Supporting Information

Scotch-tape-like exfoliation of graphite assisted with elemental sulfur and graphene/sulfur composites for high-performance lithium-sulfur batteries

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Experimental section

Synthesis of graphene oxide. Graphene oxide (GO) preparation started by exfoliating expandable graphite power (100 mesh) using a modified Hummers' method [S1, S2] at 1000 °C in forming gas for 60 s. In a typical reaction, 1.0 g graphite, 0.5 g $NaNO_3$, and 23 mL H_2SO_4 were stirred together in an ice bath until homogenized. After 3 g $KMnO_4$ was slowly added to the solution while stirring, the solution was transferred to a 40 ^{o}C water bath, stirred for about 2 h to form a thick paste. 100 mL water was added and stirred for 1 h, and then 2 mL H_2O_2 (30 wt. % aqueous solution) was added and stirred for 2 h. Inorganic anions and other impurities were removed through 15 washing cycles that included centrifugation, discarding supernatant liquid, and resuspending the solid in an aqueous mixture using stirring and ultrasonication. After drying, the loose and brown GO powder was obtained.

Table S1. Electronegativity of elements and corresponding contact angle of sulfur.

Element	Electronegativity	Contact angle of sulfur (°)
F	3.98	NaF: 67.5
Cl	3.16	NaCl: 40.1
Br	2.96	NaBr: 34.4
I	2.66	NaI: 23.8
O	3.44	SiO_2: 57.2
C	2.55	Graphene: 4.3
S	2.58	–

Table S2. Comparison of a set of electrochemical performance of carbon/sulfur composites for lithium-sulfur batteries.

Composites	S content (wt.)	Specific capacity (mAh g^{-1})				Cycle stability (mAh g^{-1})	Coulombic efficiency	Ref.
		0.1C [a]	0.5C	1C	2C			
S coating on graphene sheets	73%	1502	935	850	570	100th, 615 @ 1C	96.1%	Our work
GO/S [b]	66%	1000	735	550	370	50th, 954 @ 0.1C	96.7%	S3
mGO/PEG/S [c]	70%	960	590	–	–	100th, 520 @ 0.2C	–	S4
Sandwich-type Nafion/rGO/S	70%	–	647	505	–	50th, 750 @ 0.1C	–	S5
CHS/S [d]	70%	1160	1071	750	520	100th, 974 @ 0.5C	94%	S6
CMK-3/PEG/S	70%	1320	–	–	–	200th, 800 @ 0.1C	–	S7
PEDOT:PSS/ CMK-3/S [e]	50%	1140	–	–	–	150th, 600 @ 0.5C	96–98%	S8
BMC/S [f]	50%	–	–	995	–	100th, 550 @ 1C	–	S9
ACF/S [g]	50%	1000	–	–	–	80th, 800 @ 0.09C	95%	S10
HCF/S [h]	75%	–	950	–	–	150th, 730 @ 0.2C	99% with $LiNO_3$	S11
DCNT/S [i]	40%	930	–	–	–	100th, 680 @ 0.25C	96%	S12

[a] 1C=1675 mA g^{-1}; [b] GO: graphene oxide; [c] mGO: mildly oxidized graphene oxide; [d] CHS: Carbon hollow sphere; [e] PEDOT:PSS: Poly(3,4-ethylenedioxythiophene)-poly(styrene sulfonate); [f] BMC: bimodal mesoporous carbon; [g] ACF: activated carbon fiber cloth; [h] HCF: hollow carbon nanofiber; [i] DCNT: disordered carbon nanotubes.

