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Metal-Organic Framework Derived NiS_2 Hollow Spheres as Multifunctional Reactors for Synergistic Regulation of Polysulfides Confinement and Redox Conversion

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Experimental section

1. The preparation process of C HSs

In a typical procedure, after carbonization of Ni-MOF under 450 ℃ for 6 h, the obtained Ni/C HSs powder was etched in 2 M HCl aqueous solution under 150 ℃ for 24 [1]. After cooling to room temperature, the C HSs power was collected and washed with deionized water and ethanol for 3 times, respectively.

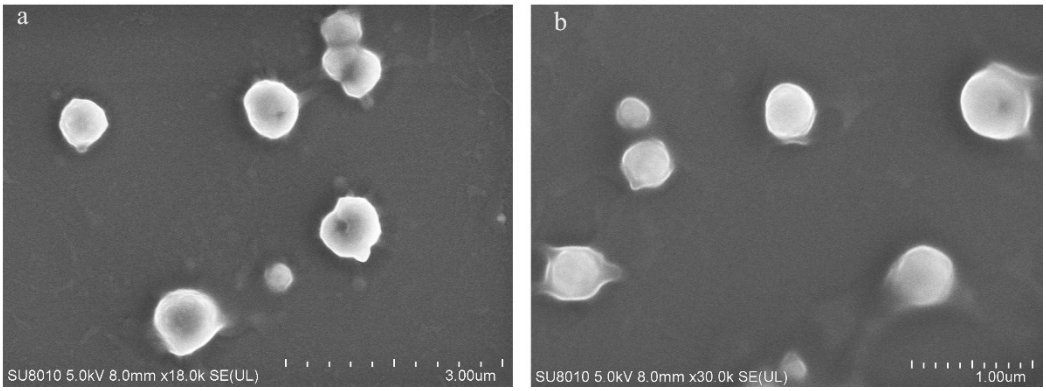

Fig. S1 SEM images of Ni-MOF.

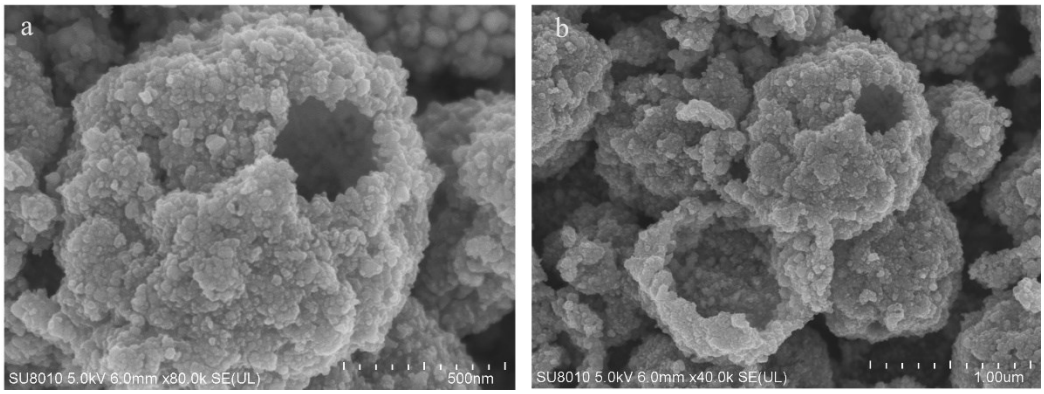

Fig. S2 SEM images of NiS_2/C HSs.

