Supporting Information

Experimental Section

1. The preparation of H-MoS₂

0.6 g $Na_2MoO_4 \cdot 2H_2O$ and 0.8 g $CS(NH_2)_2$ were dissolved into 60 mL of distilled water, and the pH value of the solution was adjusted to 1 by adding hydrochloric acid. After continuously stirring, the blue solution was transferred into the 100 mL teflon-lined stainless steel autoclave and was treated with 180°C for 24h. The obtained black powders were washed by water and ethanol, and finally treated with freeze-drying method.

2. Material Characterization

Crystallization structure was measured by X-ray diffraction (XRD, GBC MMA, Cu K_{α} radiation). The morphology and microstructure of MoS₂ were recorded by scanning electron microscopy (SEM, JEOL JSM7500F) and high resolution transmission electron microscopy (HRTEM, JEOL 2011).

3 Specimen preparation for TEM characterization

The after cycled cell was disassembled in the glove box and the electrode materials are dispersed in the tetraethylene glycol dimethyl ether and then loaded onto the holey carbon grid.

4. Electrode and coin cell preparation

4.1 Half cell

The CR2032 type coin cell was assembled in an argon-filled glove box. The working electrode was consisted of 80% active materials, 10% KS-6 and 10% sodium

carboxymethyl cellulose (CMC) on Cu foil. The electrode was dried at 110 °C for 10 h in a vacuum oven and then pressed under 30 MPa by a tablet compression machine. Lithium foil was served as anode electrode and reference electrode, and glass fiber filter separator. The electrolyte 1.0 was used as the was Μ bis(trifluoromethanesulfonyl)imide and dissolved in tetraethylene glycol dimethyl ether.

4.2 Full cell

The cathode electrode material was consisted of 80% LiCoO₂, 10% KS-6 and 10% sodium carboxymethyl cellulose (CMC) on Aluminum foil. The electrode was dried at 110 °C for 10 h in a vacuum oven and then pressed under 30 MPa by a tablet compression machine. The as-prepared H-MoS₂ was used as anode electrode material. The electrolyte and separator were the same as those using in half cell. The N/P ratio is 1.05:1, and loading of cathode and anode is 1.3 mg/cm² and 1.0 mg/cm². And the tapping density of H-MoS₂ and LiCoO₂ is 0.8 g/cm³ and 2.5 g/cm³, respectively.

5. Electrochemical characterization

Land CT2001A cell testing system was used to test galvanostatic charge/discharge cycles in the voltage range of 1.0-3.0 V and 0.1-3.0 versus Li⁺/Li for half cell and 1.5-3.5 V for full cell. Electrochemical impedance spectroscopy (EIS) was measured on Parstat 2273 electrochemical workstation (AMETEK). The ac perturbation signal was ± 5 mV and the frequency ranged from 100 mHz to 100 kHz.

Fig.s:



Fig. S1 (a) The XRD pattern of $H-MoS_2$ and $B-MoS_2$, and the HRTEM images of (b) $H-MoS_2$ and (c) $B-MoS_2$.



Fig. S2 The electrochemical performances of H-MoS₂ and B-MoS₂. (a) The galvanostatic charge and discharge curves and (b) cyclic performance of B-MoS₂ at 0.2 A·g⁻¹ in 1.0–3.0 V. (c) The galvanostatic charge and discharge curves and (d) cyclic performance of H-MoS₂ at 0.2 A·g⁻¹ in 0.1–3.0 V.



Fig. S3 GITT curve of H-MoS₂/Li battery cycling at (a) 1–3 V and (b) 0.1–3 V.



Fig. S4 The Lithium ion diffusion coefficient by GITT measurement: (a) 1st cycle and

(b) 10th cycle.



Fig. S5 Cycling performance of MoS_2 with EC-DEC as electrolyte.



Fig. S6 (a) The galvanostatic curves of graphite/LiCoO₂ cell at 0.5 C. (b) Cycling performance of graphite/LiCoO₂ cell at 0.5 C.



Fig. S7 The electrochemical impedance spectroscopy measurement of $H-MoS_2$ at 1.0 V and $B-MoS_2$ at 1.0 V cycling between 1.0–3.0 V and $H-MoS_2$ at 0.1 V cycling between 0.1–3.0 V.