Supporting Information

Insights into the dynamic catalytic effect of metal sulfide with prominent lithiation process in the application of Li-S batteries

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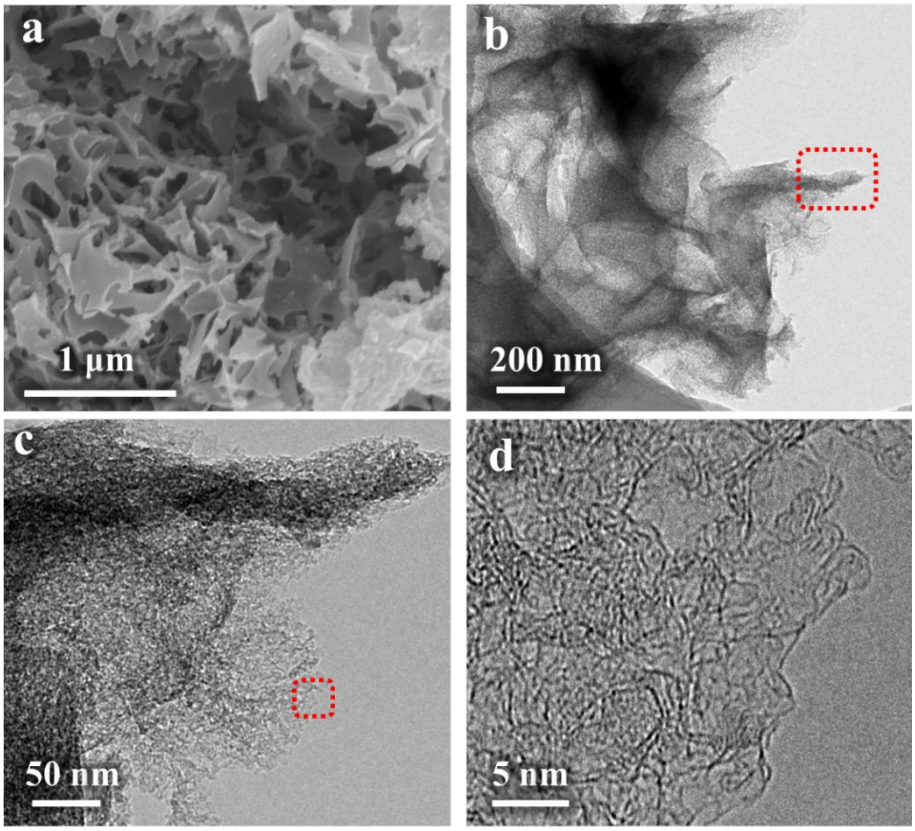

Figure S1 (a) SEM and (b-d) TEM images of HPGC.

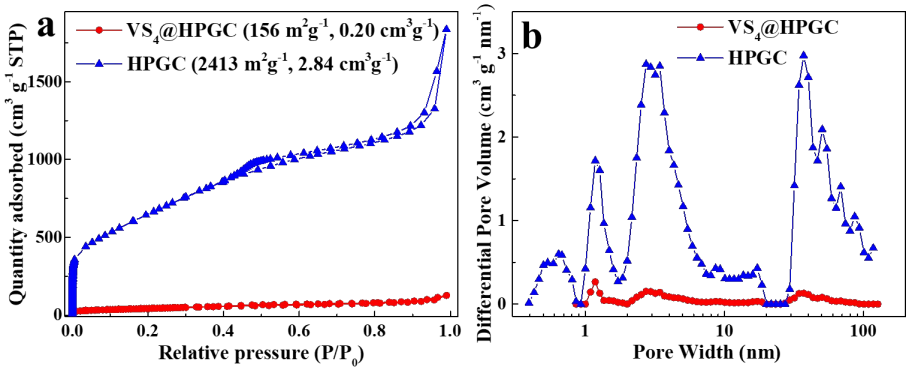

Figure S2 (a) N_2 adsorption/desorption isotherms and (b) pore-size distributions of VS_4@HPGC and HPGC.