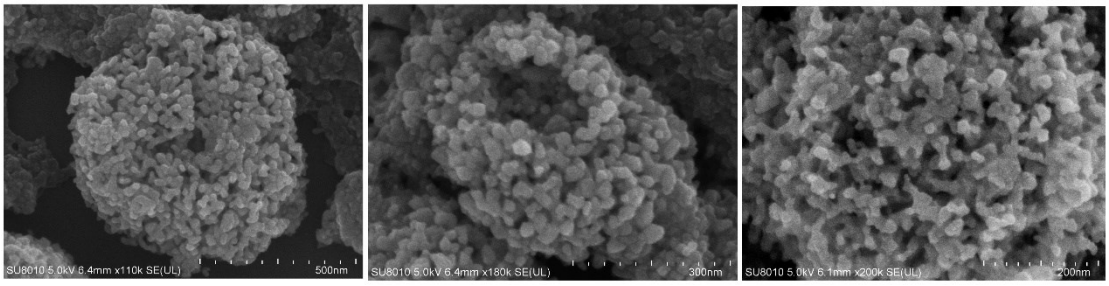

Fig. S3 SEM images of C HSs.

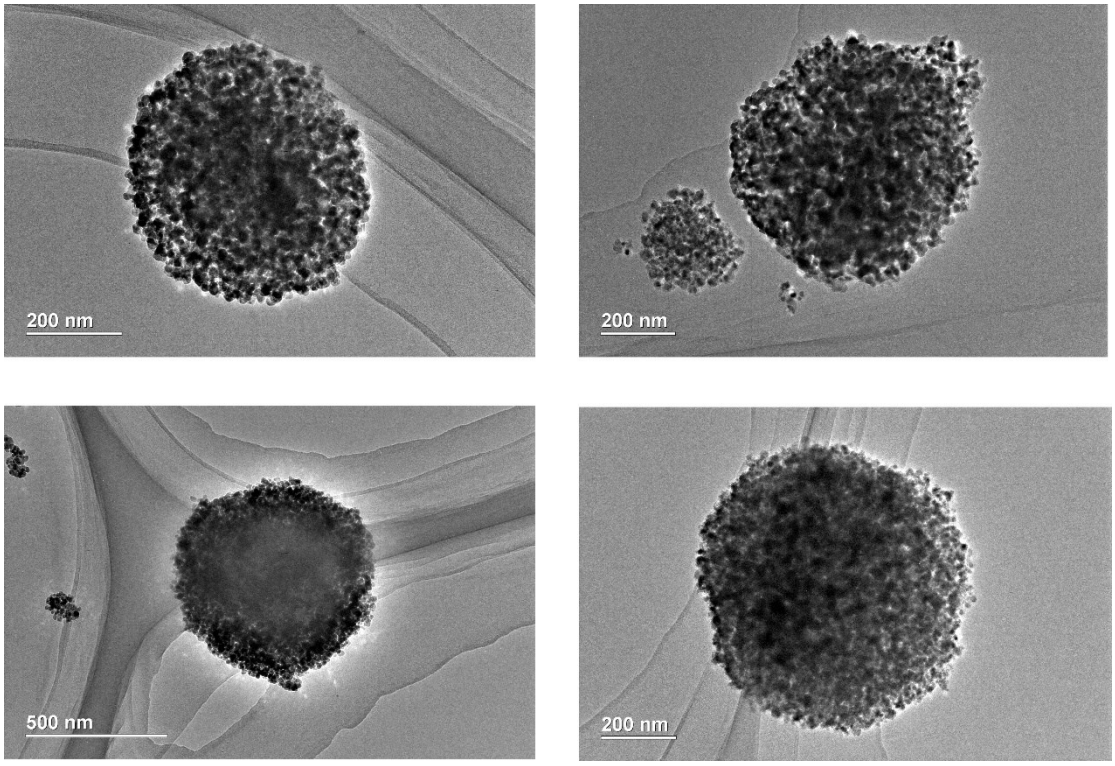

Fig. S4 TEM images of NiS_2/C HSs.

