

Supporting Information

Facile fabrication and electrochemical properties of high-quality reduced graphene oxide/cobalt sulfide composite as anode material for lithium-ion batteries

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Figure captions of Supporting Information:

Fig. S1. AFM image (left) and corresponding section analysis (right) of the as-prepared GO.

Fig. S2. (a) XRD pattern and (b) SEM of as-obtained sample in the first reaction step.

Fig. S3. (a) XRD, (b) XPS survey and (c) C1s spectra of the as-prepared GO and rGO.

Fig. S4. SEM image of pure cobalt sulfide.

Fig. S5. XRD patterns of the as-prepared composite after different electrochemical cycles.

Fig. S6. Rate-capability performance of the pure cobalt sulfide at various current densities of 100, 250, 500, 1000, and 2000 mA g⁻¹.

Synthesis of Graphene oxide (GO)

GO was synthesized from graphite powder by a modified Hummers' method [S1]. In a typical synthesis, graphite powder was first treated at 1050 °C in air for 15 s. The thermally treated graphite powder (3 g) was then put into a solution of concentrated H₂SO₄ (12 mL), K₂S₂O₈ (2.5 g), and P₂O₅ (2.5 g). And the mixture was kept at 80 °C for 4.5 hours in an oil bath. After that, the mixture was cooled to room temperature and diluted with 500 mL deionized water, then filtered and washed with deionized water to remove the residual acid. The product was dried at room temperature overnight. Subsequently, this pre-treated graphite powder was put into 120 mL cold concentrated H₂SO₄. Then, KMnO₄ (15 g) was added gradually under stirring and the temperature of the mixture was kept to be below 20 °C. The above mixture was kept at 35 °C for 2 hours. Then, deionized water (250 mL) was gradually added keeping the reaction in an ice bath. After adding the deionized water, the mixture was stirred for 2 hours. Subsequently, 700 mL deionized water was added into the mixture, and then 30% H₂O₂ solution (20 mL) was added. The mixture was washed with 5% HCl solution and followed by DI water to remove the acid, and then dialyzed for 3 weeks.

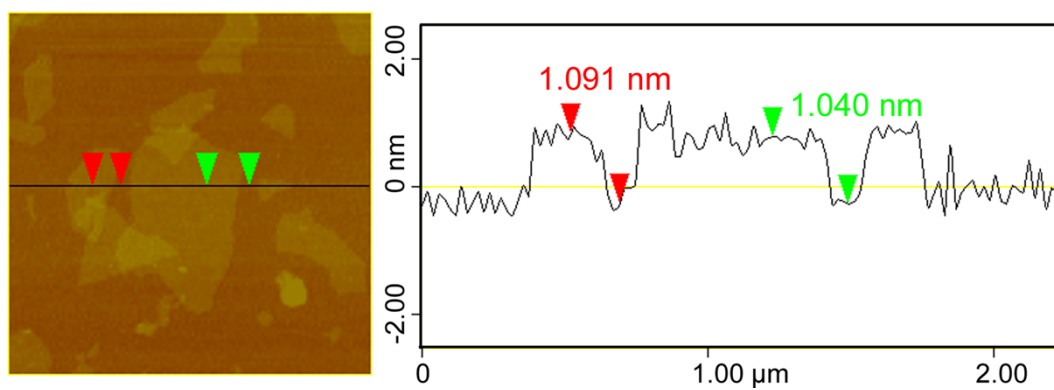


Fig. S1. AFM image (left) and corresponding section analysis (right) of the as-prepared GO.

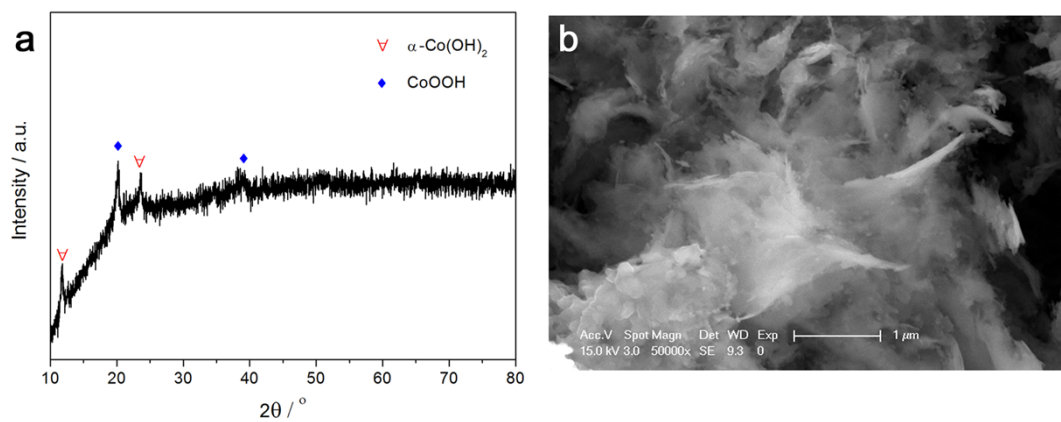


Fig. S2. (a) XRD pattern and (b) SEM of as-obtained sample in the first reaction step.

