

Sulfur-Modified Carbon-Coated CoMoO₃ Nanohybrid Electrodes for Enhanced Lithium-Storage Capacity

Bo Zhang, Yingqi. Li, Yue Ma, Bo Jin, Xingyou Lang, Yongfu Zhu,* Qing Jiang

Key Laboratory of Automobile Materials, Ministry of Education (Jilin University),
School of Materials Science and Engineering, Jilin University, Changchun 130022,
China

*Corresponding author

E-mail: yfzhu@jlu.edu.cn

Electrodes preparation and electrochemical measurements

The preparation process of our electrodes is provided as follows: the active materials, poly vinylidene fluoride (PVDF) and acetylene black were grinded with a mass ratio of 70:10:20. The addition of suitable N-methyl-2-pyrrolidone (NMP) solvent makes the mixture form uniform slurry. Then the slurry was directly pasted on the copper foil, and we then dry it at 110 °C. The average mass loading of these electrodes was ~0.93 mg cm⁻². Various electrodes were encapsulated into a typical 2016 coin-type cell. The Li foil was utilized as the counter and reference electrodes, and 1 M LiPF₄ solution (equivalent ethylene carbonate (EC), ethyl methyl carbonate (EMC) and dimethyl carbonate (DMC) were utilized to form mixed solvent) as electrolyte. The cyclic voltammetry (CV), galvanostatic charge-discharge (GCD) and electrochemical impedance spectroscopy (EIS) measurements were operated via CHI 660e electrochemical working station and CT2001A LAND test system.

For the $\text{CoMoO}_3/\text{C-S}/\text{Li}$ LIB device, the calculation equations of energy density (E) and power density (P) are presented as follow:

$$E = \frac{\int QdV}{m} = \frac{\int itdV}{m} \quad (\text{S1})$$

$$P = \frac{E}{\Delta t} \quad (\text{S2})$$

wherein the Q , V , i , t , Δt represents the capacity, potential, discharge current, time and entire discharge time.