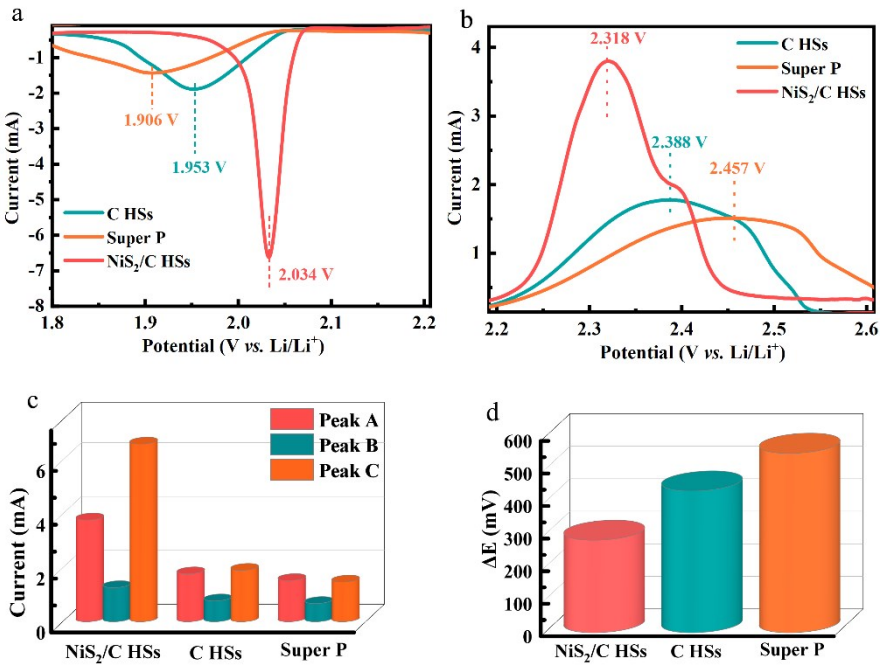

Fig. S5 The enlarged CV profiles of peak C (a) and peak A (b) of Li-S batteries with different cathodes. The current of the peak A, peak B and peak C of the CV profiles (c). The potential difference between peak A and peak C (d).

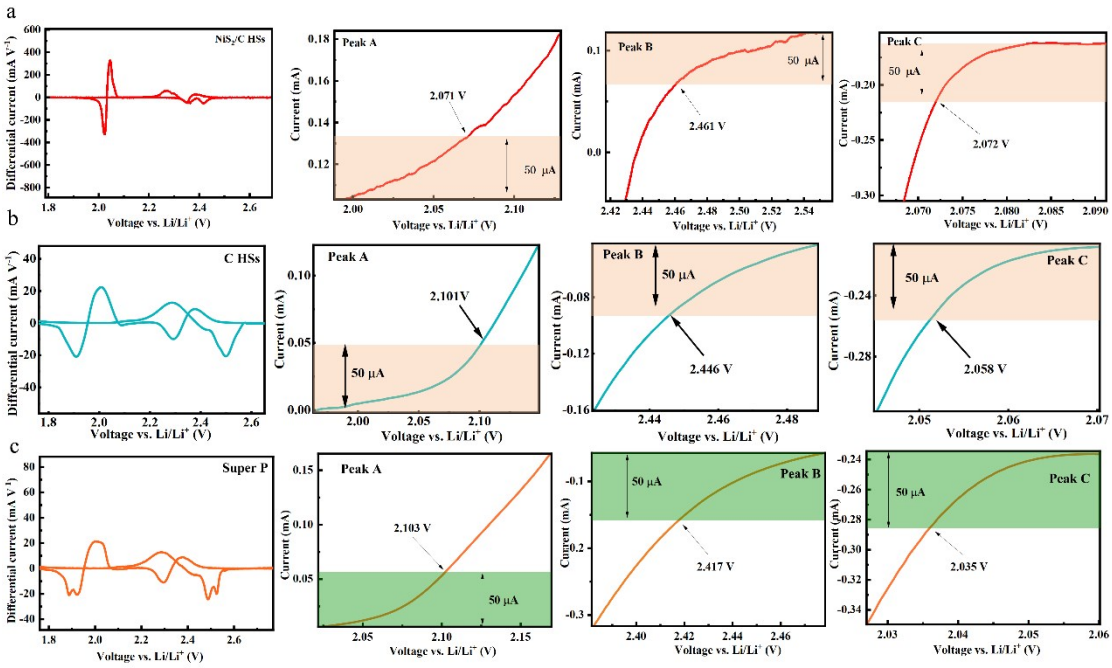

Fig. S6 Onset potential measurement of Li-S batteries with NiS_2/C HSs (a), C HSs (b) and Super P (c). The baseline current are obtained from the values before the cathodic and anodic peaks, where the value of current remain almost unchanged and the value of dI/dV≈0 [2]. The onset current is 50 μA beyond the baseline current (50 μA more negative than baseline current for cathodic peak and 50 μA more positive than baseline current for anodic peak) [2].