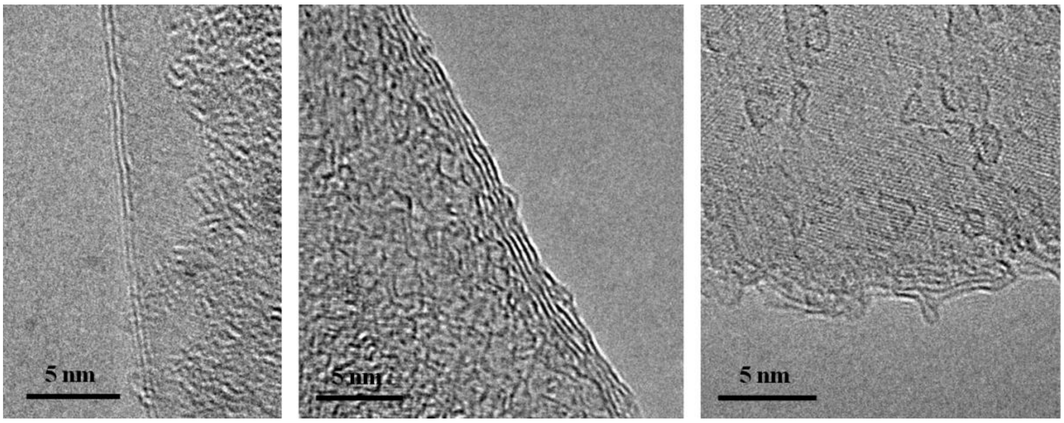

Figure S1. HRTEM images of the folded edge of freestanding graphene sheet obtained by ball milling for 6h.

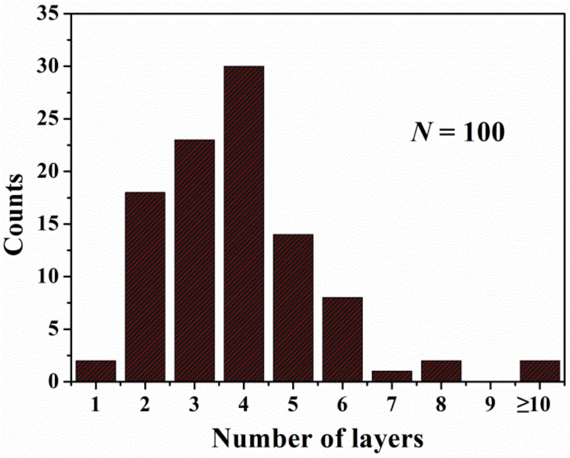

Figure S2. The thickness statistics of graphene sheets *via* ball mill for 6h by analyzing a large number of HRTEM images for the flake edges and AFM height profiles.

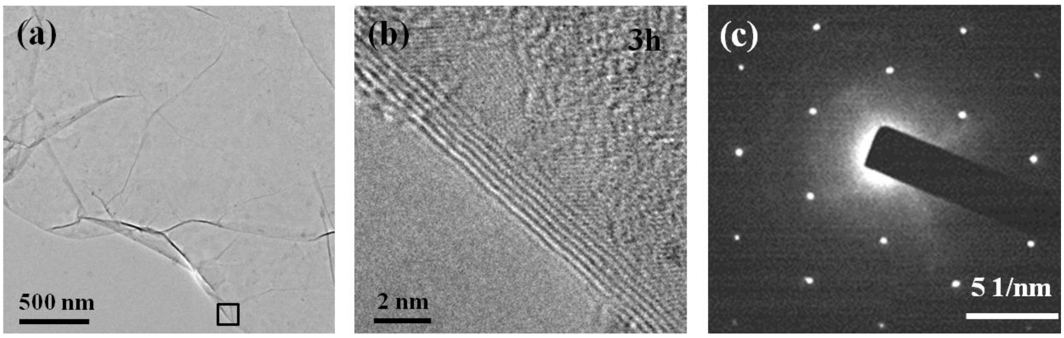

Figure S3. (a) A representative TEM image of sulfur-assisted graphene by ball milling for 3h. (b) HRTEM image of the folded edge of freestanding graphene sheet, acquired from the black square in (a). (c) SAED pattern indicating the single-crystal structure for as-prepared graphene sheets.