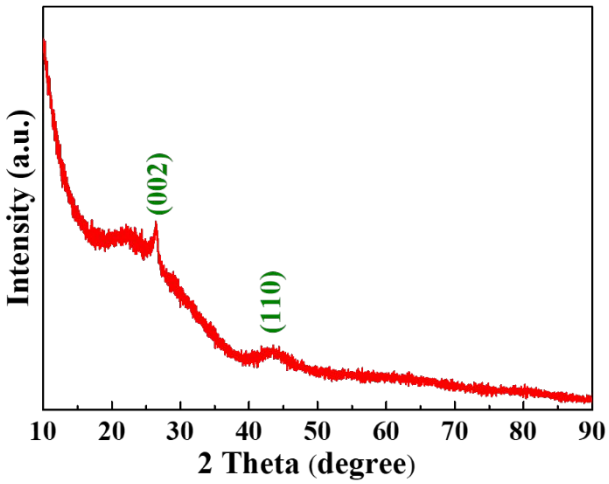

Figure S3 XRD pattern of HPGC.

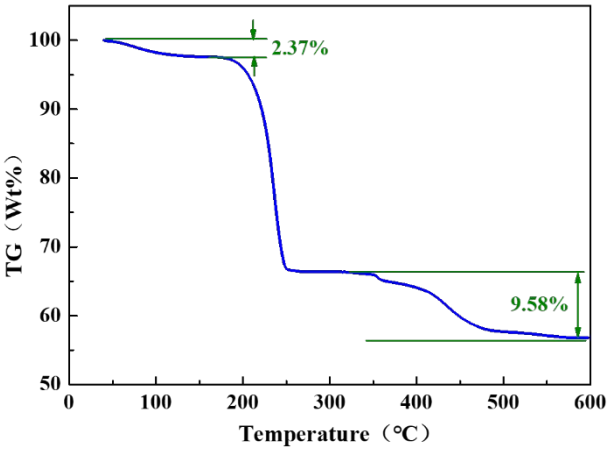

Figure S4 TG curve of VS_4@HPGC.

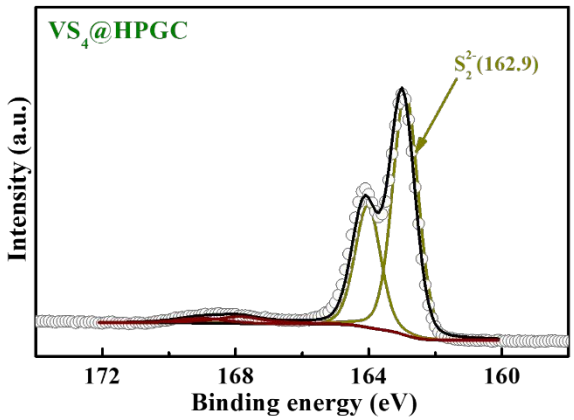

Figure S5 S 2p XPS spectra of VS_4@HPGC.

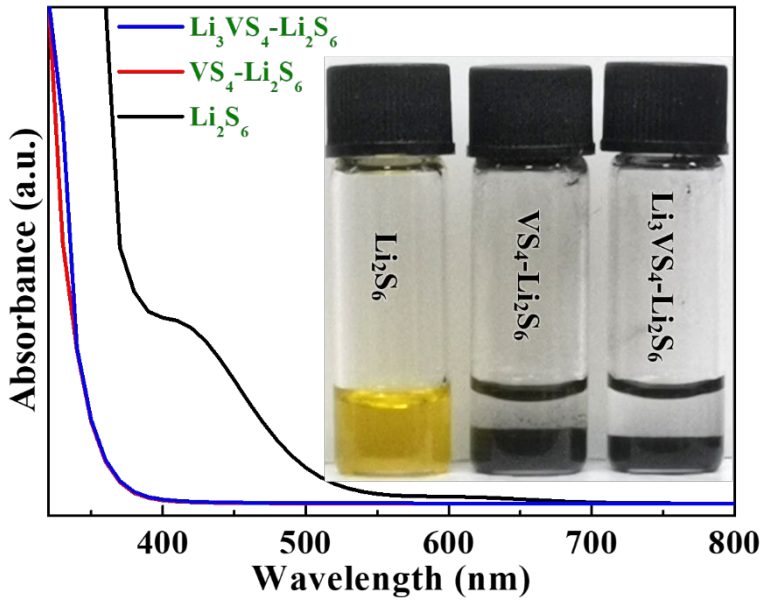

Figure S6 Photo images and UV-vis spectroscopy measurements of Li_2S_6 absorption experiments with VS_4@HPGC and Li_3VS_4@HPGC.

