Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Electronic Supplementary Information

Nitrogen-Sulfur Co-doped Porous Graphene as a Sulfur Immobilizer for Lithium Sulfur Battery

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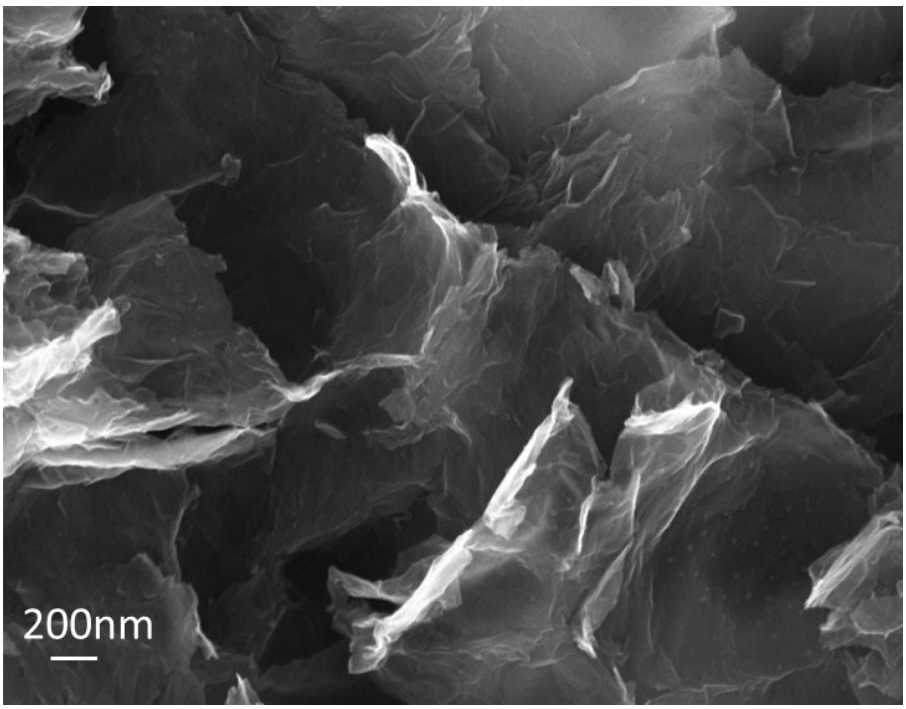

Figure S1 SEM image of NSG.