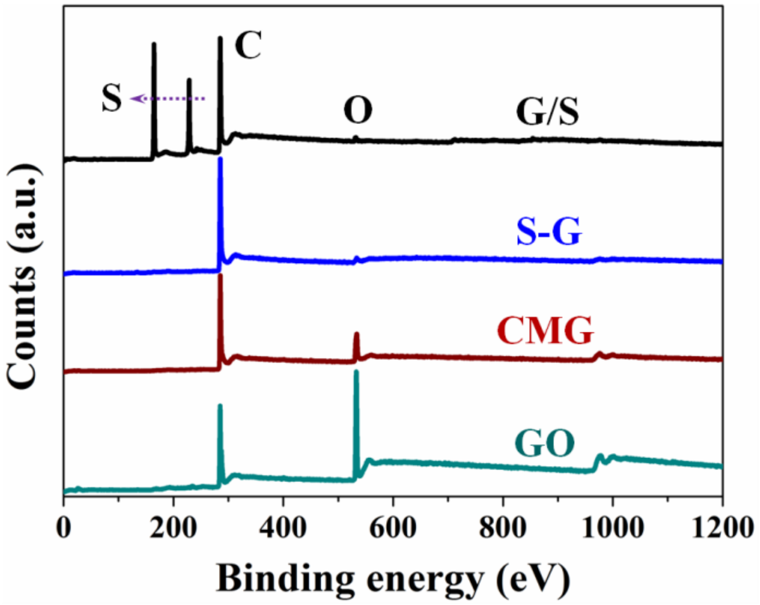

Figure S4. XPS survey spectra of graphene oxide (GO) prepared by Hummer's method, the chemically modified graphite (CMG), as-made sulfur-assisted graphene sheets (S-G), and graphene/sulfur composite (G/S).

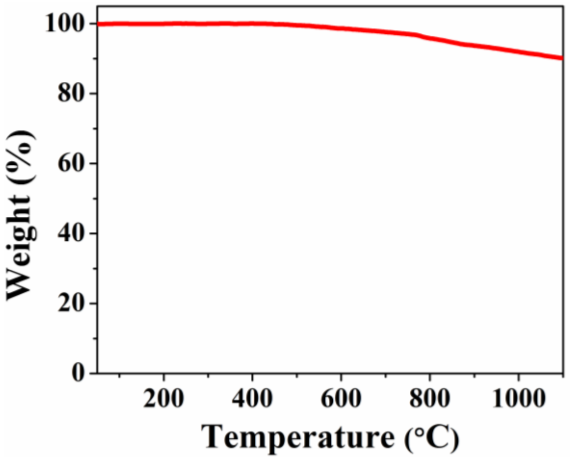

Figure S5. The thermal stability of as-made graphene sheets by ball mill for 6h confirmed by TGA. The measurement was carried out in a flowing N_2 atmosphere. The sample was heated up to 1100 °C at a rate of 2 °C/min. The TGA datum illustrates excellent thermal stability for as-made graphene sheets.

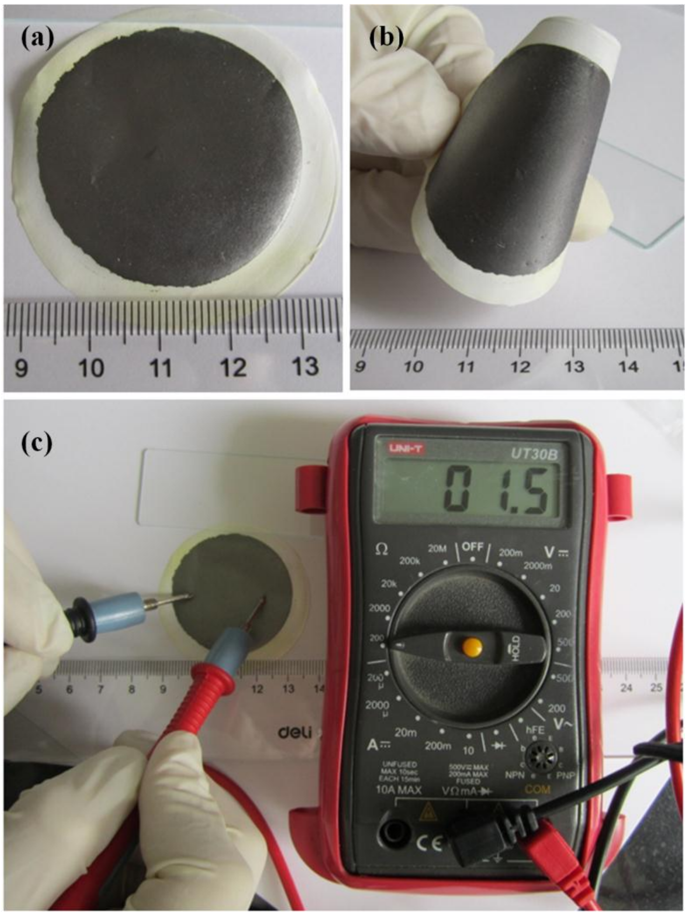

Figure S6. Photographs of a 25-μm-thick wafer-sized (ϕ 4 cm) graphene paper (a), showing outstanding flexibility (b) and extraordinary high electrical conductivity (c).