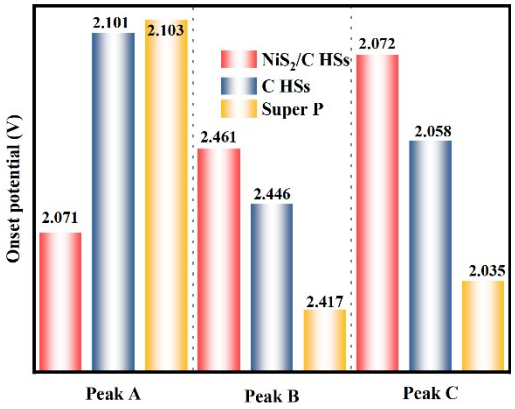

Fig. S7 The onset potential of Li-S batteries with NiS_2/C HSs, C HSs and Super P.

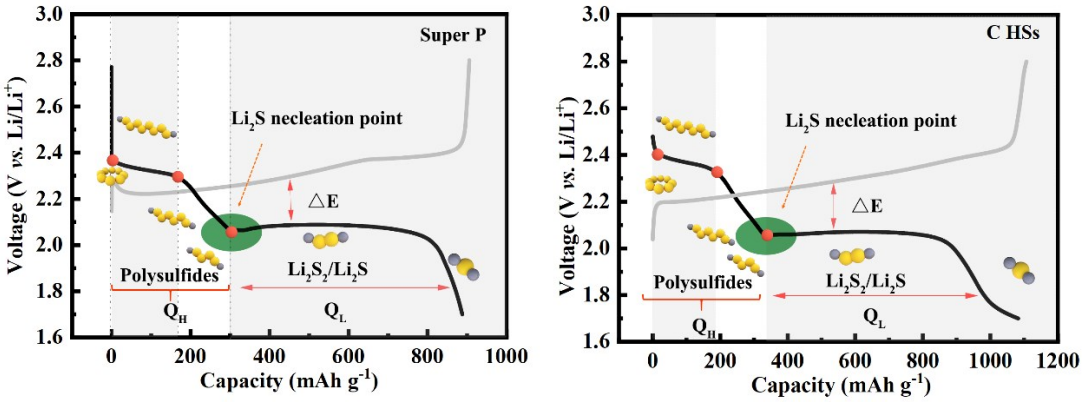

Fig. S8 The charge/discharge profiles of the cells with Super P and C HSs.

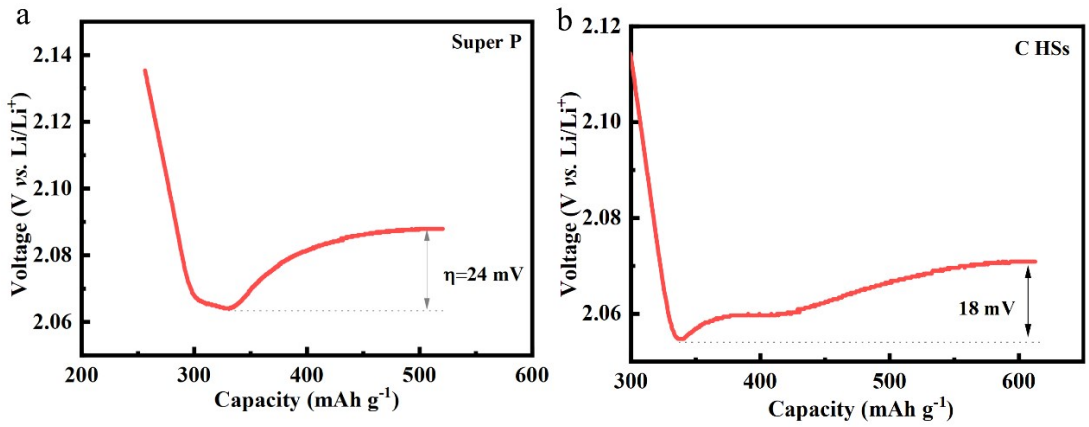

Fig. S9 The nucleation overpotential of Li_2S of the cells with Super P (a) and C HSs (b).

