Supporting Information

Glucose intercalation induced 1T-G-MoS₂ hybrids for high-

performance rechargeable aqueous zinc-ion batteries

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Experimental Section

Materials:

Ammonium molybdate tetrahydrate ((NH₄)₆Mo₇O₂₄·4H₂O, AR) and thiourea (CH₄N₂S, AR) were purchased from Sinopharm Chemical Reagent Co., Ltd. Glucose (C₆H₁₂O₆, AR), zinc trifluoromethane sulfonate (Zn(CF₃SO₃)₂, 98%) and N-methyl pyrrolidone (NMP, AR) were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. Poly(vinylidene fluoride) (PVDF) binder was purchased from Shanghai McLean Biochemical Co., Ltd. Acetylene black was purchased from Cyber Electrochemical Materials Network. Zinc foil was purchased from Hefei Wenghe Metal Materials Co., Ltd. Deionized water was obtained from Mili-Q water purification system. All chemicals for the experiments were used without any further modification.

Synthesis of $1T-C_x$ -MoS₂:

 $1\text{T-C}_{x}\text{-MoS}_{2}$ was synthesized according to previously published literature with a modification.¹ Typically, glucose (0.4 g) was ultrasonically dispersed in deionized water (20 mL) for 0.5 h. Thereafter, (NH₄)₆Mo₇O₂₄·4H₂O (0.36 g) and thiourea (0.60 g) were added into the glucose solution with continuously stirring. Subsequently, the solution was transferred into a 50 mL Teflon-lined autoclave and the reaction was continued for 12 h at 210 °C. After naturally cooling to room temperature, the product was collected by centrifugation and washed several times with deionized water and ethanol. Finally, the resultant product was dried at 60 °C in vacuum oven for 12 hours. In order to study the glucose content on the structure and composition of the obtained MoS₂, control samples with different glucose additions (0 g, 0.1 g, 0.2 g and 0.8 g) were also prepared using the same synthetic route.

Materials characterizations:

Field emission scanning electron microscopy (FE-SEM, S-400, Hitachi) and transmission electron microscopy (TEM, JEM-F200, JEOL) equipped with an energy dispersive X-ray (EDX) were used to analyze the morphologies and elemental

mapping analysis. Powder X-ray diffraction (XRD) patterns were obtained using a Bruker D8-ADVANCED X-ray diffractometer equipped with graphitemonochromated Cu K α radiation ($\lambda = 1.54056$ Å). X-ray photoelectron spectroscopy (XPS) measurements were carried out with an Escalab 250Xi system using a monochromatic Al Ka source (1486.6 eV) for the analysis of surface chemical analysis. N₂ adsorption and desorption experiments (ASAP 2460) were performed using the Brunauer-Emmett-Teller (BET) model to determine the specific surface area and pore size distribution. Raman spectra were obtained by a Renishaw inVia-Reflex Raman spectrometer. Thermogravimetric analysis (TGA) measurements were performed on Diamond TG 6300 at a heating rate of 10 °C min⁻¹ in air atmosphere in the range of 25~700 °C.

Electrochemical measurements:

The electrochemical performance was evaluated by CR2032 type coin cells. Specifically, the active materials, acetylene black and PVDF were mixed at a mass ratio of 7:2:1 using NMP as solvent. Then, the mixture was thoroughly grounded and the obtained slurry was cast onto carbon paper to fabricate the working electrode. After vacuum drying at 60 °C for 12 h, the electrode was cut into round slices (diameter = 14 mm) with the active mass loading of approximately 2.0 mg. CR2032 coin cells were assembled using the working electrode, 2.0 M Zn(CF₃SO₃)₂ aqueous solution, glass fiber separator and Zinc foil in ambient air. Rate capability, cycle performance, and galvanostatic intermittent titration technique (GITT) tests were carried out on a battery testing system (Neware CT-4008Tn, Shenzhen, China) at room temperature. Cyclic voltammetry (CV), galvanostatic charge/discharge tests (GCD), and electrochemical impedance spectroscopy (EIS) tests with the frequency range of 1 MHz to 0.01 Hz are performed on an electrochemical workstation (CHI 660E, Shanghai, China).

The capacitive contribution calculation based on the CV measurement:

The peak current (i_p) and the scan rate (v) can be studied by the following equation:²

$$i_p = av^o$$
 (Equation S1)

where *a* and *b* are two constants, v (V s⁻¹) and i_p (A) represent the scan rate and the corresponding peak current, respectively. The *b* value can be calculated from log (i_p) against log (v) curves. Specifically, the *b* value of 0.5 and 1.0 indicates that the electrochemical process is controlled by diffusion behaviors and pseudocapacitive behaviors, respectively. If the value of *b* is between 0.5 and 1, the electrochemical reaction is controlled by both internal diffusion and surface pseudo capacitance.