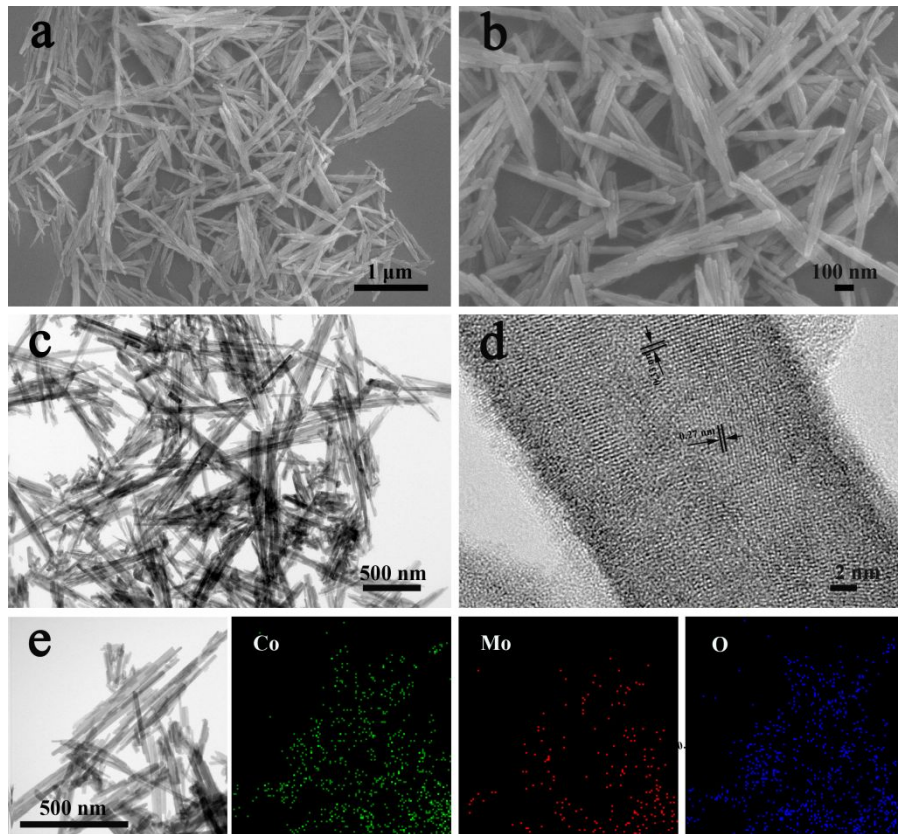


Fig. S1 (a and b) FESEM observation of CoMoO_4 sample; (c) TEM observation of CoMoO_4 sample; (d) HRTEM observation of CoMoO_4 sample; (e) element scanning of CoMoO_4 sample

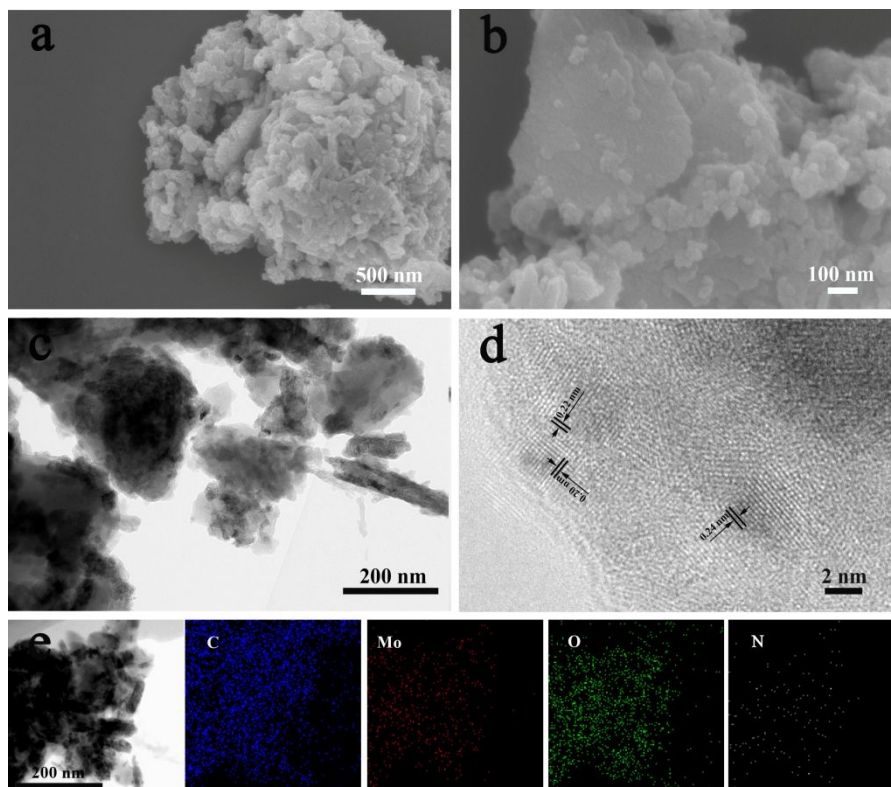


Fig. S2 (a and b) FESEM observation of CoMoO_3/C sample; (c) TEM observation of CoMoO_3/C sample; (d) HRTEM observation of CoMoO_3/C sample; (e) element scanning of CoMoO_3/C .