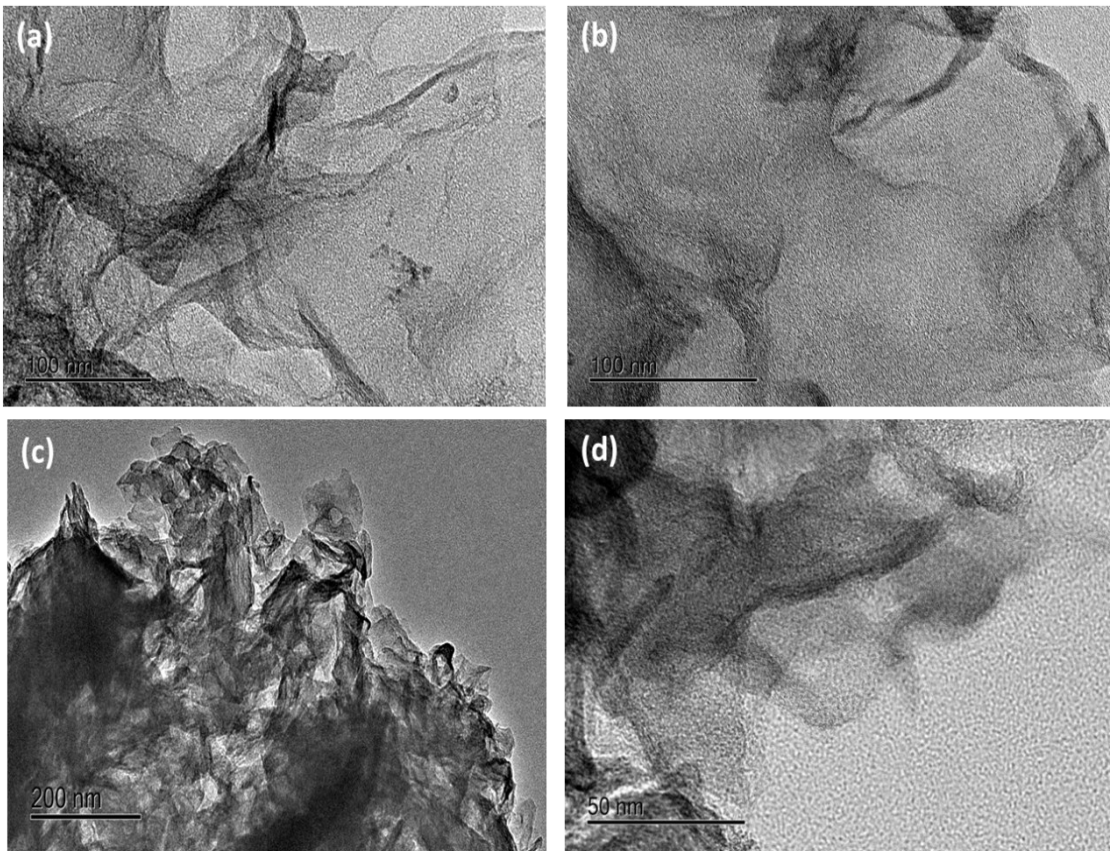

Figure S2 (a, b) TEM images of A-NSG. (c, d) High-magnification TEM images of the A-NSG@S.

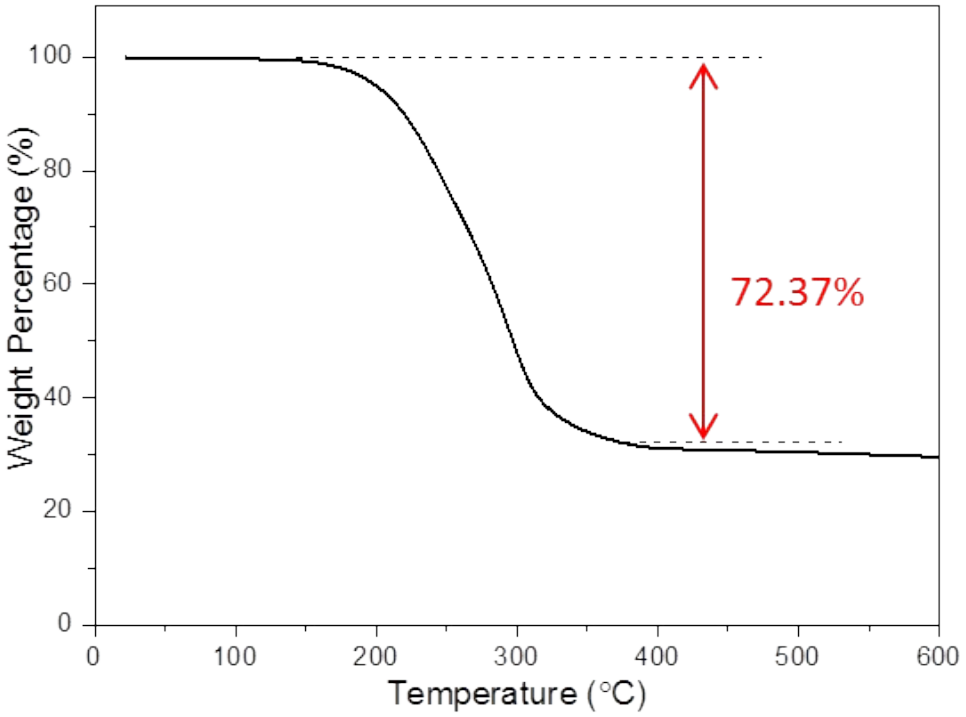

Figure S3 Thermogravimetric curves of pure sulfur powder and A-NSG@S in the N_2 with a heating rate of 10 ˚C min^{-1}.

