

Supporting Information

A Dual-Functional Fibrous Skeleton Implanted with Single-Atomic Co-N_x Dispersions for Longevous Li–S Full Batteries

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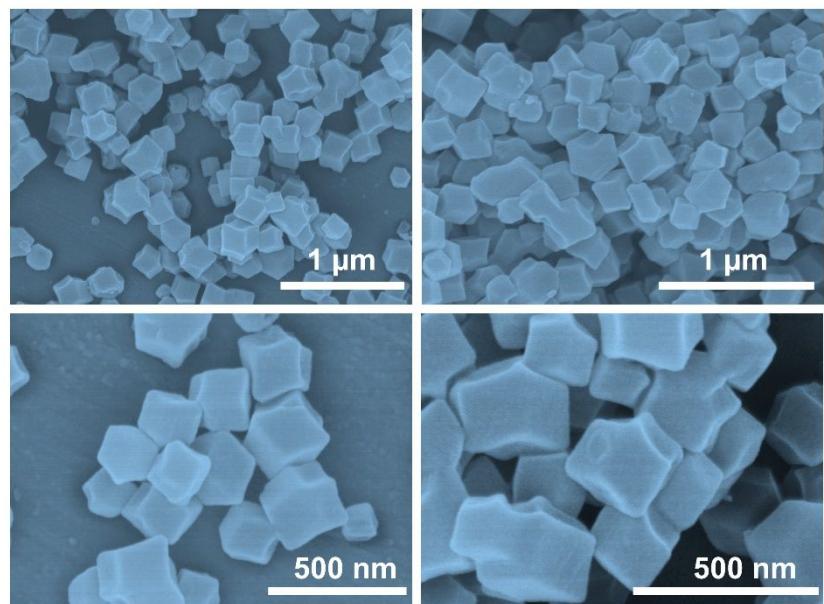


Figure S1. SEM images of as-obtained Co-ZIF8 polyhedrons.

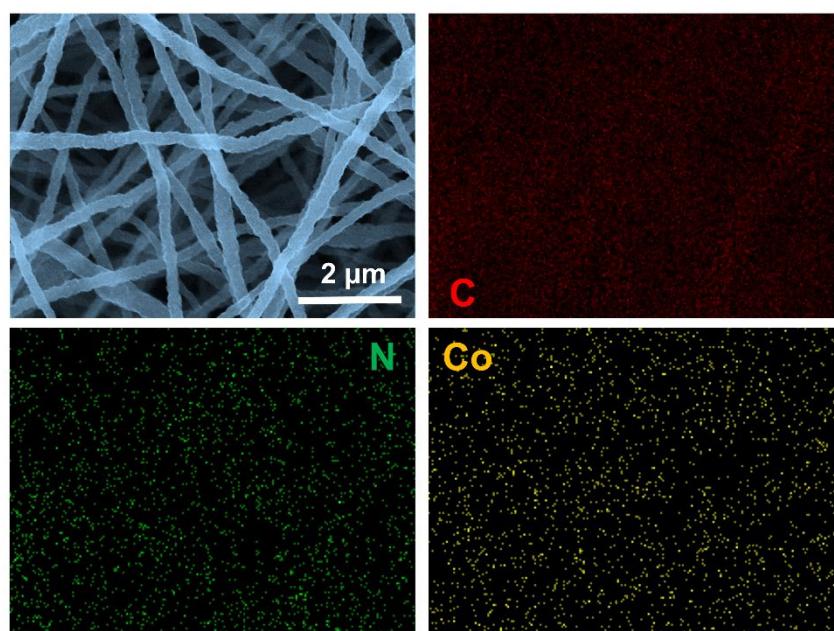


Figure S2. SEM image and corresponding elemental maps of Co-PCNF.

