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Electronic Supplementary Information (ESI) for

2H-MoS₂/carbon cloth composite for high-performance all-solid-state supercapacitor derived from molybdenum dithiocarbamate complex

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S1. Chemicals and Materials

Diethylamine (Et₂NH, 99.8%), sodium hydroxide (NaOH, 95.0%), carbon disulfide (CS₂, 98.0%), methanol, diethyl ether, molybdenum trioxide (MoO₃, 99.0%) were purchased from Energy Chemical Co., Ltd. and used without further purification. Pure water (960 k Ω cm) was produced by UNIQUE-S10 RO-DI water purification system and used for the preparation of all aqueous solutions. Carbon cloth (CC), polyvinyl alcohol (PVA) powder, hydrophilic cellulose membranes were obtained from Saibo Electrochemical Co., Ltd. and used as received, unless otherwise stated.

S2. Syntheses and Preparations

S2.1. Synthesis of sodium *N*,*N*-diethyldithiocarbamate (Na(Et₂dtc))

The sodium dithiocarbamate complex was synthesized according to literature reported procedures with some modifications. ^{S1} Under a N₂ atmosphere, Diethylamine (2.1 mL, 20.0 mmol) and NaOH (1.0 g, 25.0 mmol) were mixed and then dissolved in 20 mL of methanol with vigorous stirring for 30 minutes at room temperature. Carbon disulfide (1.7 mL, 28.0 mmol) was then added dropwise at 0 °C, and the low temperature was maintained during the addition. The resulting yellow slurry was recrystallized from diethyl ether to obtain the pale yellow crystalline Na(Et₂dtc) compound. Yield: 1.2 g (82 % based on Et₂NH).

S2.2. Synthesis of Mo₂O₂(*µ*-S)₂(Et₂dtc)₂ complex

A mixture of MoO₃ (144 mg, 1.0 mmol), as-synthesized Na(Et₂dtc) (242 mg, 1.5 mmol), and DMF/H₂O (1:1 in v/v, 10 mL) was sealed in a Teflon-lined stainless steel autoclave. The autoclave was heated and kept at 90 °C for 2 days. After cooling down to ambient temperature, golden crystals of Mo₂O₂(μ -S)₂(Et₂dtc)₂ were isolated and dried in air. Yield: 532 mg (60.8% based on Mo).

S2.3. Pyrolysis of Mo₂O₂(*µ*-S)₂(Et₂dtc)₂ complex at different temperatures

The as-synthesized Mo₂O₂(μ -S)₂(Et₂dtc)₂ complex was uniformly placed in a quartz boat and put at the heating center of the furnace tube. The sample was calcinated at a selected temperature (T = 100, 200, 250, 300, 310, 320, 330, 340, 350, 400, 500, 600, 700, 800, 900 °C) for 1 h, with a heating rate of 2 °C min⁻¹ under an N₂ atmosphere. The resultant product was collected for further characterizations and denoted as MoS₂-T.

S3. X-ray Crystallography. A golden yellow block single crystal of complex $Mo_2O_2(\mu-S)_2(Et_2dtc)_2$ was mounted on a glass fiber attached to a copper mounting pin. Crystallographic data were collected on a Rigaku XtalLAB PRO diffractometer with Cu K α radiation at 293(2) K. Empirical absorption corrections were applied using spherical harmonics, implemented in the SCALE3 ABSPACK scaling algorithm. ^{S2} The structure was solved using direct methods, which yielded the positions of all non-hydrogen atoms. Hydrogen atoms were placed in calculated positions in the final structure refinement. Structure determination and refinement were carried out using the SHELXTL software package with Olex2 graphical user interface.^{S3,S4} Detailed crystallographic data are summarized in Table S1.

