Reduced graphene oxide derived from the spent graphite anodes as a sulfur host in lithium-sulfur batteries

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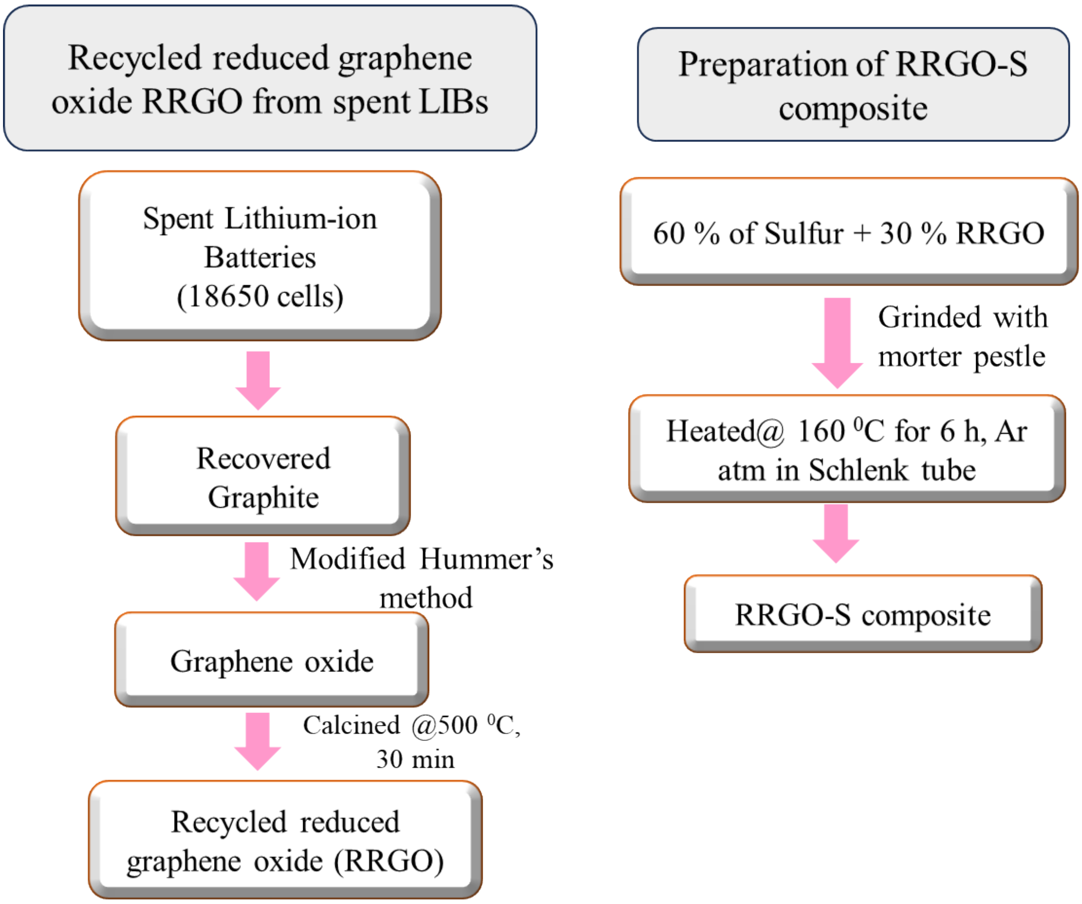

Fig.S1. Flow chart for the synthesis of RRGO and RRGO-S composite.