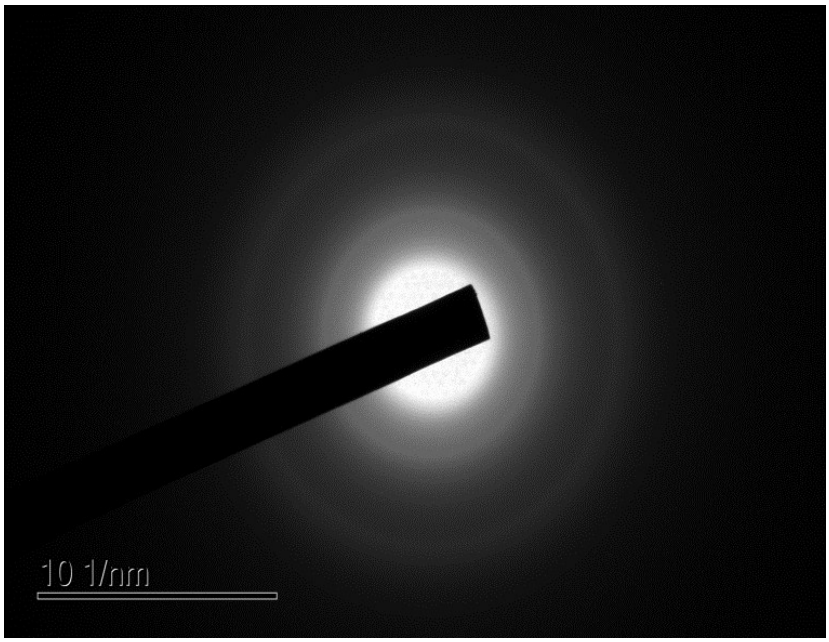

Figure S4 The SAED pattern of A-NSG@S.