According to the current (*i*) measured at a specific potential (V), the ratio between the diffusion-controlled portion and the capacitance-controlled portion of Zn^{2+} ion storage behavior can be studied by the following equation³

$$i = k_1 v + k_2 v^{1/2}$$
 (Equation S2)

where k_1 and k_2 are two constants, k_1v and $k_2v^{1/2}$ represent capacitance control and diffusion control process, respectively.

Zn²⁺ ions diffusion coefficient calculation based on the GITT measurement:

The cell was discharged or charged at 0.1 A g⁻¹ for 300 s and then relaxed for 3600 s to make the voltage reach equilibrium. The diffusion coefficient of Zn^{2+} ions $\binom{D}{Zn^{2+}}$ was obtained using Fick's second law with the follow equation:^{4,5}

$$D_{Zn^{2+}} = \frac{4}{\pi\tau} \left(\frac{n_B V_m}{A}\right)^2 \left(\frac{\Delta E_s}{\Delta E_\tau}\right)^2$$
(Equation S3)

Where τ (s) is the duration of the current pulse; n_B (mol) is the number of moles of the active substance; V_m (cm³ mol⁻¹) is the molar volume of the active substance; A (cm²) is the contact area between the electrode and the electrolyte; ΔE_s (V) is the steady-state voltage change caused by a continuous two-step current pulse, and ΔE_{τ} (V) is the total difference in the cell voltage during the constant current pulse after the iR drop.

DFT Calculations:

The Vienna Ab-initio Simulation Package (VASP) was employed to conduct all Density Functional Theory (DFT) calculations.^{6,7} The Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional, employing the generalized gradient approximation

(GGA) method with Grimme D3 dispersion correction, was utilized in this study.⁸⁻¹⁰ The projected augmented wave (PAW) method was utilized to describe core-valence interactions in all DFT calculations.¹¹ The energy cutoff for plane wave expansions was set to 450 eV, and the $2\times2\times2$ Monkhorst-Pack grid k-points were used to sample the Brillouin zone integration for structural optimization of surface structures. To compute the band structures, open-source package VASPKIT is applied to generated corresponding k-points with high symmetry and analysis the band structures.¹² Structural optimization was carried out with energy and force convergence criteria setting at 1.0×10^{-5} eV and 0.02 eV Å⁻¹, respectively.



Fig. S1 SEM images of (a, b) 1T-G-MoS₂ and (c, d) pristine 2H-MoS₂ nanoflowers.



Fig. S2 TEM images of (a) 1T-G-MoS $_2$ and (b) pristine 2H-MoS $_2$ nanoflowers.



Fig. S3 (a) N_2 adsorption and desorption isotherms and (b) pore diameter distributions of pristine 1T-G-MoS₂ and 2H-MoS₂.



Fig. S4 (a) HRTEM images of 2H-MoS₂, and (b) the corresponding IFFT simulated image for the marked rectangular area in S4 (a).



Fig. S5 (a) C 1s and (b) O 1s XPS spectra of 1T-G-MoS₂.



Fig. S6 S 2p XPS spectra of 1T-G-MoS₂ and 2H-MoS₂.



Fig. S7 TGA curves of 1T-G-MoS₂ and 2H-MoS₂ under air atmosphere.



Fig. S8 The cleavage of the Mo 4d orbitals in (a) 2H and (b) 1T phase MoS₂.



Fig. S9 (a) XRD patterns of $1T-G_x-MoS_2$ with different glucose contents. (b) The galvanostatic charge/discharge curves of AZIBs using $1T-G_x-MoS_2$ as cathodes with different glucose contents. $1T-G_{0.1}-MoS_2$, $1T-G_{0.2}-MoS_2$, $1T-G_{0.4}-MoS_2$ and $1T-G_{0.8}-MoS_2$ represent the glucose addition contents of 0.1, 0.2, 0.4, and 0.8 g, respectively.



Fig. S10 Electrochemical performance of 1T-G-MoS₂ electrode for zinc storage. (a) CV curves at the scan rate of 0.1 mV s⁻¹ for the initial three cycles. b) Galvanostatic charge/discharge profiles at 0.1 A g⁻¹ for the initial ten cycles at 0.1 A g⁻¹.



Fig. S11 Rate performance at various current density of 1T-G-MoS₂ and 2H-MoS₂ electrodes.



Fig. S12 Long-term cycling performance of 1T-G-MoS₂ electrode at 5 A g⁻¹.



