Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

CuS_2 Sheet: Hidden anode material with high capacity for sodium-ion batteries

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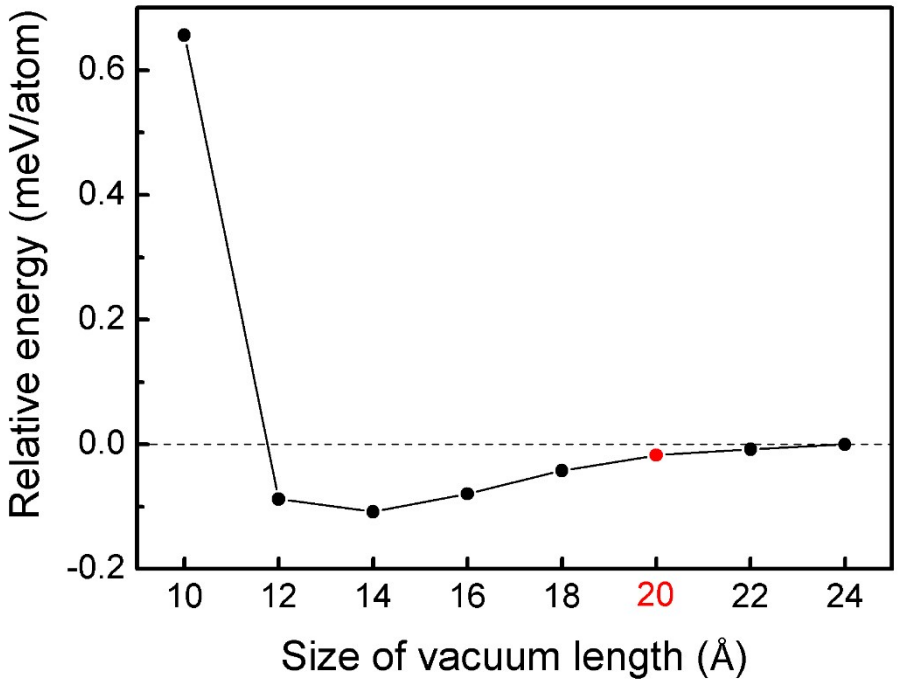

Fig. S1. The calculated total energies per atom as a function of length of the vacuum.

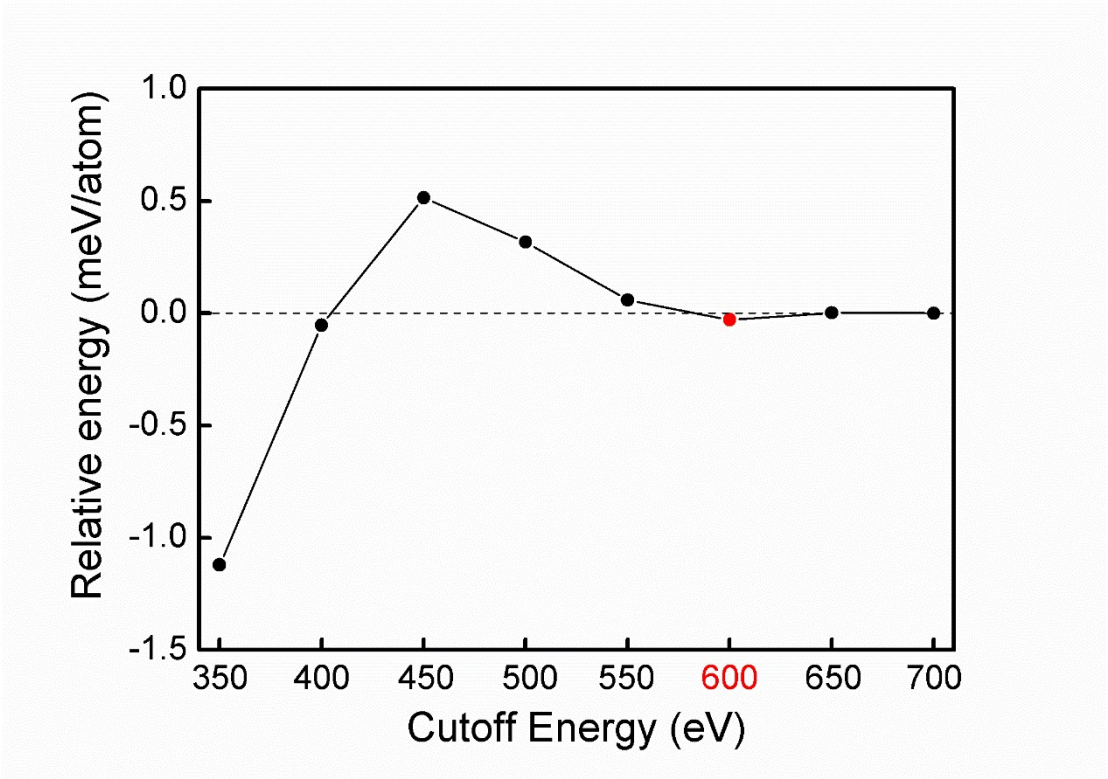

Fig. S2. The calculated total energies per atom as a function of cutoff energy.