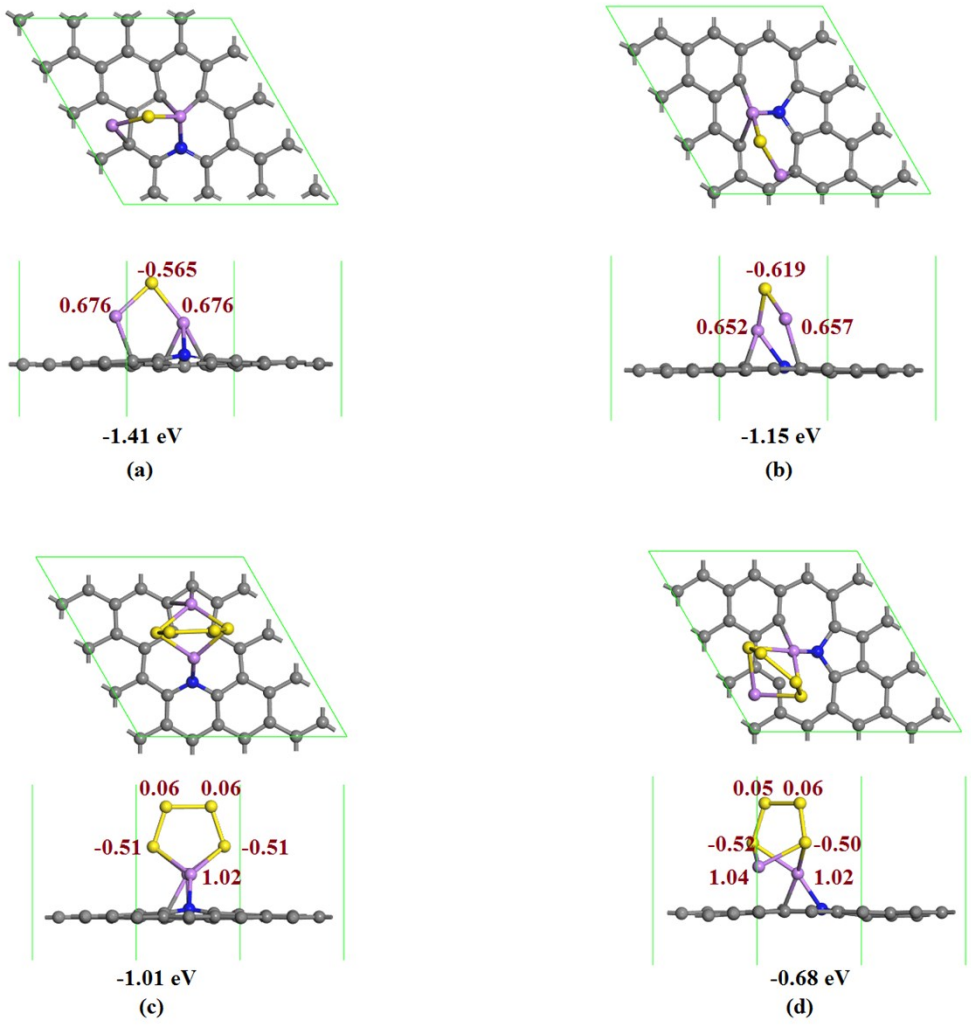

Figure S5 Optimized configurations for the adsorption of Li_2S on pyridinic and pyrrolic N sites (a, b), Li_2S_4 on pyridinic and pyrrolic N sites (c, d) with corresponding adsorption energies in eV and atom Mulliken charge. Gray, blue, yellow, and purple balls represent C, N, S, and Li atoms, respectively.

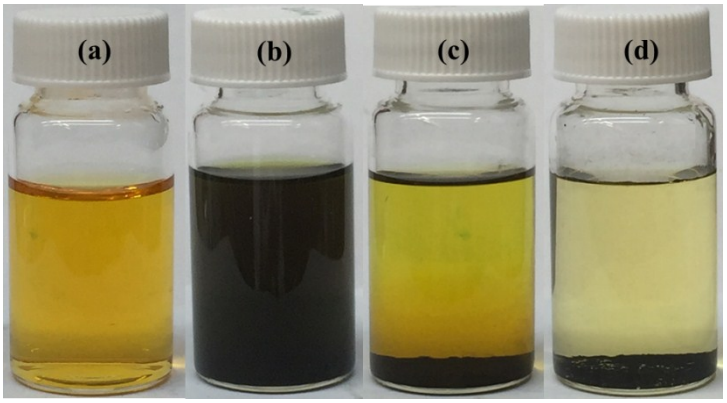

Figure S6 (a) Li_2S_4 solution (b) Initially mixed (c) Aging for 3h (d) Aging for 24h.

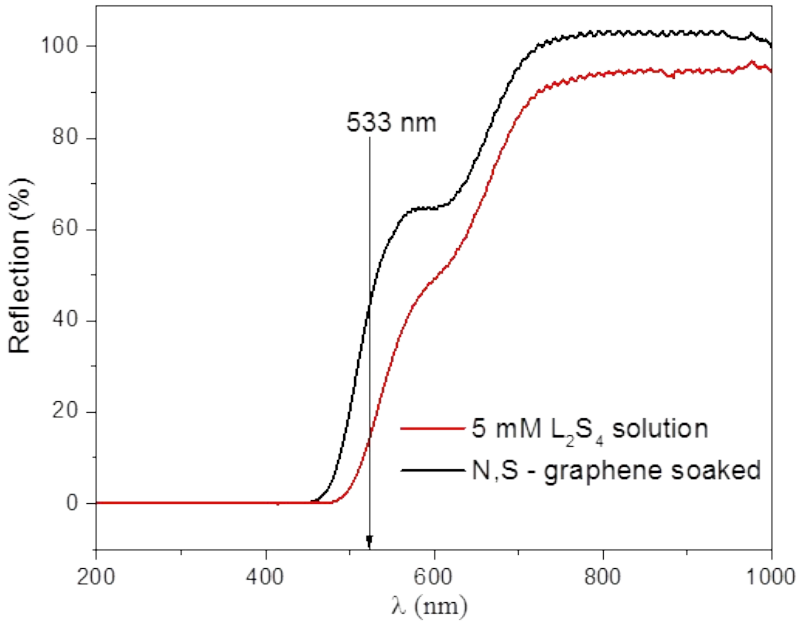

Figure S7 UV-vis spectra of 5 mM pristine Li_2S_4 solution and solution after soaking the N, S-graphene.