S4. Equations used to calculate the mass specific capacitance of 2H-MoS₂/CC electrode as well as the mass specific capacitance, energy density, power density and coulombic efficiency of ASSS device

$$C = I\Delta t / (m\Delta V) \tag{1}$$

where *I* denotes the discharge current (A), Δt represents the discharge time (s), ΔV is the potential window (V), and *m* is the mass of the active materials in the electrode (g).

$$E = 0.5C(\Delta V)^2 / 3.6 \tag{2}$$

$$P = 3600 E / \Delta t \tag{3}$$

$$\eta = \Delta t_{\rm d} / \Delta t_{\rm c} \tag{4}$$

where *C* denotes the mass specific capacitance of the device (F g⁻¹), *E* and *P* represent the energy (W h kg⁻¹) and power density (W kg⁻¹) of the supercapacitor, respectively. η stands for coulombic efficiency (%), Δt_d and Δt_c are discharging and charging times (s), respectively.

S5. Assembly of all-solid-state symmetric supercapacitor

In order to prepare the PVA-H₂SO₄ gel electrolyte, polyvinyl alcohol (PVA) powder (1 g) was dissolved into DI water (10 mL), and then the mixture was heated at 90 °C under vigorous stirring until the solution became clear. Furthermore, 10 mL of aqueous H₂SO₄ (1 M) was added dropwise into the above solution under constant stirring until the mixed solution became transparent once again.

To fabricate the all-solid-state symmetric supercapacitor, two $2H-MoS_2/CC$ flexible electrodes coated with PVA-H₂SO₄ gel electrolyte were assembled into a supercapacitor configuration via sandwiching hydrophilic cellulose membrane as a separator between them. After the PVA-H₂SO₄ gel was naturally solidified at room temperature for *ca.* 12 h, the all-solid-state symmetric supercapacitor device was obtained.

S6. Kinetic mechanism analyses:

The measured i_{total} consists of two parts: the slow diffusion-controlled process (i_{diff}) and the capacitance-dominated process (i_{cap}) including ion adsorption/desorption (i.e. EDLC process) and the fast faradaic reaction of redox species, which can be described by equations 1 and 2:

$$\mathbf{i}_{\text{total}} = \mathbf{i}_{\text{cap}} + \mathbf{i}_{\text{diff}} = \mathbf{a}\mathbf{v}^{\text{b}} \tag{5}$$

$$\log i = \log a + b \log v \tag{6}$$

where i is the measured current density in CV curves (A g^{-1}), v is the scan rates (V s^{-1}), and (a, b) are the fitting parameters obtained from log i versus log v plots. It is widely recognized that the b value near 0.5 represents the slow diffusion-controlled processes, while the b value close to 1 indicates the process are dominated by capacitive effects. In addition, further quantification of the capacitive and diffusion contributions to the total capacitances are analyzed using equations **3** and **4**:

$$i_{\text{total}} = i_{\text{cap}} + i_{\text{diff}} = k_1 \nu + k_2 \nu^{1/2}$$
 (7)

$$i_{\text{total}}/v^{1/2} = k_1 v^{1/2} + k_2 \tag{8}$$

S7. Capacitance mechanism analyses:

According to the CV curves, the calculated accumulated charge (Q) can be calculated by the following equation:

$$Q = \int i dV/2vm$$
(9)

where i is the measured current density in CV curves (A g^{-1}), v is the scan rates (V s^{-1}), and m (g) is the mass of the active material in the electrode.

The capacitance is constituted by the contributions of electrical double-layer capacitance (C_{dl}) and pseudocapacitance (C_p), which can be calculated from the plots of Q versus $v^{-1/2}$ and Q⁻¹ versus $v^{1/2}$. When $v \rightarrow 0$, the electrode will have enough time to store the maximum amount of charge to reach its maximum capacitance (C_T), conversely, when $v \rightarrow \infty$, the maximum electrical double-layer capacitance (C_{dl}) can be obtained from the intercept of the plots of Q versus $v^{-1/2}$. Pseudocapacitance (C_p) can be calculated using Equation **6**:

$$C_p = C_T - C_{dl} \tag{10}$$

Complex	$Mo_2O_2(\mu-S)_2(Et_2dtc)_2$
Empirical formula	$C_{10}H_{20}Mo_2N_2O_2S_6$
Formula weight	584.52
Temperature (K)	293(2)
Crystal system	monoclinic
Space group	C2/c
<i>a</i> (Å)	34.0378(9)
<i>b</i> (Å)	6.9406(2)
<i>c</i> (Å)	17.9645(5)
α (°)	90
β (°)	106.222(3)
γ (°)	90
$V(\text{\AA}^3)$	4075.0(2)
Ζ	8
$D_{\rm c}({\rm g~cm^{-3}})$	1.905
$\mu (\mathrm{mm}^{-1})$	15.879
<i>F</i> (000)	2320
Reflections collected	10123
Independent reflections	3957
Completeness	99.9%
R _{int}	0.0328
R_1^a , w $R_2^b(I > 2\sigma(I))$	0.0667, 0.1597
GOF on F^2	1.102

Table S1 Crystal data and structure refinements for $Mo_2O_2(\mu$ -S)₂(Et₂dtc)₂.

 $a R_1 = \sum (||F_0| - |F_c||) / \sum |F_0|. b WR_2 = \sum [W(F_0^2 - F_c^2)^2] / \sum [W(F_0^2)^2]^{1/2}$



Fig. S1 Experimental (top) and simulated (bottom) PXRD patterns for $Mo_2O_2(\mu-S)_2(Et_2dtc)_2$.



Fig. S2 SEM images of $Mo_2O_2(\mu$ -S)₂(Et₂dtc)₂.at randomly selected areas.



Fig. S3 TGA profile of $Mo_2O_2(\mu$ -S)₂(Et₂dtc)₂ under N₂.



Fig. S4 The digital photos of MoS_2 -T samples (T = 310 to 350).



Fig. S5 PXRD pattern of MoS_2 -T (T = 310, 320, 330, 340 and 350).



Fig. S6 PXRD pattern of MoS_2 -T (T = 400 to 900).



Fig. S7 SEM images of MoS_2 -300 at different magnification.



Fig. S8 SEM images of MoS_2 -400 at different magnification.



Fig. S9 SEM images of MoS₂-500 at different magnification.



Fig. S10 SEM images of MoS_2 -600 at different magnification.



Fig. S11 SEM image of MoS_2 -700.



Fig. S12 SEM images of MoS₂-800 at randomly selected areas.



Fig. S13 SEM images of MoS_2 -900 at randomly selected areas.



Fig. S14 (a) N₂ adsorption and desorption isotherm curves of MoS₂-T (T = 400, 600, 800 and 900); (b) BJH desorption pore size distribution of MoS₂-600.



Fig. S15 N₂ adsorption and desorption isotherm curves of MoS₂-T (T = 250, 300, 310 and 330).

Table S2 Specific surface areas for MoS_2 -T materials obtained from BETmeasurements.

Materials	Specific surface area
	$(m^2 g^{-1})$
MoS ₂ -250	11.6
MoS ₂ -300	47.6
MoS ₂ -310	50.3
MoS ₂ -330	56.8
MoS ₂ -400	76.4
MoS ₂ -600	77.3
MoS ₂ -800	72.7
MoS ₂ -900	71.6



Fig. S16 XPS (**a**) survey spectra and high-resolution scans of (**b**) C 1s, (**c**) N 1s, and (**d**) O 1s electrons of MoS₂-600.



Fig. S17 Digital images of $Mo_2O_2(\mu$ -S)₂(Et₂dtc)₂/CC (left) and 2H-MoS₂/CC (right).



Fig. S18 PXRD patterns for 2H-MoS₂/CC and Mo₂O₂(μ -S)₂(Et₂dtc)₂/CC (orange sparse-line region represents diffraction peaks from Mo₂O₂(μ -S)₂(Et₂dtc)₂).



Fig. S19 SEM images of $Mo_2O_2(\mu$ -S)₂(Et₂dtc)₂/CC at different magnification.