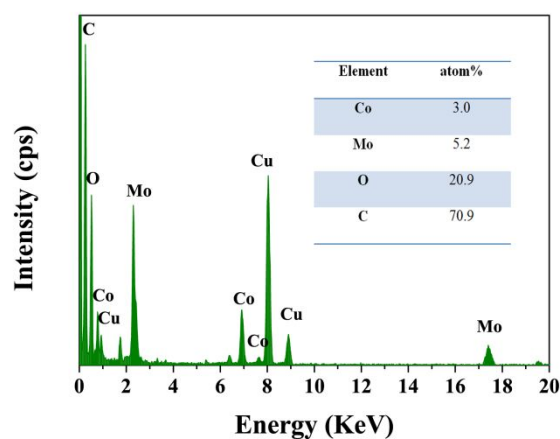


Fig. S3 EDS data of CoMoO₃/C

On the basis of the mapping of CoMoO₃/C (Fig. S2e), the N signal is pretty weaker, suggesting a pretty lower N content in C structure. Also, the EDS data in Fig. S3 could not show obvious N content, revealing that the N content is negligible. Therefore, we didn't highlight the N doped C. Instead, we state the C coating in our work.

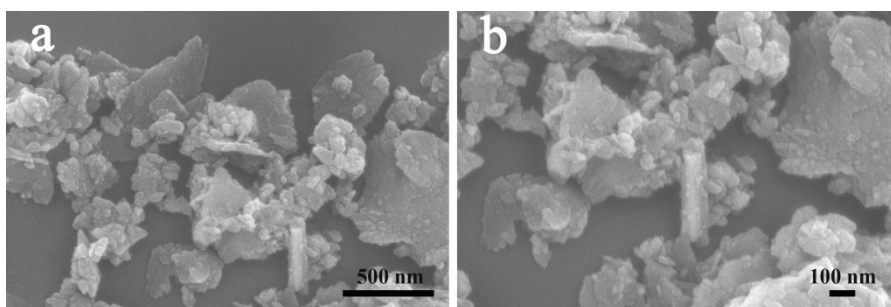


Fig. S4 (a and b) FESEM observation of CoMoO₄-PDA sample

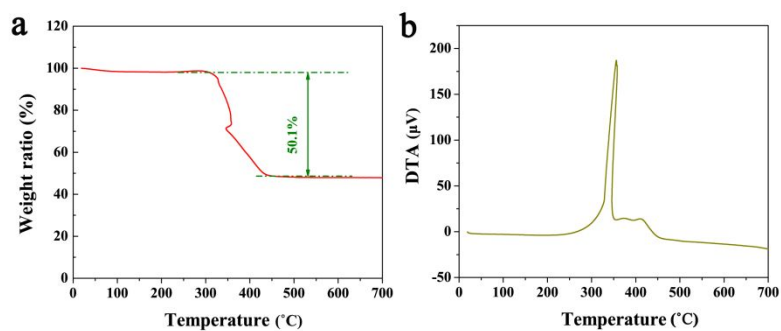


Fig. S5 (a) TG curve of CoMoO₃/C; (b) DTA curve of CoMoO₃/C

To investigate the weight ratios of C and MoS₂ in CoMoO₃/C-S, we have made TGA and ICP test. In Fig. S5, the TGA result of CoMoO₃/C was shown. The declined weight of the sample during the heating process is 50.1%. Assuming that the CoMoO₃ was oxidized to CoMoO₄ in the TG test, and the mass ratio of C is calculated as 46.25% of CoMoO₃/C. Because there are many factors (MoS₂, C, CoMoO₃) in the CoMoO₃/C-S, the calculation of C content seem difficult via the TG result of CoMoO₃/C-S. In view of high C content and partial S modification, we approximately utilized the 46.25% (C content of CoMoO₃/C) as the C content of CoMoO₃/C-S.

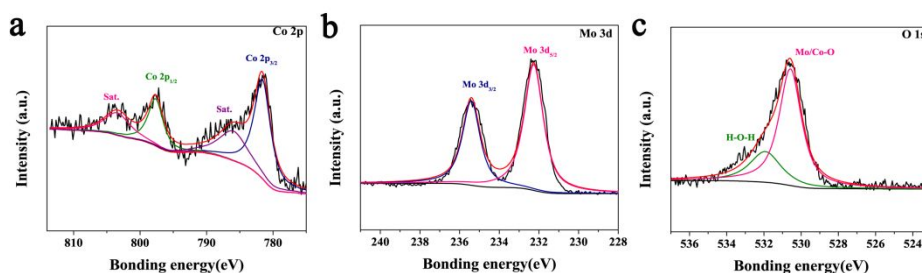


Fig. S6 (a-c) XPS spectra of CoMoO₄ for different elements: (a) Co 2p; (b) Mo 3d; and (c) O 1s

