Supporting Information

Two-Dimensional phosphorus carbide (*β*-PC) as Highly Efficient Metal-free Electrocatalyst for Lithium−Sulfur Batteries: A first-principles study

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Table S1 The convergence of energy cutoff of the plane waves for β_0-PC.

ENCUT (eV)	Energy (eV/atom)
440	-6.70152
460	-6.70123
480	-6.70081
500	-6.70064
520	-6.70060
540	-6.70064

Table S2 The convergence of energy cutoff of the plane waves for β_1-PC.

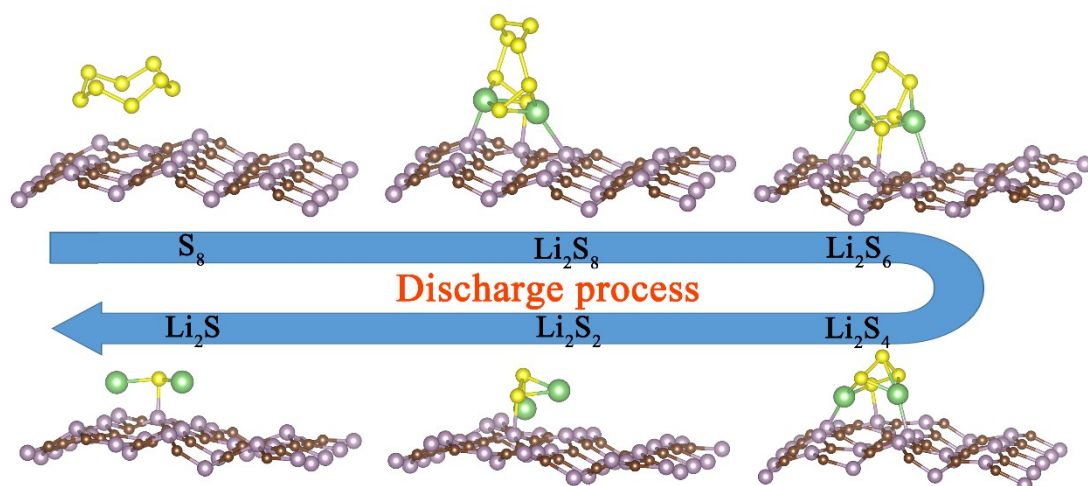| ENCUT (eV)	Energy (eV/atom)
440	-6.95240
460	-6.95218
480	-6.95174
500	-6.95153
520	-6.95147
540	-6.95141