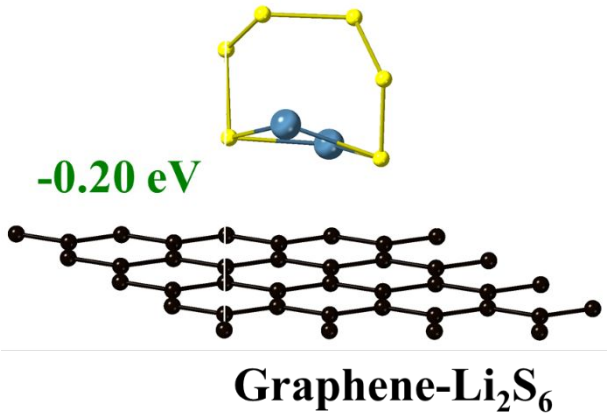

Figure S7 Optimized adsorption configuration for Li_2S_6 on graphene surface by DFT calculation.

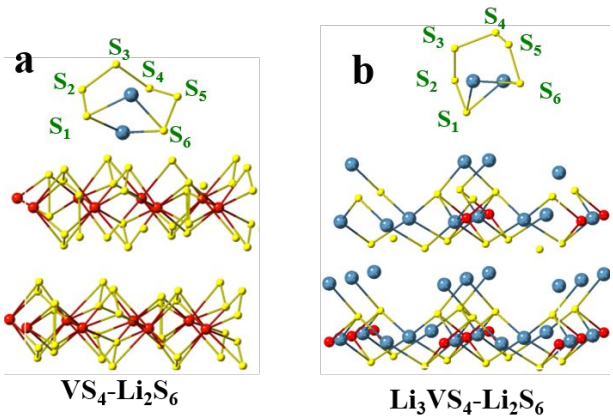

Figure S8 Index of sulfur atoms in Li_2S_6 with optimized adsorption configuration on (a) VS_4 (-111)

surface, (b) Li_3VS_4 (001) surface by DFT calculation.

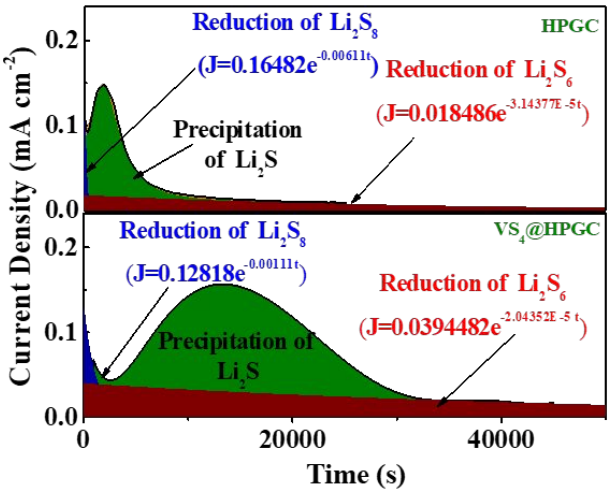

Figure S9 Fitting results of current vs. time curve on HPGC and VS_4@HPGC.

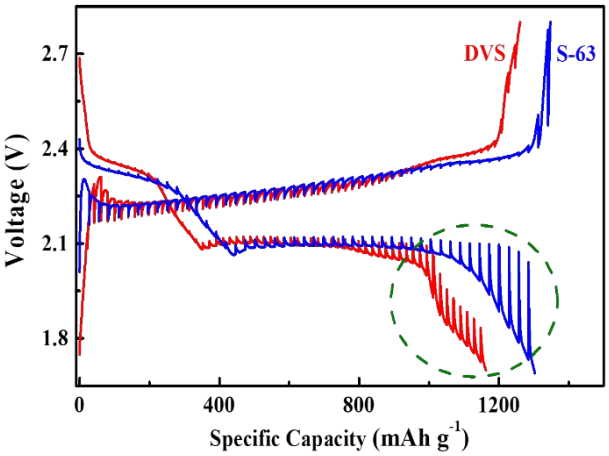

Figure S10 GITT curves of DVS and S-63.

