Supporting Information

A novel porous C_4N_4 monolayer as potential anchoring materials for lithium-sulfur battery design

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Supporting Figures

Up to now, a few carbonized nitrogen monolayers (e.g. $C_{22}N_4$, C_3N, C_2N, *g*-CN and CN_2) monolayers have been reported. To determine the relative stability of our predicted C_4N_4, we fully relax the other C_mN_{8-m} (m=1-8) monolayers and calculate the relative formation energy, as shown in Figure S1. The C_mN_{8-m} monolayer, sitting on the solid line, is thermodynamically stable with respect to decomposition into other C-N monolayers and elements (C or N), and it is experimentally synthesizable. Based on this, our predicted C_4N_4 monolayer is also stable with respect to the other C_mN_{8-m} monolayers.

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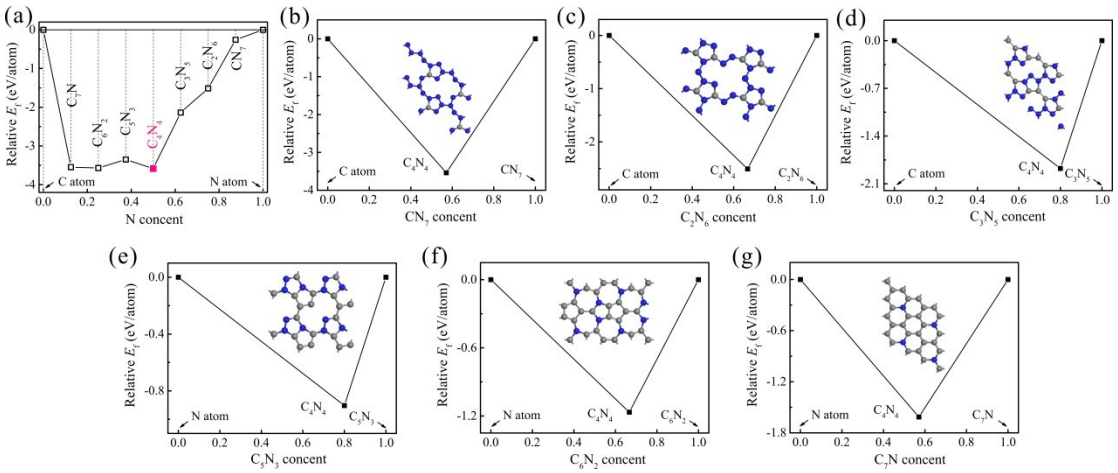

Figure S1. (a) Relative formation energies of C_mN_{8-m} (m=1-8) monolayers with respect to C and N atoms at 0 K. To clearly show the stability of C_4N_4 monolayer, we show all the possible decomposition paths. Relative formation energy of C_4N_4 monolayer with respect to C atom and (b) CN_7, (c) C_2N_6 or (d) C_3N_5 as well as N atom and (e) C_5N_3, (f) C_6N_2 or (g) C_7N at 0 K. The insert shows the structure of C_mN_{8-m} (m=1-8) monolayers.