Figure S1 The structural configurations of adsorbed intermediates on the β_0-PC.
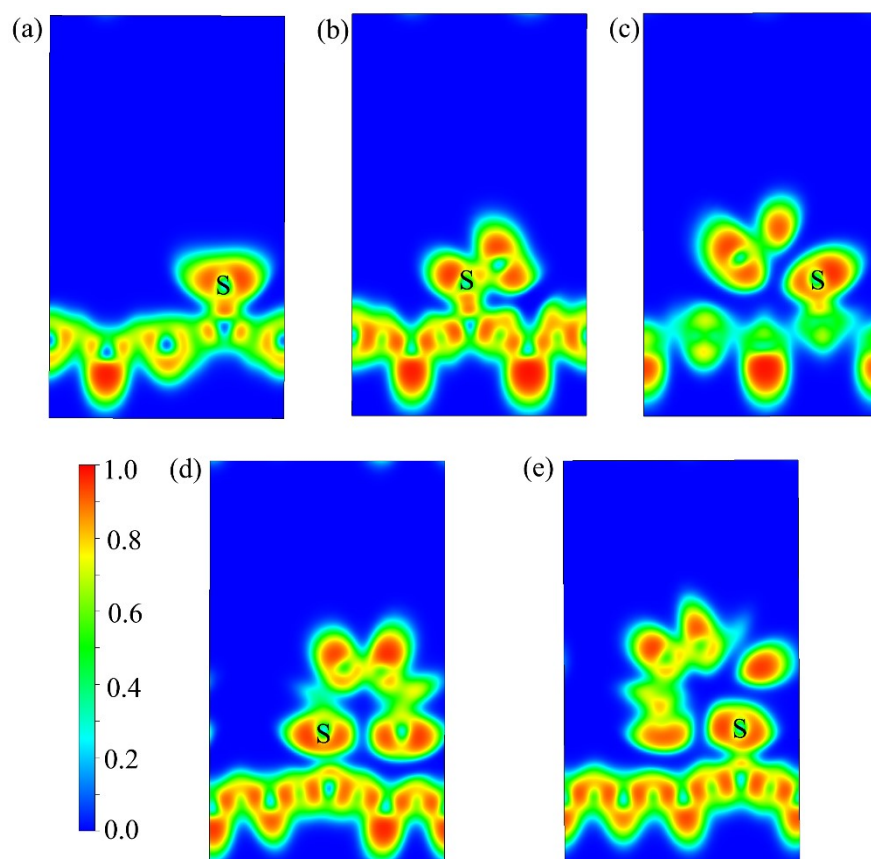
Figure S2 Sliced electron localization function of (a) Li_2S, (b) Li_2S_2, (c) Li_2S_4, (d) Li_2S_6, (e) Li_2S_8 adsorbed on β_0-PC surfaces between the S-P and Li-P atoms.
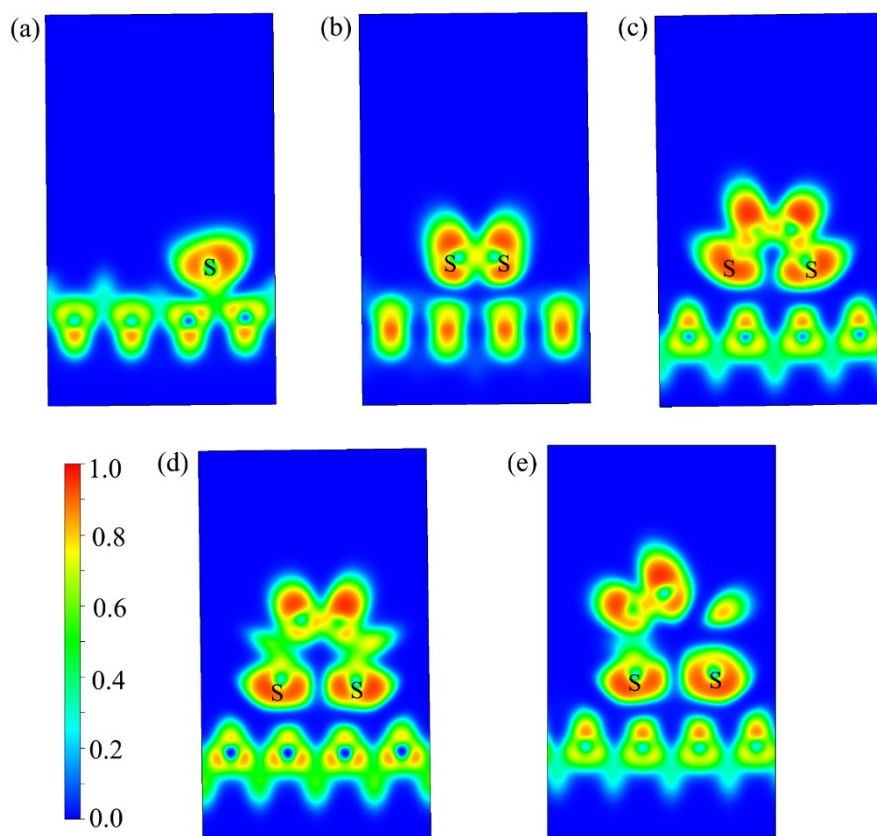
Figure S3 Sliced electron localization function of (a) Li_2S, (b) Li_2S_2, (c) Li_2S_4, (d) Li_2S_6, (e) Li_2S_8 adsorbed on β_1-PC surfaces between the S-P and Li-P atoms.
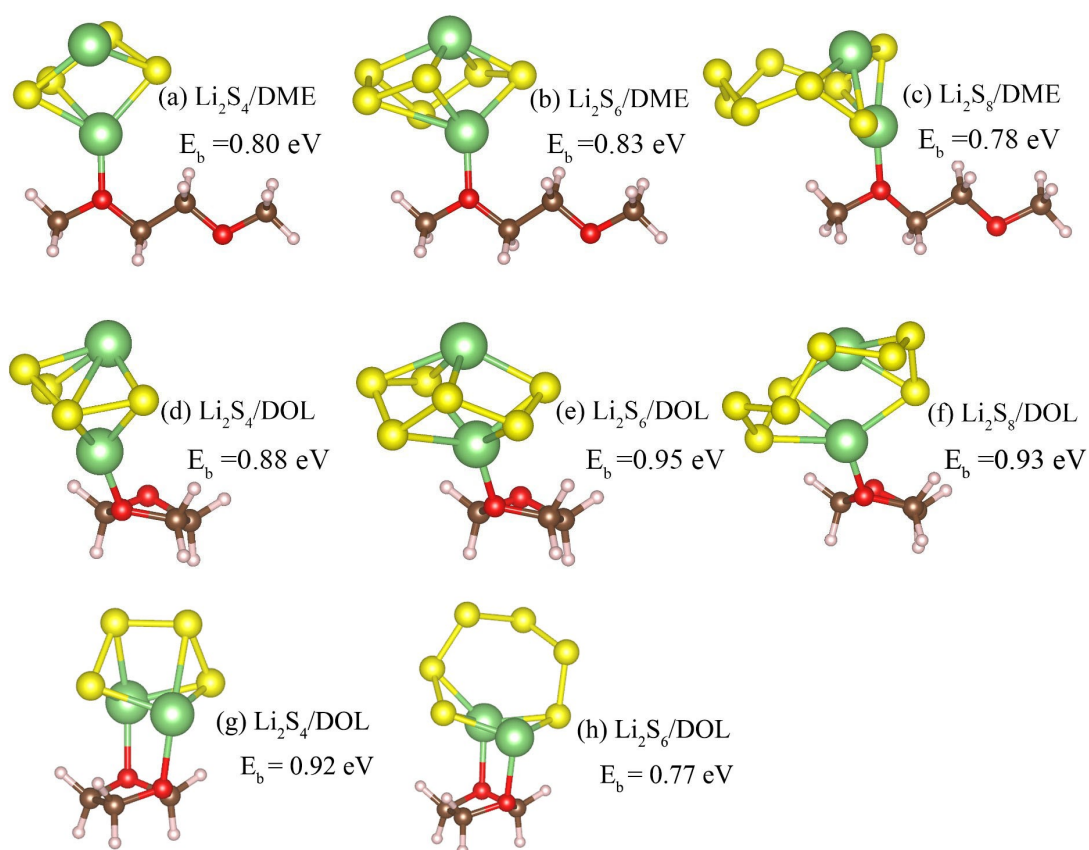
Figure S4 The structures of (a) Li_2S_4, (b) Li_2S_6 and (c) Li_2S_8 adsorbed on DME, (d) Li_2S_4, (e) Li_2S_6 and (f) Li_2S_8 adsorbed on DOL and (g) Li_2S_4, (h) Li_2S_6 adsorbed on DOL through 2Li−O bonds simulated with vdW functional. The