Electronic Supplementary Information

Experimental section

Materials.

Ammonium chloride (NH_4Cl), ammonium molybdate tetrahydrate ((NH_4)$_6Mo_7O_{24}$·$4H_2O$), thiourea (CH_4N_2S), and poly(vinyl alcohol) (Mw ~67000), were purchased from Aladdin Ltd. (Shanghai, China). Carbon cloth (CC) was obtained from Titan Technology Co., Ltd. All the chemicals were used as received without further purification. The deionized water (18.25 MΩ/cm) used in all experiments was prepared by passing water through an ultra-pure purification system.

Synthesis of MoS_2/CC, T-MoS_2/CC, and Exp-MoS_2/CC.

The Exp-MoS_2/CC was prepared through a reported solvothermal method.[1] Specifically, 0.2 g (NH_4)$_6Mo_7O_{24}$·$4H_2O$ (the Mo source) and 0.3 g CH_4N_2S (the S source) were firstly dissolved in a mixture of water and ethanol (2:3) with a total volume of 40 mL under vigorous stirring. Then the above solution together with the pre-treated CC was transferred into an autoclave and heated at 200°C for 20 h. After cooling to room temperature, the product grown on CC was taken out, washed, and finally dried at 60°C in a vacuum oven. MoS_2/CC was prepared by the same method but only used water as a solvent. T-MoS_2/CC was prepared through an ethanol post-treatment with MoS_2/CC.

Synthesis of MnO_2/CC.

First, 0.32 g of $KMnO_4$, and 1 mL of concentrated HCl were added to 80 ml of deionized water and stirred vigorously for 30 min to obtain a homogeneous solution. Then, the mixed solution and the CC were transferred into a Teflon-lined autoclave (100 mL) and underwent heating treatment at 85°C for 0.5 h.

Characterizations.

X-ray diffraction (XRD) patterns were obtained from a Shimadzu XRD-7000 diffractometer with Cu Kα radiation (40 kV, 30 mA). Scanning electron microscopy (SEM, ZISS 300) and transmission electron microscopy (TEM, JEM-F200, JEOL Ltd.)

were carried out to reveal the morphology information of samples. X-ray photoelectron spectroscopy (XPS) measurements were performed on an ESCALABMK II X-ray photoelectron spectrometer using Mg as the exciting source. In-situ Raman spectroscopy was recorded on the Lab RAM HR Evolution confocal microscope with 532 nm laser.

Electrochemical measurements.

Electrochemical tests were carried out by cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) using a CHI660b electrochemical workstation. The cycling durability was examined on a CT2001A LAND tester. The performance of the samples was examined in a conventional three-electrode setup using the carbon rod counter electrode, Ag/AgCl reference electrode, Exp-MoS$_2$/CC working electrode, and 2.0 M NH$_4$Cl aqueous electrolyte. The areal capacitances (C, mF cm^{-2}) were calculated based on the GCD curves according to the following equation (1):

$$C = It/SV \tag{1}$$

where I/S is the current density (mA cm^{-2}) and t is the discharge time (s). V is the potential window (V).

Fabrication of I-ASC and S-ASC.

I-ASC was fabricated with Exp-MoS$_2$/CC (anode), MnO$_2$/CC (cathode), and PVA/NH$_4$Cl through in situ polymerization to achieve integrated structure. S-ASC was fabricated by the traditional method with a sandwich-like structure. The areal energy densities (E, mWh cm^{-2}) and power densities (P, mW cm^{-2}) were calculated from equations (2) and (3):

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

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

where ΔV is the discharge voltage range (V) and Δt is the discharge time (s).

Computational details

The COMSOL Multiphysics software was employed to simulate the intrinsic

piezoelectric field from 1T MoS$_2$. The simplified model of the 1T MoS$_2$ nanosheet was set to 50 nm × 20 nm × 40 nm (length × width × height) with the specific parameters of density: 4.8 g cm^{-3}, dielectric constant: ε_r = 11.8, Young modulus: 265 Gpa.[2] The deformation ratio of the NH$_4^+$ intercalation was ≈13.1%, which was based on the estimation results of the in situ XRD spectra.

DFT calculation details.