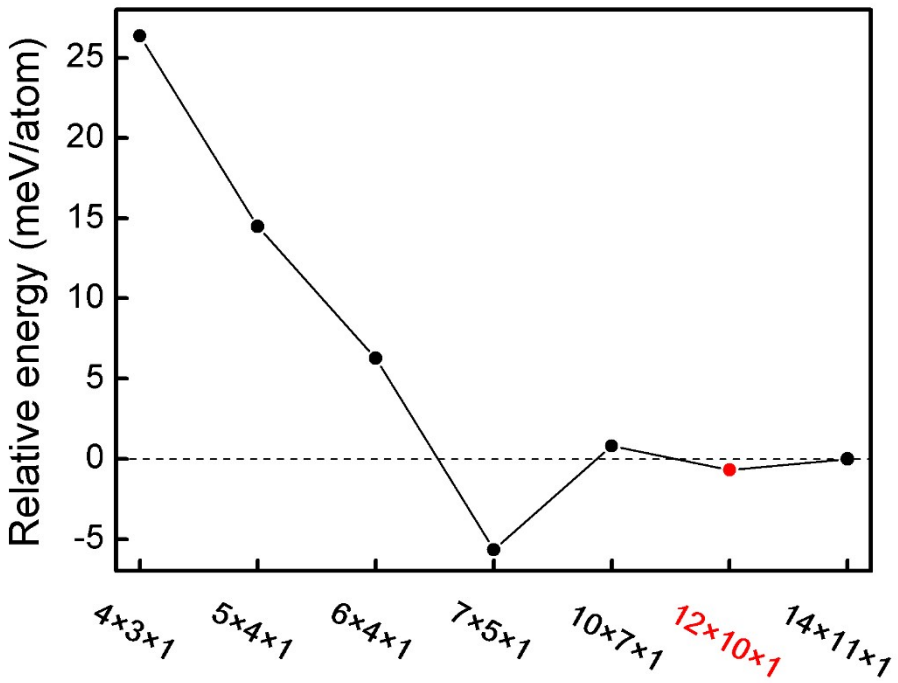

Fig. S3. The calculated total energies per atom as a function of K point mesh.

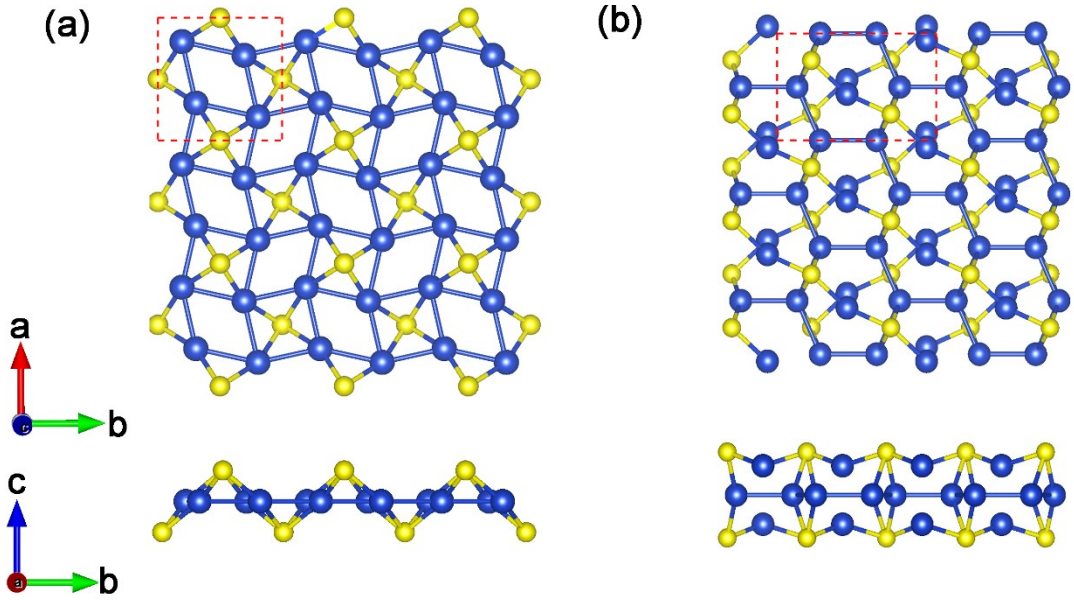

Fig. S4. Top and side views of two predicted Cu_2S monolayers ((a) Cu_2S-0(δ-Cu_2S) and (b) Cu_2S-1) with low formation energy.

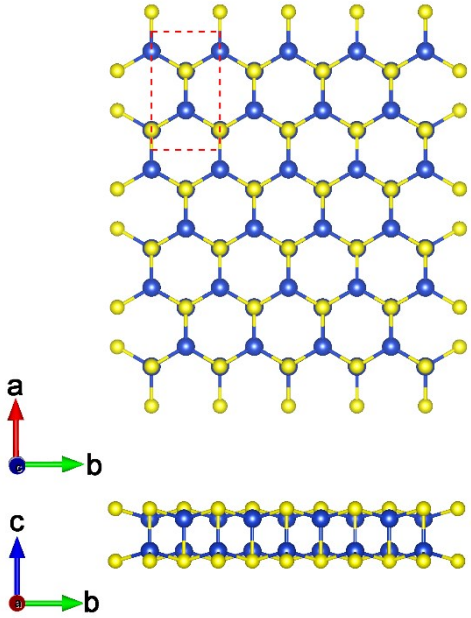

Fig. S5. Top and side views of the predicted h-CuS monolayer with lowest formation energy.