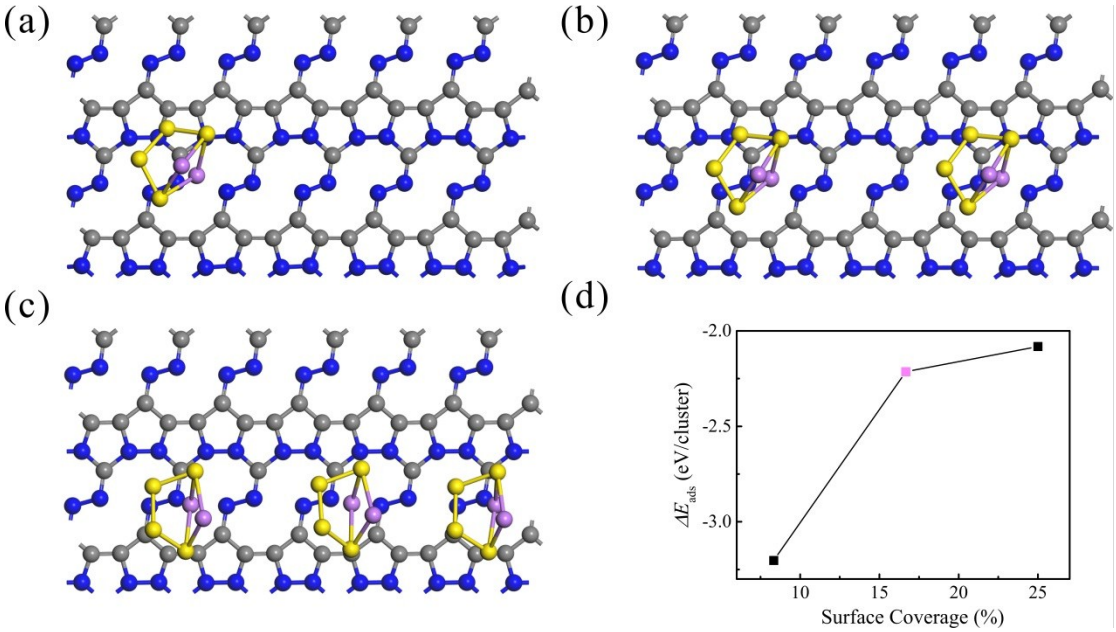

Figure S2. The most stable adsorption structures for the adsorption of Li2S4 on C4N4 at (a) 8.33 %, (b) 16.67 % and (c) 25 % surface coverage, respectively. (d) The relationship between average adsorption energy (ΔEads) and surface coverage.

