Electronic Supplementary Material (ESI) for RSC Advances.

Electronic Supplementary Information

Room-Temperature Synthesis of Water-Dispersible Sulfur-Doped Reduced Graphene Oxide without Stabilizers

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1. Elemental data of reduced graphene oxide by XPS spectra.

Table S1. Elemental data of reduced graphene oxide prepared using different amounts of NaSH • H_2O

$R_{NaSH/GO}$	**C** (wt%)	**S** (wt%)	**O** (wt%)	**C/S**	**C/O**
0:1 (GO)	65.84	0	29.55	--	2.23
0.5:1	72.18	1.96	22.20	36.83	3.25
1:1	78.37	2.29	17.85	34.22	4.39
2:1	84.03	2.46	12.71	34.16	6.71
5:1	84.26	2.53	12.45	33.30	6.77
10:1	85.06	2.39	11.97	35.59	7.11
20:1	83.94	2.62	12.69	32.04	6.61
40:1	84.55	2.45	12.34	34.51	6.85

2. Comparison of S-rGO prepared by different methods

Table S2. Comparison of S-rGO prepared by different methods

No	**Method**	**Precursor**	**Reaction Conditions**	**C/O ratio** (XPS data)	**Sulfur Element**	**Ref.**
1	CVD method	Sulfur/hexane vapor	950℃, 2.5min	＞99	0.60 at.%	[1]
2	CVD method	Graphene/H_2S	1000℃, 5min	/	0.96 at.%	[2]
3	Thermal Annealing	GO/Benzyl disulfide (BDS)	600℃, 30min 900℃, 30min 1050℃, 30min	/ / /	1.53 wt% 1.35 wt% 1.30 wt%	[3]
4	Thermal Annealing	GO/BDS	1050℃, 30min	/	2.54 wt%	[4]
5	Thermal Annealing	GO/Phenyl disulfide	1000℃, 30min	6.72-40.71	0.35-3.95 at.%	[5]
6	Thermal Annealing	GO/PEDOT	800℃, 3h	/	2.02 wt%	[6]
7	Thermal Annealing	Graphite oxide/ SO_2/H_2S/CS_2 gas	600-1000℃, 12min	9.30-24.09	0.10-11.99 wt%	[7]
8	Thermal Annealing	GO/H_2S gas	250℃, 10-30min 650℃, 10-30min 1000℃, 10-30min	5.94-6.69 6.77-7.03 22.72-30.85	No 0.83-1.89 at.% 2.02-2.19 at.%	[8]
9	Hydrothermal Treatment	GO/Na_2S	200℃, 10h	9.17*	2.22 at.%*	[9]
10	Ethanol-thermal reaction	GO/BDS/Ethanol	180℃, 10h	8.68	1.20 at.%	[10]
11	Refluxing	GO/P_4S_{10}	120℃, 12h	/	2.20 at.%	[11]
12	This work	GO/NaSH	RT, 10min-2h	6.71	2.50 wt%	

*measured by EDX.

3. PH of GO and S-rGO dispersions

Table S3. The PH of GO and rGO dispersions

$NaSH{\cdot}xH_2O$：GO by weight ratio	0:1	0.5:1	1:1	2:1	5:1	10:1	20:1	40:1
PH	5.46	5.94	7.20	8.67	10.04	9.68	9.75	11.79

4. ^{13}C MAS NMR spectra of S-rGO

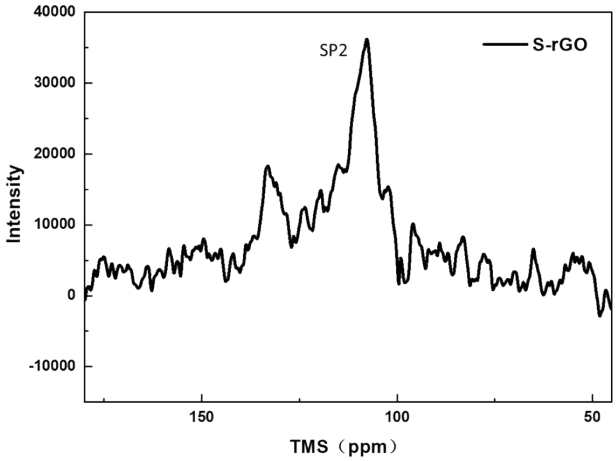

Fig.S1 ^{13}C NMR spectra of S-rGO in this work.