brown, red, pink, yellow and green spheres represent the C, O, H, S and Li atoms, respectively.
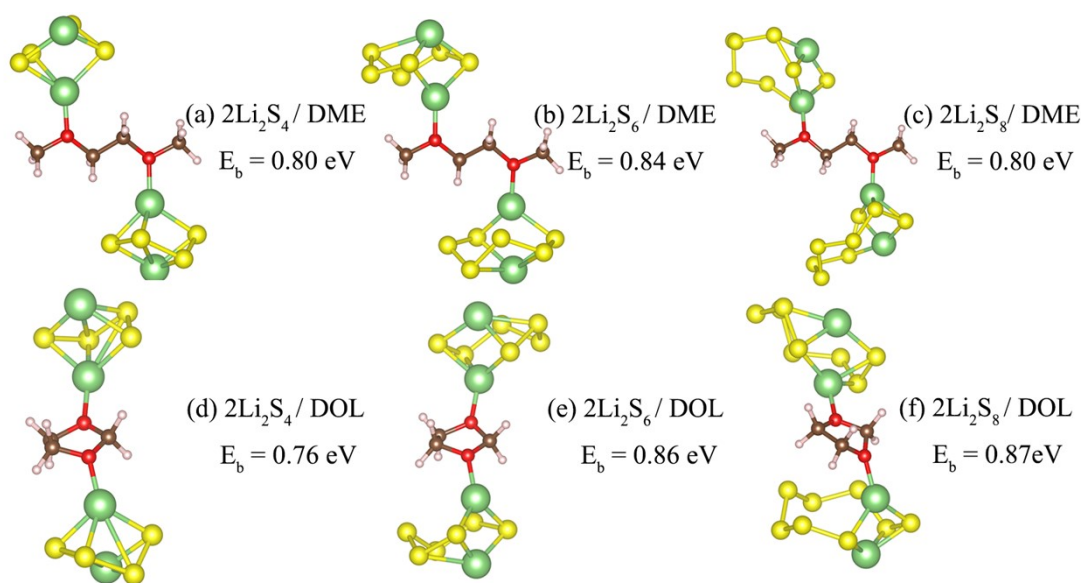
Figure S5 The structures of two (a) Li_2S_4, (b) Li_2S_6 and (c) Li_2S_8 molecules adsorbed on DME and two (d) Li_2S_4, (e) Li_2S_6 and (f) Li_2S_8 molecules adsorbed on DOL simulated with vdW functional. The brown, red, pink, yellow and green spheres represent the C, O, H, S and Li atoms, respectively.