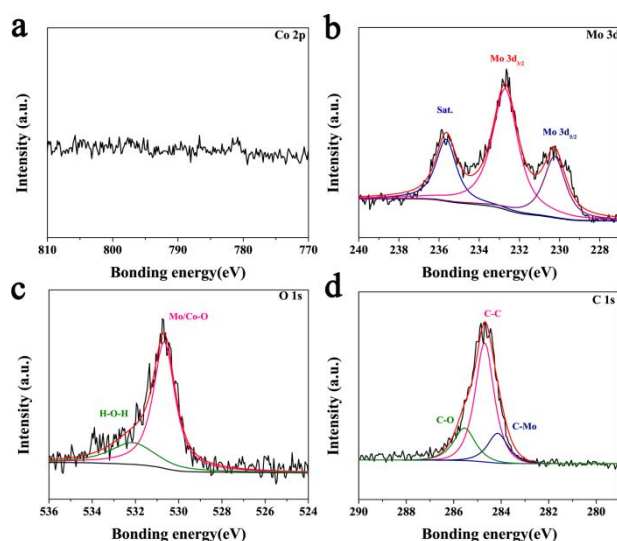


Fig. S7 (a-d) XPS spectra of CoMoO₃/C for different elements: (a) Co 2p; (b) Mo 3d; and (c) O 1s; (d) C 1s.

Table S1 ICP data of CoMoO₃/C-S sample

	Co	Mo	S
mass concentration (mg L ⁻¹)	0.2466	0.4398	0.3627
molar concentration (mmol L ⁻¹)	4.17E ⁻³	4.58E ⁻³	1.13E ⁻³

Also, the ICP results CoMoO₃/C-S was shown in Table S1, the mole ratio of Co:Mo:S is 4.22: 4.63: 1.15. Because the outer nanosheet is low-crystallinity MoS₂, we approximately assess the content of MoS₂ (Co doped MoS₂ is considered as

MoS₂) via the ICP result of S element. By an approximate treatment, we use the Co element to assess the content of CoMoO₃. Thus, the mole ratio of CoMoO₃ and MoS₂ is 4.22:0.575. Based on the 46.25% C content and MoS₂:CoMoO₃, the ratio of CoMoO₃:C:MoS₂ is about 47.30: 46.25: 6.45.

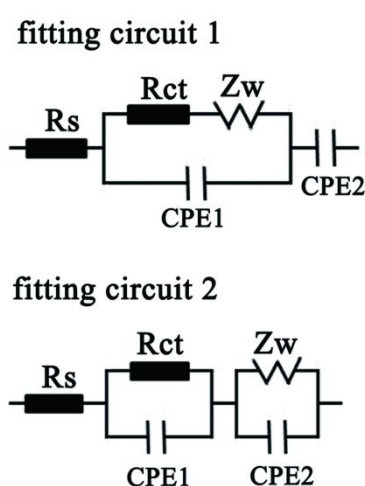


Fig. S8 Fitting circuit 1 for CoMoO₄ and CoMoO₃/C and fitting circuit 2 for CoMoO₃/C-S

Table. S2 R_s and R_{ct} values of CoMoO₄, CoMoO₃/C and CoMoO₃/C-S;

	R_s (Ω)	R_{ct} (Ω)
CoMoO ₄	9.51	179.10
CoMoO ₃ /C	16.87	51.77
CoMoO ₃ /C-S	14.01	70.95

