Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information

A Facile Synthesis of Three Dimensional Graphene Sponge Composited with Sulfur Nanoparticles for Flexible Li-S Cathodes

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Fig. S1 Digital camera image of the scaled up synthesis for precursor on a scale of gram per batch.

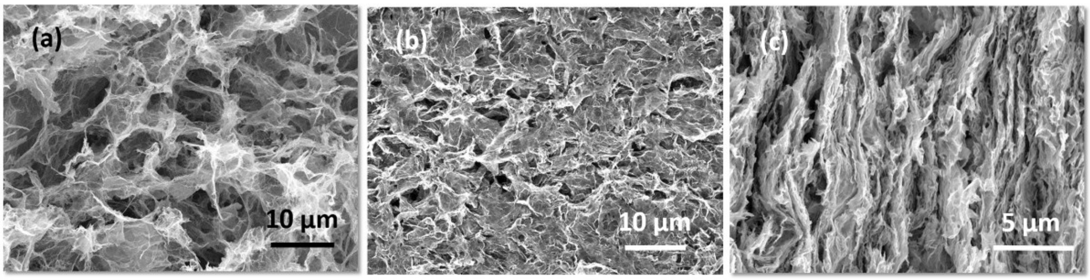

Fig. S2 The SEM images: 3DGS before compressed (a), front view (b) and cross section (c) of 3DGS after compressed.

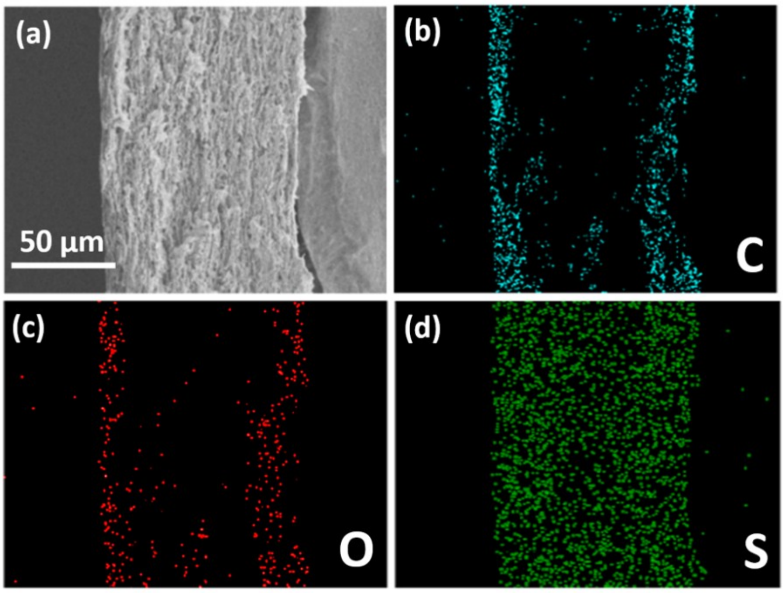

Fig. S3 The cross-sectional SEM image of the compressed 3DGS (a) and the corresponding EDS mapping, carbon (b), oxygen (c), sulfur (d) elements.

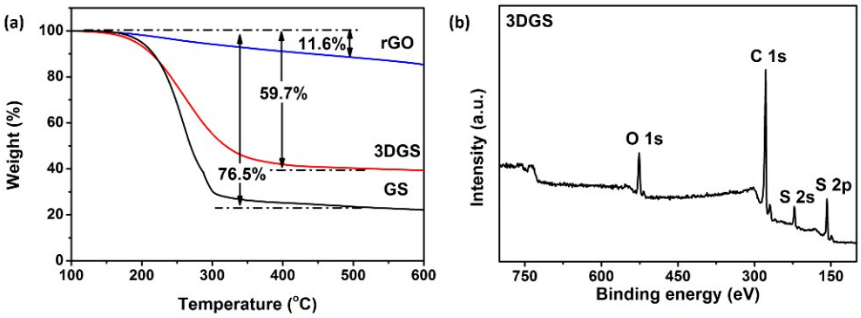

Fig. S4 TG curves of rGO, 3DGS and GS tested from 100 ^{o}C to 600 ^{o}C at a heating rate of 10 ^{o}C min^{-1} under N_2 atmosphere (a), XPS full scan spectrum of 3DGS (b).

