Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Heterointerface Effect on Catalytic Performance of Single Atom Catalysts for Sulfur Chemistry in Lithium-sulfur Batteries

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Data1. For Section 3.1

Table S1 Cell parameters (cp), the corresponding supercells (cs), the supercell of graphene (sg) for SACs, the lattice mismatch (lm), and the total number of atoms contained in the heterostructure materials (tma).

	cp(Å)	cs	sg	lm	tma
TiS_2	a = b = 3.377	5 × 5 × 1	7 × 7 × 1	2.354%	172
C_3N_4	a = b = 7.150	2 × 2 × 1	6 × 6 × 1	1.777%	127
BN	a = b = 2.513	5 × 5 × 1	5 × 5 × 1	1.749%	99
G[#]	a = b = 2.468	6 × 6 × 1	6 × 6 × 1	0.000%	99
rGO	a = b = 12.188	1 × 1 × 1	5 × 5 × 1	1.211%	106

The 6 × 6 × 1 supercell of graphene is used to construct SACs.

Table S2 E_{bin}, interval-distance and charge transfer of two components of different heterostructures.

heterostructures	E_{bin} (meV Å^{-2})	Interlayer distance (Å)	Charge transfer (e/cell)[#]
VN_4@G/TiS_2	−18.84	3.32	−1.48
VN_4@G/C_3N_4	−18.06	3.24	−0.76
VN_4@G/BN	−21.32	3.25	−0.02
VN_4@G/G	−19.28	3.26	−0.11
VN_4@G/rGO	−12.15	3.95	−0.43
MoN_4@G/TiS_2	−10.81	3.36	−1.11
MoN_4@G/C_3N_4	−7.49	3.29	−0.20
MoN_4@G/BN	−24.85	3.26	−0.05
MoN_4@G/G	−19.19	3.20	−0.16
MoN_4@G/rGO	−12.71	3.96	−0.51
WN_4@G/TiS_2	−14.82	3.37	−1.11
WN_4@G/C_3N_4	−8.15	3.29	−0.19
WN_4@G/BN	−26.39	3.25	−0.07
WN_4@G/G	−20.79	3.21	−0.19
WN_4@G/rGO	−14.36	3.95	−0.53

For charge transfer, a negative value means that the MN_4@G layer loses charge.