Fig. S20 TEM images of $2H-MoS_2$ at randomly selected areas.



Fig. S21 The SAED image of 2H-MoS₂.







Fig. S23 CV partition analysis for the comparison of capacitive and diffusion-controlled contribution at different scan rates: (a) 50 mV s⁻¹, (b) 20 mV s⁻¹, (c) 10 mV s⁻¹, and (d) 5 mV s⁻¹.



Fig. S24 Dependence of (**a**) Q on $v^{-1/2}$ and (**b**) Q^{-1} on $v^{1/2}$ for 2H-MoS₂/CC electrode in 1 M H₂SO₄.

Electrode materials	Capacitance (F g ⁻¹)	Electrolyte	Reference
2H-MoS ₂ nanoflowers	$382 \text{ F g}^{-1} \text{ at } 2 \text{ A g}^{-1}$	1 M Na ₂ SO ₄	S5
2H-MoS ₂ /GNF/CNT	104 F g ⁻¹ at 0.5 A g ⁻¹	1 M Na ₂ SO ₄	S 6
Ammoniated 1T/2H MoS ₂	346 F g^{-1} at 1 A g^{-1}	2 M KOH	S7
1T/2H MoS ₂	259 F g ⁻¹ at 5 mV s ⁻¹	1 M KCl	S 8
Water coupled 1T MoS ₂	380 F g^{-1} at 5 mV s^{-1}	0.5 M Li ₂ SO ₄	S 9
High defect density 1T MoS ₂	379 F g ⁻¹ at 1 A g ⁻¹	1 M KOH	S10
1T MoS ₂ /TiO ₂ /Ti	428 F g^{-1} at 0.2 A g^{-1}	1 M Na ₂ SO ₄	S11
Carbon nanoparticle/MoS ₂	394.2 F g ⁻¹ at 5 mV s ⁻¹	1 M Na ₂ SO ₄	S12
1T-MoS ₂ /Ti ₃ C ₂ MXene	386.7 F g ⁻¹ at 1 A g ⁻¹	$1 \text{ M H}_2\text{SO}_4$	S13
MoS ₂ /N-doped graphene	245 F g^{-1} at 0.25 A g^{-1}	1 M KOH	S14
1T-MoS ₂ nanotubue	328.5 F g ⁻¹ at 1 A g ⁻¹	3 M KCl	S15
Highly defective MoS ₂ nanosheets	442.0 F g^{-1} at 1 A g^{-1}	1 M H ₂ SO ₄	S16
Electrostatically charged MoS ₂ /graphene	154 E a-l at 1 A a-l	$2 M N_{\odot} SO$	S17
oxide	154 F g · at 1 A g ·	$2 \text{ IM } \text{Na}_2 \text{SO}_4$	517
Tubular MoS ₂ /PANI	520 F g^{-1} at 1 A g^{-1}	$1 \text{ M H}_2 \text{SO}_4$	S18
MoS ₂ /PPy nanowire	462 F g^{-1} at 1 A g^{-1}	0.5 M Na ₂ SO ₄	S19
Layered MoS ₂ /PPy nanotube	307.5 F g ⁻¹ at 1 A g ⁻¹	1 M KCl	S20

 Table S3 Comparison of different MoS₂-based supercapacitor electrodes in three-electrode system.



Fig. S25 GCD curves of pristine CC in three cycles.



Fig. S26 GCD curves of 2H-MoS₂/CC electrode before and after 5000 cycles.

Table S4 Parameters of EI	S of 2H-MoS ₂ /CC electrode	before and after 5000 cycles.
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Items –	High frequency		Low frequency
	Rs	R _{ct}	W
Before 5000 cycles	0.12	< 10 ⁻⁵	0.31
After 5000 cycles	0.11	0.35	0.31

(Unit: Ω)



Fig. S27 Four all-solid-state symmetric supercapacitor device in series: (a) CV curves of a single device and four devices connected in series; (b) GCD curves of a single device and four devices connected in series.

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