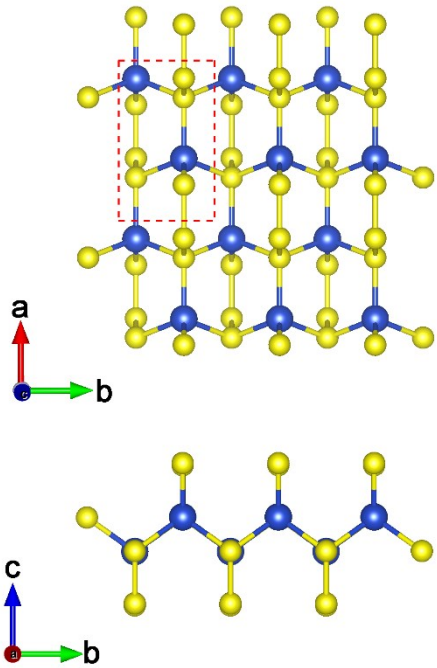

Fig. S6. Top and side views of obtained CuS_3 monolayer with lowest formation energy.

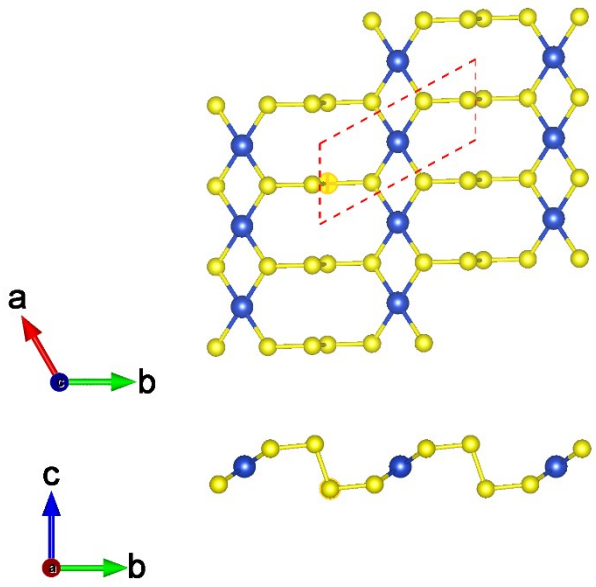

Fig. S7. Top and side views of obtained CuS_4 monolayer with lowest formation energy.

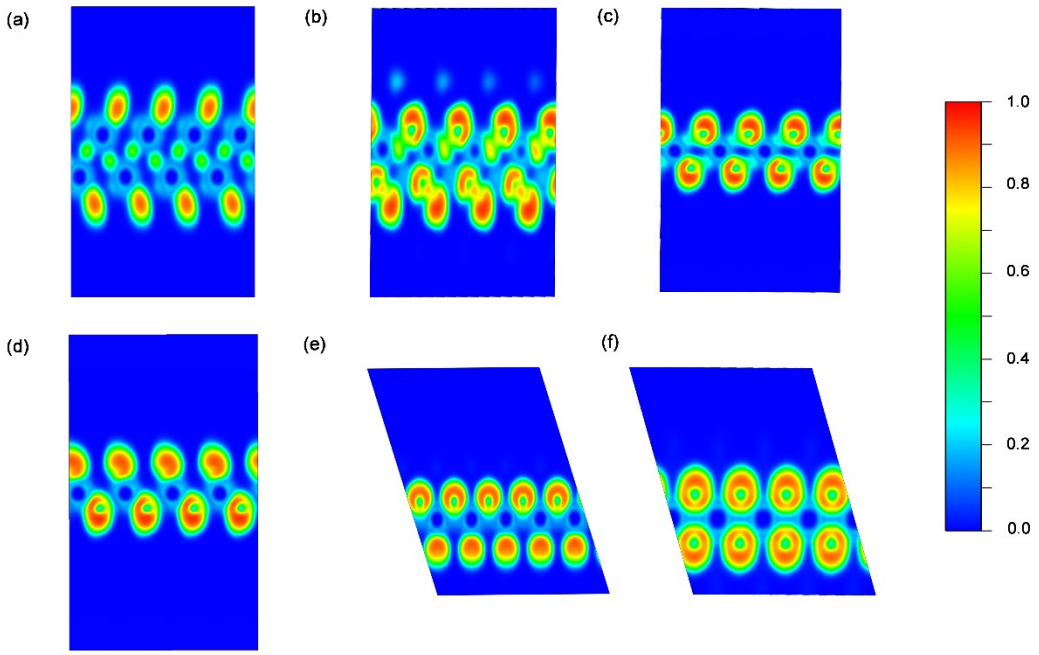

Fig. S8. 2D electron localization functions maps(side views) of (a)α-CuS_2, (b)β-CuS_2, (c)γ-CuS_2, (d)δ-CuS_2, (e)CuS_2(2H-MoS_2) and (f) CuS_2(1T-MoS_2) monolayer; ELF = 0 (blue) and 1 (red) illustrate electron vanishing and accumulated density, respectively.

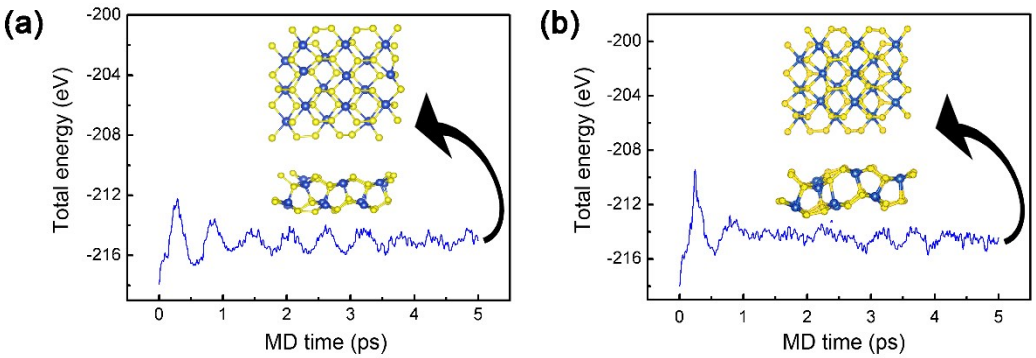

Fig. S9. The final MD images (viewed from top and side) and total energy curves with molecular dynamics simulated time of α-CuS_2 at (a) 400K, (b) 500K, the total simulated time is 5 ps and the time step is set at 1 fs.