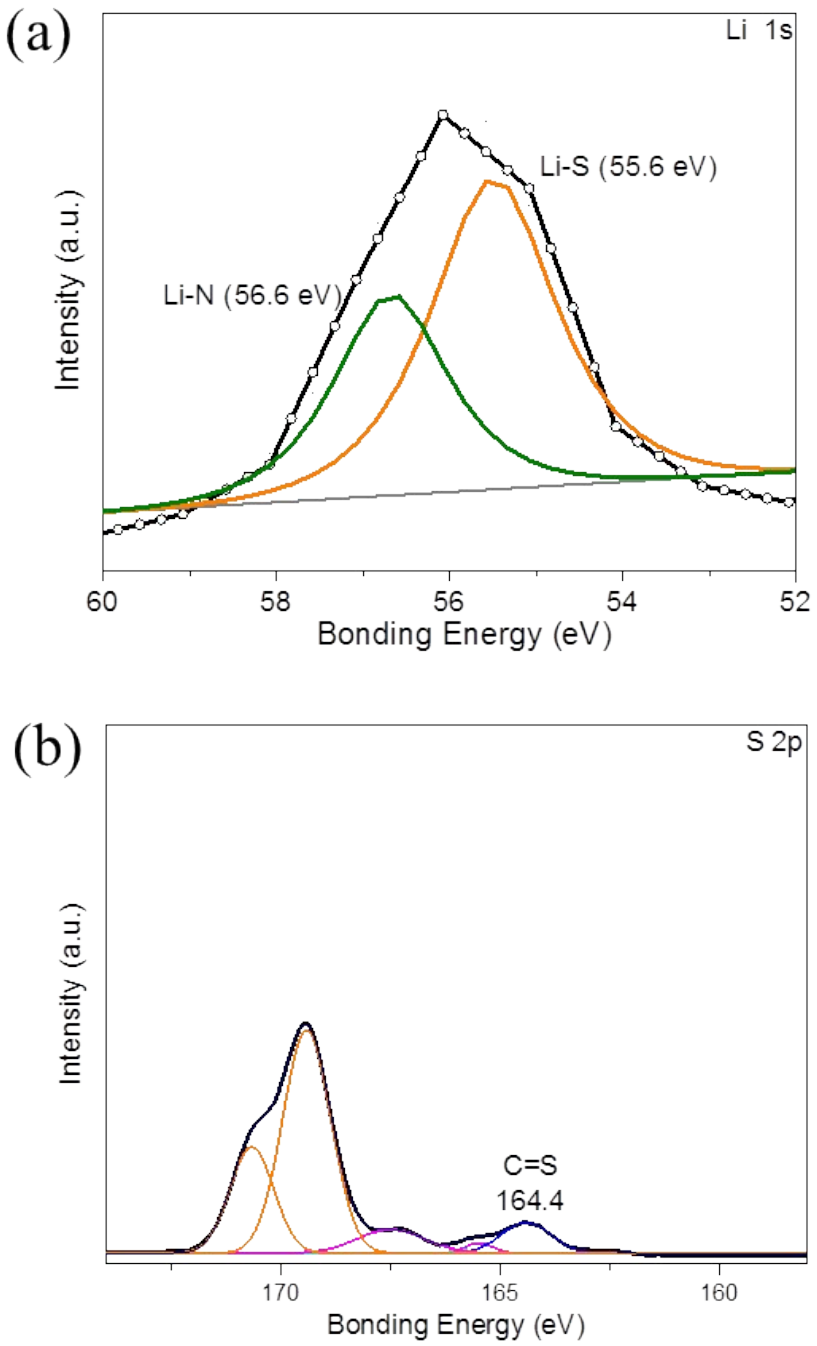

Figure S8 High-resolution XPS of the N,S co-doped electrode after discharged to 2.1 V. (a) Li 1s, (b) S 2p spectra of N,S graphene-Li_2S_x.

