Single-Atom Cobalt Encapsulated in Carbon Nanotubes as an Effective Catalyst for Enhancing Sulfur Conversion in Lithium-Sulfur Batteries

Theoretical computations

The VASP software was utilized for quantum density functional theory (DFT) investigations to determine the binding energy (E_b) between substrates and Li_2S_n (n=1, 2, 4, 6, 8). The binding energy was estimated using the following equation:

$$E_b = E_{Li_2S_n + substrate} - E_{Li_2S_n} - E_{substrate}$$

The energy of Li_2S_n, substrate, and Li_2S_n-substrates are respectively associated with E_b, $E_{substrate}$, and $ELi_2S_{n+substrate}$.

We performed calculations for each LiPS and catalyst combination using a minimum of 10 distinct initial configurations in order to determine the most favorable total SCF energy. Due to the presence of Co atoms as catalysts, we utilized unrestricted Kohn-Sham DFT to accurately represent the open-shell system. The exchange-correlation potential was chosen to be the extended gradient approximation, as proposed by Perdew, Burke, and Ernzerhof. The plane wave's cut-off energy was established at 400 eV. The energy threshold for the iterative solution of the Kohn-Sham equation was established at 10^-5 eV. The structures were relaxed until the residual stresses on the atoms decreased to below 0.05 eV $Å^{-1}$. The weak interaction was characterized using the DFT+D3 approach, which incorporates an empirical correction based on Grimme's scheme. The vacuum space was adjusted to a size greater than 15 Å, ensuring that there was no contact between periodic pictures. The Brillouin zones of Co[0001] materials and CoN_4 materials were sampled using a 3 × 3 × 1 k points grid. An ascending-nudged elastic band approach was employed to ascertain the minimum energy pathway for the transitional state scanning process.

Table S1. EXAFS fitting parameters at the Co K-edge for various samples						
Sample	Shell	N[a]	R (Å)[b]	σ^2 (Å^2·10^{-3}) [c]	ΔE_0 (eV)[d]	R factor(%)
COSAC@CNT	Co-N	4.5	1.91	9.8	-6.0	0.6
CONP@CNT	CO-CO	8.7	2.51	6.3	5.9	0.2