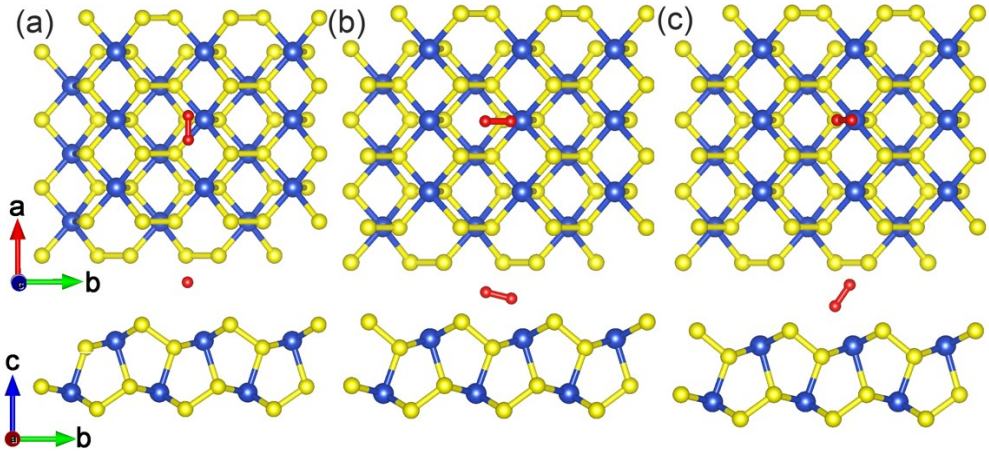

Fig. S10. Three optimized configurations of O_2 adsorbed on α-CuS_2 sheet. The adsorption energy is 0.48, -0.01, and 0.33 eV for (a), (b), and (c).

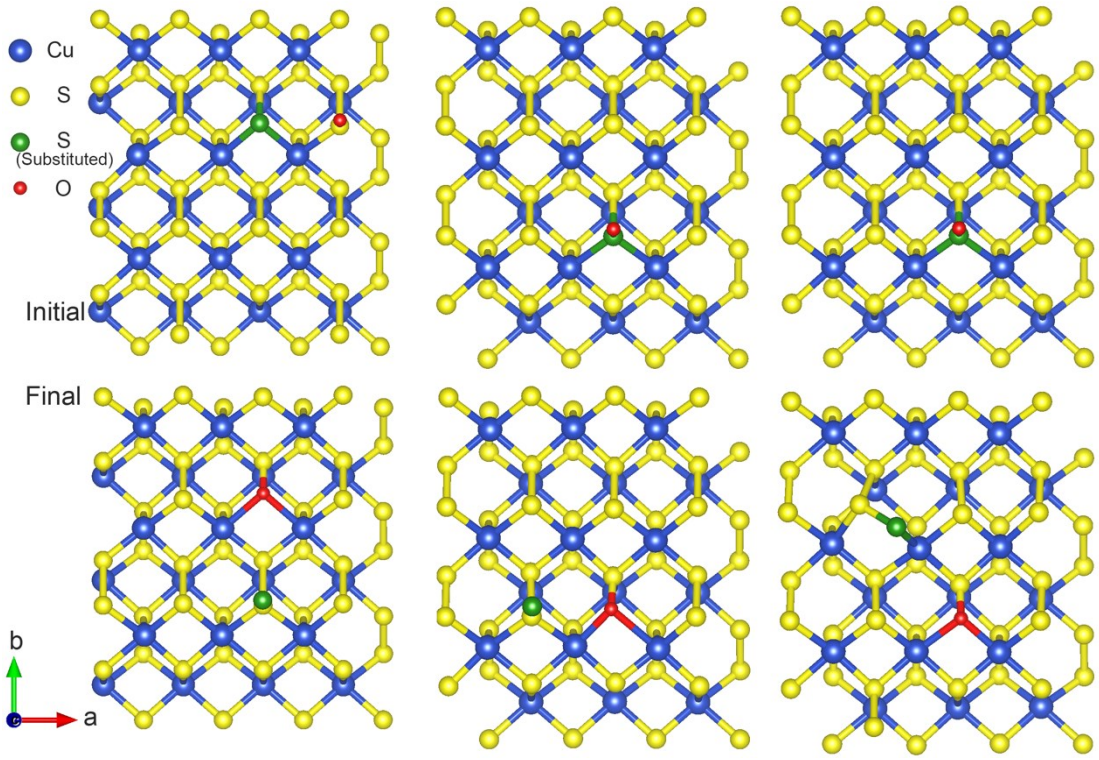

Fig. S11. Three initial (top panel) and three final (bottom panel) states prepared for NEB calculation of the O replace S chemical reactions.