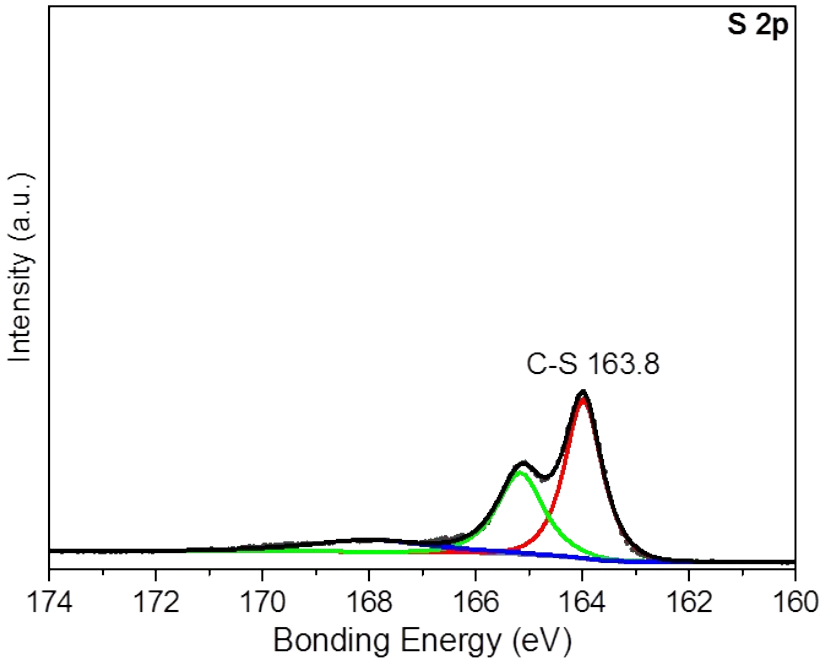

Figure S9. High-resolution S 2p spectra of N,S graphene@ S

Table S1 Percentage of Carbon atoms and Full width at Half-Maximum Values of C 1s Peaks in Graphene, A-NSG.

	carbon % (atom %)	fwhm (eV)
Graphene	93.4%	1.13
A-NSG	89.81 %	1.21

Table S2 Carbon, oxygen, nitrogen, sulfur atomic percent of GO, A-NSG materials.

Sample	C (at%)	O (at%)	N (at%)	S (at%)	C/O
GO	70.2	29.8	_	_	2.4
A-NSG	89.81	5.16	4.18	0.85	17.25

Table S3a Comparisons of the adsorption energies of graphene, N-graphene and N,S-graphene with Li_2S, and Li_2S_4.

E_{ads} (eV)	Graphene	Pyridinic like graphene	Pyrrolic like graphene	N, S doped Graphene
Li_2S	-0.94	-1.41	-1.15	-1.85
Li_2S_4	-0.49	-1.01	-0.68	-1.25

Table S3b Comparisons of the atomic charge transfers (QT) of graphene, N-graphene and N,S-graphene with Li2S, and Li2S4.

Q_T (e)	Graphene	Pyridinic N	Pyrrolic N	N, S doped Gr
Li_2S	0.227	0.787	0.69	0.555
Li_2S_4	0.019	0.12	1.15	0.317