All spin-polarized computations were performed under the framework of density functional theory (DFT). Perdew-Burke-Ernzerhof (PBE) functional among a generalized gradient approximation (GGA) was introduced to describe the exchange and correlation effect of electron. The projector augmented wave (PAW) method was employed to describe the electron-ion interaction. The kinetic energy cutoff for plane wave was set to 400 eV. The Brillouin zone was sampled by 3×3×1 grid for structural relaxation and 9×9×1 grid for density of states calculation. Cl-NEB method combined with dimer method was employed to search the transition states of the diffusion process. The energy criterion was set to 10^{-5} eV and force criterion was set to 0.02 eV/Å for structure relaxation and 0.1 eV for transition state computation. DFT-D3 method was employed for the correction of vdWs force. Pristine 2H-MoS$_2$ and 1T-MoS$_2$ were modeled by 4×4×1 supercell containing two layers and the ammonium-intercalation 1T-MoS$_2$ was constructed by adding three ammonium ions between double-layer 1T-MoS$_2$.

Figure S1. (a) Low- and (b) high-magnification SEM images of Exp-MoS$_2$/CC.

Figure S2. SEM and energy-dispersive X-ray elemental mapping images of Exp-MoS$_2$/CC.

Figure S3. (a) TEM and (b) HRTEM images of Exp-MoS$_2$.

Figure S4. XPS survey spectrum of Exp-MoS$_2$.

Figure S5. XPS spectra of MoS_2, $T-MoS_2$, and $Exp-MoS_2$ in N 1s region.

Figure S6. GCD curves of Exp-MoS$_2$/CC in different electrolytes at 1 mA cm^{-2}.

Figure S7. Nyquist plots of Exp-MoS$_2$/CC in different cation electrolytes.

Figure S8. The phase angle vs. frequency plots of Exp-MoS$_2$/CC in different cation electrolytes.

Figure S9. GCD curves of (a) MoS_2/CC, (b) T-MoS_2/CC, and (c) Exp-MoS_2/CC at a range of 1–50 mA cm^{-2}.

Figure S10. CV curves of (a) MoS_2/CC, (b) T-MoS_2/CC, and (c) Exp-MoS_2/CC at a range of 5–50 mV s^{-1}.

Figure S11. Fitted Warburg impedance at the low frequency for (a, b) MoS_2/CC, (c, d) $T\text{-}MoS_2/CC$, and (e, f) $Exp\text{-}MoS_2/CC$ during GCD process.

Figure S12. (a) Low- and (b) high-magnification SEM images of MnO$_2$/CC.

Figure S13. XRD pattern of MnO_2/CC.

Figure S14. (a) CV curves, (b) GCD curves, and (c) Nyquist plots of MnO_2/CC.

Figure S15. (a) CV curves of S-ASC at various potential windows. GCD curves of (b) I-ASC and (c) S-ASC at various potential windows. GCD curves of (d) I-ASC and (e) S-ASC at various current densities. (f) Nyquist plots of I-ASC and S-ASC.

Figure S16. GCD curves of (a) I-ASC and (b) S-ASC at various bending states.

Table S1. Comparison of electrochemical performance of the I-ASC and S-ASC with previously reported energy storage devices.

Device	Electrolyte	Energy density ($mWh\ cm^{-2}$)	Power density ($mW\ cm^{-2}$)	Ref.
I-ASC	**PVA/NH₄Cl**	**223.7**	**900.1**	**This work**
S-ASC	**PVA/NH₄Cl**	**186.5**	**899.6**	
δ-MnO₂//ACC	1 M (NH₄)₂SO₄	0.86	20	3
α-MnO₂//(NH₄)ₓWO₃	2 M NH₄Ac	1.01	18	4
MnO₂/CNTF//MoS₂@TiN/CNTF	PVA/NH₄Cl	0.195	1.1	5
ACC@VPP//PTCDI	PVA/NH₄Cl	0.32	0.09	6
Cu-HHB/I₂//P-MXene	1 M (NH₄)₂SO₄	31.5	400	7
Ti₃C₂Tₓ//Zn	PVA/ZnCl₂	20	0.5	8
Ti₃C₂Tₓ/CNF//Ti₃C₂Tₓ/CNF	PVA/H₂SO₄	0.1	0.299	9
AC/CNT/MXene-N/GO//AC/CNT/MXene-N/GO	PVA/H₂SO₄	0.42	-	10
MoS₂//Na-MnO₂	Na₂SO₄	178.4 μWh cm⁻²	1.1 μW cm⁻²	11
Ti₃C₂Tₓ//Ti₃C₂Tₓ	PVA/H₂SO₄	1.64 μWh cm⁻²	0.7783 μW cm⁻²	12

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