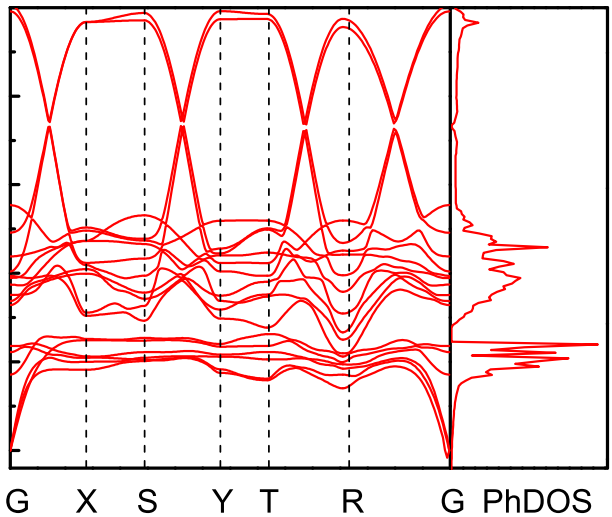

Fig. S12. The phonon band structure and DOS of the bulk CuS_2.

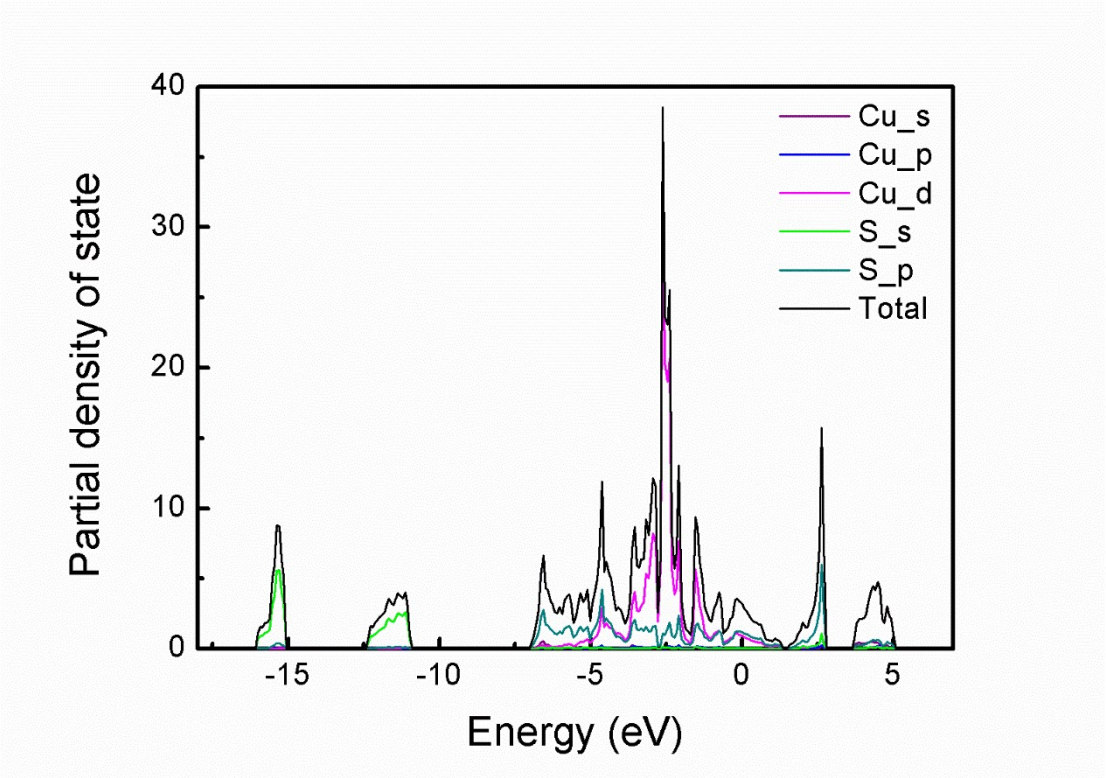

Fig. S13. The partial density of states of α-CuS_2 monolayer are calculated by PBE method, the Fermi level is set at 0 eV.