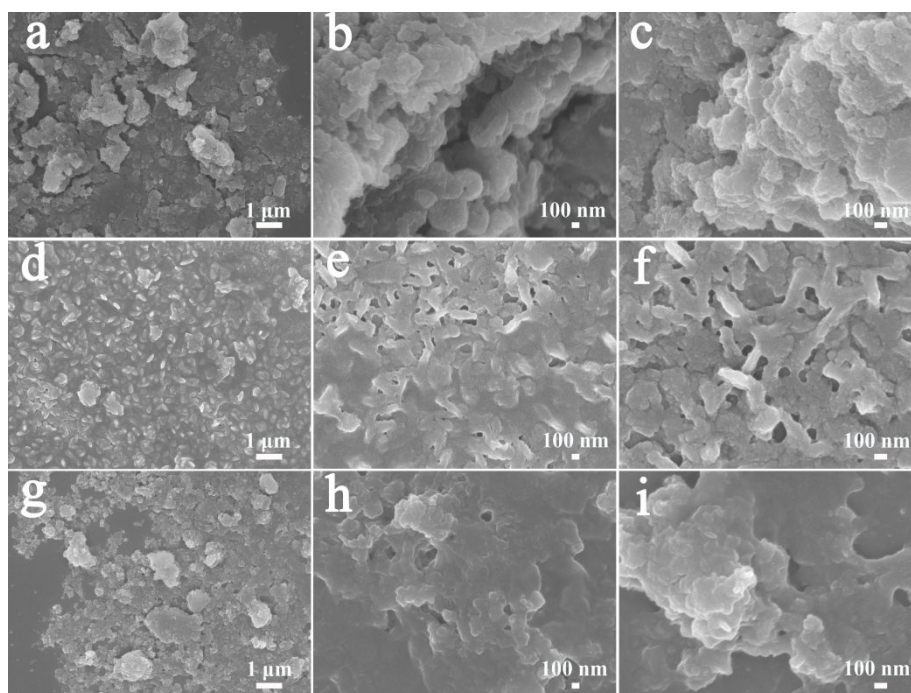


Fig. S9 (a-c) FESEM observation of $\text{CoMoO}_3/\text{C-S}$ sample after 500 cycles; (d-f) FESEM observation of CoMoO_4 sample after 500 cycles; (g-i) FESEM observation of CoMoO_3/C sample after 500 cycles.

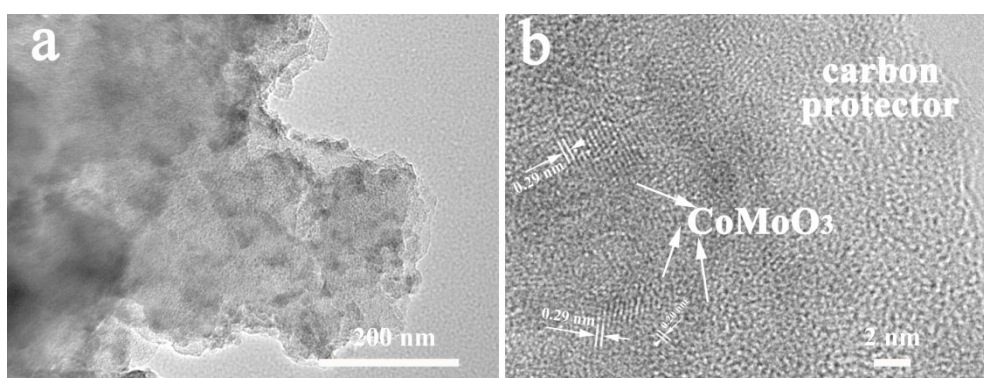


Fig. S10 (a) TEM observation of $\text{CoMoO}_3/\text{C-S}$ sample after 500 cycles; (b) HRTEM observation of $\text{CoMoO}_3/\text{C-S}$ sample after 500 cycles