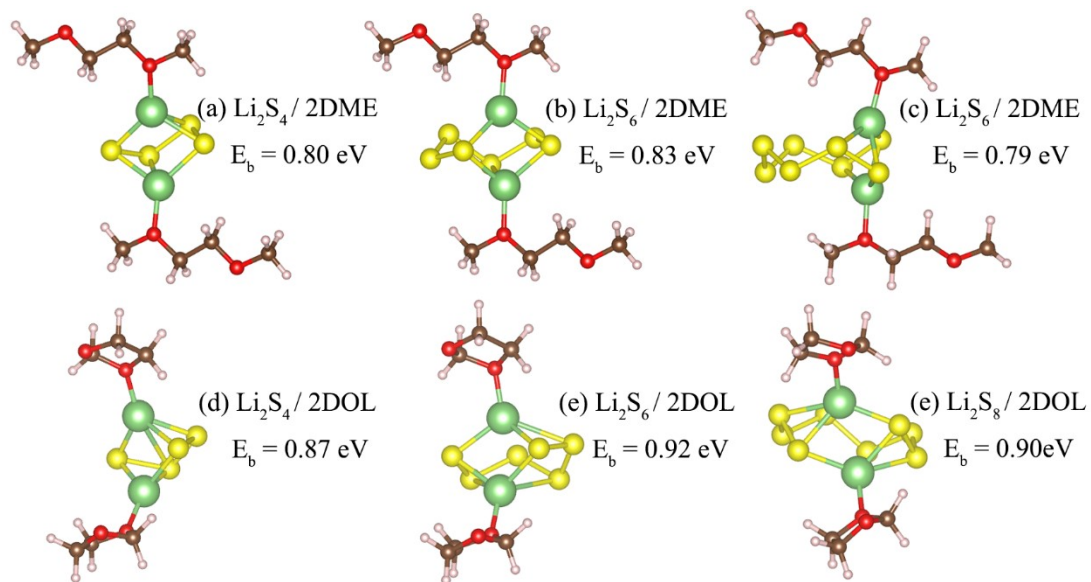
Figure S6 The structures of (a) Li_2S_4, (b) Li_2S_6 and (c) Li_2S_8 adsorbed on two DME molecules and (d) Li_2S_4, (e) Li_2S_6 and (f) Li_2S_8 adsorbed on two DOL molecules simulated with vdW functional. The brown, red, pink, yellow and green spheres represent the C, O, H, S and Li atoms, respectively.
