Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Hydrogen-Substituted Graphdiyne/Graphene as sp/sp^2 Hybridized Carbon Interlayer for Lithium−Sulfur Batteries

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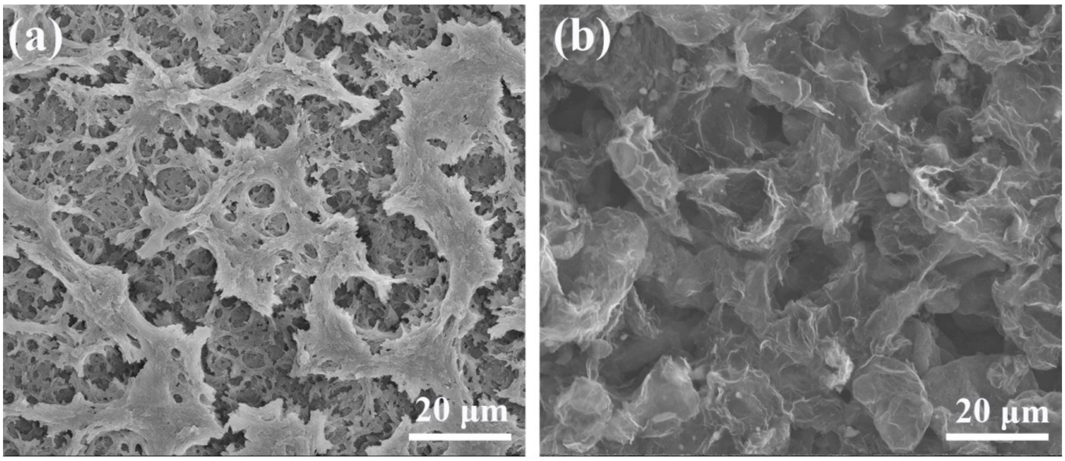

Fig. S1 The morphology of the HsGDY a) and Gra-HsGDY b) by SEM images. Crosslinked structures are observed for the Gra-HsGDY sample, implying better film-forming property.

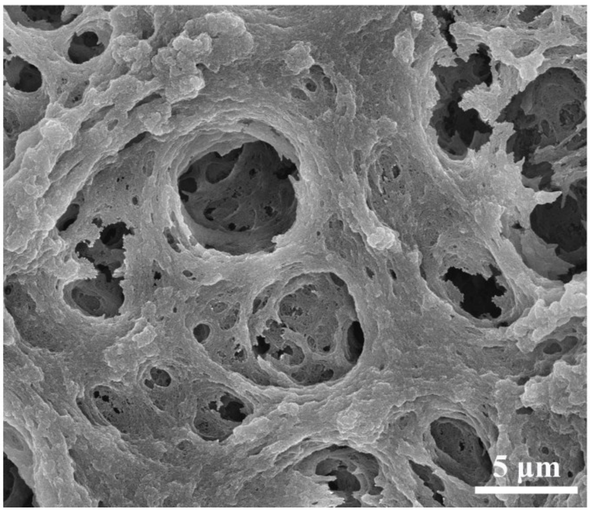

Fig. S2 SEM image of HsGDY.

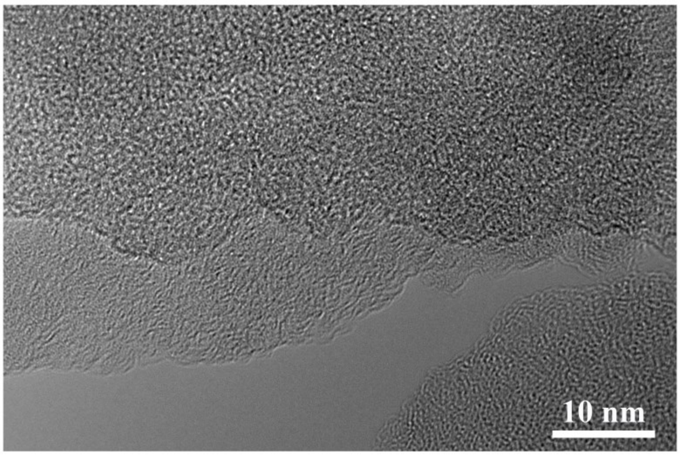

Fig. S3 TEM image of HsGDY.