5. XRD pattern of GO and S-rGO sheets.

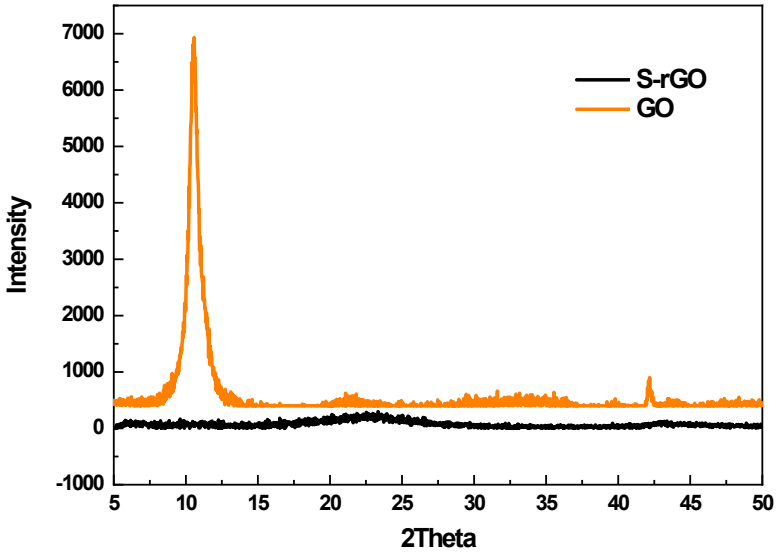

Fig. S2 XRD patterns of GO and rGO sheets

6. The fabrication of S-rGO laminate.

This simple method will facilitate the implementation of S-rGO into relevant applications. To proof the feasibility of this method, an S-rGO laminate was prepared base on this work.

Ref. 8 reported the S-rGO laminate for EMI shielding application, in which the S-rGO was prepared by thermal annealing process, and the laminate was fabricated by pressing the S-rGO powder. Based on our work, an S-rGO laminate with the sheet resistance of 4096 Ω/m^2 can be obtained by directly filtering S-rGO dispersion at room temperature.

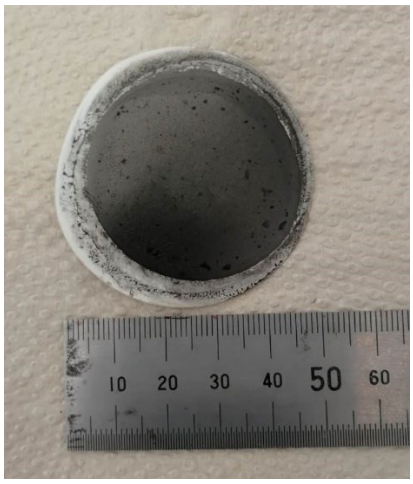

Fig. S3 The digital photograph of the S-rGO laminate.