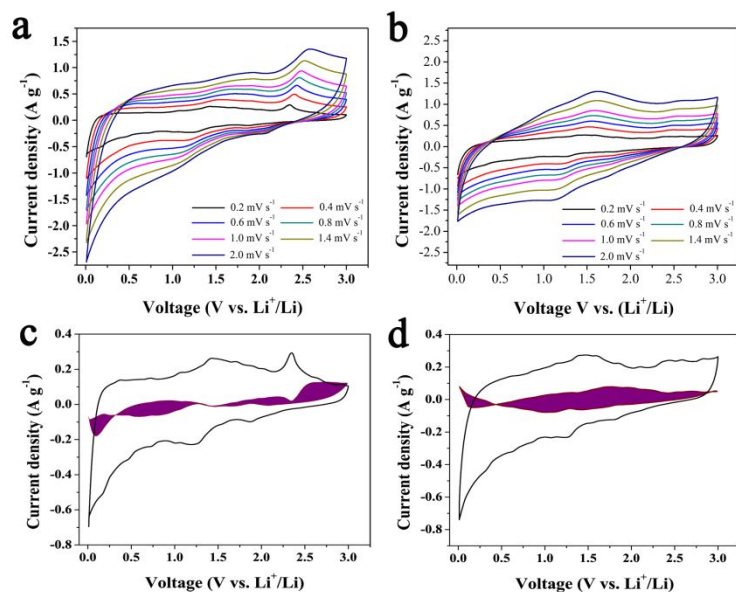


Fig. S11 (a and b) CV curves of CoMoO₃/C-S at different scanning rates before cycles and after 500 cycles; (c and d) Extraction of the capacitive contribution in the entire CV at 2 mV s⁻¹ for CoMoO₃/C-S electrode before cycles and after 500 cycles.

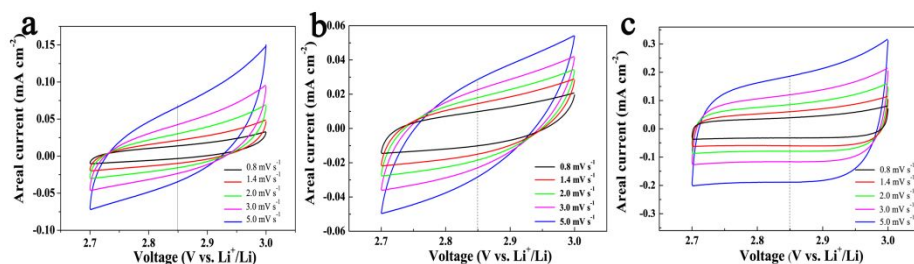


Fig. S12 (a) CV curves in the potential region 2.7-3.0V of CoMoO₃/C-S before cycles; (b) CV curves in the potential region 2.7-3.0V of CoMoO₃/C-S after 100 cycles; (c) CV curves in the potential region 2.7-3.0V of CoMoO₃/C-S after 500 cycles.

