

Supporting Information

## **Precise Plasma Engineering of Few-Layer 1T-MoS<sub>2</sub> Self-Supported Electrodes for Bifunctional Energy Storage and Electrocatalysis**

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### **List of Texts and Figures**

**Text S1.** Experimental section.

**Text S2.** Electrochemical measurement and evaluations.

**Text S3.** Preparation of flexible symmetric supercapacitors (SSC) devices.

**Text S4.** Material characterizations.

**Figure S1.** SEM images of MoS<sub>2</sub>-0, MoS<sub>2</sub>-50, MoS<sub>2</sub>-100 and MoS<sub>2</sub>-150.

**Figure S2.** TEM images of MoS<sub>2</sub>-0 at different magnification.

**Figure S3.** Nitrogen absorption–desorption isotherms and Pore diameter distributions.

**Figure S4.** The specific capacitances at 1 mA cm<sup>-2</sup> to 20 mA cm<sup>-2</sup> of MoS<sub>2</sub>-0, MoS<sub>2</sub>-50, MoS<sub>2</sub>-100 and MoS<sub>2</sub>-150.

**Table S1.** Comparison of capacitance retention after cycling based on M-Co(1.5%) with recently reported supercapacitor electrode materials.

### **Text S1. Experimental section**

*Chemicals:* Sodium molybdate (Na<sub>2</sub>MoO<sub>4</sub>·2H<sub>2</sub>O), thioacetamide (C<sub>2</sub>H<sub>5</sub>NS), sodium borohydride (NaBH<sub>4</sub>), and anhydrous sodium sulfate were (Na<sub>2</sub>SO<sub>4</sub>, 99%) were purchased from Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China). All chemicals were used without any further purification.

### **Text S2. Electrochemical measurement and evaluations**

The electrochemical properties are tested by using the three-electrode test method. The three electrodes consist of a counter electrode, a reference electrode and a working electrode. The counter electrode is MoS<sub>2</sub>-0, MoS<sub>2</sub>-50, MoS<sub>2</sub>-100 and MoS<sub>2</sub>-150. Cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) are all measured in a 1 M Na<sub>2</sub>SO<sub>4</sub> solution using a Dutch Ivium (Vertex C. DC) electrochemical station.

To evaluate the electrocatalytic performance of the as-prepared materials, electrochemical tests are conducted using a standard three-electrode system. A mercury/mercury oxide (Hg/HgO) electrode serves as the reference electrode, a

platinum plate electrode serves as the counter electrode, and the as-prepared MoS<sub>2</sub>-0, MoS<sub>2</sub>-50, MoS<sub>2</sub>-100 and MoS<sub>2</sub>-150 is directly used as the working electrode. All electrochemical measurements are performed in a 0.5 M sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) electrolyte. The linear sweep voltammetry (LSV) and electrochemical impedance spectroscopy (EIS) curves are both recorded using a Dutch Ivium (Vertex C.DC) electrochemical workstation.

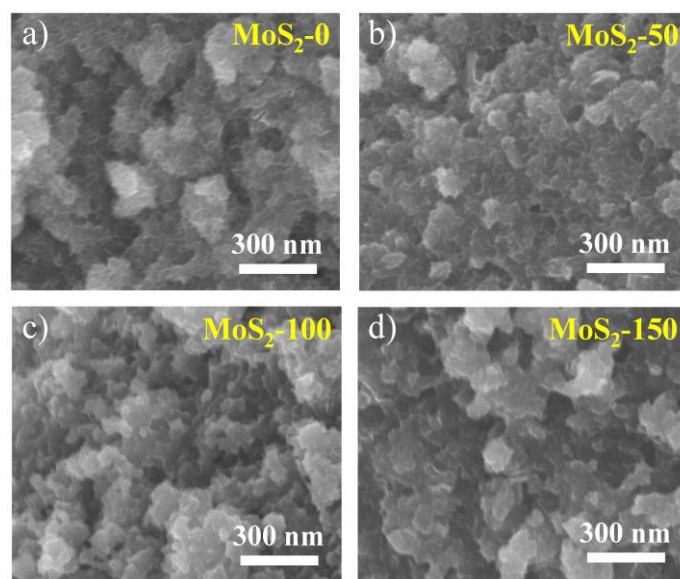
### **Text S3. Preparation of flexible symmetric supercapacitors (ASC) devices**