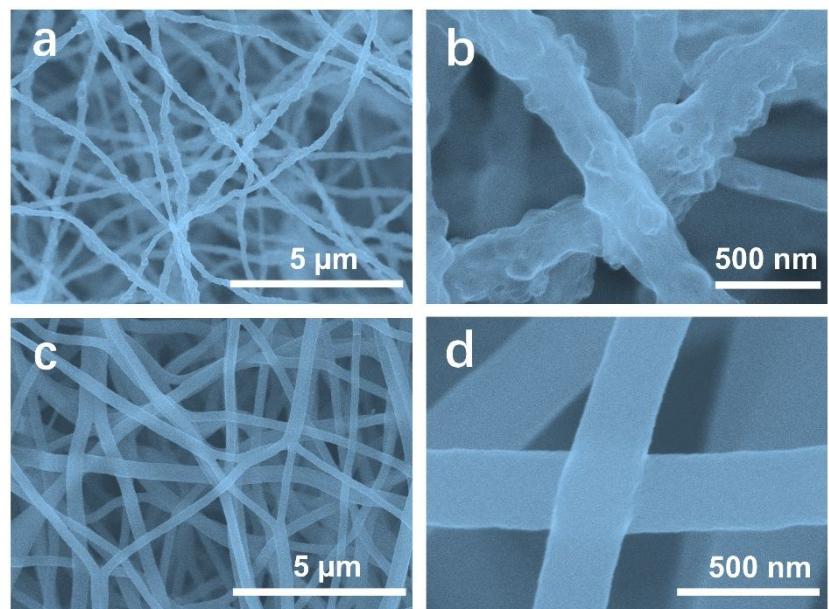


Figure S3. SEM images of (a,b) PCNF and (c,d) CNF.

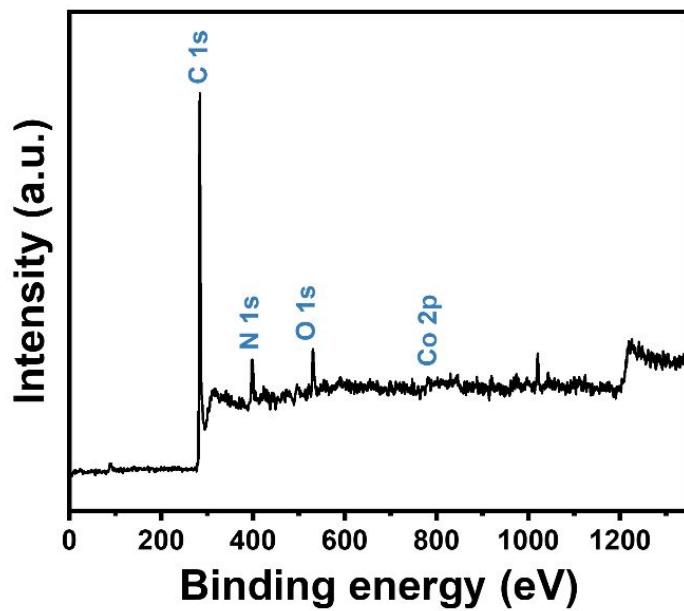


Figure S4. XPS survey spectrum of as-prepared Co-PCNF.