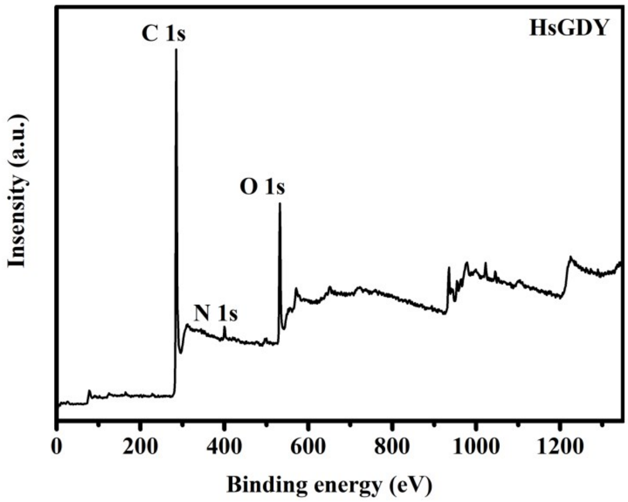

Fig. S4 XPS survey spectra of HsGDY.

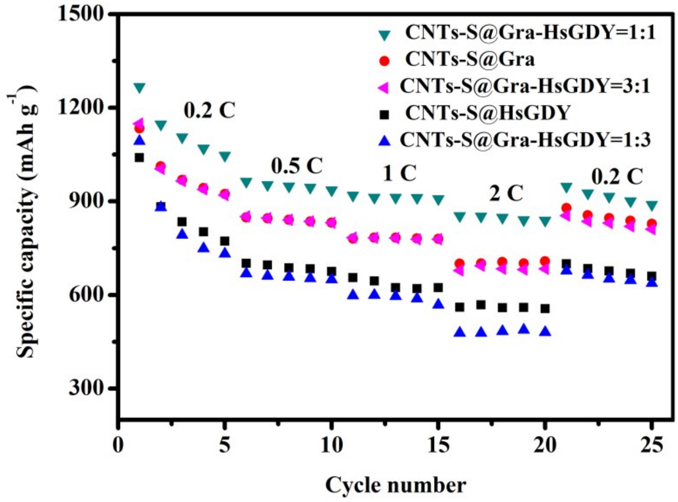

Fig. S5 The rate performances of CNTs-S@Gra-HsGDY with different mass ratios of Gra to HsGDY.

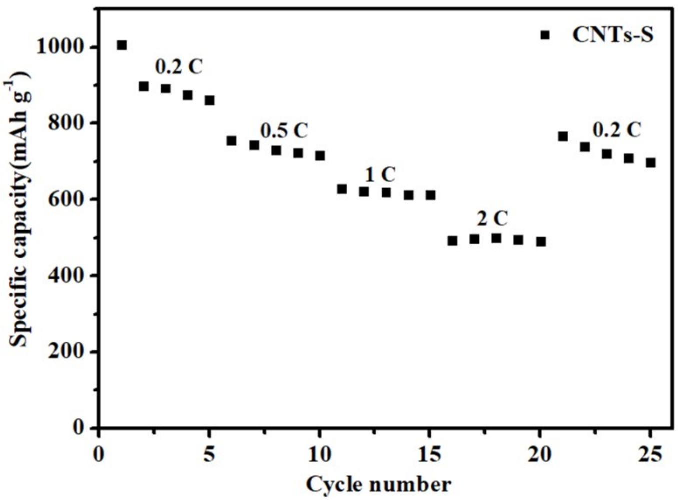

Fig. S6 The rate performance of the CNTs-S cathode at 0.2 to 2 C.