In the process of fabricating a flexible symmetric supercapacitor (ASC) device, 1 g of polyvinyl alcohol (PVA) powder is first dissolved in 10 mL of deionized water. The resulting solution is then heated to 90 °C under continuous stirring, and 2 mL of 0.5 M Na<sub>2</sub>SO<sub>4</sub> solution is added. MoS<sub>2</sub>-100 is used as both the positive and negative electrode material in the PVA-Na<sub>2</sub>SO<sub>4</sub> gel electrolyte. After curing at room temperature, the flexible ASC device is successfully assembled.

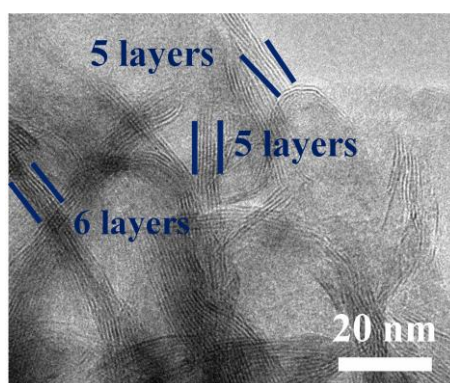
### **Text S4. Material characterizations**

XRD spectra were collected on X-ray diffraction (PANalytical Empyrean) with Cu K radiation ( $\lambda = 0.154$  nm). Raman spectra were obtained by a spectrometer (TACHI Regulus8220) with 532 nm laser excitation. Materials nanostructure characterizations come from Field emission scanning electron microscope (FE-SEM, Hitachi Regulus8220) and Energy-dispersive X-ray spectroscopy (EDX, Oxford EDX, with INCA software), and transmission electron microscope (TEM, JEM-2100) with configured EDX. Atomic Force Microscope (AFM). Valence structure was collected by X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB 250Xi equipped with

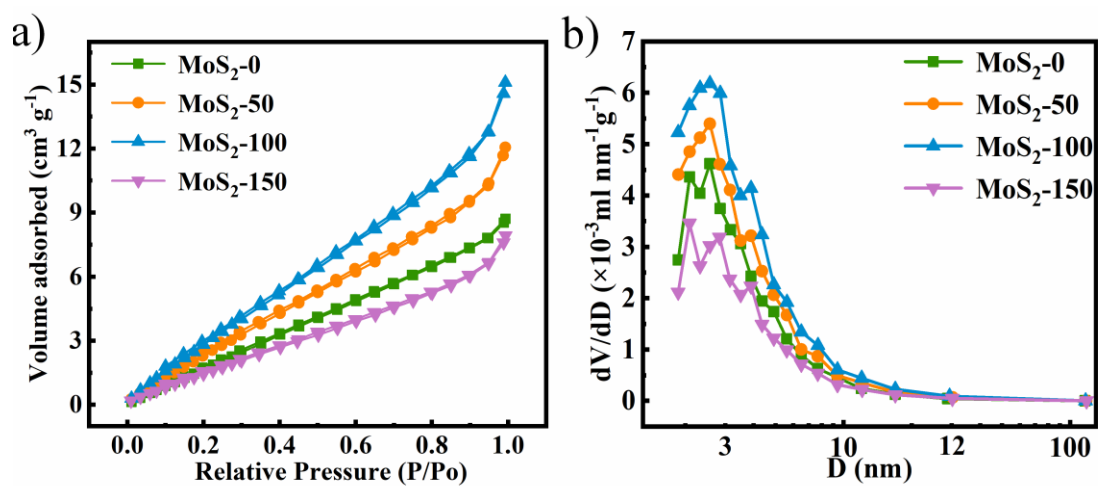
monochromatic Al K $\alpha$  source). Fourier Transform infrared spectroscopy (FTIR).