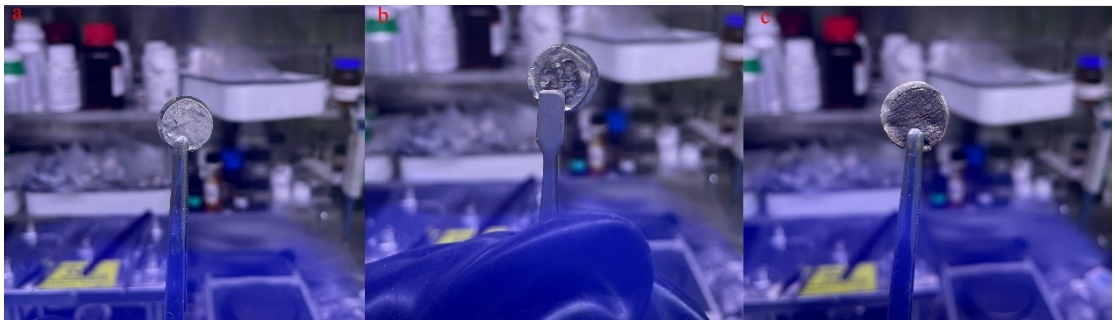

Fig. S10 The digital photographs of Li anode disassembled from the Li-S cells with NiS_2/C HSs (a), C HSs (b) and Super P (c).