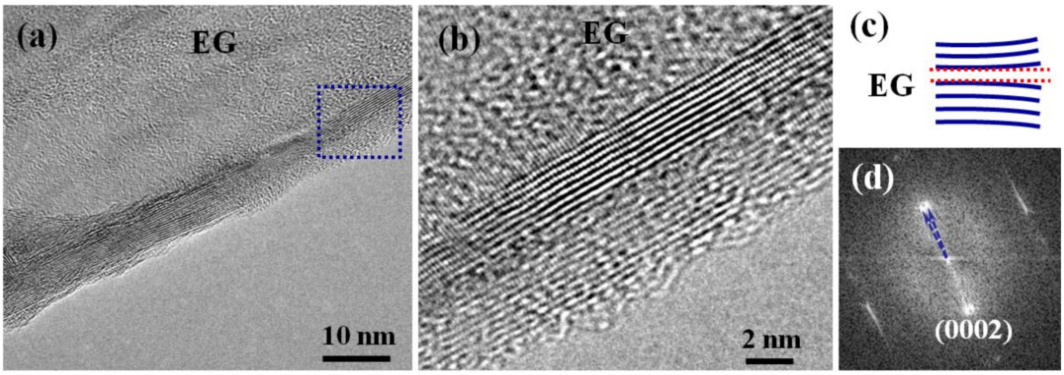

Figure S7. (a–b) HRTEM images of edge-opened graphite (EG) showing cleavage along the basal plane and its structural diagram (c). (f) 2D fast Fourier transform (FFT) taken from (a), blue lines indicate angle of rotation.