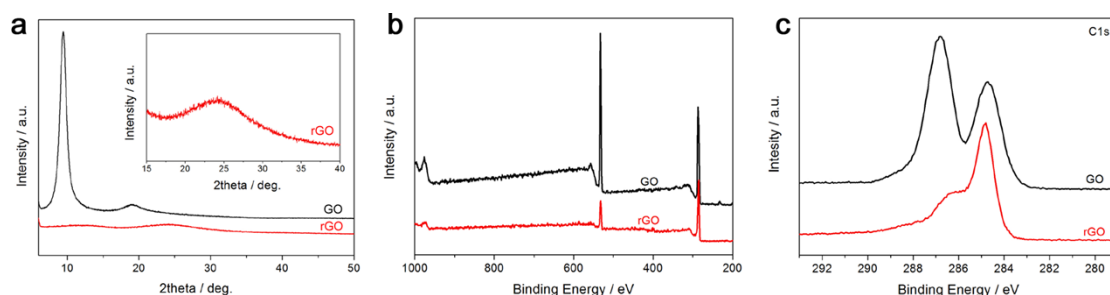


Fig. S3. (a) XRD, (b) XPS survey and (c) C1s spectra of the as-prepared GO and rGO.

Characterization of GO and rGO. Fig. S3a shows the XRD patterns of the as-prepared GO and rGO samples. XRD pattern of GO reveals an intense and sharp peak located at about $2\theta = 10^\circ$, corresponding to the characteristic diffraction peak of GO powder. After the reduce processing, the sharp peak at 10° disappears and a new broad diffraction peak at $2\theta = 24^\circ$ appears in the XRD pattern of rGO, confirming the GO is reduced in this process [S2]. Furthermore, the reduction of GO can be substantiated directly by XPS analysis. Fig. S3b shows the XPS survey spectra of GO and rGO. The reduction effect is clear since the content of oxygen content of rGO significantly reduces compared with that of GO. The high-resolution C 1s XPS spectra of GO and rGO are presented in Fig. S3c, a significant decrease of oxygenated carbon related signals at 286-289 eV reveals that most of the hydroxyl, epoxide, and carboxyl functional groups are removed after the reduction.

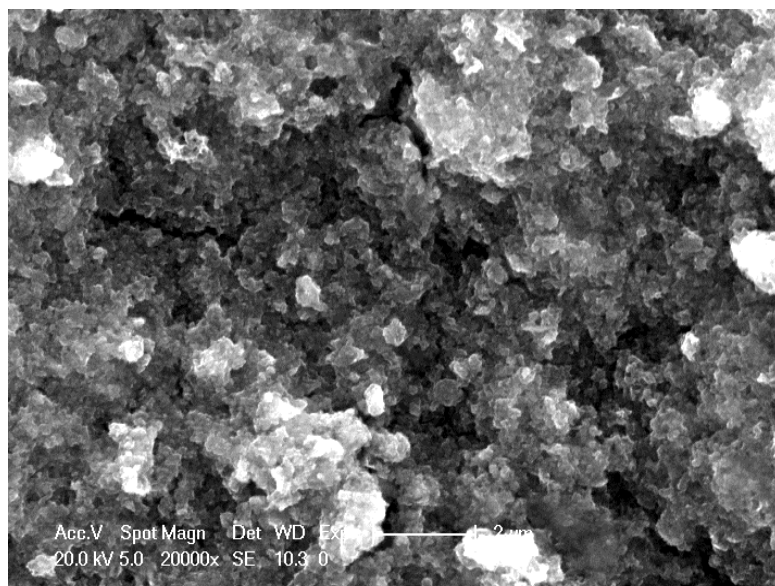


Fig. S4. SEM image of pure cobalt sulfide.