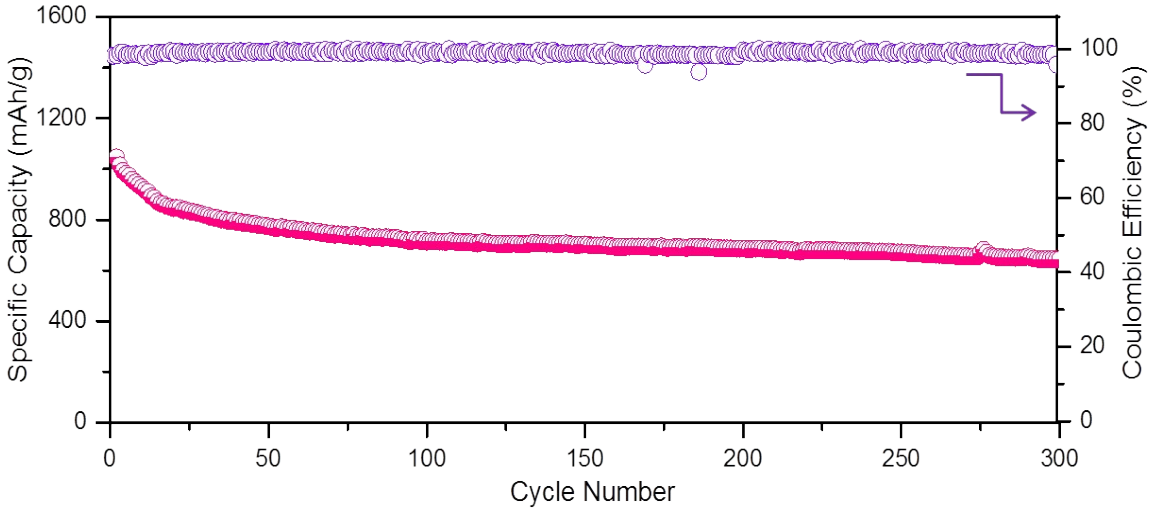

Figure S10 Long-term cycling performance test of the A-NSG@S electrode at 1C discharge rate and the corresponding Coulombic efficiency

Table S4 Parameters identification by modeling the impedance spectra

in Figure 9b.

Cycle number	R_e (ohm)	R_{ct} (ohm)	R_g (ohm)	$Y_1(\Omega^{-1}\cdot cm^{-2}\cdot s^{-n})$	n 1	$Y_2(\Omega^{-1}\cdot cm^{-2}\cdot s^{-n})$	n 2
Pristine	7.23	34.71	-	0.000027	0.757	-	-
1	7.98	36.23	18.23	0.00003	0.84	0.087	0.93
600	8.329	41.56	23.23	0.0009	0.6523	0.1094	0.865

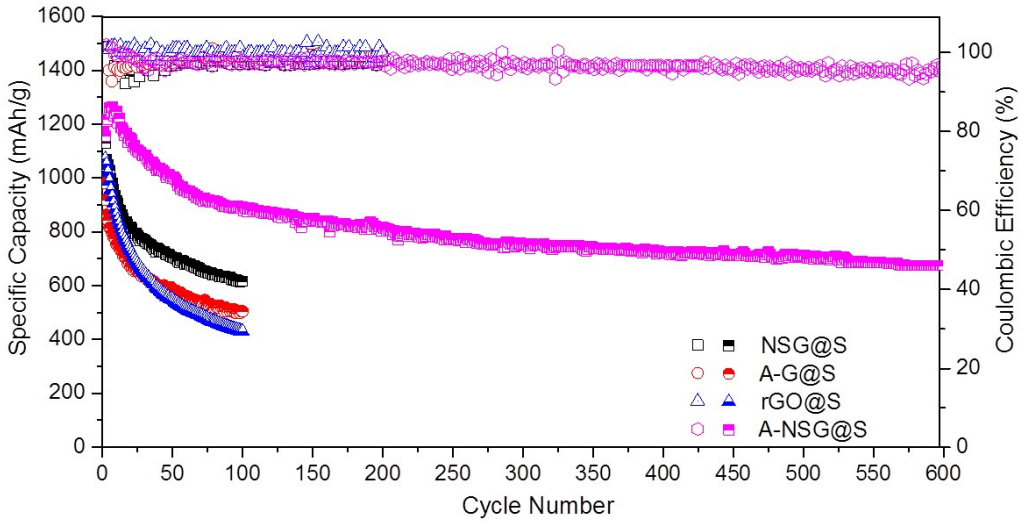

Figure S11 Cycling performance of A-NSG@S material cycled at 0.2C, in comparison with NSG@S, A-G@S, rGO@S material. Specific capacity values were all calculated based on the mass of sulfur.