Fig. S13 The LED board lighted by the soft pack battery assembled using 1T-G-MoS₂ as cathode.



Fig. S14 (a) Linear fitting from the log (*i*) vs. log (v) plots of 1T-G-MoS₂ electrode at cathodic/anodic peaks. (b) Normalized contribution proportions from the capacitance (shaded region) and diffusion at 1.0 mV s⁻¹ for 1T-G-MoS₂ electrode.



Fig. S15 (a) CV curves of the 2H-MoS₂ electrode at different scan rates. (b) The corresponding linear fitting from log(i) vs. log(v) plots of 2H-MoS₂ electrode at cathodic/anodic peaks. (c) The capacitive contribution ratios of 2H-MoS₂ electrode at different scan rates.



Fig. S16 Nyquist plots of 1T-G-MoS₂ and 2H-MoS₂ electrodes.



Fig. S17 Contact angle tests with water for (a) 1T-G-MoS₂ and (b) 2H-MoS₂.



Fig. S18 The density of states of (a) 1T-G-MoS₂ and (b) pristine 2H-MoS₂.



Fig. S19 SEM images of 1T-G-MoS₂ electrode at (a) fully discharged (0.2 V) and (b) charged (1.3 V) states.



Fig. S20 HRTEM images of 1T-G-MoS₂ at (a) fully discharged (0.2 V) and (c) fully charged (1.3 V) states. (b, d) The IFFT simulated images of the marked rectangular area in S20 (a) and (c), respectively.

Materials	Electrolyte	Current density (A g ⁻¹)	Discharged specific capacity (mAh g ⁻¹)	Cycling stability (cycles)	Reference
1T MoS ₂ /MWCNT	3 M Zn(CF ₃ SO ₃) ₂	0.1	160.3	84.6% over 500 cycles	13
1T-MoS ₂	3 M Zn(CF ₃ SO ₃) ₂	0.1	164.1	at 1 A g ⁻¹ 83.1% after 100 cycles	14
MoS ₂ 250	3 M Zn(CF ₃ SO ₃) ₂	0.1	138.6	at 1 A g ⁻¹ 87.8% after 1000 cycles	15
MoS ₂ -PEG	3 M	0.1	139.5	at 1 A g ⁻¹ 83.5% after 200 cycles	16
TH-MoS ₂	2 M	0.1	156.0	at 1 A g ⁻¹ 97.3% after 500 cycles	17
N doned 1T MoS-	2n(CF ₃ SO ₃) ₂ 3 M	0.1	149.6	at 1 A g ⁻¹ 97.1% after	18
N doped 11 MoS ₂	$Zn(CF_3SO_3)_2$	0.1	149.0	at 1 A g^{-1} 98.6% after	10
F/P-MoS ₂	$Zn(CF_3SO_3)_2$	0.1	147.7	200 cycles at 1 A g ⁻¹ 98% after	17
VS_2	1 M ZnSO ₄	0.1	159.1	200 cycles at 0.5 A g ⁻¹	20
VS ₂ @VOOH	3 M ZnSO ₄	0.1	165	82% after 400 cycles at 1 A g ⁻¹	21
VSe ₂	2 M ZnSO ₄	0.1	131.8	80% after 500 cycles at 0.1 A g^{-1}	22
KMnHCF	30 M KFSI + 1 M	0.2	138.0	71% after 400 cycles	23

Table S1 Summarized table of the electrochemical performance of the AZIBs usingMoS2, vanadium-based compounds, and Prussian blue analogues cathodes.

	Zn(CF3SO3)2			at 0.2 A g ⁻¹	
1T-G-MoS ₂	2 M Zn(CF ₃ SO ₃) ₂	0.1	182.15	72.0% after 500 cycles at 1 A g ⁻¹	This work

State	Mo ⁶⁺ (%)	1T (%)	2H (%)
Original	9.4	51.4	39.2
Fully discharged	19.6	50.3	30.1
Fully charged	10.5	51.6	37.9

Table S2 The proportion of Mo^{6+} , 1T-MoS₂, and 2H-MoS₂ at the initial, fully discharged and charged states of 1T-G-MoS₂.

According to the peak shapes and literatures, Mo 3d was fitted with peaks including the Mo⁶⁺ peak (235.85 eV), the 1T phase Mo $3d_{3/2}$ and Mo $3d_{5/2}$ peaks (232.05 and 228.8 eV), and the 2H phase Mo $3d_{3/2}$ and Mo $3d_{5/2}$ peaks (233.1 and 229.95 eV). The proportion calculated from the peak area are listed in the Table S2.

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