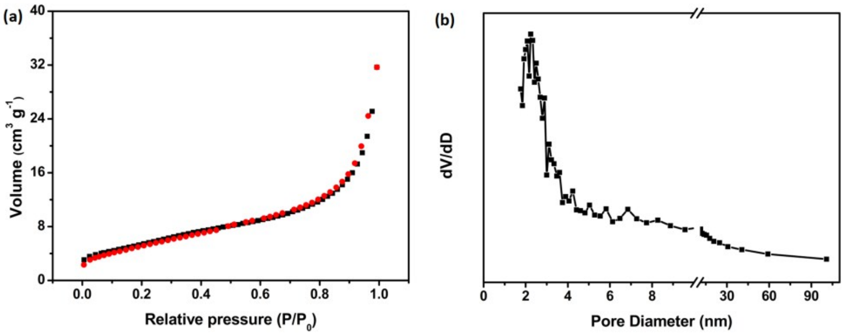

Fig. S5 N_2 adsorption-desorption isotherms (a), and the corresponding pore size distributions of the 3DGS sponge (b).

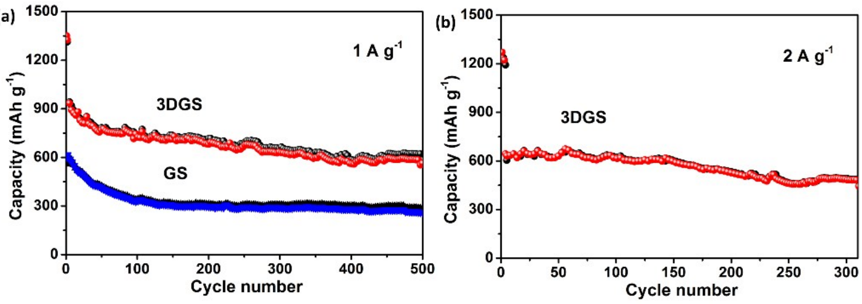

Fig. S6 The cyclic performance of the 3DGS and GS electrodes at 1 A g^{-1} for 500 cycles (a), the cyclic performance of the 3DGS electrode at 2 A g^{-1} for 300 cycles (b).

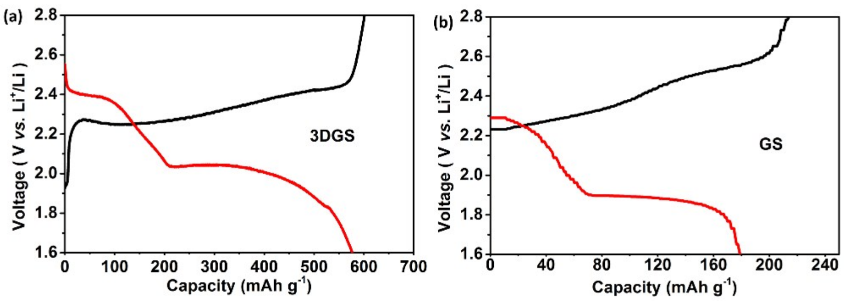

Fig. S7 The discharge and charge profiles of 3DGS (a) and GS (b) electrodes after 500 cycles at 1.5 A g^{-1}.

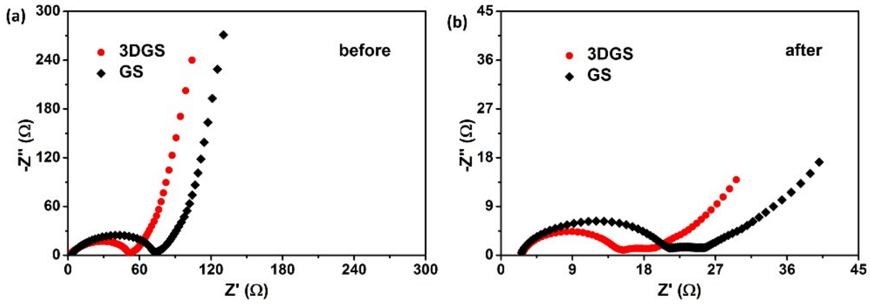

Fig. S8 The EIS plots of 3DGS and GS electrodes before (a) and after cycling (b) for 500 cycles at 1.5 A g^{-1}.