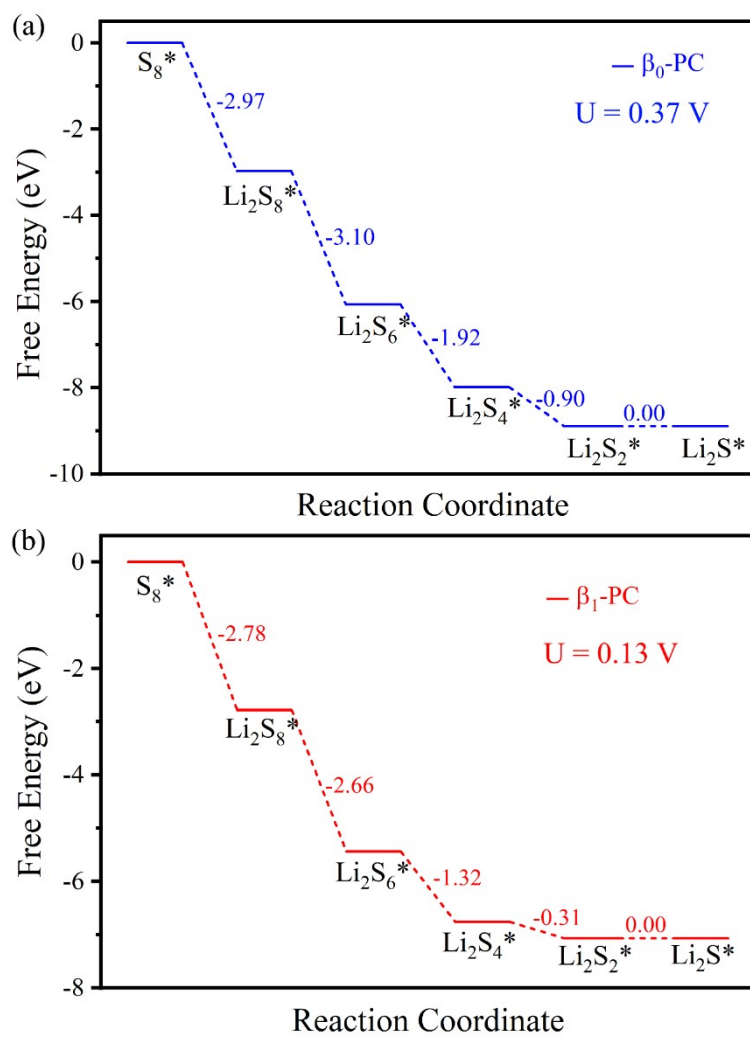
Figure S7 Calculated SRR Gibbs free energy diagram of β_0-PC with a potential of U=0.37 V. (b) Calculated SRR Gibbs free energy diagram of β_1-PC with a potential of U=0.13 V.
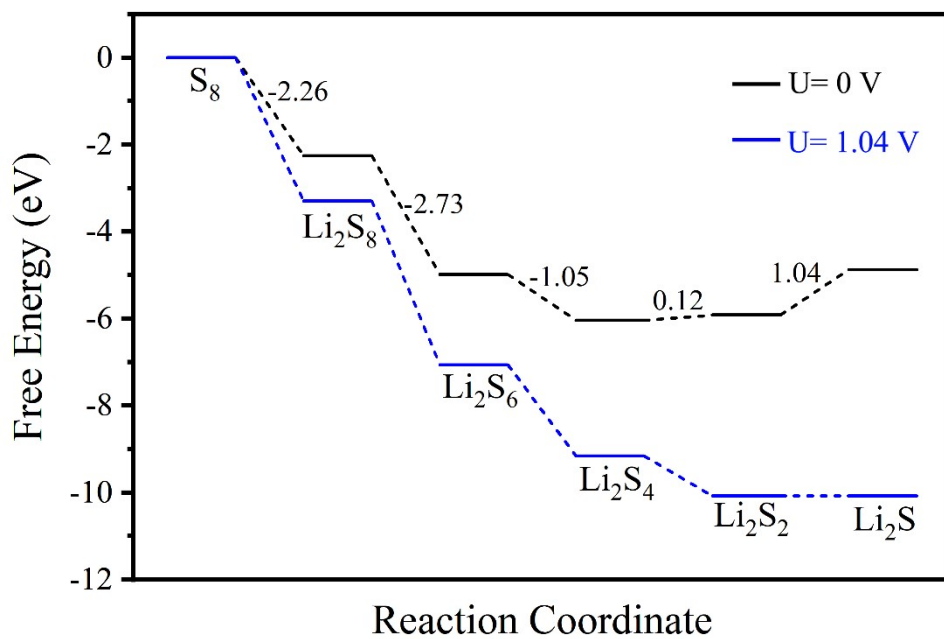
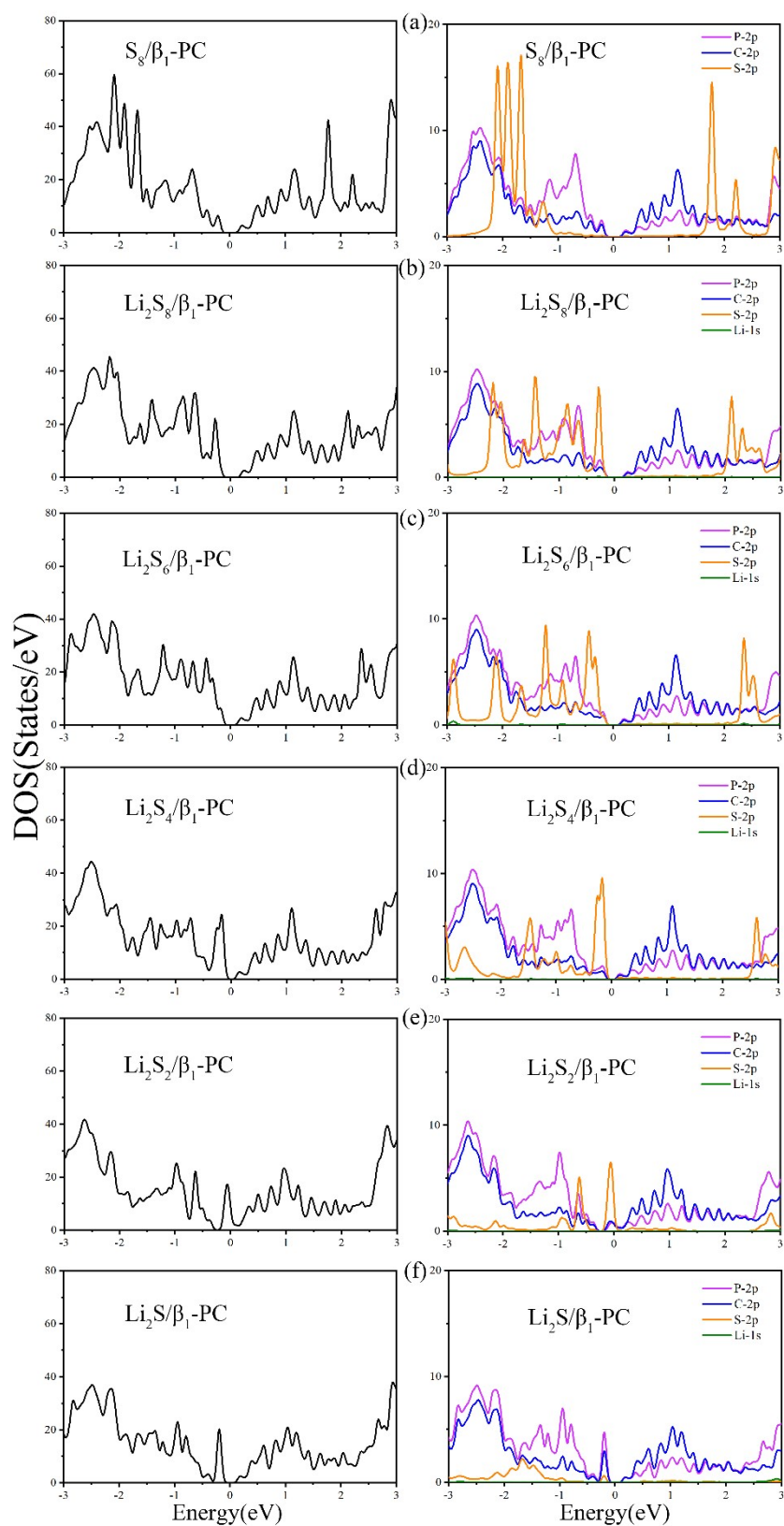Figure S8 The Gibbs free energy diagram of sulfur reduction reaction in vacuum under potentials U=0 V and U=1.04 V.