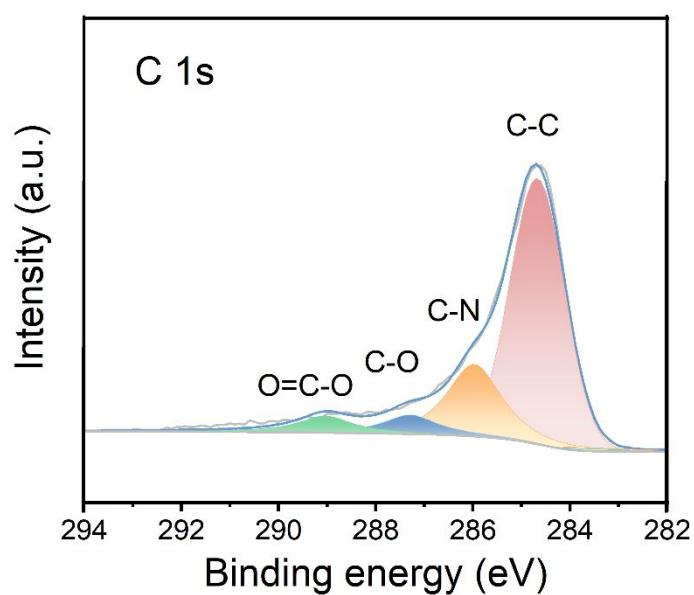


Figure S5. High-resolution XPS C 1s spectrum of Co-PCNF.

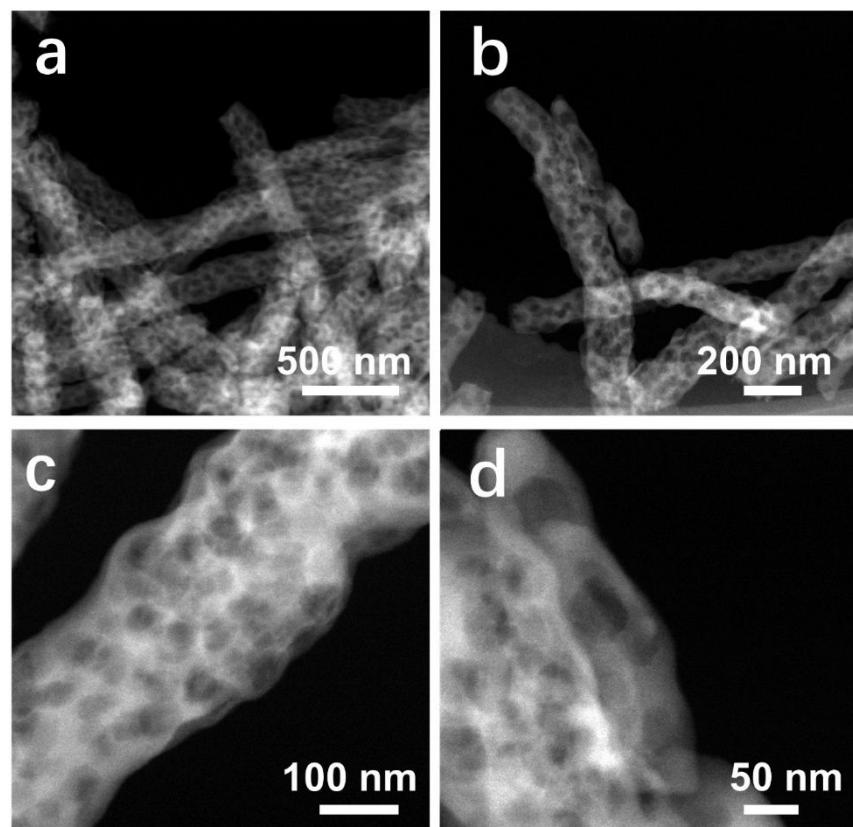


Figure S6. HAADF-STEM images of Co-PCNF.