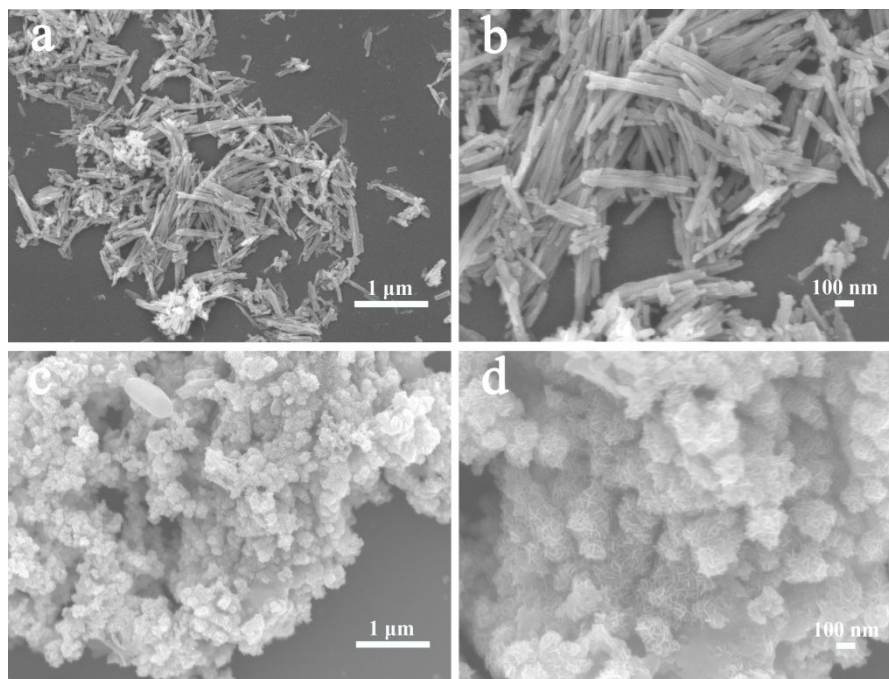


Fig. S13 (a and b) FESEM observation of CoMoO₄-H sample; (c and d) FESEM observation of CoMoO₄-S sample