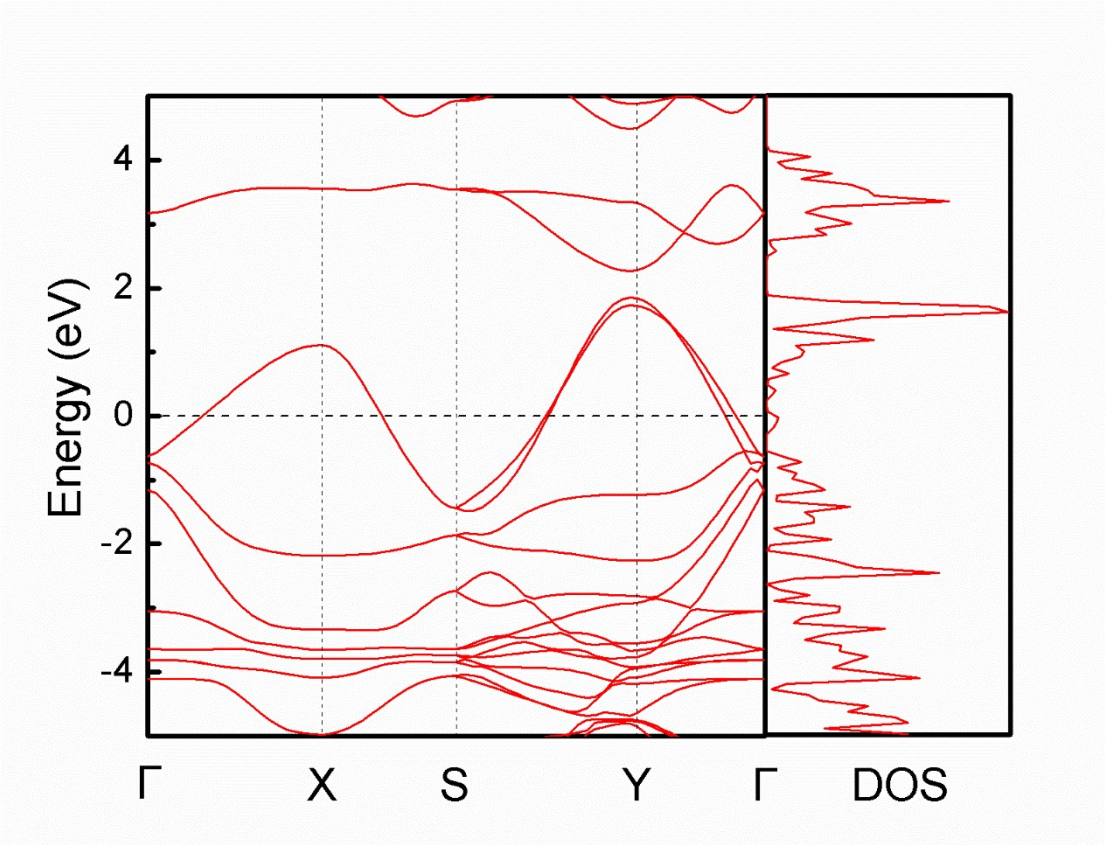

Fig. S14. The band structure of α-CuS_2 monolayer is calculated by HSE06 method, the Fermi level is set at 0 eV.

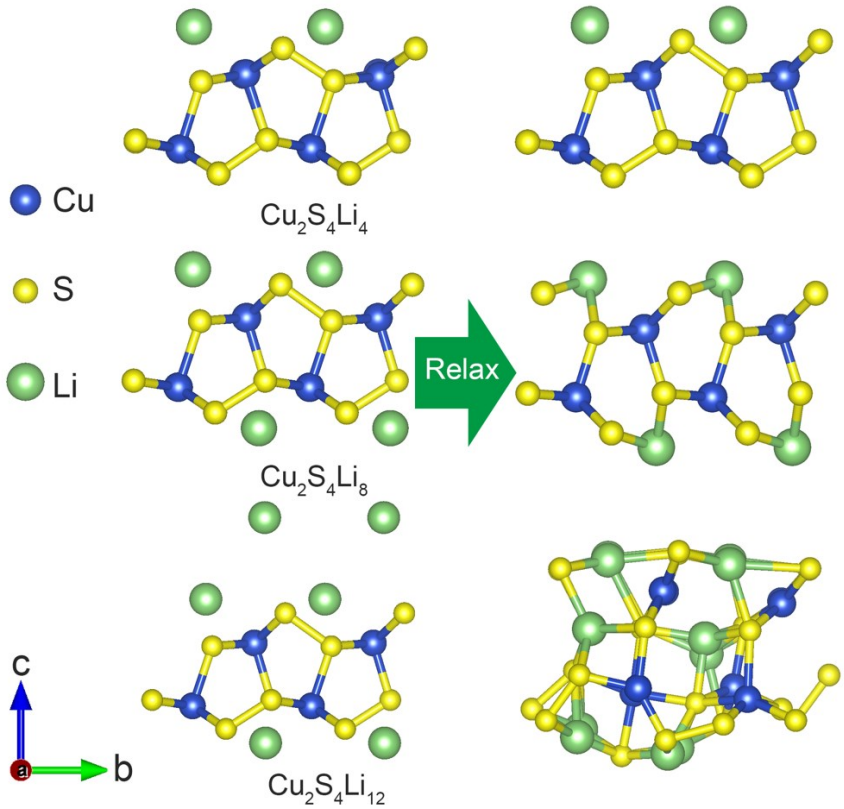

Fig. S15. The initial (left panel) and relaxed (right panel) structures of Li adsorbed on α-CuS_2 sheet.