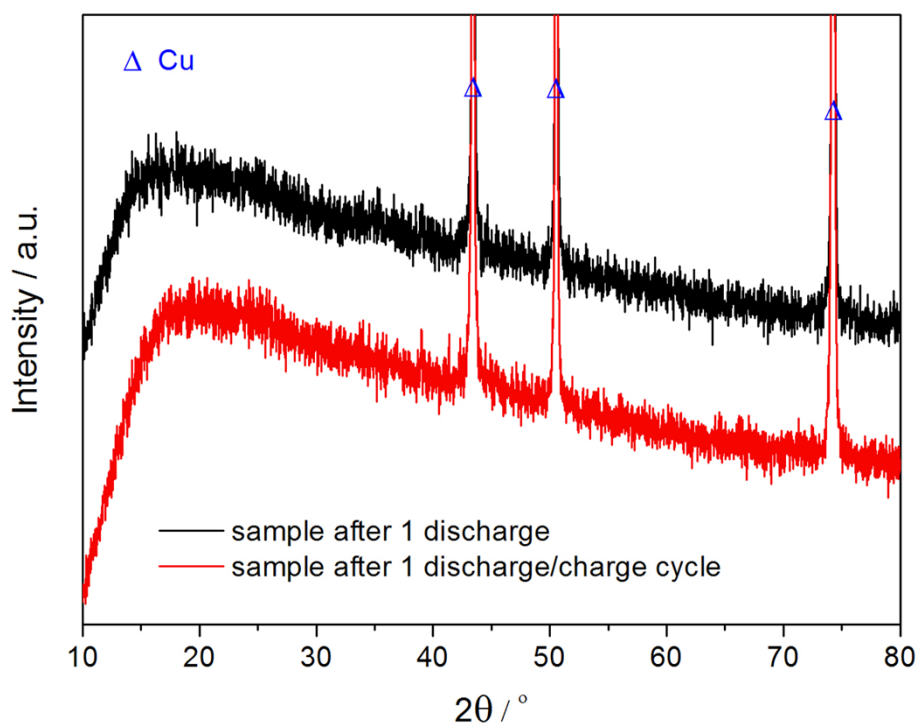


Fig. S5. XRD patterns of the as-prepared composite after different electrochemical cycles.

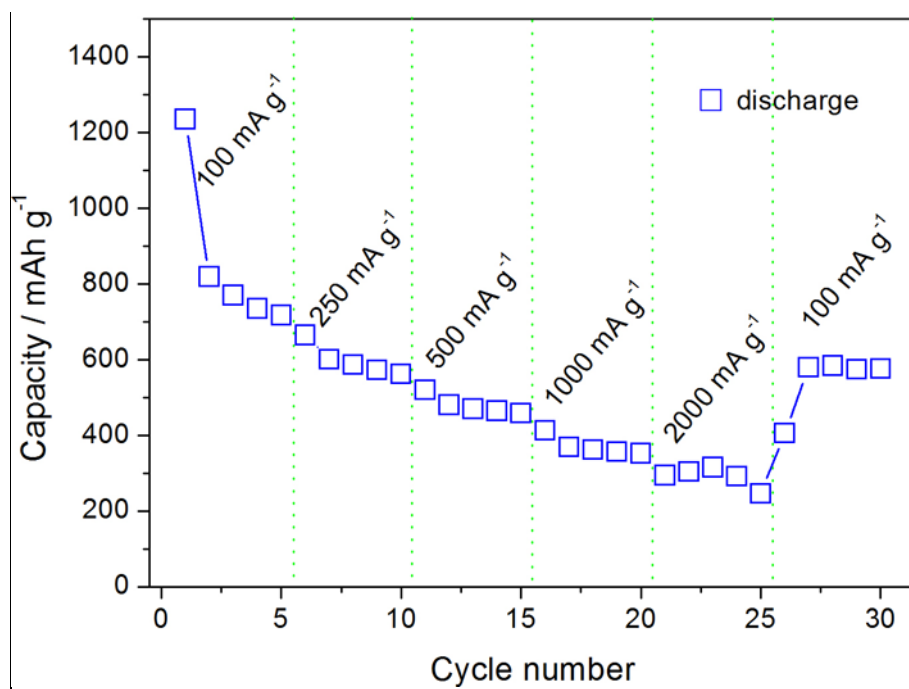


Fig. S6. Rate-capability performance of the pure cobalt sulfide at various current densities of 100, 250, 500, 1000, and 2000 mA g⁻¹.

Table S1 Comparison on electrochemical properties of cobalt sulfide-graphene composites

Phase of cobalt sulfide in the composites	Methods	Current density (mA g ⁻¹)	Initial Coulombic efficiency (%)	Cycle number	Retained capacity (mAh g ⁻¹)	Ref.
CoS ₂ /Co ₉ S ₈ /CoS	Solvothermal	100	55	50	954	S3
CoS ₂	Hydrothermal	50	67	40	~650	S4
Co ₃ S ₄	Hydrothermal	140.56	67	100	720	S5
CoS	Solvothermal	62.5	63	40	749	S6
Co ₉ S ₈ /Co _{1-x} S	Ultrasonic-assistant wet chemical method	200	74	150	994	This work

References

- S1. a) W. S. Hummers, R. E. Offeman, *J. Am. Chem. Soc.* **1958**, *80*, 1339; b) N.I. Kovtyukhova, P.J. Ollivier, B.R. Martin, T.E. Mallouk, S.A. Chizhik, E.V. Buzaneva, A.D. Gorchinskiy, *Chem. Mater.* **1999**, *11*, 771-778.
- S2. L. Tang, Y. Wang, Y. Li, H. Feng, J. Lu, J. Li, *Adv. Funct. Mater.* **2009**, *17*, 2782-2789.
- S3. G. Huang, T. Chen, Z. Wang, K. Chang, W. Chen, *J. Power Sources* **2013**, *235*, 122-128.
- S4. J. Xie, S. Liu, G. Cao, T. Zhu, X. Zhao, *Nano Energy* **2013**, *2*, 49-56.
- S5. N. Mahmood, C. Zhang, J. Jiang, F. Liu, Y. Hou, *Chem. Eur. J.* **2013**, *19*, 5183-5190.
- S6. Y. Gu, Y. Xu, Y. Wang, *ACS Appl. Mater. Interfaces* **2013**, *5*, 801-806.