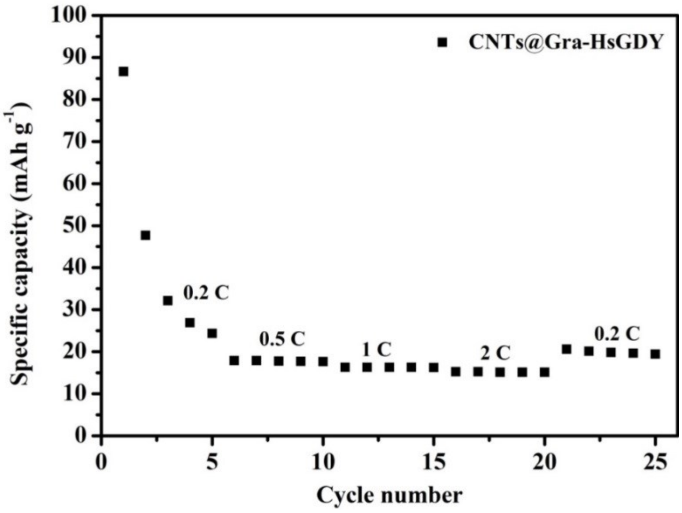

Fig. S7 Rate performance of the CNTs@Gra-HsGDY electrode without any sulfur loading.

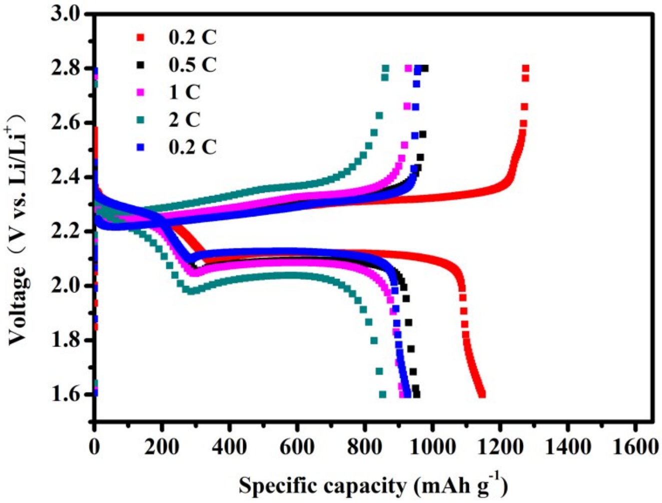

Fig. S8 The galvanostatic discharge-charge plots of CNTs-S@Gra-HsGDY under various current rates.

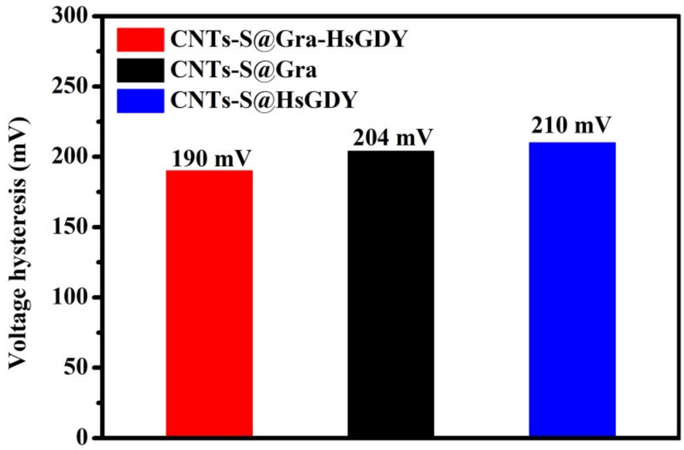

Fig. S9 Comparison of voltage hysteresis of the three samples obtained from the galvanostatic charge-discharge profiles at 0.2 C in Fig. 2d