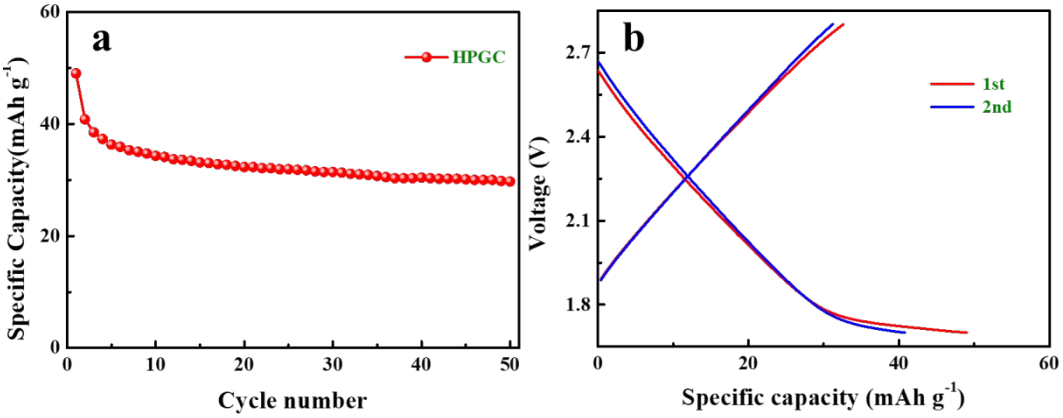

Figure S11 Cycling performance of HPGC and the charge/discharge curves in the first three cycles.

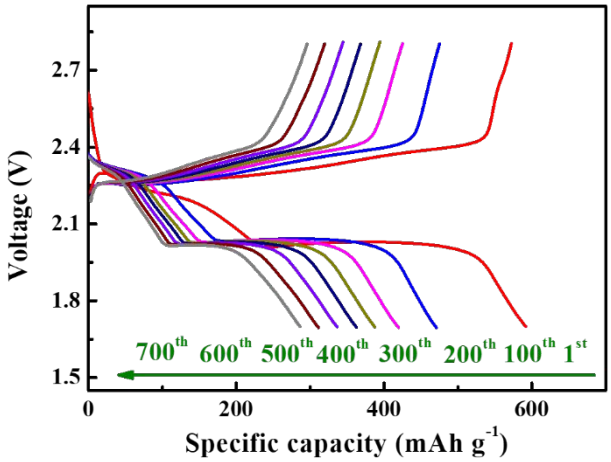

Figure S12 Charge/discharge curves of DVS at different cycles at 3C.

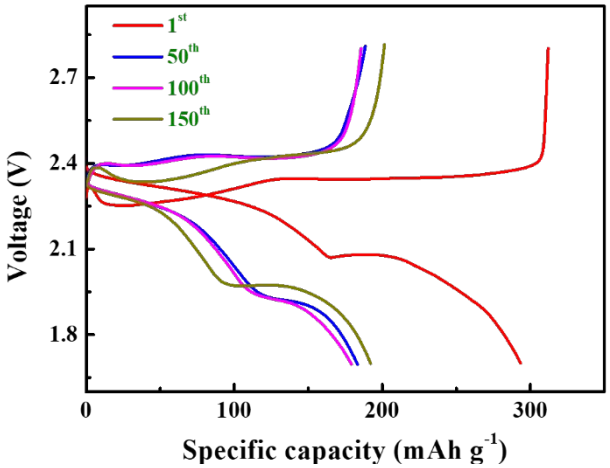

Figure S13 Charge/discharge curves of S-90 at different cycles at 3C.

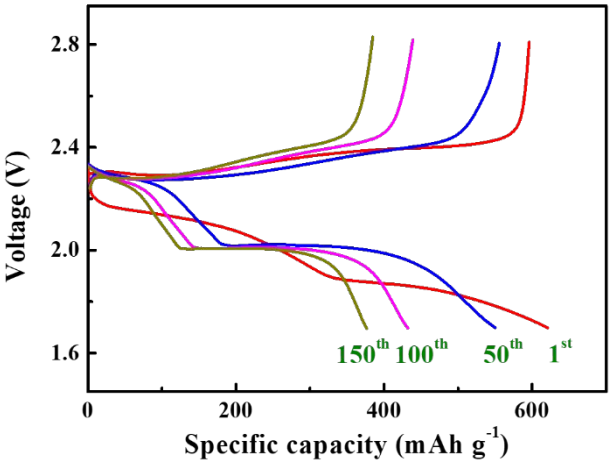

Figure S14 Charge/discharge curves of S-63 at different cycles at 3C.