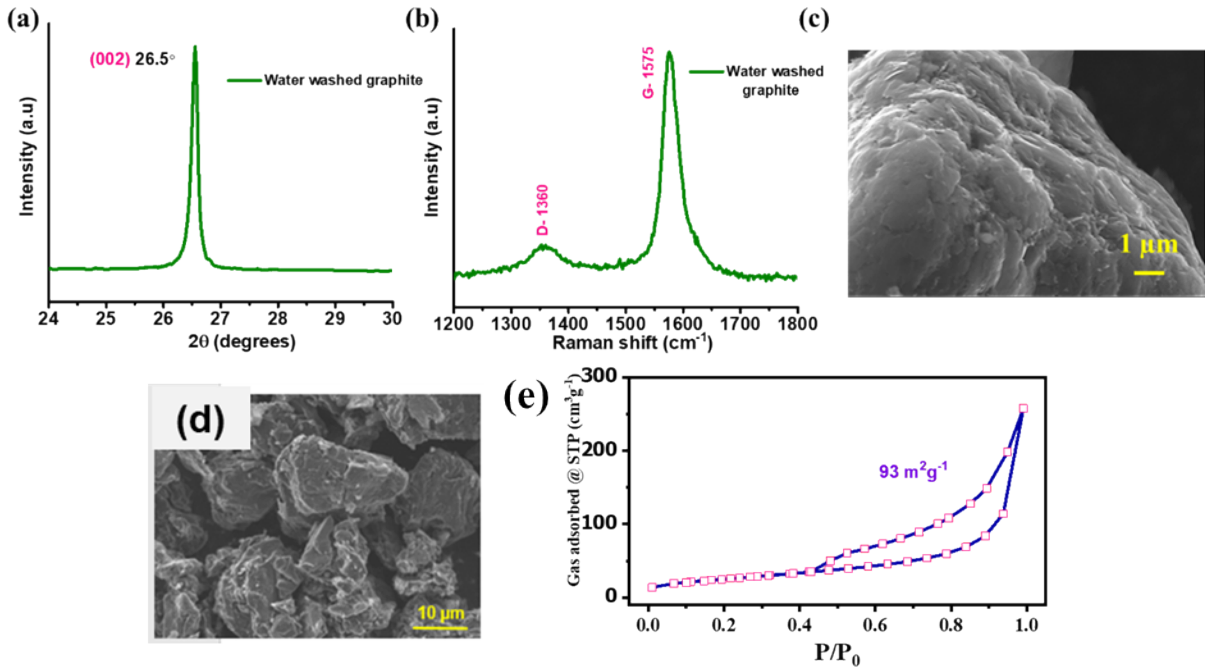

Fig.S2. (a) XRD, (b) Raman spectra, and (c-d) SEM of water-washed graphite, (e) BET adsorption-desorption curve of RRGO.

The textural properties of water-washed graphite were analyzed by SEM, XRD, and Raman spectra, as shown in Fig. S2. Fig.S2a, the XRD clearly shows a sharp (002) peak at 26.5° for a hexagonal crystal lattice of the graphite, and the Raman spectra demonstrates (Fig. S2b) a flat D band at 1360 and G band at 1575 cm^{-1}, I_D/I_G ratio of ~2 thus depicting a more disordered graphitic phase. Fig. S2c,d displays SEM of water-washed graphite having a smooth surface. Fig. S2(e) represents the BET adsorption-desorption curve of RRGO with a BET surface area of 93 m^2 g^{-1}.

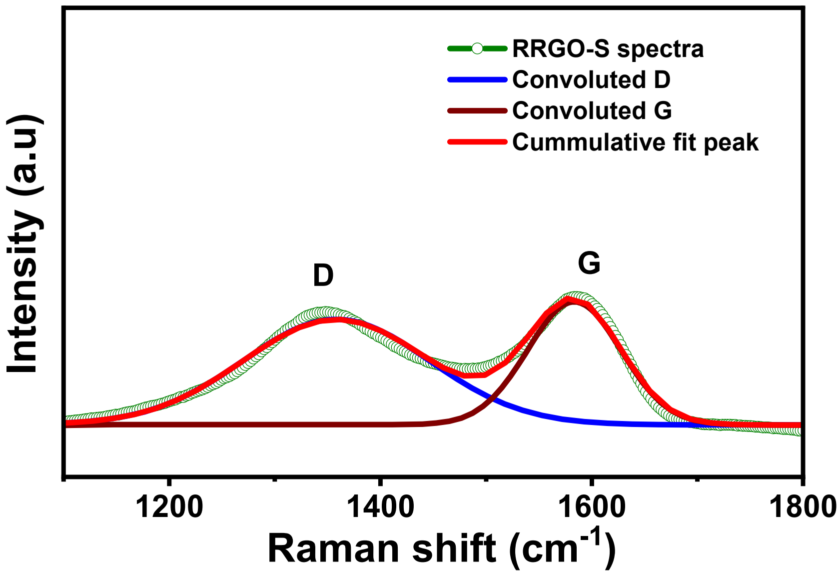

Fig.S3. Deconvoluted peaks of Raman spectra of RRGO-S composite.