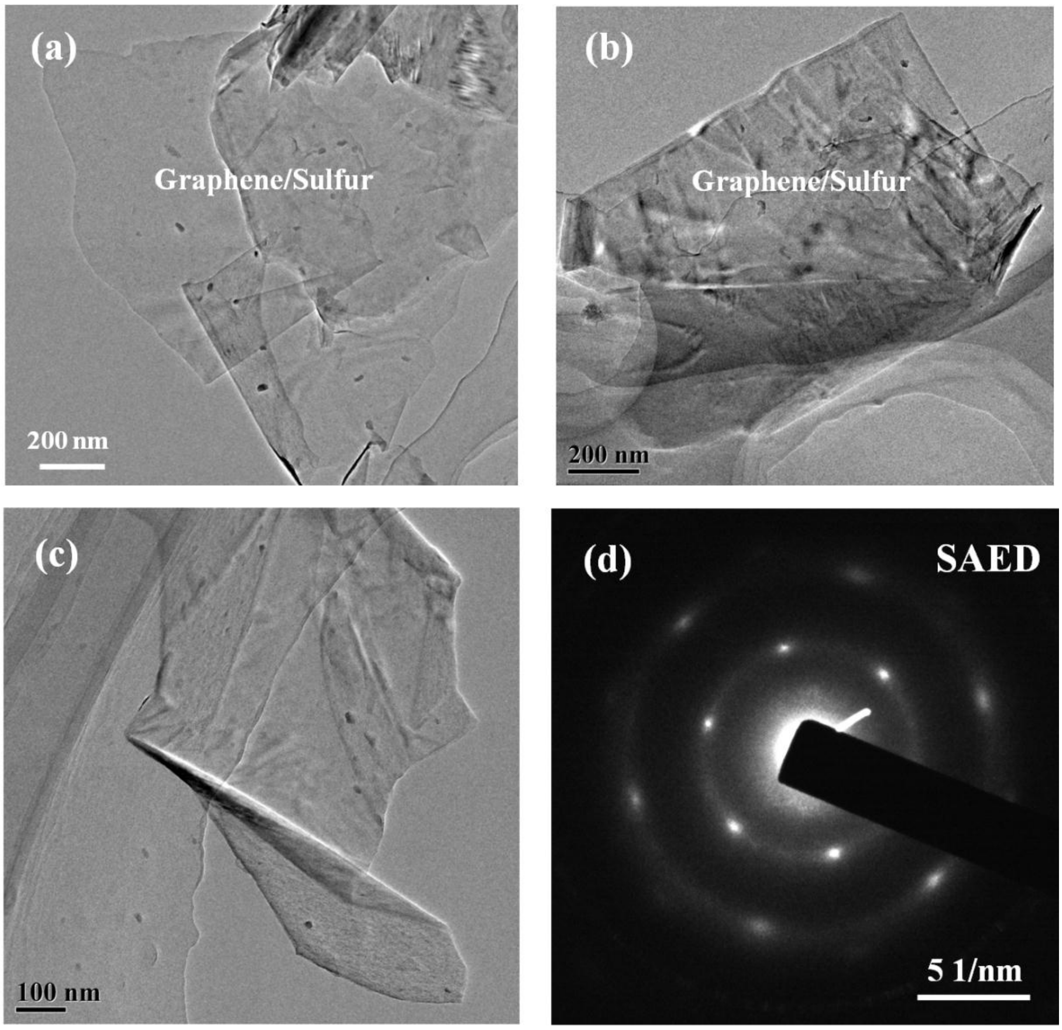

Figure S8. TEM images and SAED pattern of graphene/sulfur composite showing a uniform and thin sulfur layer coating on the graphene sheet.

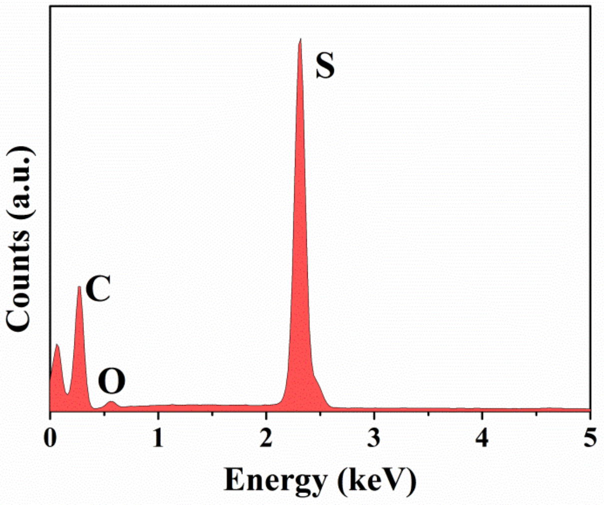

Figure S9. Energy dispersive spectroscopic (EDS) spectrum of graphene/sulfur composite captured for the region shown in Figure 5a.

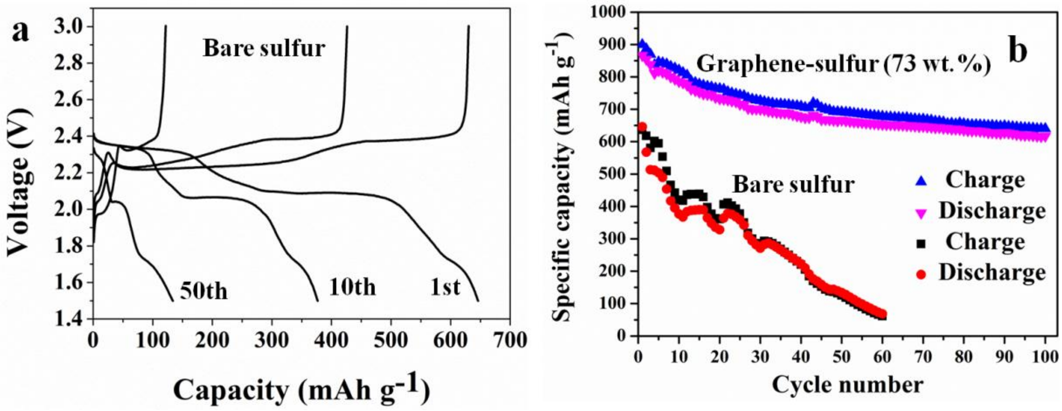

Figure S10. (a) Galvanostatic discharge/charge profiles at the rate of 1C for bare sulfur. (b) Cycling performance of charge/discharge at rate of 1C for graphene/sulfur composite and bare sulfur.