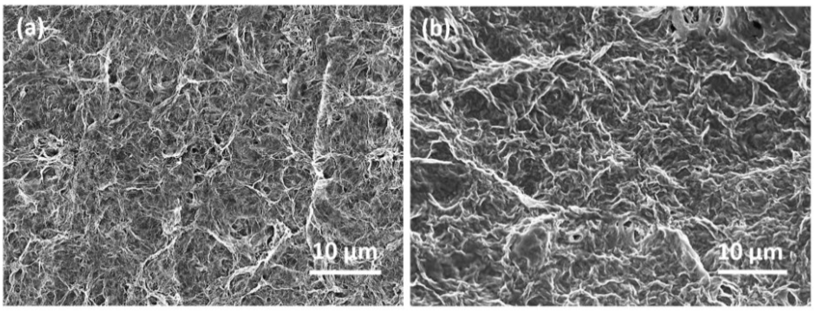

Fig. S9 The morphologies of the 3DGS electrode before (a) and after (b) cycling for 500 cycles at 1.5 A g^{-1}.

Table S1 A survey of electrochemical performance for 3D graphene and sulfur composites.

Host material	sulfur content (%)	sulfur mass loading (mg cm^{-2})	discharge capacity (mAh g^{-1})	cyclic capacity retention	Reference
3DGS	54.4/54.4	2	740 , 1.5 A/g	94.77%, 100th 94.05%, 200th 84.14%, 300th 83.17%, 400th	This work
G-S63 hybrid	63/63	2	700, 0.75 A/g	77.3%, 100th	1
S-rGO paper	63/63	1	657, 1.5 A/g	90%, 167th	2
3D-GNS	86.7/60.69	1	785, 1.5 A/g	85%, 200th	3
GCS	70/56	1.2	613, 1.67 A/g	85%, 450th	4
BGA-S	59/47.2	0.135	882, 3.3 A/g	68%, 200th	5
Dense S-G	70.6/49.42	0.8-1	920, 0.83 A/g	83%, 300th	6
3DCGS	80.1/72	1	1180, 0.33 A/g	83%, 200th	7

Note：The sulfur content in Table S1 is presented as the sulfur content in composite/the sulfur content in the electrode (calculated based on the composite, conductive additive and binder).

The calculation process of the sulfur content in 3DGS and GS:

According to the TGA curves, the total mass loss of 3DGS is 60%, which is attributed to the sublimation of sulfur and the further reduction of rGO. The weight loss of rGO between 150 °C – 350 °C is 9%. If we presume the weight of 3DGS as 1mg, the sulfur mass in the composite as X mg. then the following equation is founded:

$(1 - X) \cdot 11.6\% + X = 59.7\%$

Through calculation, X = 0.544, therefore, the sulfur content in 3DGS composite is 54.4wt%. Similarly, the sulfur content in GS composite is 73.4wt%.

The calculation method and details of electrolyte uptake ability:

The electrolyte uptake (EU) ability was evaluated by the following equation:

$\mathrm{EU}\ (\%) = (W_1 - W_2)/W_1 \times 100\%$

Where W_1 and W_2 are the weights of the electrodes before and after soaking in the electrolyte, respectively. Both the 3DGS and GS electrodes were immersed in the electrolyte overnight. After removing the electrolyte on the surface of the electrodes, the weights were measured and used to calculate the electrolyte uptake value through the equation. The electrolyte uptake abilities of 3DGS and GS electrodes are 2500% and 40%, respectively.

Reference

1. G. Zhou, L.-C. Yin, D.-W. Wang, L. Li, S. Pei, I. R. Gentle, F. Li and H.-M. Cheng, *ACS Nano*, 2013, **7**, 5367-5375.
2. C. Wang, X. Wang, Y. Wang, J. Chen, H. Zhou and Y. Huang, *Nano Energy*, 2015, **11**, 678-686.
3. C. Wang, K. Su, W. Wan, H. Guo, H. Zhou, J. Chen, X. Zhang and Y. Huang, *J. Mater. Chem. A*, 2014, **2**, 5018-5023.
4. S. Niu, W. Lv, C. Zhang, Y. Shi, J. Zhao, B. Li, Q.-H. Yang and F. Kang, *J. Power Sources*, 2015, **295**, 182-189.
5. Y. Xie, Z. Meng, T. Cai and W. Q. Han, *ACS Appl. Mater. Interfaces*, 2015, **7**, 25202-25210.
6. H. Li, X. Yang, X. Wang, M. Liu, F. Ye, J. Wang, Y. Qiu, W. Li and Y. Zhang, *Nano Energy*, 2015, **12**, 468-475.
7. J. He, Y. Chen, P. Li, F. Fu, Z. Wang and W. Zhang, *J. Mater. Chem. A*, 2015, **3**, 18605-18610.