The I_D/I_G ratio was calculated by FWHM which is ~2.

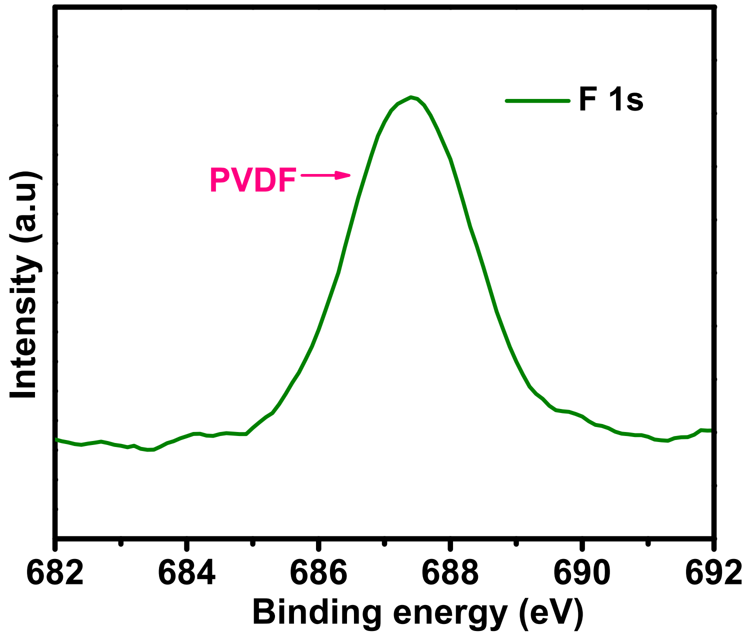

Fig.S4. High resolution F 1s spectra of RRGO-S@CF electrodes**.**

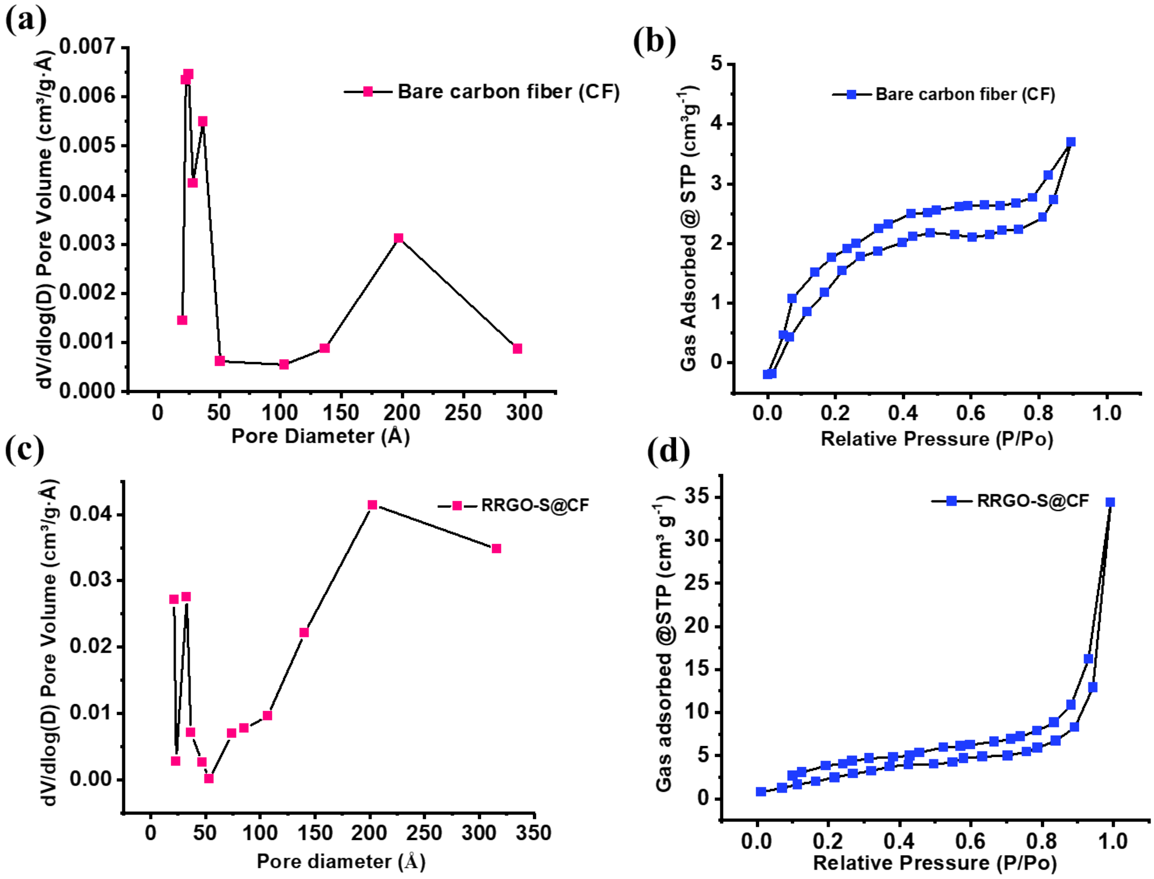