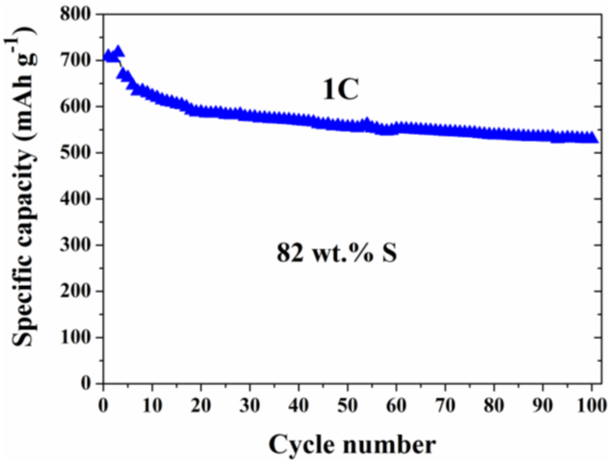

Figure S11. Discharge capacities versus cycle number for the graphene/sulfur composite with 82 wt.% S.

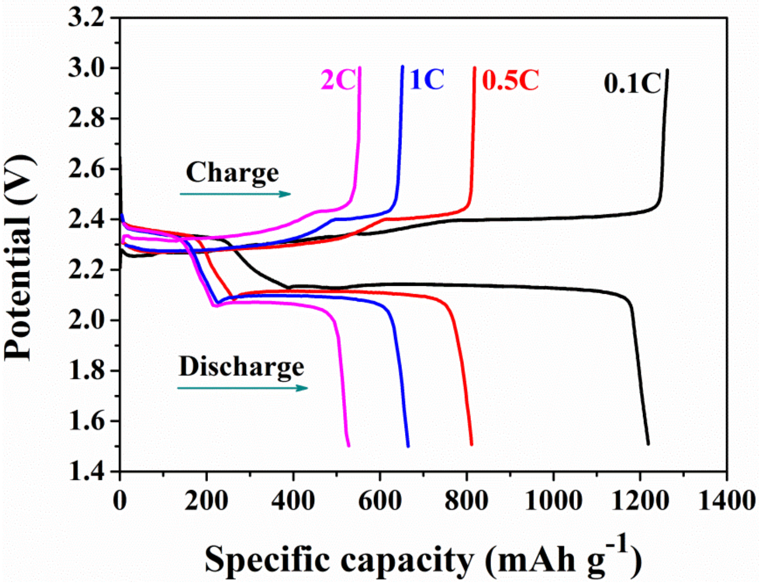

Figure S12. The first cycle charge and discharge voltage profiles of the graphene/sulfur composite with 82 wt.% sulfur at various rates.

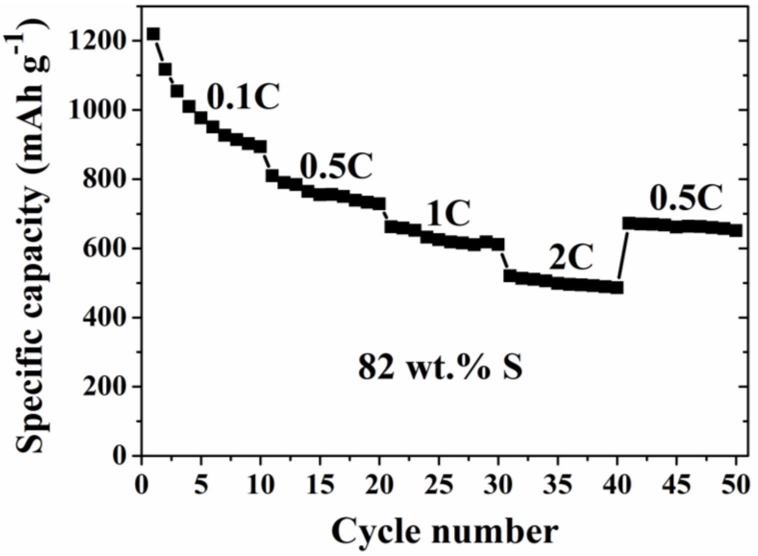

Figure S13. Reversible capacity *vs* current density (rate capability) of graphene/sulfur composite with 82 wt.% sulfur.

References

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