no significant difference in the binding energy position for the fresh and aged samples.

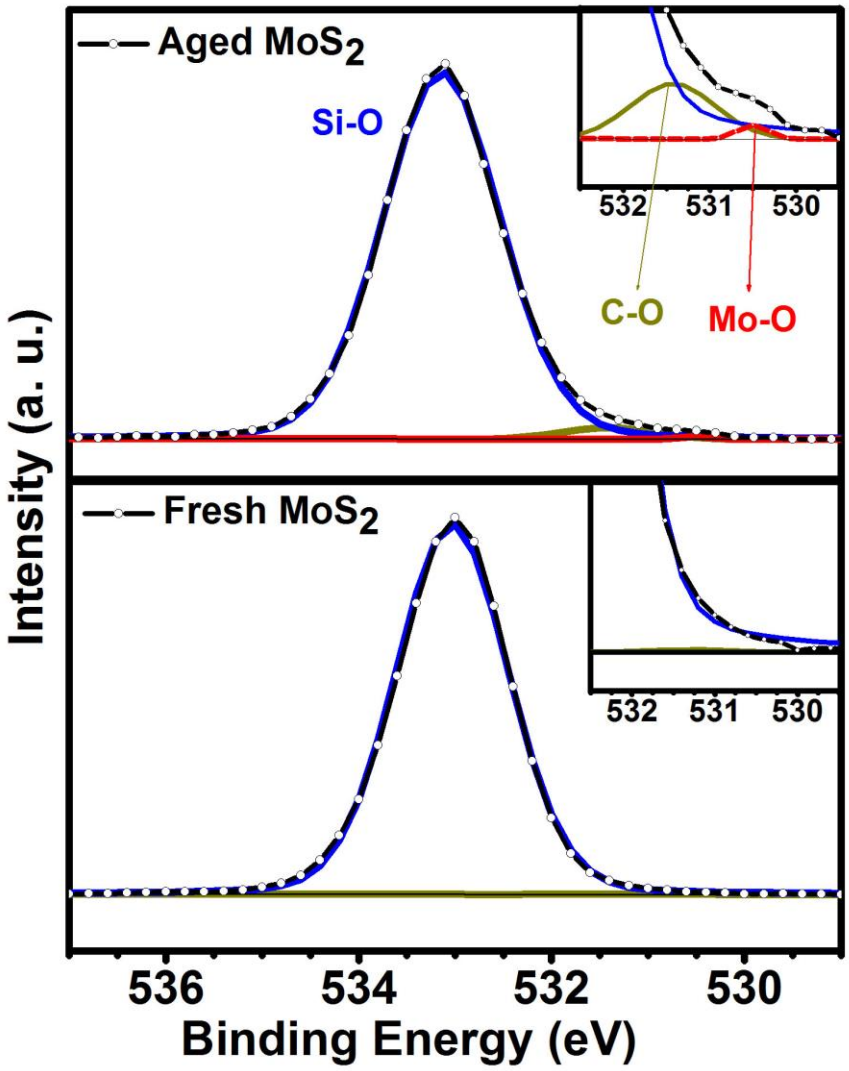

Figure S5: XPS O1s spectra of the fresh (top) and 1-year aged (bottom) CVD-grown monolayer MoS_2 sheet. The aging was performed under ambient conditions (room temperature and atmospheric pressure).