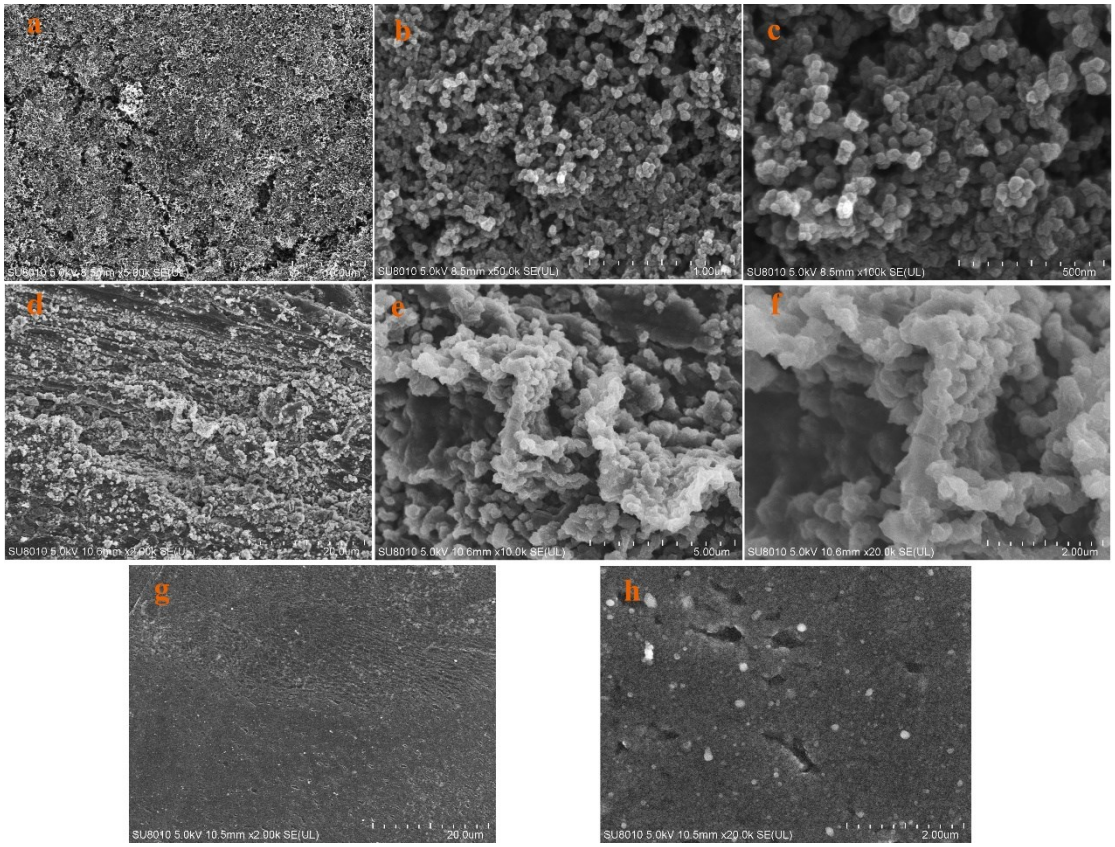

Fig. S11 SEM images of Li anode of the cells with C HSs (a-c), Super P (d-f), and NiS_2/C HSs (g, h).

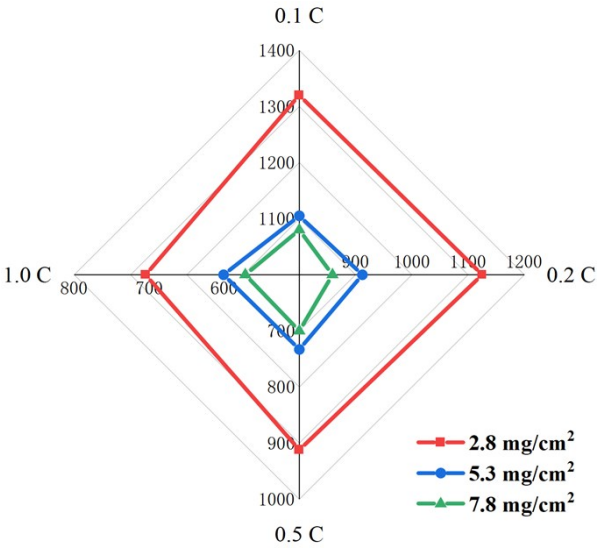

Fig. S12 The rate performance of the cells with NiS_2/C HSs under 2.8 mg/cm^2, 5.3 mg/cm^2 and 7.8 mg/cm^2 sulfur loadings.

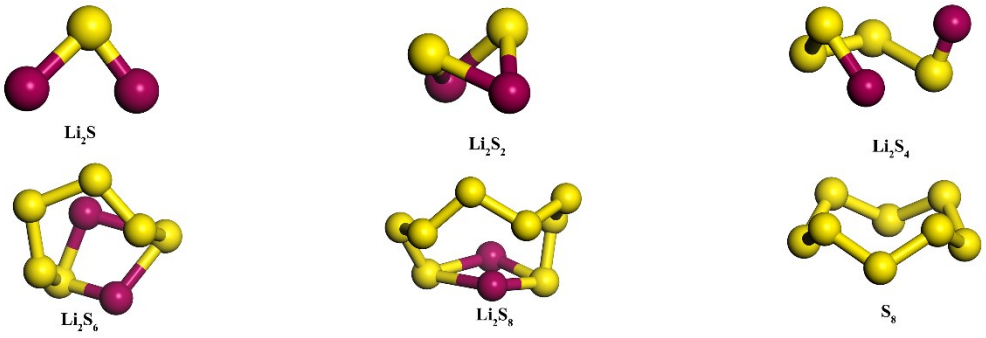

Fig. S13 The optimized geometries of Li_2S, Li_2S_2, Li_2S_4, Li_2S_6, Li_2S_8 and S_8.