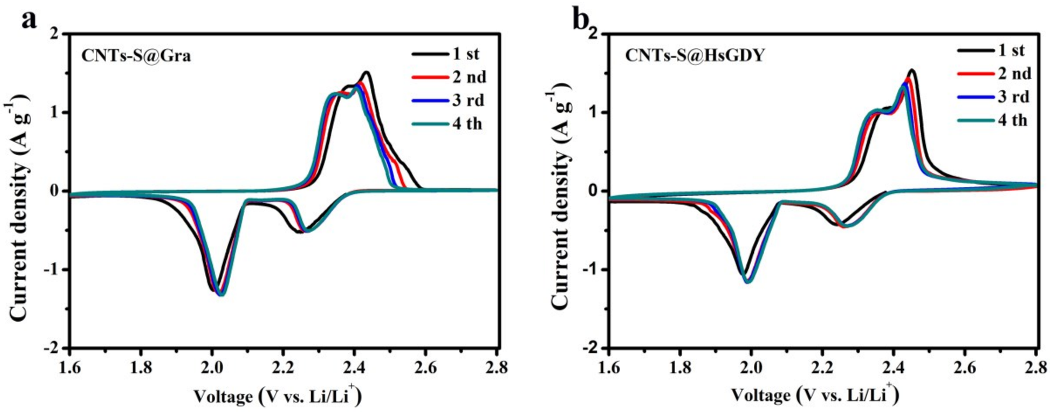

Fig. S10 (a-b) CVs of CNTs-S@Gra and CNTs-S@Gra-HsGDY between 1.6 and 2.8 V for the initial four cycles, respectively, recorded at a scan rate of 0.1 mV s^{-1}.

Table S1 Voltage hysteresis and collection coefficient of the three samples obtained from the CV profiles in Fig. 3d.

Sample	Polarized voltage (ΔV)	Collection coefficient (I_L/I_H)
CNTs-S@ HsGDY	0.38	2.44
CNTs-S@Gra	0.35	2.49
CNTs-S@Gra-HsGDY	0.32	2.68

Table S2 Characteristics of various materials reported in literatures.

Materials	Sulfur loading (mg cm^{-2})	Initial performance		Cycling performance			Ref.
		Rate (C)	Capacity (mAh g^{-1})	Rate (C)	Cycles	Capacity Decay (% per cycle)	
Sulfonated GO	1.2~1.6	0.2	1240	0.5	250	0.18	1
MWCNT	1.1	0.1	1100	0.5	150	0.22	2
Graphene	1.21	0.1	1100	0.1	100	0.283	3
Super P	0.7	0.2	1151	0.2	200	0.185	4
rGO	1.1	0.2	1070	0.2	200	0.185	5
Porous Carbon	1.5	0.2	1166	0.5	200	0.20	6
Acetylene Black	1.0-1.3	0.5	939	1	200	0.20	7
PPy nanotube	2.5-3	0.2	1101	0.5	300	0.11	8
High-flux GO	1-1.2	0.5	1182	0.5	100	0.29	9
B-RGO	1.5	0.1	1227	0.1	300	0.15	10
S/CoOOH	1.0-1.3	0.1	1199	1	500	0.09	11
CNT@TiO_{2-x}-S	2.2	0.1	1187	0.2	100	0.3	12
$CoSe_2$/GO	1.0	0.2	1331	6	500	0.1	13
Gra-HsGDY	1.2	0.2	1267	1	500	0.089	This work

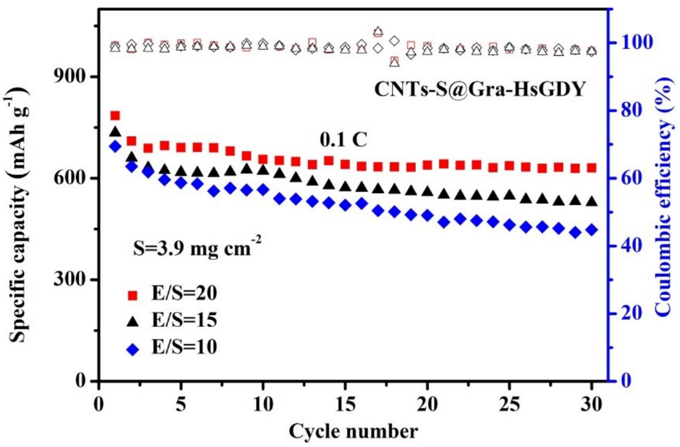

Fig. S11 Cycling stability of CNTs-S@Gra-HsGDY at 0.1 C with sulfur loading of ~3.9 mg cm^{-2} under various E/S ratios.