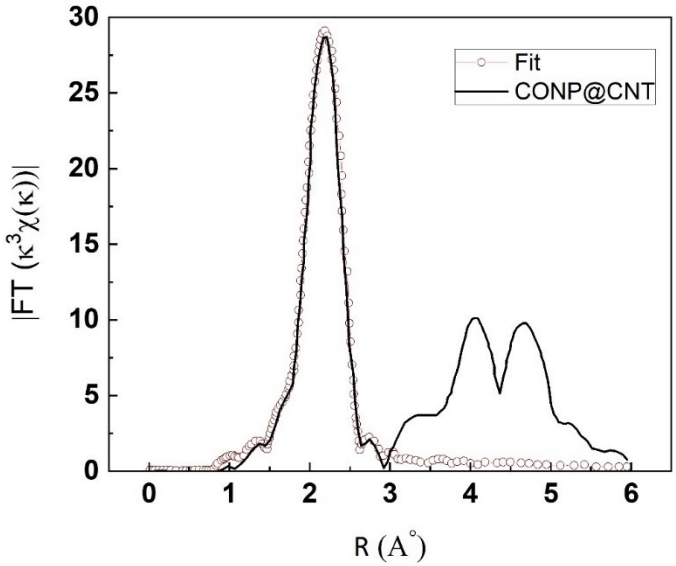

Fig.S1 EXAFS R space fitting curves of CONP@CNT

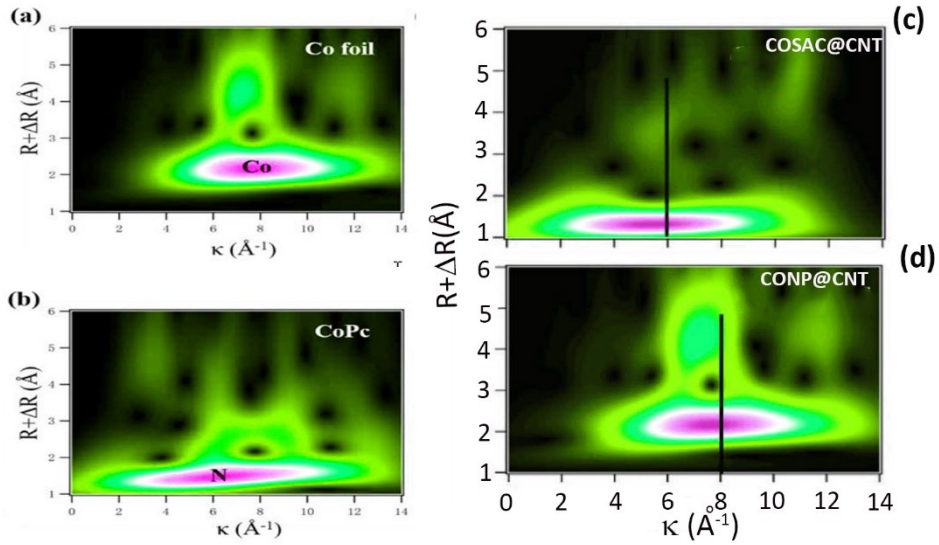

Fig.S2 wavelet

transform (FT) of (a) Co foil, (b) Co PC (c) CoSAC@CNT and (d) CoNP@CNT.

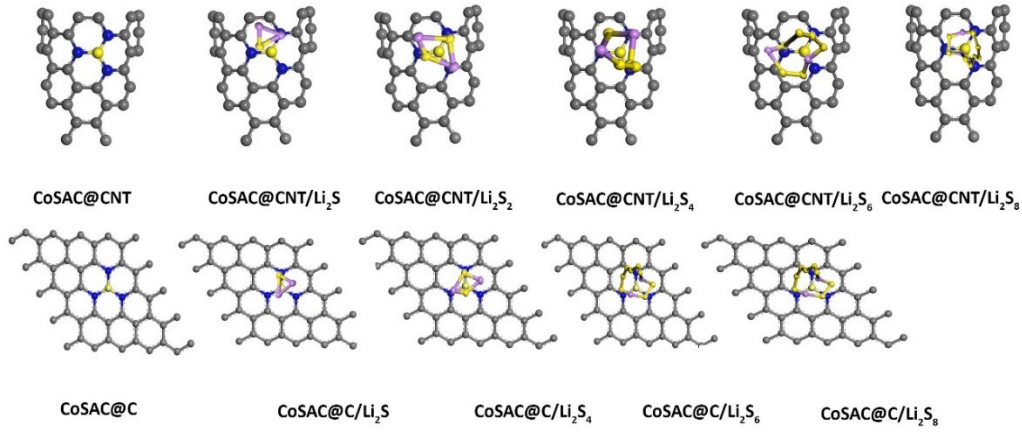

Fig.S3 Optimized LiPS structures on CNT and plain carbon sheet

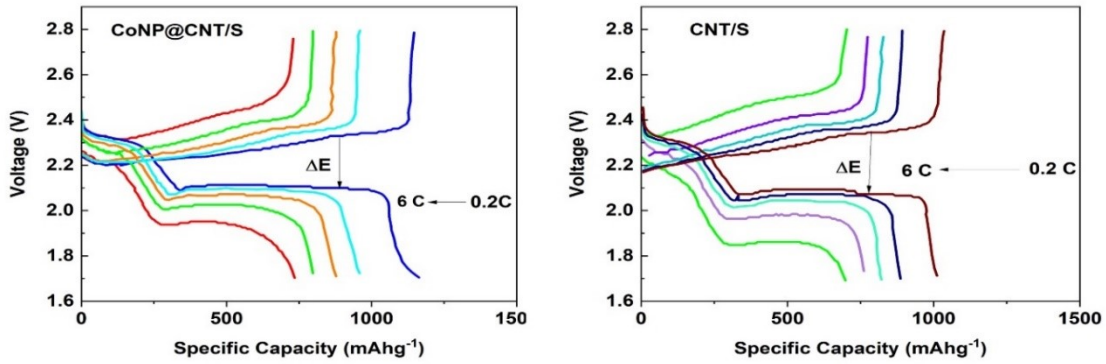

Fig.S4 Co/NGDY and NGDY electrodes charge/discharge profiles captured at 0.2 C, , 1, 2, 3.6 C, and 3.6 C.