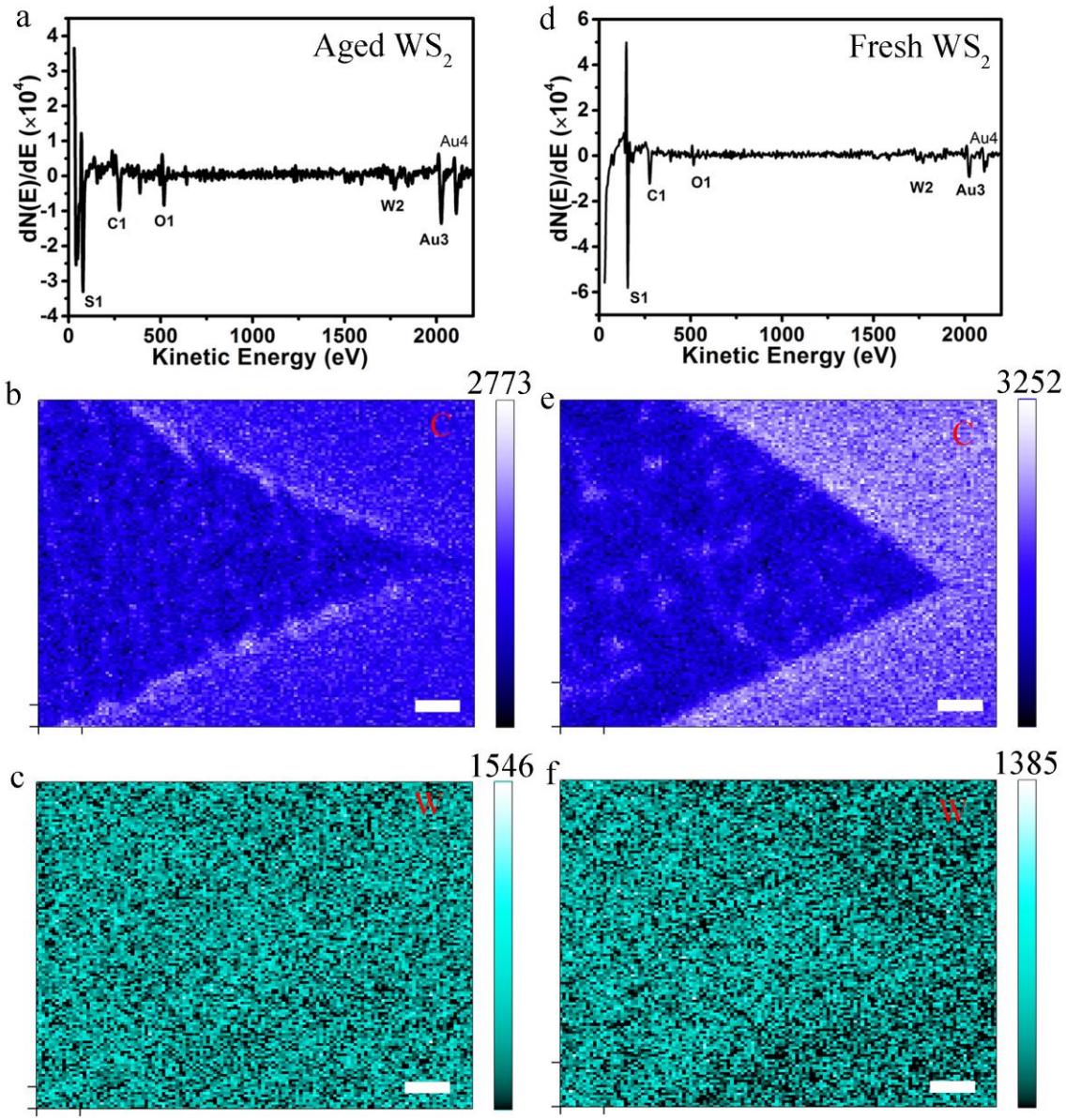

Figure S6: (a) Auger survey, AES mapping for (b) Carbon, (c) Tungsten of aged CVD-grown monolayer WS_2. (d) Auger survey, AES mapping for (e) Carbon, (f) Tungsten of fresh CVD-grown monolayer WS_2. The scale bar is 1 μm.