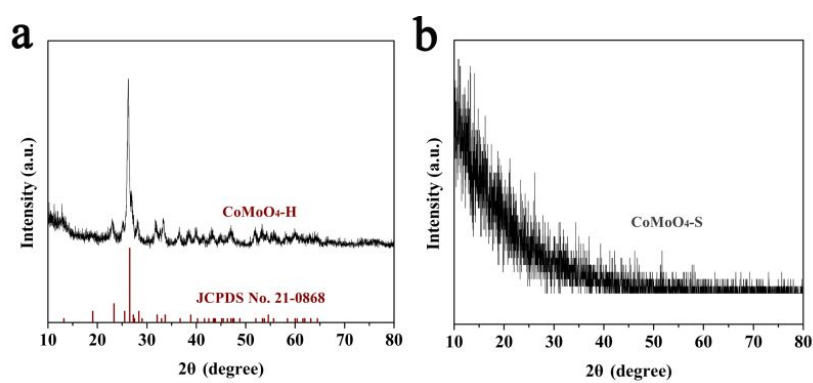


Fig. S14 (a and b) XRD curves of CoMoO₄-H and CoMoO₄-S samples

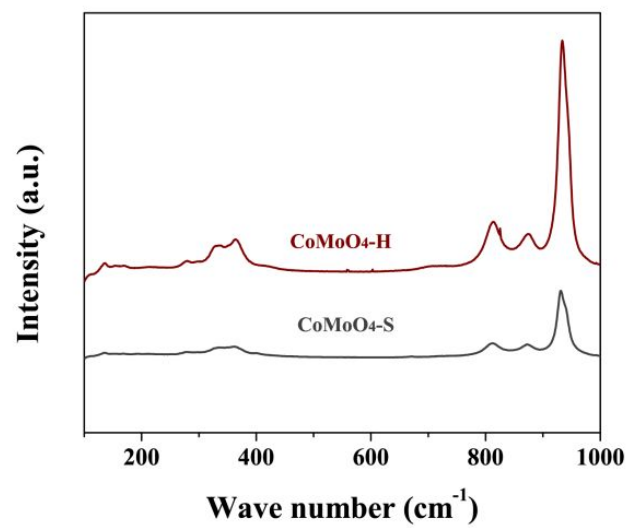


Fig. S15 Raman spectra of CoMoO₄-H and CoMoO₄-S samples

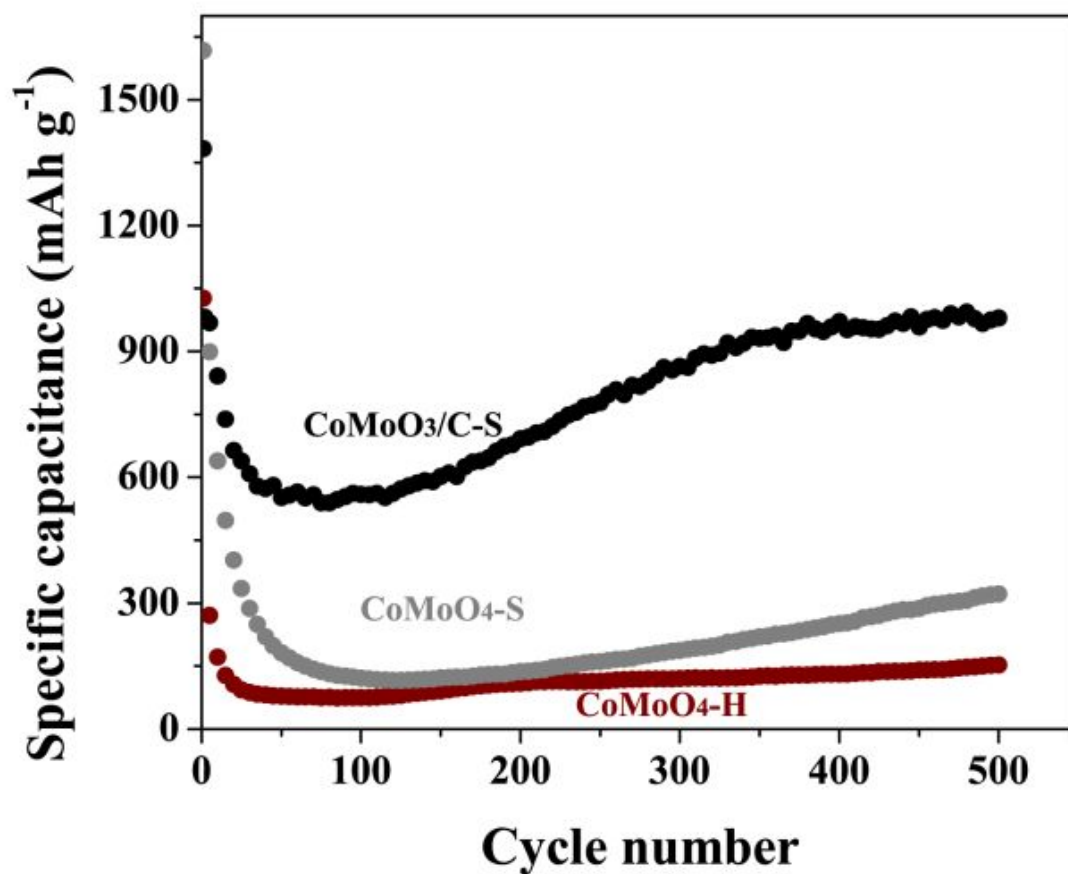


Fig. S16 Cycling performance of CoMoO₃/C-S, CoMoO₄-H and CoMoO₄-H-S

electrodes.

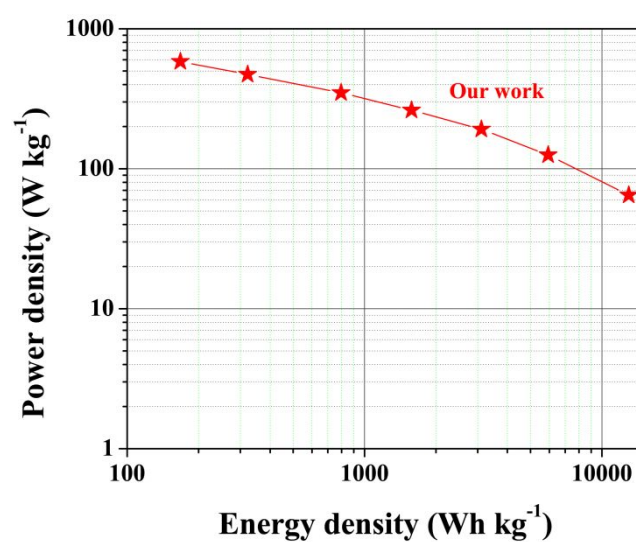


Fig. S17 Ragone plot for CoMoO₃/C-S//Li plate