Figure S9 (Right panel) TDOSs and (left panel) PDOSs projected onto the different atomic orbitals of (a) S_8 @ β_1-PC and Li_2S_n @ β_1-PC with (b) n = 8, (c) n = 6, (d) n = 4, (e) n = 2, and (f) n = 1. The Fermi level is set to 0.

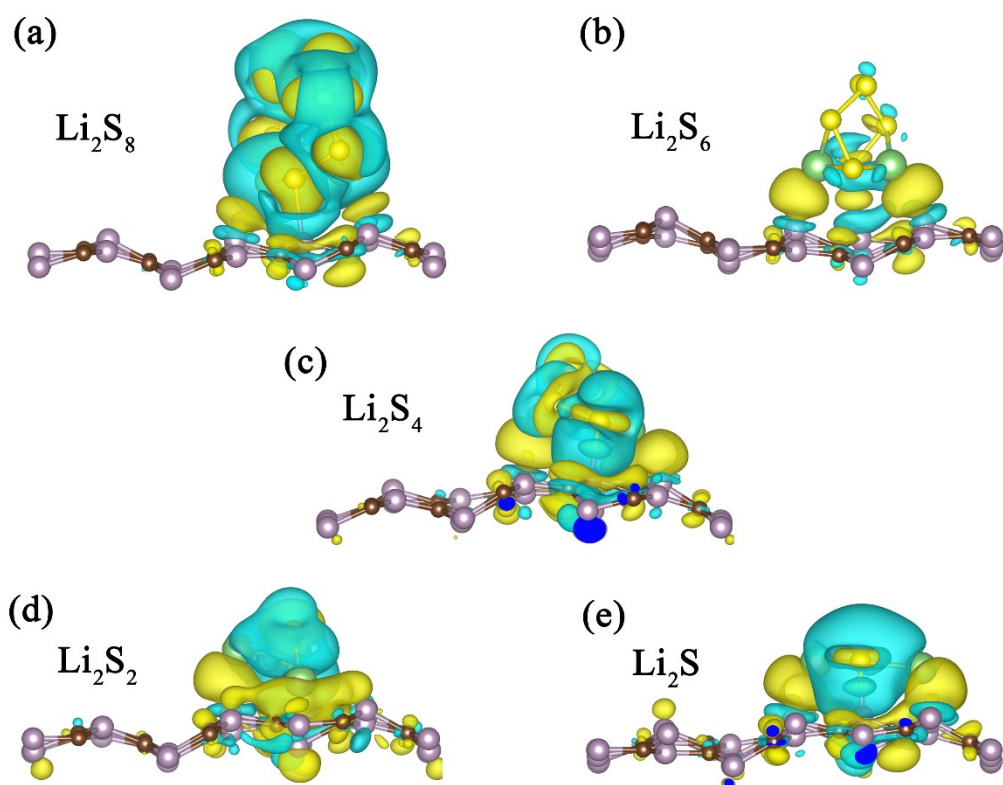Figure S10. Differential charge density distribution for (a) Li_2S_8、(b)Li_2S_6、(c)Li_2S_4、(d)Li_2S_2 (e)Li_2S molecules adsorbed on the β_0-PC. Yellow and blue colors indicate the electron accumulation and depletion, respectively.

Coordinates of β_0-PC

POSCAR of β_0-PC

1.00000000000000

    5.0570000000000004   0.0000000000000000   0.0000000000000000

    0.0000000000000000   2.9190000381000001   0.0000000000000000

    0.0000000000000000   0.0000000000000000   20.0000000000000000

  P   C

  2   2

Direct

  0.0356262804186827 0.9971171071842875 0.1518894184959692

  0.5356309672806532 0.4973582542839807 0.2032574553146063

  0.2093512523693093 0.4971457224172937 0.1715866483905728

  0.7093615059313620 0.9974189331144387 0.1835365107988878

Coordinates of β_0-PC

POSCAR of β_1-PC

1.00000000000000

     4.7020000000000008   0.0000000000000000   0.0000000000000000

     0.0000000000000000   2.9120000000000008   0.0000000000000000

     0.0000000000000000   0.0000000000000000   20.0000000000000000

  P   C

  2   2

Direct

  0.0177654991436995 0.0000000000000000 0.9997884943248252

  0.6782124076314275 0.0000000000000000 0.0852162910794760

  0.2175005326161852 0.5000000000000000 0.0273980921654555

  0.4785215606086837 0.5000000000000000 0.0575971224302521