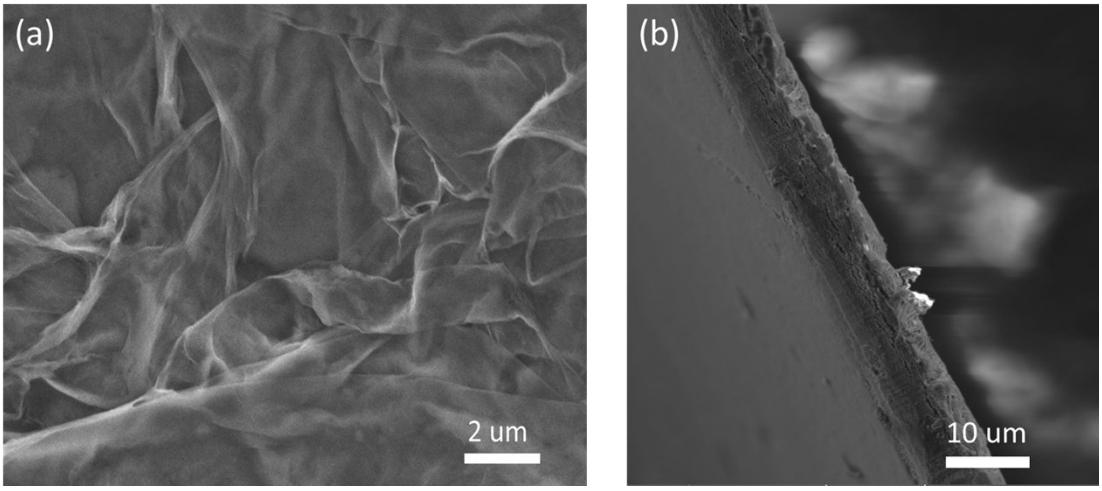

Fig. S4 The SEM images of the S-rGO laminate, (a)surface, (b)cross-section.

References

[1] Gao H, Liu Z, Song L, Guo W, Gao W, Ci L, et al. Synthesis of S-doped graphene by liquid precursor. Nanotechnology. 2012;23:275605.

[2] Liang C, Wang YL, Li T. Sulfur-doping in graphene and its high selectivity gas sensing in NO_2. 2015 Transducers - 2015 18th International Conference on Solid-State Sensors, Actuators and Microsystems (TRANSDUCERS); p. 1421-1424.

[3] Yang Z, Yao Z, Li G, Fang G, Nie H, Liu Z, et al. Sulfur-Doped Graphene as an Efficient Metal-free Cathode Catalyst for Oxygen Reduction. ACS Nano. 2012;6(1):205-211.

[4] Xia L, Yang J, Wang H, Zhao R, Chen H, Fang W, et al. Sulfur-doped graphene for efficient electrocatalytic N_2-to-NH_3 fixation. Chem Commun. 2019;55(23):3371-3374.

[5] Hoque MA, Hassan FM, Seo M-H, Choi J-Y, Pritzker M, Knights S, et al. Optimization of sulfur-doped graphene as an emerging platinum nanowires support for oxygen reduction reaction. Nano Energy. 2016;19:27-38.

[6] Cai B, Shao C, Qu L, Meng Y, Jin L. Preparation of sulfur-doped graphene fibers and their application in flexible fibriform micro-supercapacitors. Frontiers of Materials Science. 2019;13(2):145-153.

[7] Poh HL, Šimek P, Sofer Z, Pumera M. Sulfur-Doped Graphene via Thermal Exfoliation of Graphite Oxide in H_2S, SO_2, or CS_2 Gas. ACS Nano. 2013;7(6):5262-5272.

[8] Shahzad F, Kumar P, Yu S, Lee S, Kim Y-H, Hong SM, et al. Sulfur-doped graphene laminates for EMI shielding applications. Journal of Materials Chemistry C. 2015;3(38):9802-9810.

[9] Tian ZS, Li JT, Zhu GY, Lu JF, Wang YY, Shi ZL, et al. Facile synthesis of highly conductive sulfur-doped reduced graphene oxide sheets. Phys Chem Chem Phys. 2016;18(2):1125-1130.

[10] Wang Z, Li P, Chen Y, He J, Zhang W, Schmidt OG, et al. Pure thiophene–sulfur doped reduced graphene oxide: synthesis, structure, and electrical properties. Nanoscale. 2014;6(13):7281-7287.

[11] Klingele M, Pham C, Vuyyuru KR, Britton B, Holdcroft S, Fischer A, et al. Sulfur doped reduced graphene oxide as metal-free catalyst for the oxygen reduction reaction in anion and proton exchange fuel cells. Electrochemistry Communications. 2017;77:71-75.