Figure.S5. BET surface area of (a,b) bare carbon fiber (CF) and (c,d) RRGO-S@CF electrode.

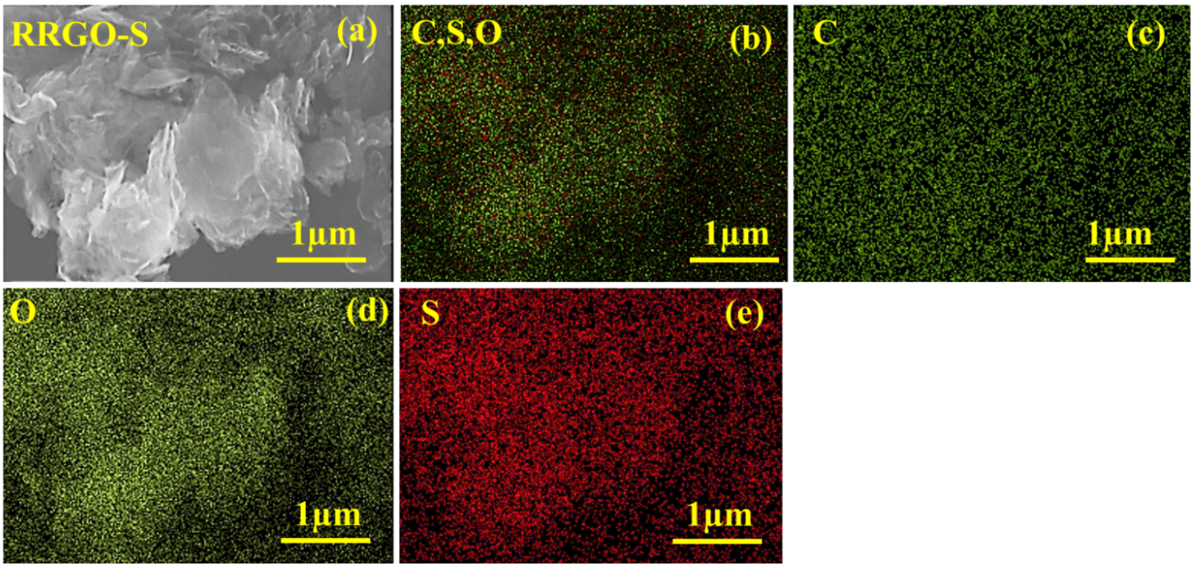

Fig.S6. EDAX images of the RRGO-S composite (a) SEM image (b) C, S, and O, (c) C, (d) O, and (e) S.