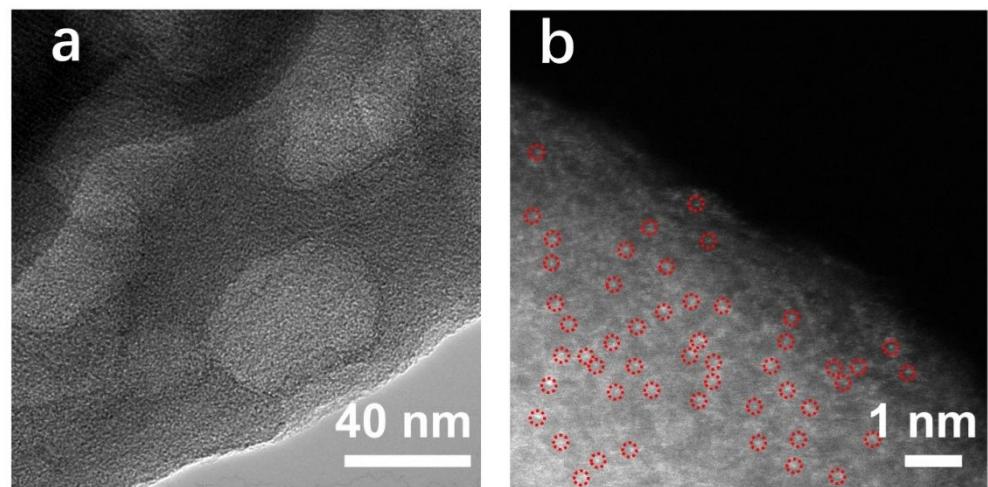


Figure S7. (a) TEM image of Co-PCNF. (b) AC-HAADF-STEM image of Co-PCNF.

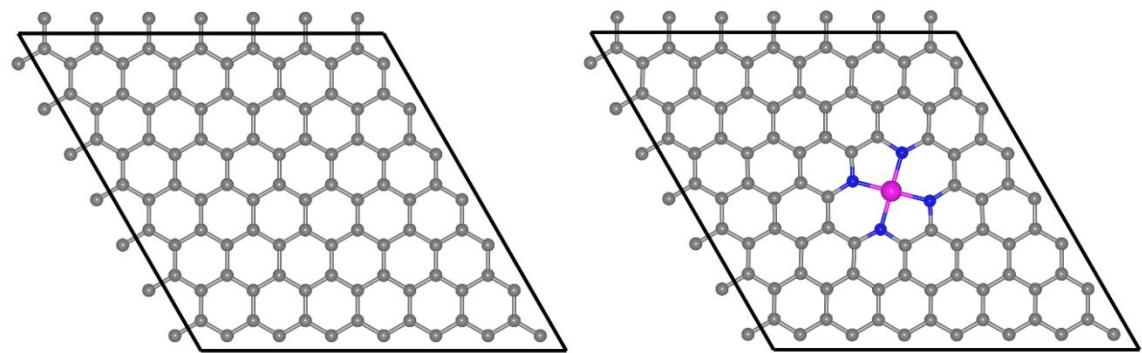


Figure S8. CNF (left panel) and Co-PCNF (right panel) models used in DFT calculations.

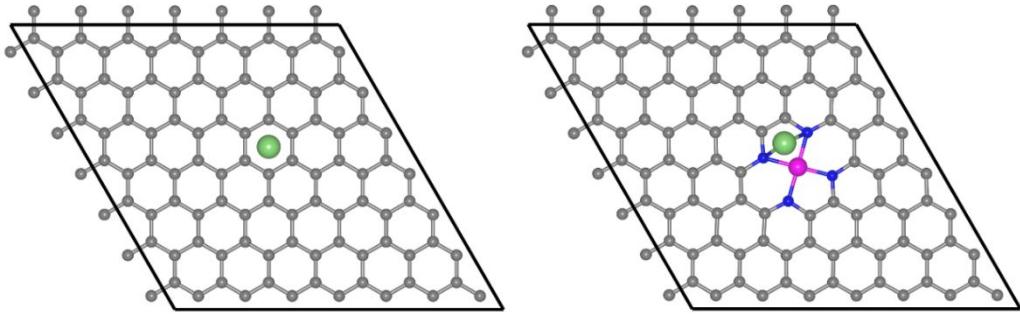


Figure S9. The optimized adsorption configurations of Li atoms on CNF (left panel) and Co-PCNF (right panel).