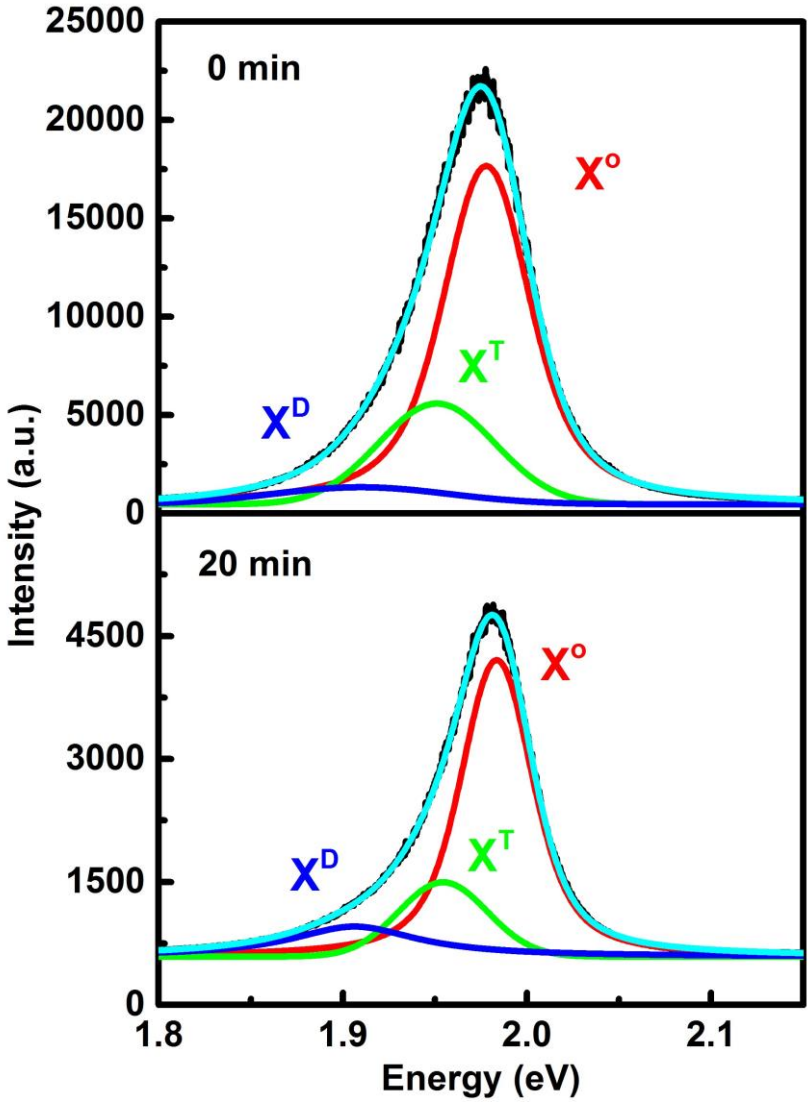

Figure S7: PL spectra with Lorentzian-Gaussian fitting of bare WS_2 before and after 20 min of accelerated aging treatment. Note that the WS_2 sheet is 5-to-6 fold lower in PL intensity after aging.