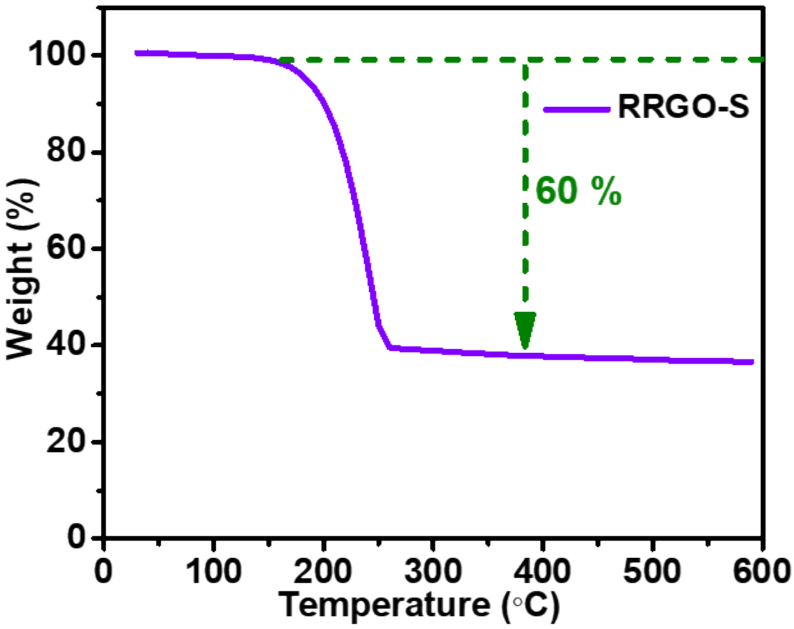

Fig.S7. TGA curves of RRGO-S composite.