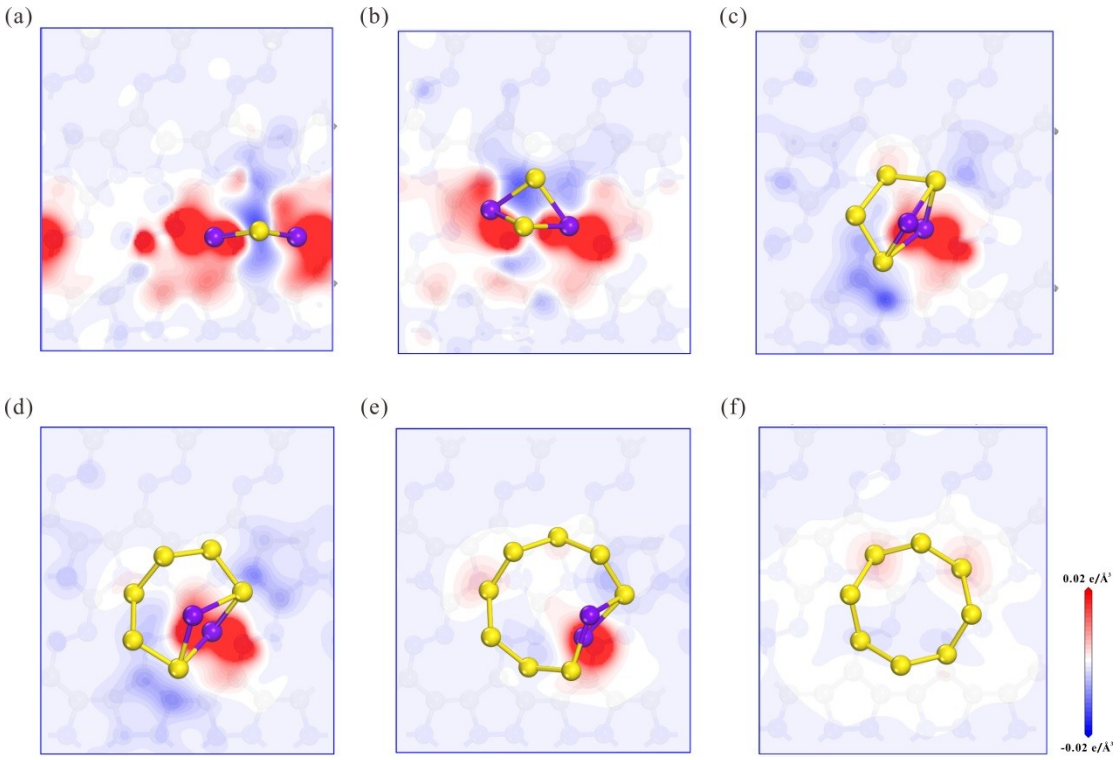

Figure S3. The charge density differences for (a) Li_2S, (b) Li_2S_2, (c) Li_2S_4, (d) Li_2S_6, (e) Li_2S_8 and (f) S_8 adsorbed on C_4N_4 monolayer are plotted. The red indicates an electron gain, while the blue represents an electron loss.

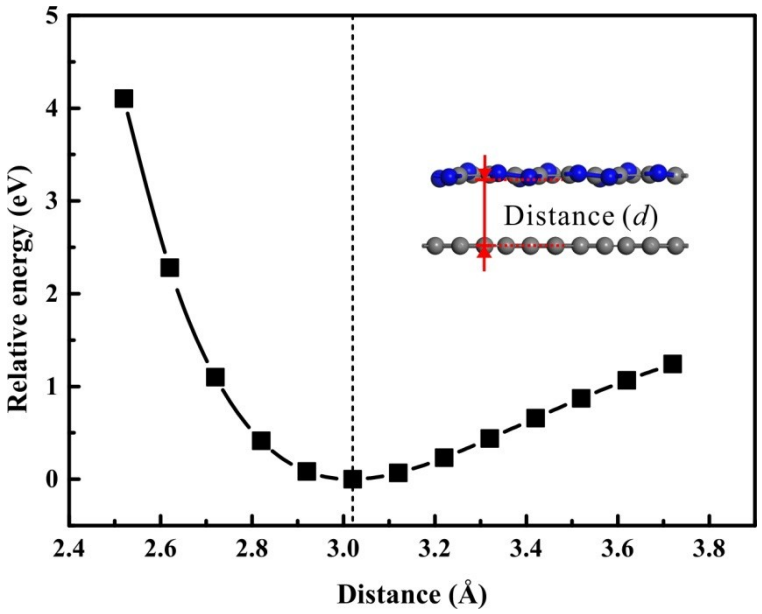

Figure S4. The relative energies as a function of lattice constant between C_4N_4 and graphyne. The carbon and nitrogen atoms are denoted as spheres in gray and blue, respectively.