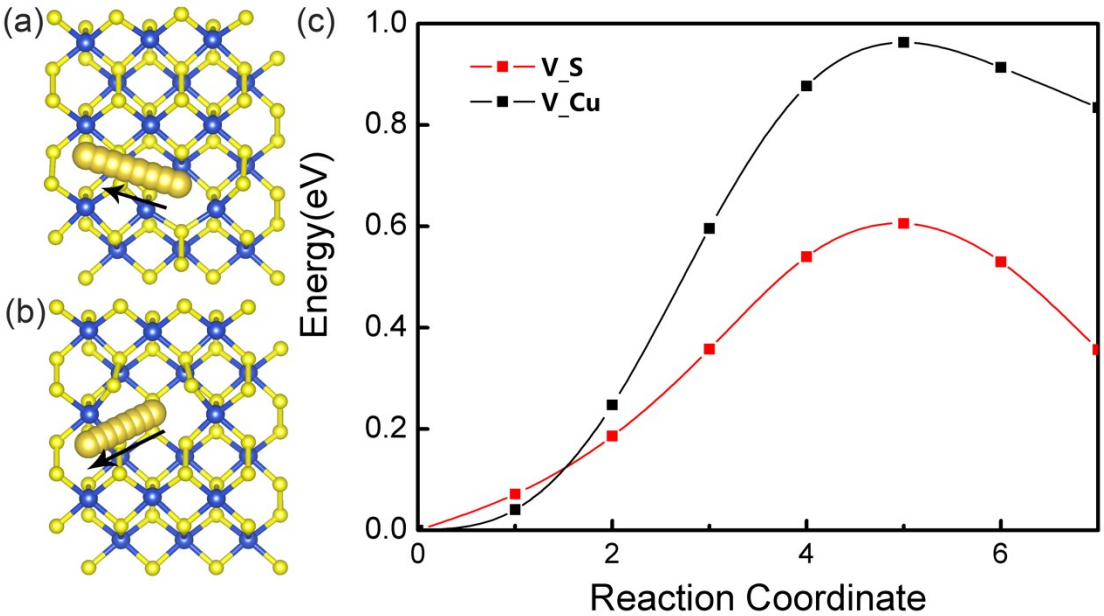

Fig. S16. The relative energy profile of Na escape from Cu-V (a) and S-V (b) diffusion path (c).

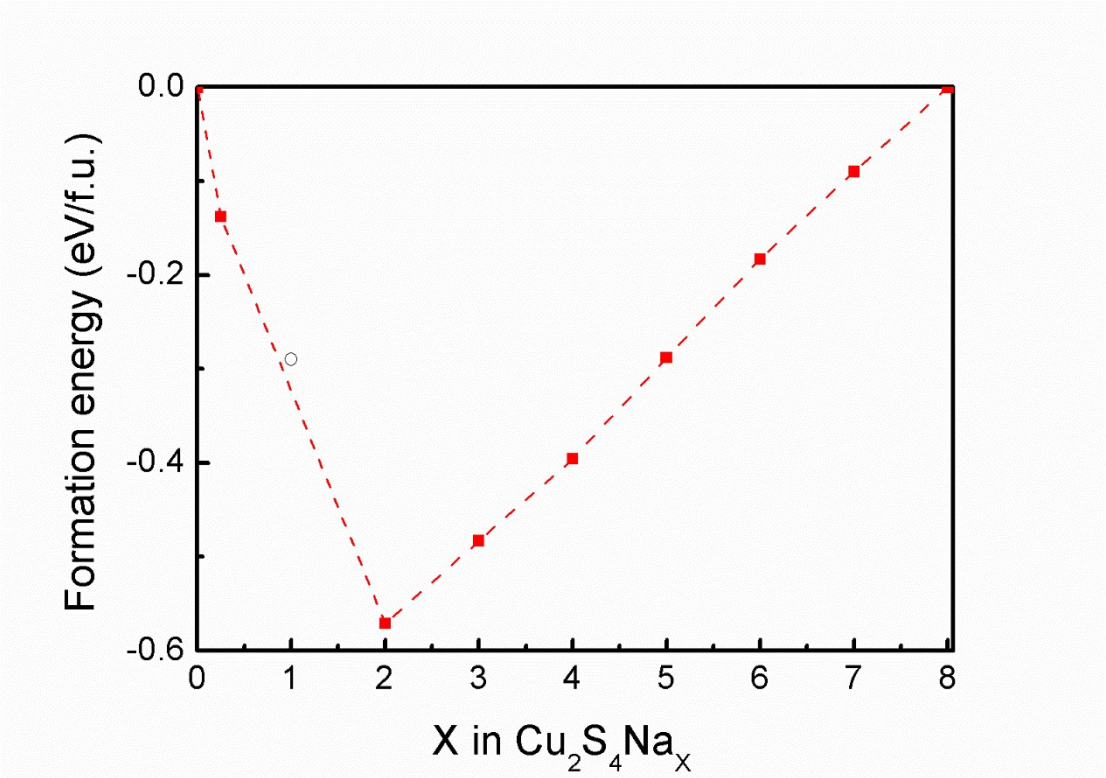

Fig. S17. The fluctuations of formation energy as a function of X in $Cu_2S_4Na_x$. The red dash lines form the convex hull for stable phases in the whole composition range of $Cu_2S_4Na_x$.