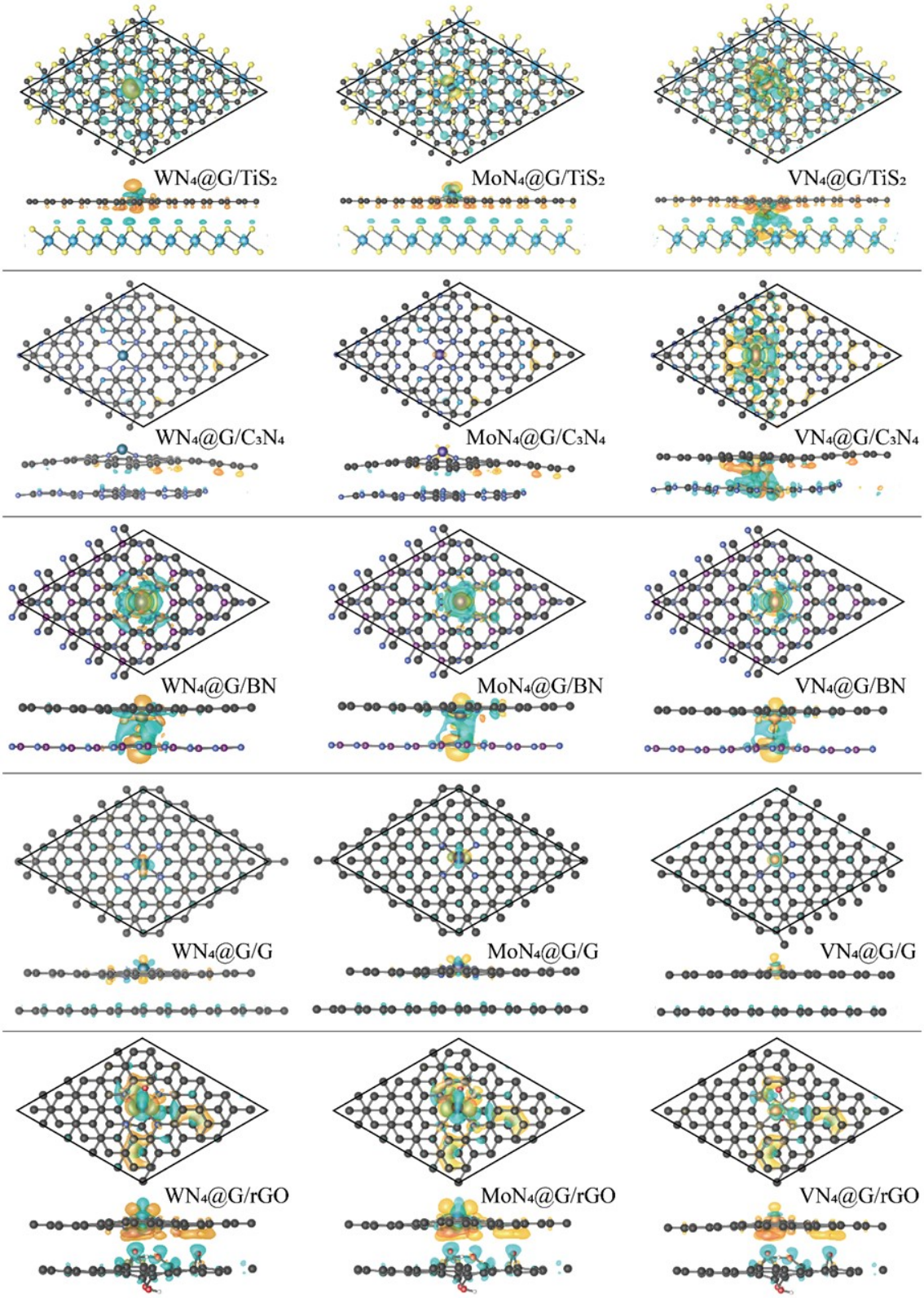

Figure S1 Structural configurations and charge density difference of heterostructures. Blue and origin isosurface indicate electron accumulation and depletion, respectively. The value of isosurface is 0.001 e/Bohr^3.

Data2. For Section 3.2

Table S3 Predicted values of E_b (eV) of Li_2S on the plane of MN_4@G in heterostructure materials according to three different linear relations (ΔE-E_b、ΔE(*LiS)-E and ICOHP-E_b) as well as those obtained from CI-NEB method.

heterostructure	E_b (ΔE)	E_b (ΔE(*LiS))	E_b (ICOHP)	E_b (CI-NEB)
VN_4@G/TiS_2	0.55	1.07	1.31	0.54
VN_4@G/C_3N_4	2.24	2.01	1.85	-
VN_4@G/BN	0.81	0.98	1.68	0.75
VN_4@G/G	0.79	0.91	1.65	-
VN_4@G/rGO	0.79	0.93	1.55	-
MoN_4@G/TiS_2	0.40	0.94	0.68	0.44
MoN_4@G/C_3N_4	0.57	0.61	0.72	-
MoN_4@G/BN	0.63	0.81	0.86	-
MoN_4@G/G	0.74	0.82	1.02	
MoN_4@G/rGO	0.56	0.74	0.83	0.58
WN_4@G/TiS_2	0.43	0.92	0.54	0.41
WN_4@G/C_3N_4	0.57	0.62	0.57	0.50
WN_4@G/BN	0.62	0.80	0.74	-
WN_4@G/G	0.72	0.79	0.96	0.83
WN_4@G/rGO	0.54	0.72	0.70	-

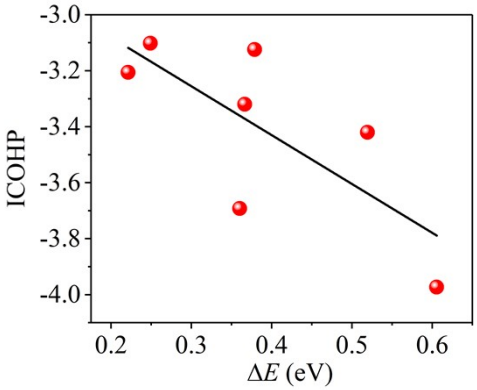

Figure S2 Relationship between ΔE and ICOHP for several heterostructures.

Data3. For Section 3.3

Table S4 Adsorption energies of S_8 and LiPs on different substrates as well as that on DOL and DME for comparison.[1]

substrates	Li_2S (eV)	Li_2S_2 (eV)	Li_2S_4 (eV)	Li_2S_6 (eV)	Li_2S_8 (eV)	S_8 (eV)
MoN_4@G/TiS_2	−4.67	−3.72	−3.71	−3.35	−3.28	−2.11
TiS_2/MoN_4@G	−3.54	−2.42	−1.65	−0.94	−1.16	−0.43
Mo_4@G	−4.06	−3.36	−3.46	−3.14	−3.71	−2.10
WN_4@G/TiS_2	−5.21	−5.15	−4.13	−4.17	−4.33	−2.52
TiS_2/WN_4@G	−3.62	−2.43	−1.65	−0.84	−1.18	−0.45
WN_4@G	−4.50	−3.92	−4.16	−4.29	−3.99	−2.63
VN_4@G/TiS_2	−1.73	−1.64	−0.96	−1.04	−1.14	−0.59
TiS_2/VN_4@G	−3.15	−2.53	−1.78	−1.00	−1.29	−0.48
VN_4@G	−3.93	−3.22	−3.00	−2.97	−3.19	−1.58
G/TiS_2	−1.57	−1.69	−1.03	−0.94	−1.01	−0.55
TiS_2/G	−3.79	−2.72	−1.80	−0.98	−1.22	−0.47
G	−1.30	−1.16	−0.75	−0.83	−0.85	−0.49
TiS_2	−3.60	−2.45	−1.77	−0.90	−1.08	−0.32
DOL [1]	-	-	−0.87	−0.90	−0.92	-
DME [1]	-	-	−0.92	−0.95	−0.98	-

References

[1] T. Li, C. He, W. Zhang, A novel porous C4N4 monolayer as a potential anchoring material for lithium–sulfur battery design, Journal of Materials Chemistry A, 7 (2019) 4134-4144.