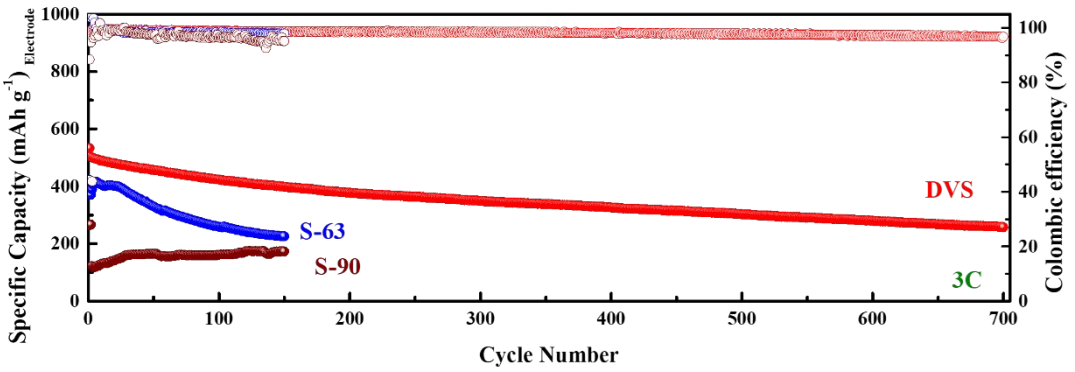

Figure S15 Cycling performances for DVS, S-63, and S-90 at 3C rate, with the specific capacities calculated based on the weight of electrode.

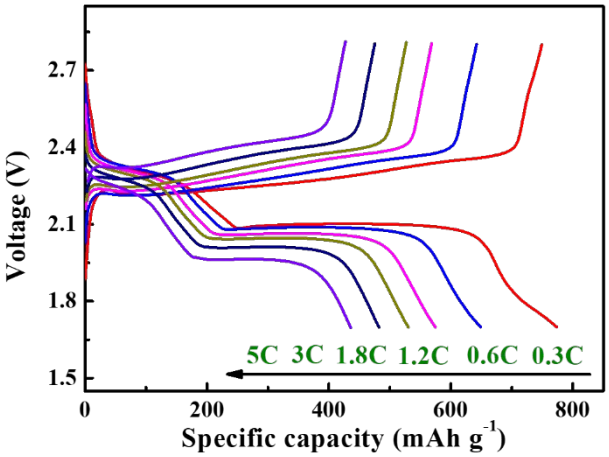

Figure S16 Charge/discharge curves of DVS at different C-rates.

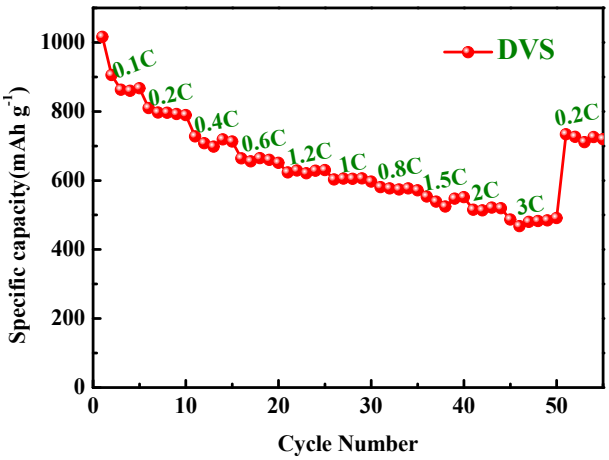

Figure S17 Rate performances of DVS at different C-rates.

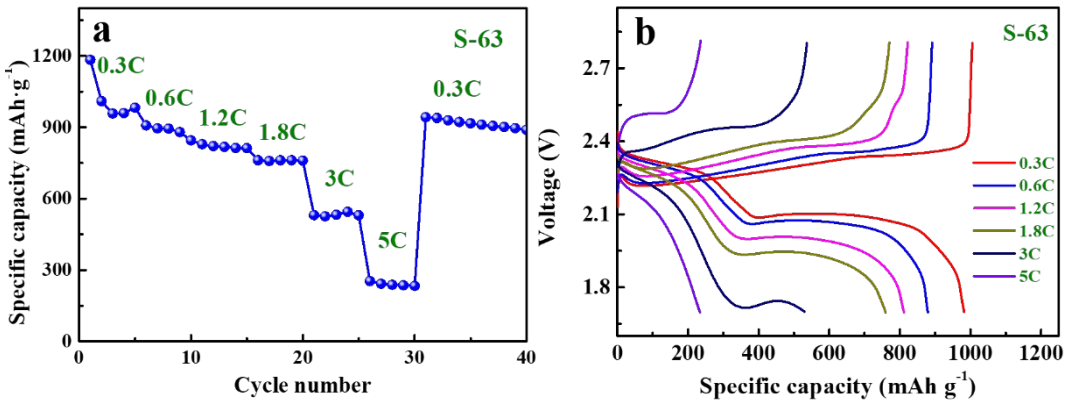

Figure S18 (a)Rate performances of S-63 and (b) the corresponding charge/discharge curves at different C-rate.