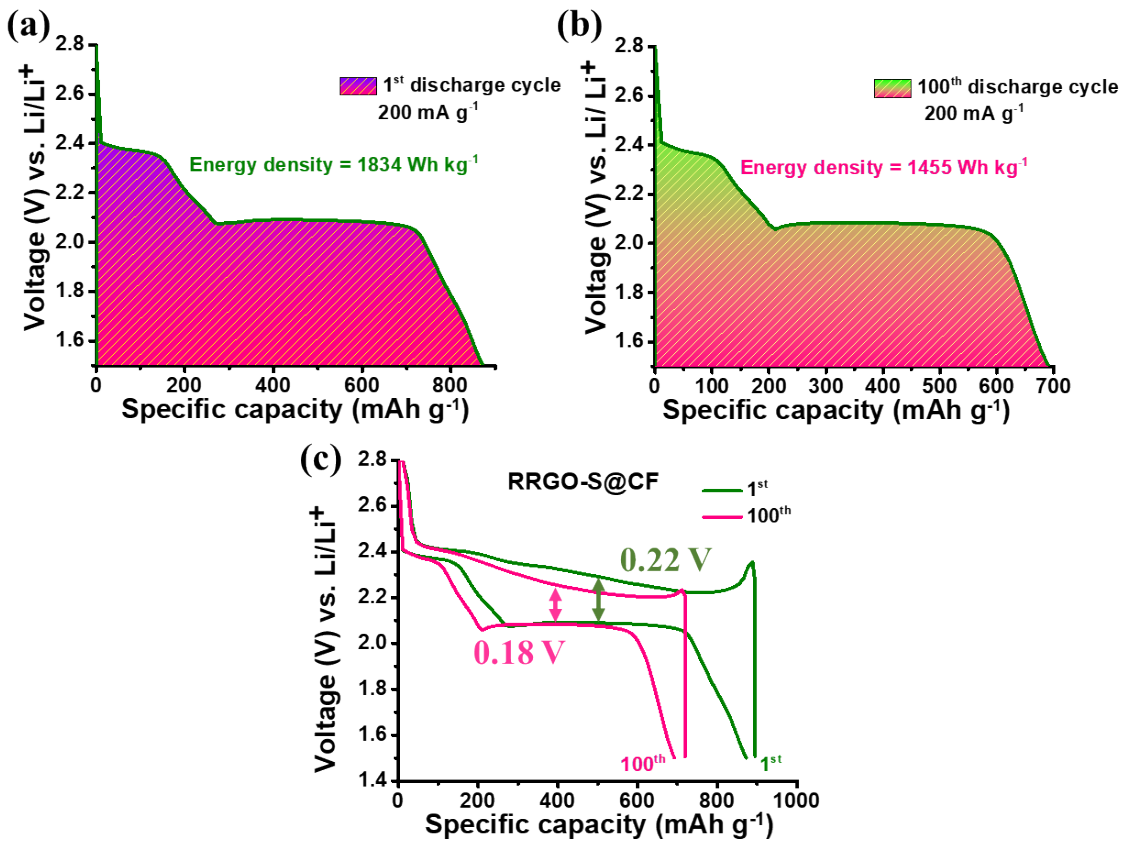

Fig.S8. Energy densities and the hysteresis plot of RRGO-S@CF during1st and 100th cycles at 200 mA g^{-1}.

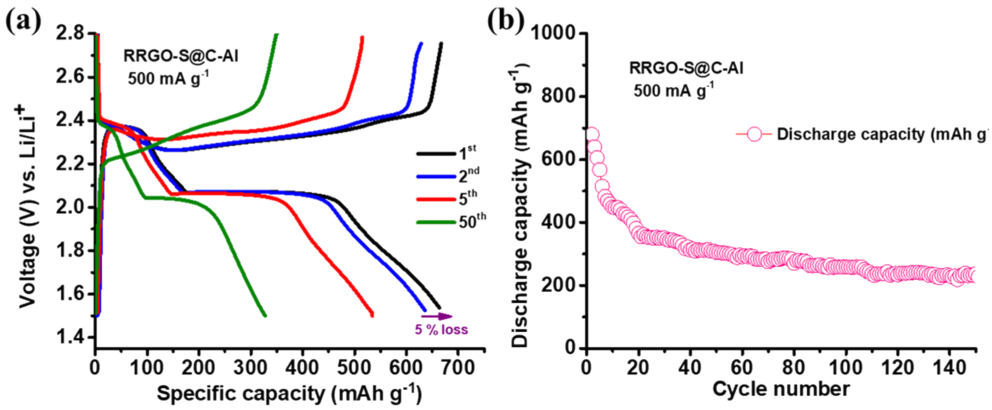

Fig.S9. (a,b) GCD and cycling stability plot of RRGO-S@C-Al at 500 mA g^{-1}.