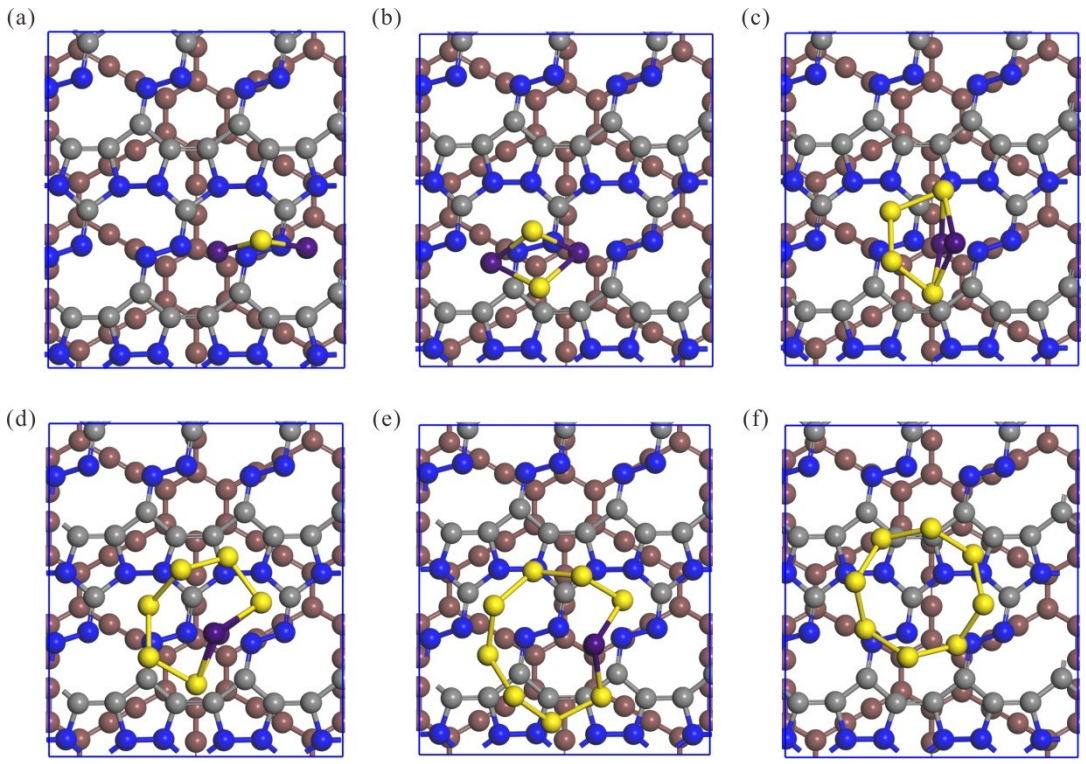

Figure S5. Fully optimized molecular structures of (a) Li_2S, (b) Li_2S_2, (c) Li_2S_4, (d) Li_2S_6, (e) Li_2S_8 and (f) S_8 adsorbed on C_4N_4/graphyne heterostructure.

Supporting POSCAR data

The atomic coordinates of C_4N_4 primitive cell.

POSCAR

C_4N_4
1.0

3.5866999626	0.0000000000	0.0000000000
0.0000000000	6.1030998230	0.0000000000
0.0000000000	0.0000000000	20.0000000000

C N
4 4
Direct

0.467709993	0.956339994	0.508199978
0.679409999	0.325170004	0.506219959
0.296649995	0.319770007	0.504390001
0.998889968	0.465469991	0.503380013
0.408060013	0.742640045	0.515820026
0.059310000	0.684740007	0.495249987
0.175180009	0.102860005	0.502210045
0.786200013	0.098839995	0.510349989

Supporting Table

Table S1. The computed diffusion constants (*D*) of S_8 cluster and LiPSs along three paths on C_4N_4 monolayer.

Clusters	Diffusion Constant (*D*)		
	1 Path	2 Path	3 Path
S_8	1.904×10^{-6}	4.689×10^{-13}	2.983×10^{-10}
Li_2S_8	6.885×10^{-11}	2.704×10^{-18}	5.872×10^{-12}
Li_2S_6	3.740×10^{-22}	1.002×10^{-25}	5.151×10^{-15}
Li_2S_4	2.298×10^{-10}	3.998×10^{-23}	3.548×10^{-16}
Li_2S_2	4.771×10^{-28}	5.400×10^{-36}	4.328×10^{-27}
Li_2S	5.275×10^{-26}	1.168×10^{-39}	1.678×10^{-30}

Table S2. The calculated of adsorption parameters and diffusion energy barrier (E_a) between S_8 cluster as well as LiPSs and C_4N_4/graphyne heterostructure.

Clusters	C_4N_4/graphyne heterostructure			
	E_{ads} (eV)	e (e)	$\Delta d_{C\text{-}X}$ (Å)	E_a (eV)
S_8	-1.863	0.007	2.839	0.259
Li_2S_8	-2.162	0.012	1.732	0.445
Li_2S_6	-2.075	0.007	1.631	0.466
Li_2S_4	-2.221	0.06	1.539	0.514
Li_2S_2	-2.547	0.346	1.358	0.823
Li_2S	-2.805	0.376	1.447	0.853