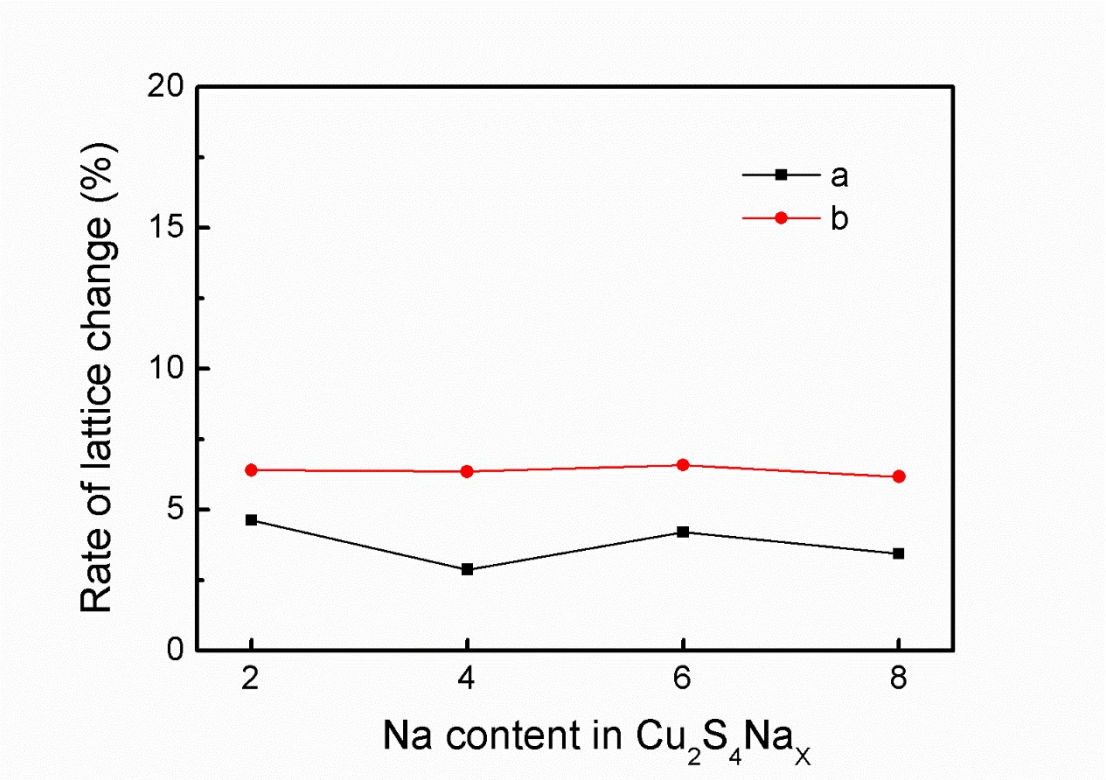

Fig. S18. Rate of lattice change in a and b (red) direction as the function of Na content in $Cu_2S_4Na_x$.

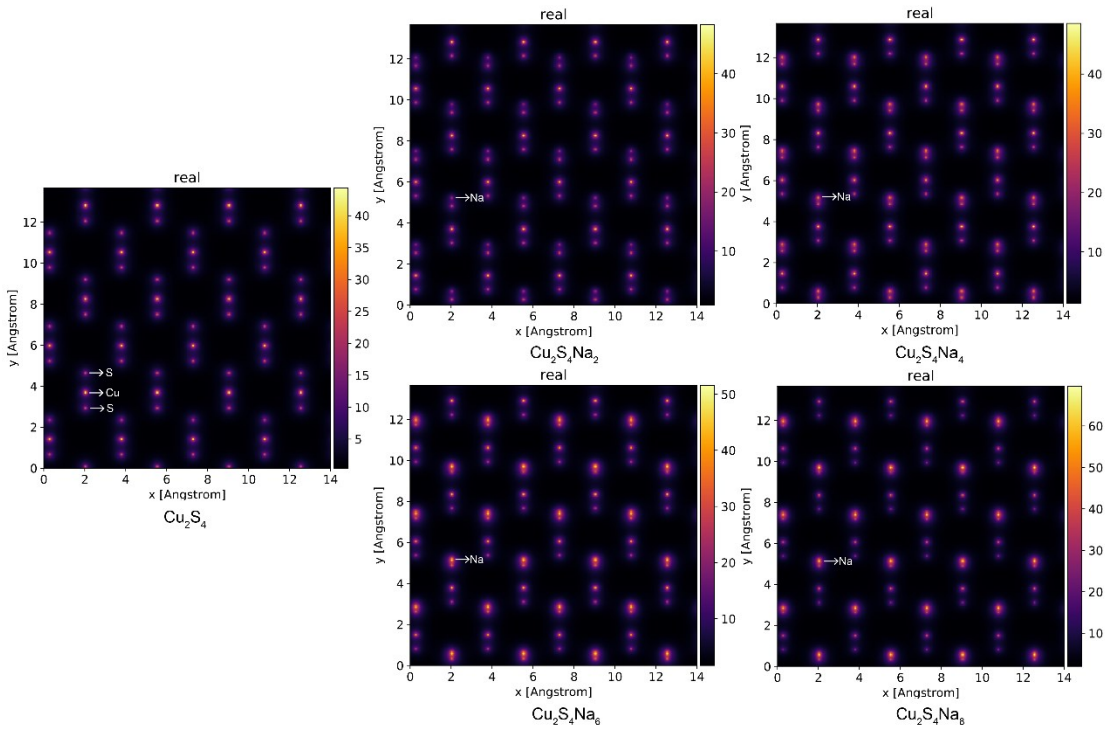

Fig. S19. The simulation of transmission electron microscope（TEM）with the stoichiometries of Cu_2S_4, $Cu_2S_4Na_2$, $Cu_2S_4Na_4$, $Cu_2S_4Na_6$, and $Cu_2S_4Na_8$.

Tab. S1. Integrated decomposed DOS for S-s, S-p, Cu-s, Cu-p, and Cu-d orbitals, in units of states per cell.

	S-s	S-p	Cu-s	Cu-p	Cu-d
Cu_2S_4	0.650	8.669	0.525	0.736	18.949
$Cu_2S_4Na_8$	0.684	8.227	1.011	0.868	18.314

Tab. S2. Bond length and slab thickness of the α-CuS_2 sheet at various Na coverage and after de-sodiation.

X in $Cu_2S_4Na_x$	S-S bond length (Å)	S-Cu bond length (Å)	slab thickness (Å)
0	2.042	2.360	4.336
1	2.151	2.386	4.386
2	2.175	2.408	4.564
3	2.167	2.406	4.547
4	2.166	2.418	4.559
5	2.166	2.403	4.542
6	2.176	2.406	4.544
7	2.168	2.406	4.537
8	2.171	2.408	4.545
After de-sodiation	2.041	3.367	4.335