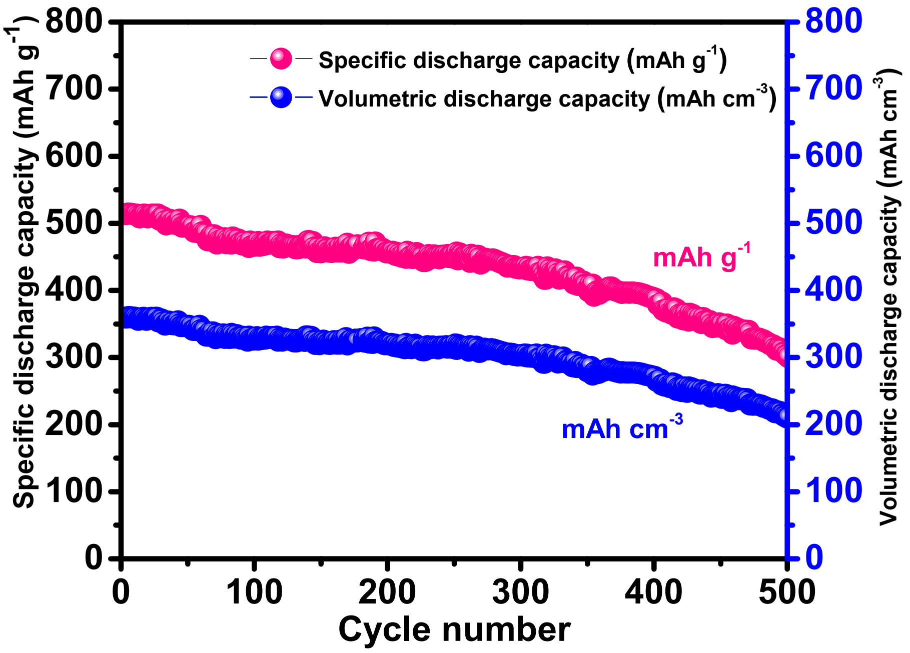

Fig.S10. Cyclic stability plot of specific and volumetric discharge capacities at 800 mA g^{-1}.

The volumetric capacity has been calculated using the formula $C_v = C_g$ x m/t. C_v stands for volumetric capacity (mAh cm^{-3}), C_g stands for gravimetric capacity (mAh g^{-1}), m is the active mass loading in g cm^{-2}, and t is the thickness of the electrode in cm. For the RRGO-S composite electrode, the active mass of the 1.13 cm^2 circular electrodes is 2 mg, and the coating thickness of electrodes is t = 35 x 10^{-4} cm.

The volumetric capacities are ~30% lower than gravimetric capacities.

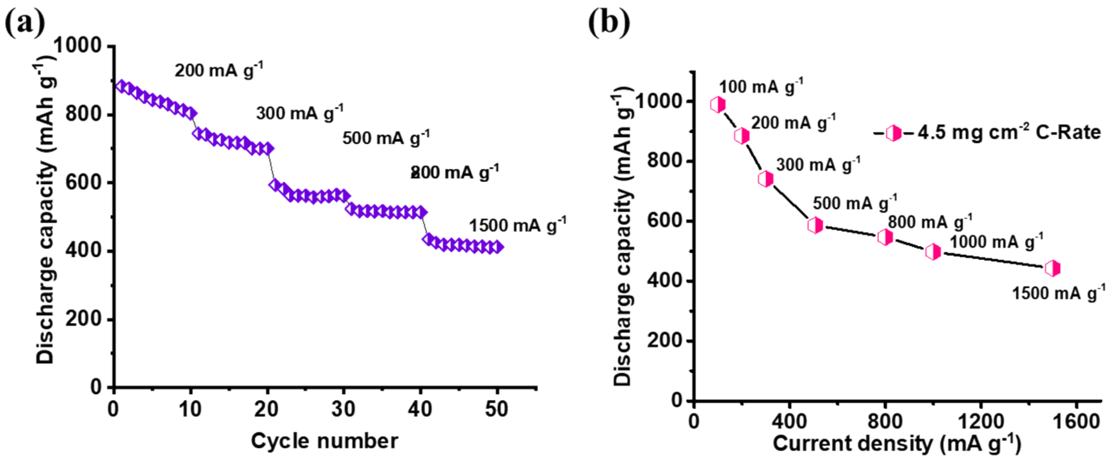

Fig. S11. Rate capability plot at different current densities (a) recorded 10 cycles for each current density (as depicted in Fig.5. (a,b) and (b) 4.5 mg cm^{-2} active material loading of sulfur.

