Electronic Supporting Information for

Enhanced pseudocapacitance contribution to outstanding Li-storage performance for a reduced graphene oxide-wrapped FeS composite

anode

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Figure S1. The XRD pattern of rGO obtained from same method.



Figure S2. XRD analysis of non-goal products under changed reaction condition (without Vitamin C and at 210 °C, respectively).



Figure S3. TG curves of three samples: Pristine FeS, FeS@rGO and FeS@rGO-M.

The weight increase (4.5%) from around 200 °C to 400 °C could be attributed to a comprehensive result of partial conversion reaction (FeS \rightarrow FeSO₄) ¹ and volatilization of absorbed oleylamine (**Figure S3**). The weight loss (19.5%) from 420 °C to 550 °C responded to a deep oxidation of FeSO₄ to Fe₂O₃. ² Thus, the practical weight loss is about ~15% for pristine FeS. Considering theoretical loss (about 9%) for a complete conversion process of FeS to Fe₂O₃, it could be estimated to be ~6% absorbed mass for oleylamine. As to FeS@rGO, the experienced process is similar with above mentioned results excepting for additional rGO combustion ranging from 450 °C to 550 °C ². According to above analysis, the rGO contents in the FeS@rGO and FeS@rGO-M could be deduced to be ~31.2% and 41.5%, respectively.



Figure S4. Raman spectrum of rGO.



Figure S5. XPS data of rGO. (a) full spectrum; (b) C 1s spectrum; (c) N 1s spectrum and (d) O 1s spectrum.

Figure S5 showed XPS data of rGO. From full spectrum, carbon, nitrogen and oxygen elements were detected in the sample of rGO obtained from oleylamine-filled reaction system. In C 1s spectrum, the main content was fitted to graphitic carbon (at 284.6 eV), only a little part was ascribed to C=O bond (at binding energy of ~288.5 eV) resulting from incomplete reduction. ³ We observed that there was a small quantity of nitrogen element (2.53 wt%) from N 1s spectrum, which may be a result

of adsorbed oleylamine solvent in the final rGO. O 1s XPS spectrum showed two types oxygen: the fitting peak at binding energy of \sim 533.1 eV corresponded to adsorbed oxygen, another peak at \sim 531.2 eV could be attributed to C=O. In brief, by this reaction system, graphene oxide could be preferably reduced to rGO.



Figure S6. SEM image of pristine FeS particles.



Figure S7. Electrochemical performance of rGO at a current density of 200 mA g⁻¹.



Figure S8. Galvanostatic charge/discharge profiles with different cycles of pristine FeS.



Figure S9. Rate capabilities at different current densities from 100 to 1200 mA g⁻¹ of three samples: Pristine FeS, FeS@rGO and FeS@rGO-M.

Rate capability tests showed that FeS@rGO composite kept very high capacity retention rate (**Figure S9**), where capacities of 675, 594, 542, 507, 481, 457 and 440 mAh g⁻¹ were retrievable as current density increased from 100 mA g⁻¹ to 200, 400, 600, 800, 1000, 1200 mA g⁻¹, respectively. Similar to above long-term cycle, FeS@rGO-M also delivered relatively low capacity even though highly few damping under wide current density range because of an excess of rGO content in the composite. As we expected, pristine FeS anode almost can't afford significative capacity retention at high rate tests.

Morphology	Synthesis method	FeS content (%)	Current rate (mA g ⁻¹)	Cycle number	Capacity (mAh g ⁻¹)	Ref.	
TiO ₂ modified FeS	Hydrothermal method	/	200	100	510	<u>4</u>	
FeS anchored rGO	Direct-precipitation approach	/	60	30	288.6	<u>3</u>	
RGO wrapped FeS	Direct-precipitation approach	88	200 1000	10	660 200	<u>5</u>	
Ultrathin C@FeS Nanosheets	Surfactant-assisted solution-based synthesis	93	100	100	615	<u>6</u>	
FeS microsheet networks	Hydrothermal method	100	100	20	697	<u>7</u>	
Graphene-wrapped FeS-graphene nanoribbons	Reduction and vulcanization	69	400	100	536	<u>8</u>	
Interconnected porous FeS/C composite	Freeze- drying/carbonization method	89.5	1000	150	703	<u>9</u>	
FeS@rGO composite	One-pot method by thermal-solvent	68.8	200	150	887		
			1000	200	662	This work	
			5000	1000	325		

 Table S1. The electrochemical performances of previous reported FeS-based anode materials and FeS@rGO composite in our work

Table S2. Kinetic parameters of pristine FeS and FeS@rGO composite electrodes

Sample	$R_{\mathrm{e}}\left(\Omega ight)$	$R_{\rm ct}\left(\Omega\right)$	$R_{ m sf}\left(\Omega ight)$	$Z_{ m w}\left(\Omega ight)$
Pristine FeS	6.838	77.37	7.501	0.003863
FeS@rGO	6.153	36.55	2.687	0.002571



Figure S10. TEM images of (a) pristine FeS after only 50 cycles at 200 mA g^{-1} and (b) FeS@rGO composite after experiencing 150 cycles at 200 mA g^{-1} .



Figure S11. N₂ adsorption/desorption isotherms and pore diameter distribution (inset). (a) Cycled pristine FeS; (b) cycled rGO; (c) raw FeS@rGO and (d) cycled FeS@rGO.

To further investigate structure change of materials including pristine FeS, rGO and FeS@rGO, BET specific surface area and pore distribution have been measured by nitrogen adsorption/desorption method at 77 K, as shown in Figure S11. For cycled FeS and rGO electrode materials, the specific surface area were 27.76, 15.93 m² g⁻¹, respectively. In view of higher specific surface area for FeS material than that of rGO, there may be two main reasons: (1) FeS flake cracked into smaller particles but without the emergence of pore structure; (2) pure rGO nanosheets tended to form a stack so that their specific surface areas reduced. Compared with raw FeS@rGO composite (specific surface area: 21.30 m² g⁻¹, no pore structure), the specific surface area of cycled FeS@rGO material increased to 43.71 m² g⁻¹, which was also higher than those of cycled pristine FeS and rGO. The cycled FeS@rGO was detected to be formation of mesoporous (average pore size: 22 nm). This result is consistent with TEM observation, thereby demonstrating the reason for of appearance pseudocapacitance.



Figure S12. (a) STEM image of FeS@rGO composite after 150 charge/discharge cycles; (b-f) corresponding EDX elemental mappings of carbon, iron, sulfur, phosphorus, fluorine at that region in (a), respectively.



Figure S13. EDX pattern of FeS@rGO composite after 150 charge/discharge cycles.



Figure S14. The line relationship of $i/v^{1/2}$ with $v^{1/2}$, where slope and intercept are equal to k_1 and k_2 , respectively.



Figure S15. The capacitance contribution (shadow region) to total capacity at different sweep rates of (a) 0.1, (b) 0.3, (c) 1.0 and (d) 1.5 mV s^{-1} , respectively.

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