Article

Supplementary Materials: One-Pot Synthesis of Graphene-Sulfur Composites for Li-S Batteries: Influence of Sulfur Precursors

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Polarisation voltages

Table S1. Polarisation voltages of charge and discharge curves at rate of C/10.

Cycle number	10	20	30	40	50
Potential at 50 % Discharge (V)	2.087	2.086	2.085	2.083	2.086
Potential at 50 % Charge (V)	2.297	2.298	2.299	2.301	2.292
Polarisation voltage (V)	0.210	0.212	0.214	0.218	0.206
Average (V)	0.212				

High-resolution spectra

Table S2. Fitting results for carbon species contribution of C 1s high-resolution XP spectra.

Spectrum	Band	Position	Height	Area	%Area
GO	1	284.5	12024	20457	47
	2	286.5	10857	16860	38
	3	288.4	2725	5370	12
	4	291.4	384	1174	3
CRGO	1	284.4	12796	16668	55
	2	285.7	3202	6101	20
	3	288.2	1548	5904	19
	4	291.4	491	1447	5
	5	294	171	372	1
GS2	1	284.4	20332	26745	61
	2	285.6	4454	8487	19
	3	288.2	1932	7368	17
	4	291.4	381	1122	3

Table S3. Element concentrations from the high-resolution XP spectra.

Sample	C 1s at%	N 1s at%	O 1s at%	Na 1s at%	S 2p at%
GO	56.8	1.8	37.7	0.1	3.6
CRGO	82.2	0.1	16.4	0.5	0.8
GS2	73.8	0.2	16	1.2	8.8

High-resolution S 2p XPS

Table S4. Sulphur species determination from S 2p high resolution spectra for the sulphur-doped samples.

Spectrum	Band	Position	Height	Area	%Area
GO	1	163.1	36	51	1
	2	164.3	16	26	0.5
	1*	168.2	2002	3384	66
	2*	169.4	1001	1692	33
CRGO	1	163.6	307	383	60
	2	164.8	160	191	30
	1*	168	25	46	7
	2*	169.2	14	23	3
GS2	1	163.5	5566	6314	62
	2	165.7	2896	3157	31
	1*	167.9	227	524	5
	2*	169.1	119	262	2.5
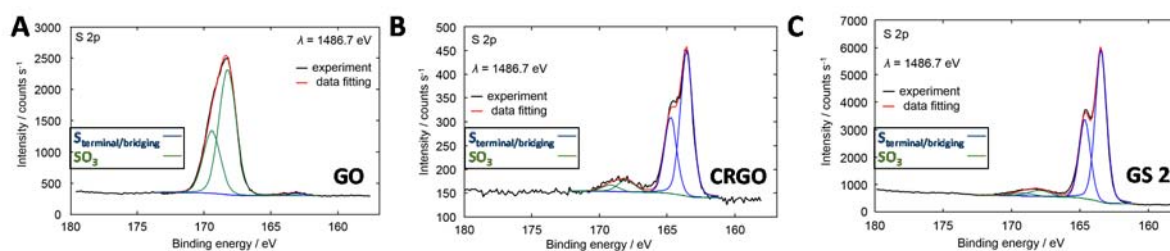
Figure S1 High resolution S 2p spectra with sulphur species fitting of the composite materials. **A** GO. **B** CRGO. **C** GS2.

All samples were fitted with two sulphur species with the S 2p doublet and a spin-orbit splitting of 1.2 eV.

The sulphur species visible in all three samples significantly differ from the GO to the CRGO and GS2. As for the GO just very small amount of terminal/bridging sulphur species typical for elemental sulphur. Large amount of S-O groups is found which are attributed to $-SO_3$ species.[1] For the reduced graphene oxides of CRGO and GS2, the opposite is true. Elemental sulphur is the dominating species. It is noted that the GS2 sample shows the highest sulphur concentration and the CRGO the lowest even lower than the GO but with other species distribution as described above. GS2 is the sample with the highest ability to entrap elemental sulphur while GO features the highest amount of sulfonate groups.[2]

Table S5. Details of the Rietveld analysis of the graphene-sulphur composite GS2.

Phase type	Space group	Lattice parameters	R_{Bragg}	$R_{f\text{-}factor}$
α-sulphur	*Fddd* (70)	a = 10.4634(1) Å	6.12	4.82
		b = 12.8662(1) Å		
		c = 24.4819(2) Å		
		$\alpha=\beta=\gamma= 90°$		
Graphitic	*P6$_3$/mmc* (194)	a = 3.4963 Å	29.7	32.2
		b = 3.4963 Å		
		c = 7.7303 Å		
		$\alpha=\beta= 90°\ \gamma= 120°$		

References

1. Balach, J.; Jaumann, T.; Klose, M.; Oswald, S.; Eckert, J.; Giebeler, L. Functional Mesoporous Carbon-Coated Separator for Long-Life, High-Energy Lithium–Sulfur Batteries. *Adv. Funct. Mater.* **2015**, *25*, 5285-5291.
2. Zhang, L.; Ji, L.; Glans, P.-A.; Zhang, Y.; Zhu, J.; Guo, J. Electronic structure and chemical bonding of a graphene oxide-sulfur nanocomposite for use in superior performance lithium-sulfur cells. *Phys. Chem. Chem. Phys.* **2012**, *14*, 13670-13675.