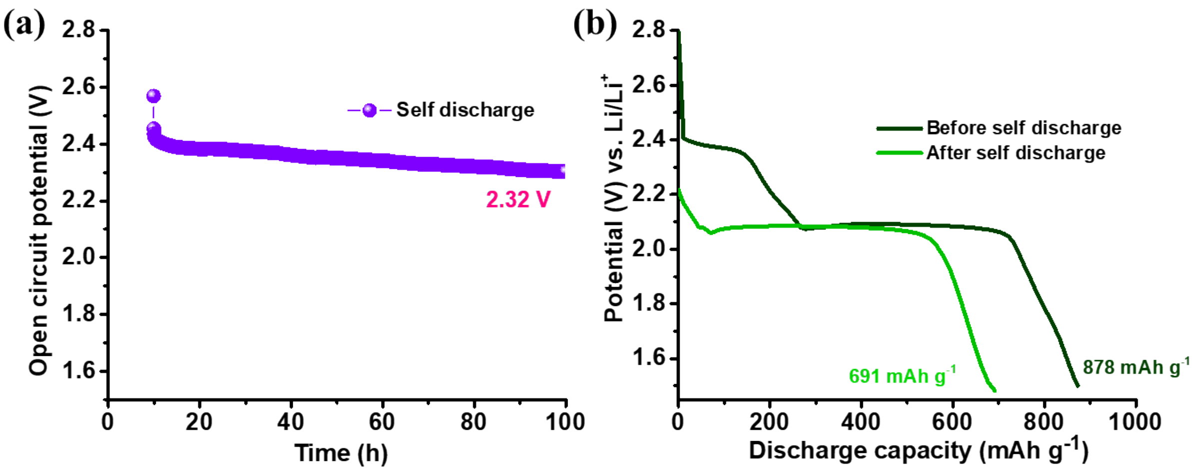

Fig.S12. (a) OCV during storage for 100 h after a full charge and (b) comparison of GCD before and after 100 h of storage (self-discharge). 20 % loss of capacity after storage has been observed.

Table.ST1. The literature survey of RGO-S composite electrode electrochemical performances in terms of capacities, capacity retention, and cycle number in comparison to the present work.

Graphene-sulfur composite	Weight % - Electrode area of S loading	Current density/ C-rate	Initial discharge capacity (mAh g^{-1})	Capacity retention @ cycles	References
GWS (Graphene wrapped sulfur)	-	0.1 C	808 mAh g^{-1}	93.7 % CE @100th cycle	[1]
g-C/S	80 % 1.2-1.5 mg cm^{-2}	0.2 C	1113	89 % @ 300th cycle	[2]
G/S-G (Graphene/ sulfur@graphene)	56%	0.1 C	1036 mAh g^{-1}	60 % @ 200th cycle	[3]
S@GO	92 %	0.2 C 1 C	1301 mAh g^{-1} 704 mAh g^{-1}	86 % @ 115th cycle	[4]
RGO@S	55%	0.1 C 0.2 C	1381 mAh g^{-1} 792 mAh g^{-1}	70 % @150th cycle 84 % @ 300th cycle	[5]
3D-NGS	88 % 1mg cm^{-2}	0.4 C	792 mAh g^{-1}	93 % @ 145th cycle	[6]
NGS/S	80 %	1 C	1140 mAh g^{-1}	72 % @ 500th cycle	[7]
NanoS@G	-	0.2 C	1400 mAh g^{-1}	51 % @ 100th cycle	[8]

Leaf like GO/S	70 % 2.7 mg cm^{-2} 4 mg cm^{-2}	0.5 C	600 mAh g^{-1} 600 mAh g^{-1}	70 % @ 600th cycle 60 % @ 600th cycle	9
SG/S Spent graphite/sulfur composite	78 % ~ 1 mg cm^{-2}	0.2 C 0.5 C	1377 mAh g^{-1} 765 mAh g^{-1}	70 % @ 500th cycle	10
S-KB / Spent graphite interlayer	60 % 0.7-1.2 mg cm^{-2}	1 C	968	~ 65 % 500th cycle	11
<u>Our work</u> RRGO-S@CF (Recycled RGO-S composite onto 3D-carbon fiber)	**60 % ~ 2mg cm^{-2} ~ 4.5 mg cm^{-2}**	**(0.03 C of theoretical capacity) 50 mA g^{-1} (C/3 of theoretical capacity) 500 mA g^{-1} (C/2 of theoretical capacity) 800 mA g^{-1} 500 mA g^{-1}**	**1113 mAh g^{-1} 552 mAh g^{-1} 535 mAh g^{-1} 575 mAh g^{-1}**	**81 % @ 150th cycle 73 % @ 400th cycle 81% @ 150th cycle**	**-**

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