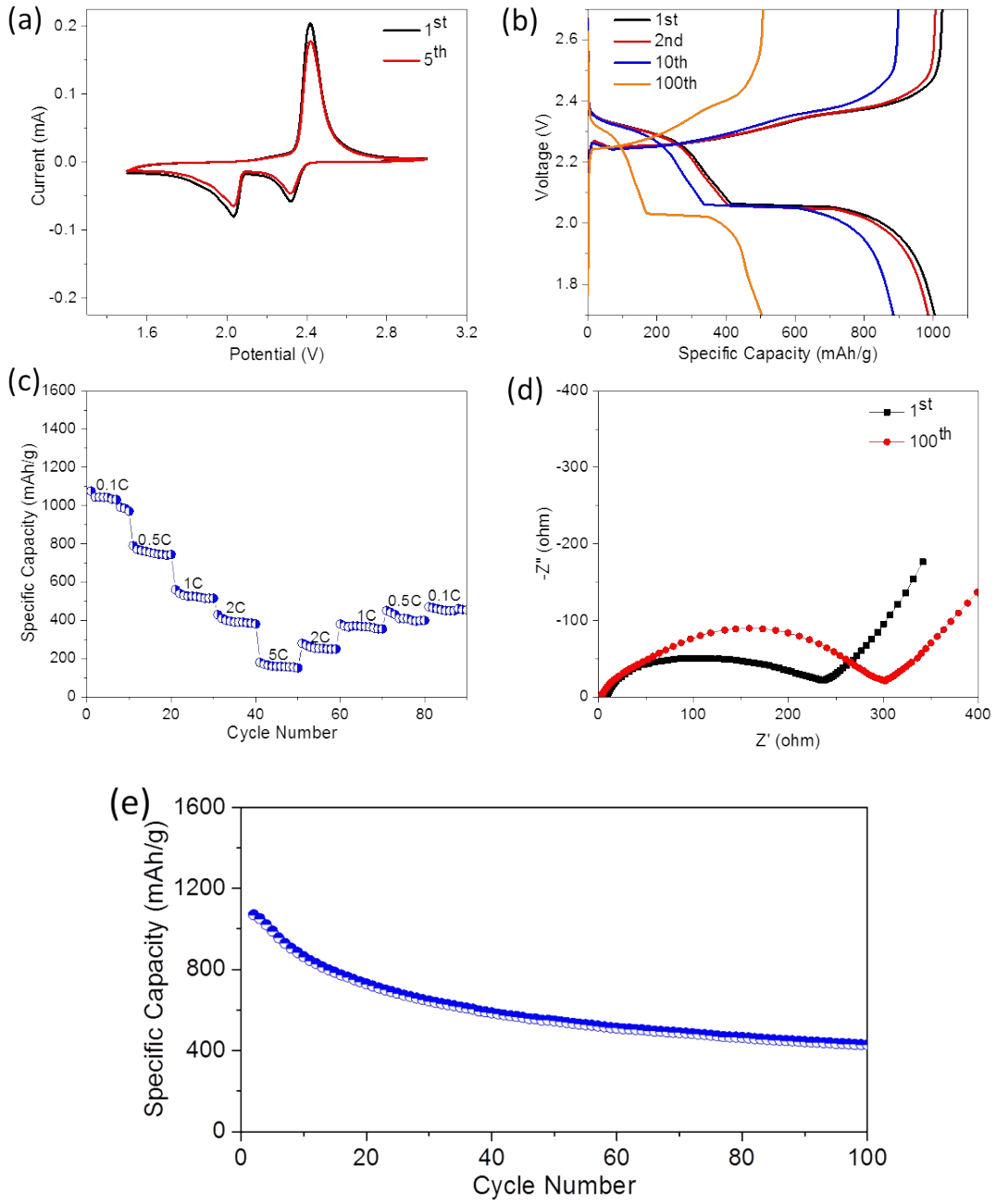

Figure S12 Electrochemical characterization of graphene as the cathode of a Li-S battery. (a) Cyclic voltammetry (CV). (b) Galvanostatic charge and discharge profiles for different cycles at 0.2C. (c) Discharge/Charge capacity cycled at various rates from 0.1 C, 0.5C, 1C, 2C, 5C (d) Electrochemical impedance spectra of graphene@S lithium-sulfur battery at different cycles.(e) Long term cycling performance of graphene@S.

Table S5 Recent advance in the dual doped carbon framework to host sulfur for Li-S batteries

Carbon Composites	Cycle life(cycles)	Decay Rate(per cycle)	Ref
A-NSG(N,S doped porous graphene)	600	0.056%	This work
N,S-graphene	500	0.078%	*Nature Commun. 2015, 6, 7760*
N,S doped carbon	500	0.065%	*Adv Mater. 2015, 27, 6021-6028*
N,O doped CNTs	200	0.15%	*Adv Mater Interfaces. 2014, 1, 1400227*
N,O-graphene	350	0.057%	*Nature Commun. 2014, 5, 5002*
N,O-doped porous carbon	200	0.05%	*Angew Chem Int Ed. 2015, 54, 4325–4329*