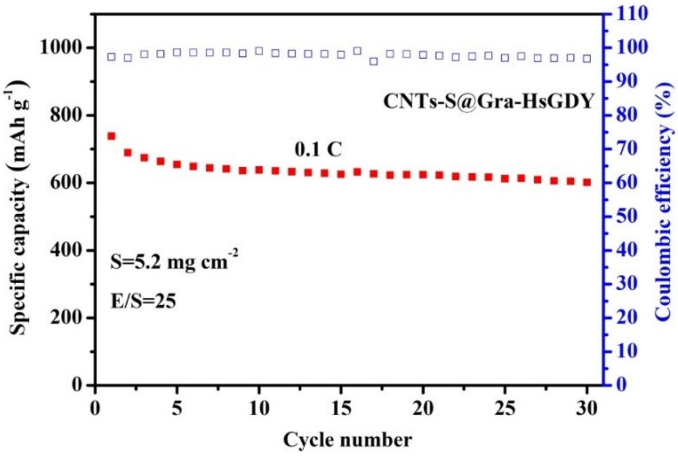

Fig. S12 Cycling peformance of the CNTs-S@Gra-HsGDY at 0.1 C with a higher sulfur loading of 5.2 mg cm^{-2} for 30 cycles.

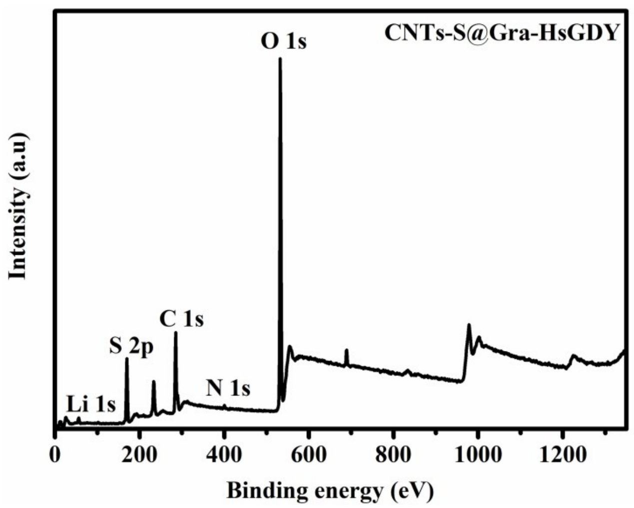

Fig. S13 XPS survey spectra of CNTs-S@Gra-HsGDY during the fifth cycle.

Fig. S14 LiPSs adsorption tests of HsGDY. 0.5 mM Li_2S_8 solution was prepared by mixing Li_2S and S_8 powder in DMSO. Afterwards, HsGDY (3 mg) were added into 1 mL Li_2S_8 (0.5 mM) solution and kept standing for 48 hours.

Table S3 Adsorption energy of Li_2S_n (n=2,4,6,8) molecule on HsGDY flake, wherein "I" means the Li_2S_n molecule adsorbed on the sp hybridized carbon of HsGDY, and "II" represents the Li_2S_n molecule adsorbed on the sp^2 hybridized carbon of HsGDY.

	E_{total}	E_{HsGDY}	E_{Li2S2}	E_{Li2S4}	E_{Li2S6}	E_{Li2S8}	E_{ads} (eV)
HsGDY-Li_2S_2-I	-46032.845	-45460.596	-570.954				-1.295
HsGDY-Li_2S_2-II	-46032.644	-45460.596	-570.954				-1.094
HsGDY-Li_2S_4-I	-46589.189	-45460.596		-1127.452			-1.141
HsGDY-Li_2S_4-II	-46589.078	-45460.596		-1127.452			-1.030
HsGDY-Li_2S_6-I	-47145.076	-45460.596			-1683.143		-1.337
HsGDY-Li_2S_6-II	-47144.979	-45460.596			-1683.143		-1.240
HsGDY-Li_2S_8-I	-47700.379	-45460.596				-2238.573	-1.209
HsGDY-Li_2S_8-II	-47700.099	-45460.596				-2238.573	-0.929

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