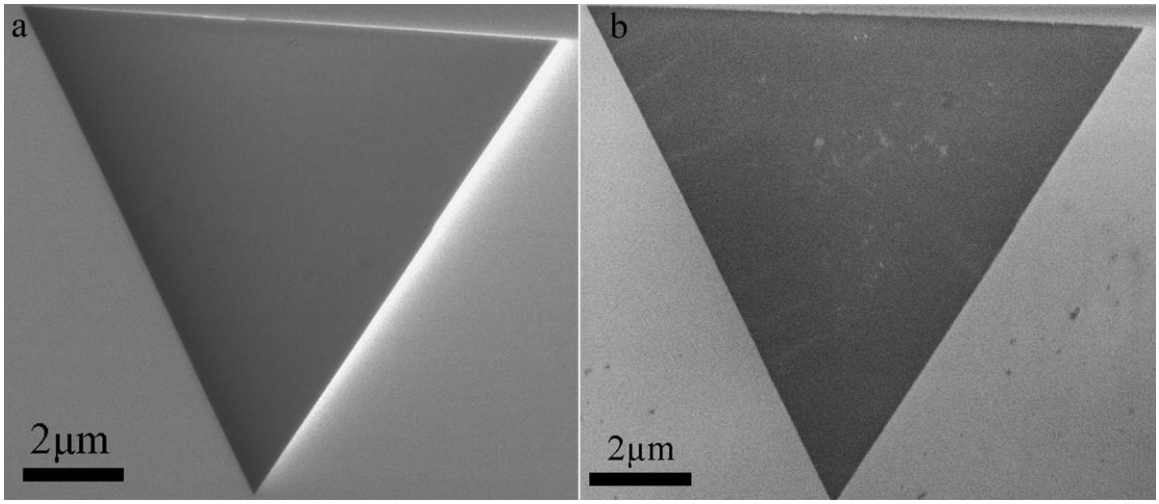

Figure S8: Monolayer WS_2 on sapphire before (a) and after (b) accelerated aging at ~80 °C for ~20 min and humidity of ~ 65%. The appearance of cracks is evident after aging.

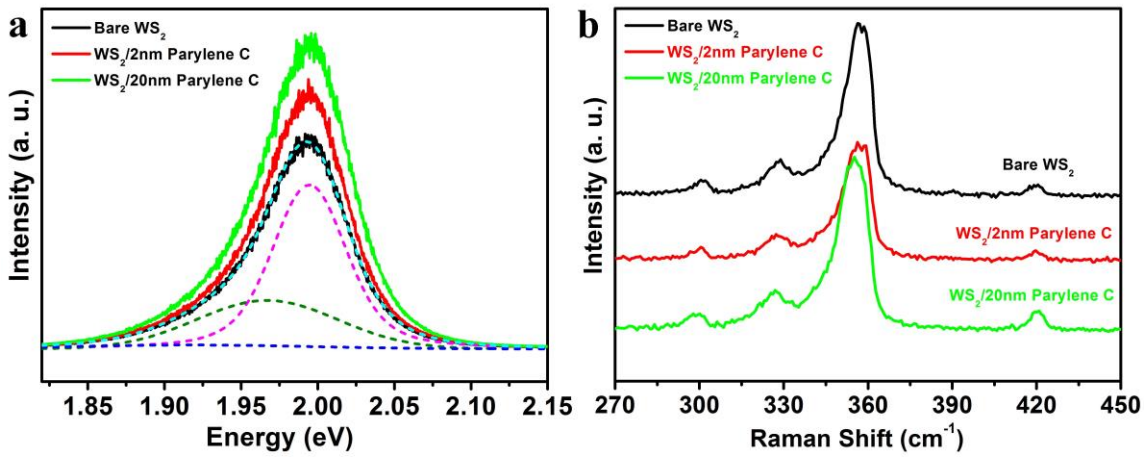

Figure S9: PL spectra (a) and Raman spectra (b) of WS_2 with and without Parylene C coating, before oxidation treatment.