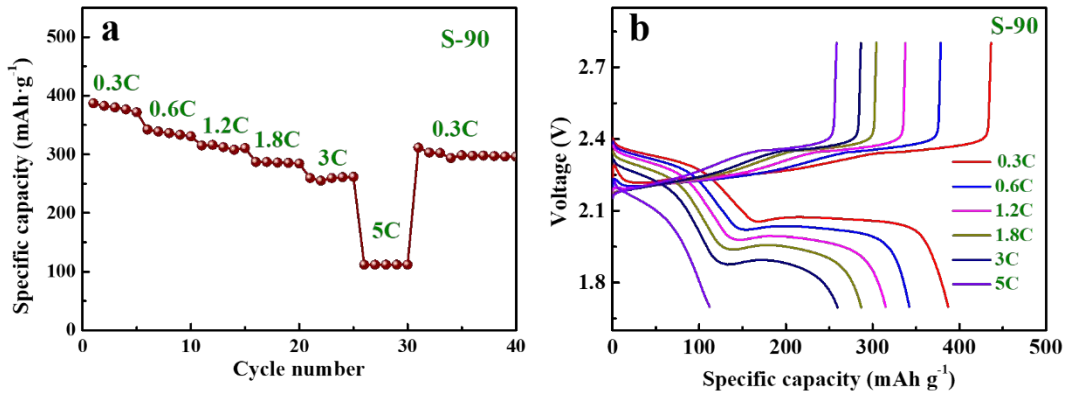

Figure S19 (a)Rate performances of S-90 and (b) the corresponding charge/discharge curves at different C-rate.

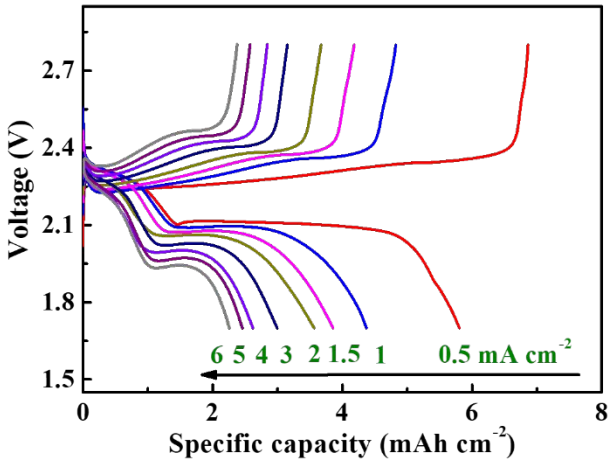

Figure S20 Charge/discharge curves for the high-loading DVS at different currents.

Table S1 The EDS results of VS_4@HPGC

Element	Atomic fraction	Mass fraction
C	26.47	11.79
S	55.72	62.80
V	13.96	23.09
O	3.85	2.33

Table S2 The electrochemical comparison of different metal sulfides in the voltage of 1.7~2.8 V

Metal sulfides	Specific capacity (mAh g^{-1})		Reference
	First cycle	10^{th} cycle	
VS_4	433.7	249.4	Our work
Co_9S_8	36	25	[1]
TiS_2	215	167	[2]
VS_2	54	22	[3]
MoS_2	6	92	[4]
SnS_2	50	5	[5]
Mo_6S_8	128	---	[6]
CoS_2	126	75	[7]
Sb_2S_3	40	---	[8]
SnS_2	70	---	[9]

Table S3 The length change of S-S bonds for Li_2S_6 b interact with VS_4 and Li_3VS_4

Band	Li_2S_6 (Å)	VS_4-Li_2S_6 (Å)	Li_3VS_4- Li_2S_6 (Å)
S_1—S_2	2.058	2.055	2.076
S_2—S_3	2.045	2.041	2.052
S_3—S_4	2.191	2.189	2.242
S_4—S_5	2.048	2.044	2.043
S_5—S_6	2.057	2.064	2.072

Table S4 The fitting results of EIS for DVS and S-63

	R_L (ohm)	Rint (ohm)	Rct (ohm)
DVS	3.80	4.65	13.69
S-63	5.55	8.17	15.99

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