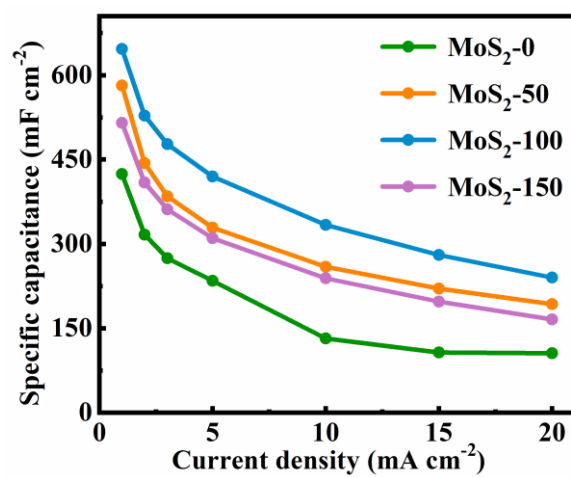
**Figure S1.** (a-d) SEM images of MoS<sub>2</sub>-0, MoS<sub>2</sub>-50, MoS<sub>2</sub>-100 and MoS<sub>2</sub>-150 are shown in sequence.



**Figure S2.** (a-b) TEM images of MoS<sub>2</sub>-0 at different magnification.



**Figure S3.** (a) Nitrogen absorption–desorption isotherms. (b) Pore diameter distributions.



**Figure S4.** The specific capacitances at 1 mA cm<sup>-2</sup> to 20 mA cm<sup>-2</sup> of MoS<sub>2</sub>-0, MoS<sub>2</sub>-50, MoS<sub>2</sub>-100 and MoS<sub>2</sub>-150.

**Table S1.** Comparison of energy density and power density based on MoS<sub>2</sub>-100 with recently reported supercapacitor symmetrical device[1–5].

| Flexible device                                                                                                          | Energy and power density                                                                                               | Refs.                                                   |
|--------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------|
| MoS <sub>2</sub> -100// MoS <sub>2</sub> -100                                                                            | 135.6 $\mu\text{Wh cm}^{-2}$ at 800 $\mu\text{W cm}^{-2}$<br>45.2 $\mu\text{Wh cm}^{-2}$ at 4000 $\mu\text{W cm}^{-2}$ | This work                                               |
| s-MnO <sub>2</sub> /MoS <sub>2</sub> // s-MnO <sub>2</sub> /MoS <sub>2</sub>                                             | 3.8 $\mu\text{Wh cm}^{-2}$ at 50 $\mu\text{W cm}^{-2}$<br>1.1 $\mu\text{Wh cm}^{-2}$ at 500 $\mu\text{W cm}^{-2}$      | <i>J. Power Sources.</i><br><b>2020</b> , 450, 227616.  |
| MoS <sub>2</sub> /AIP//MoS <sub>2</sub> /AIP                                                                             | 12.4 $\mu\text{Wh cm}^{-2}$ at 6043 $\mu\text{W cm}^{-2}$                                                              | <i>Adv. Mater.</i><br><b>2021</b> , 22, 875-884         |
| 1T-MoS <sub>2</sub> /Ti <sub>3</sub> C <sub>2</sub> MXene//<br>1T-MoS <sub>2</sub> /Ti <sub>3</sub> C <sub>2</sub> MXene | 17.4 $\mu\text{Wh cm}^{-2}$ at 600 $\mu\text{W cm}^{-2}$                                                               | <i>Adv. Funct. Mater.</i><br><b>2020</b> , 30, 1910302  |
| MnO <sub>2</sub> @Ni-mesh//<br>MoS <sub>2</sub> @Ni-mesh                                                                 | 0.86 $\mu\text{Wh cm}^{-2}$ at 16 $\mu\text{W cm}^{-2}$                                                                | <i>J. Power Sources</i><br><b>2016</b> , 336, 99-106.   |
| 3D NM@ NiCo BH//<br>3D NM NPC                                                                                            | 51.46 $\mu\text{Wh cm}^{-2}$ at 750 mW cm <sup>-2</sup>                                                                | <i>Adv. Funct. Mater.</i><br><b>2021</b> , 31, 2100290. |

## References

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