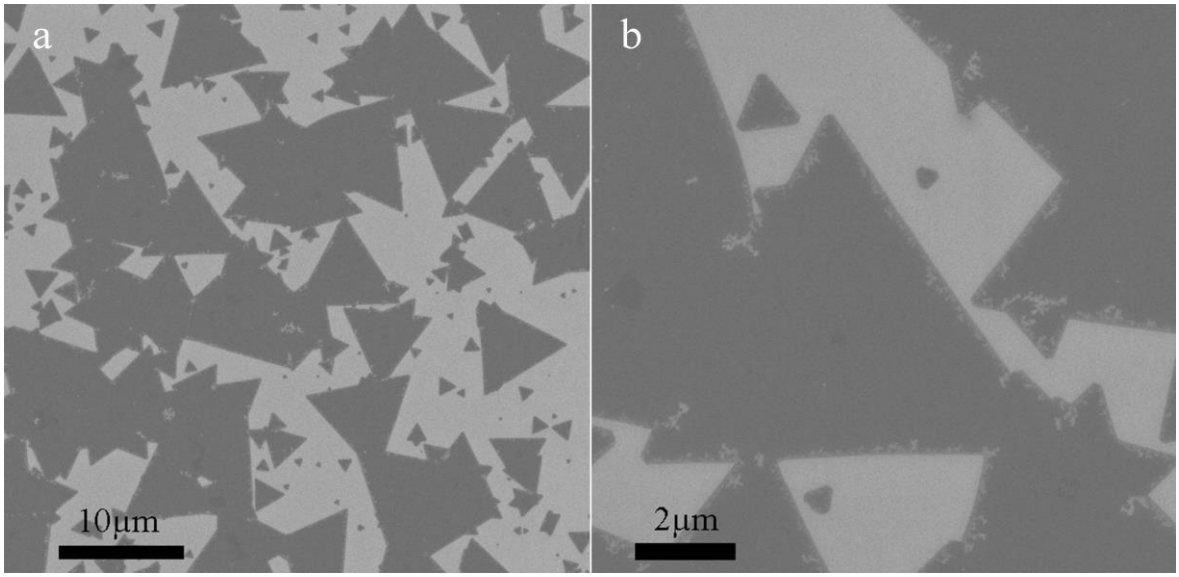

Figure S10: SEM images of MoS_2 on SiO_2 covered by PMMA, after one year in air. (a) Low magnification, (b) high magnification.

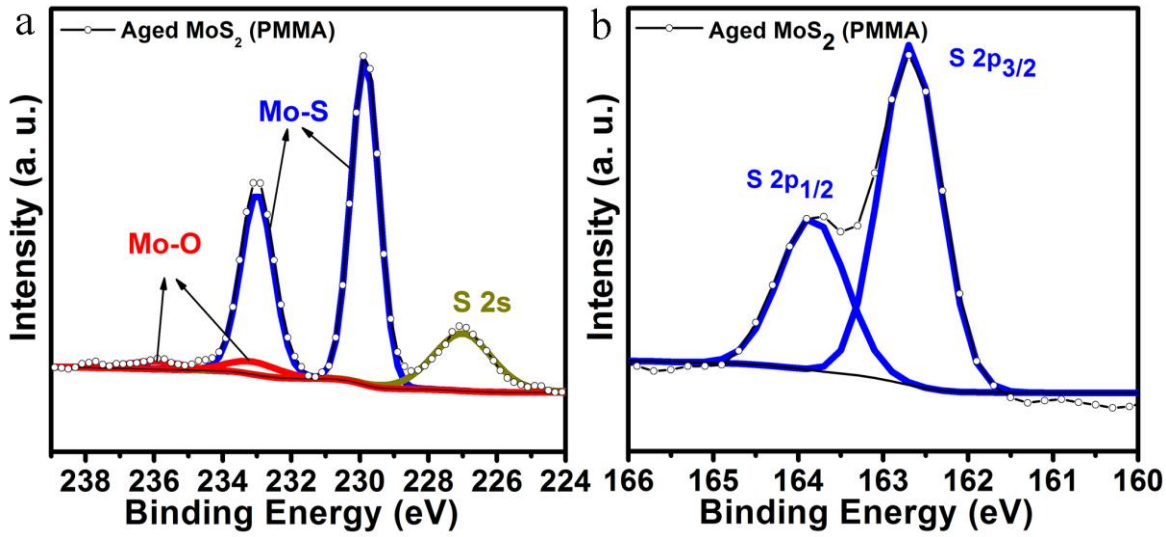

Figure S11: XPS spectra of the core-level binding energies of Mo 3d (a) and S 2p (b) of the one-year-old MoS_2 protected by a layer of PMMA. One more set of Mo 3d peaks is revealed by Lorentz-Gaussian fitting, corresponding to the oxide state. The percentage of oxide Mo|O state is 5.8%, which is much smaller than that of unprotected MoS_2.

Supplementary References:

1 Hess, S., Demir, M. M., Yakutkin, V., Baluschev, S. & Wegner, G. Investigation of Oxygen Permeation through Composites of PMMA and Surface-Modified ZnO Nanoparticles. *Macromol Rapid Comm.* **2009**, *30*, 394-401.

2 Spivack, M. A. & Ferrante, G. Determination of Water Vapor Permeability and Continuity of Ultrathin Parylene Membranes. *J Electrochem Soc* **1969**, *116*, 1592.