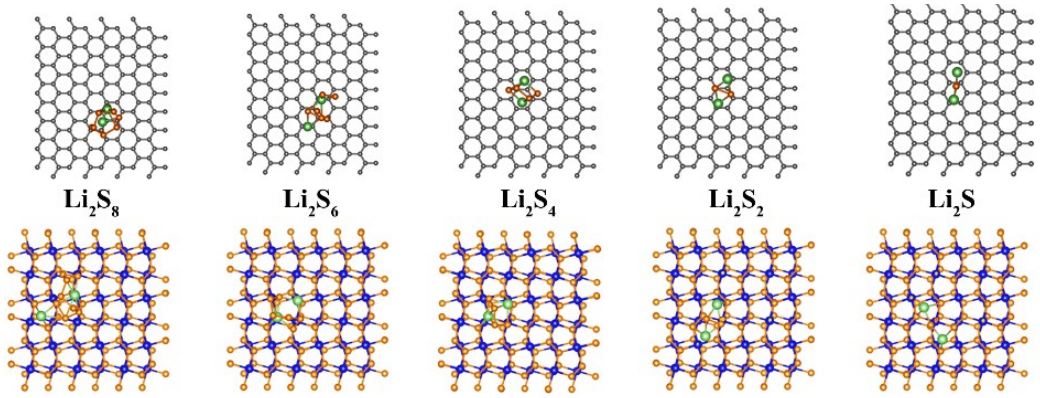

Fig. S14 Optimized configurations of polysulfides adsorption on graphene and NiS_2.

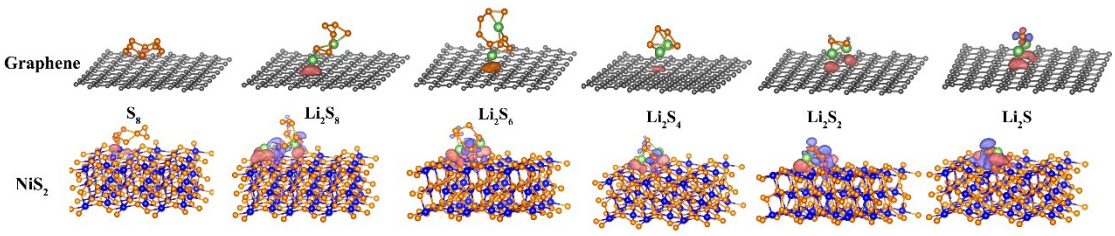

Fig. S15 The charge density difference of polysulfides adsorbed at graphene and NiS_2, in which, blue denoted the decrease of charge and red denoted charge accumulation.

Table. S1 The electrochemical performance of various cathodes of Li-S batteries.

Cathode	Current rate (C)	Area loading (mg cm^{-2})	Areal capacity (mAh cm^{-2})	Ref.
NiS_2/C HSs	0.2	7.8	6.6	This work
NiS_2/C HSs	0.2	5.3	4.8	This work
NiS_2/C HSs	0.2	2.8	3.1	This work
Co,N-G@CNT/S	0.2	4.2	4.4	[3]
S@CPZC	0.2	9.2	7.1	[4]
S@CPZC	0.2	5.1	4.7	[4]
H-S@Co-CNCs	0.2	4.0	2.7	[5]
CNT-NC@GC/S	0.2	3.36	3	[6]
S@H-LDH	0.2	1.5	1.3	[7]
CNF/LPS/CNT	0.2	7.64	3.5	[8]
S/CoNi@PNCFs	0.2	4.5	4	[9]
Sb_2Se_{3-x}/rGO	0.2	3	2.9	[10]
NC/MoS_3-S NBs	0.2	5.5	4.5	[11]
COF-MF@S	3.43 mA cm^{-2}	4.1	3.7	[12]
N-PC@uCo/S	0.2	5.9	4.8	[13]
3WO_3-WS_2/S	0.5	5.0	4.4	[14]
3WO_3-WS_2/S	0.5	10.0	4.7	[14]
ZnS_{1-x}-CC/S	0.1	5.0	3.5	[15]
$NiCoO_4$/CNF/S	0.1	6.9	4.6	[16]

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