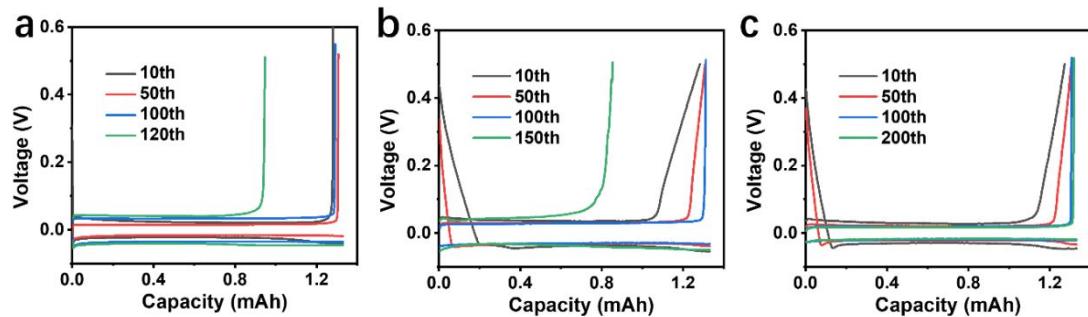


Figure S10. Voltage-capacity curves of (a) Cu foil, (b) PCNF and (c) Co-PCNF at different cycles.

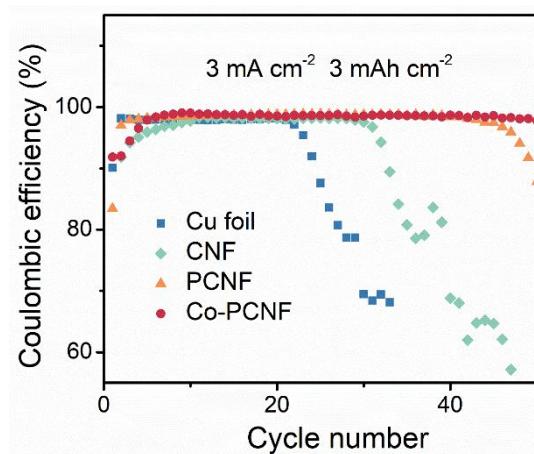


Figure S11. Coulombic efficiencies of different substrates at a current density of 3.0 mA cm^{-2} under a capacity of 3.0 mAh cm^{-2} .

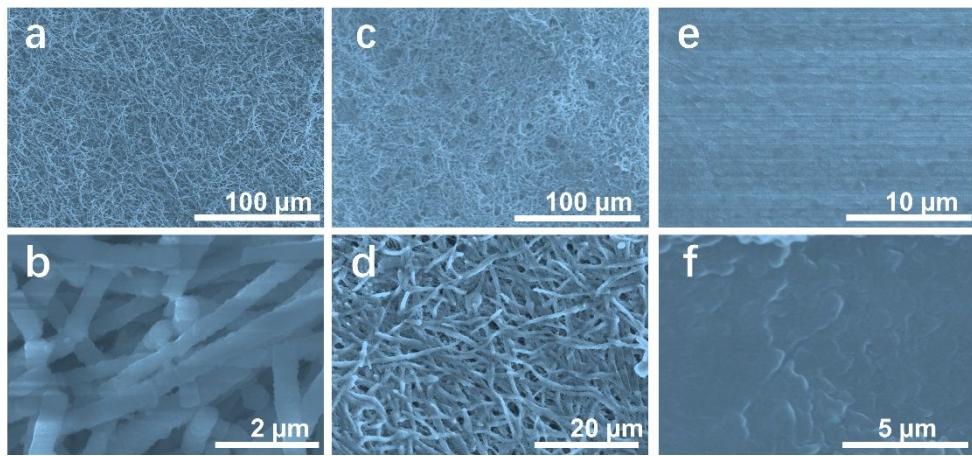


Figure S12. Post-mortem SEM images of the Co-PCNF electrodes with different capacities of Li plating: (a,b) 0.5 mAh cm^{-2} ; (c,d) 1.0 mAh cm^{-2} ; (e,f) 5.0 mAh cm^{-2} .

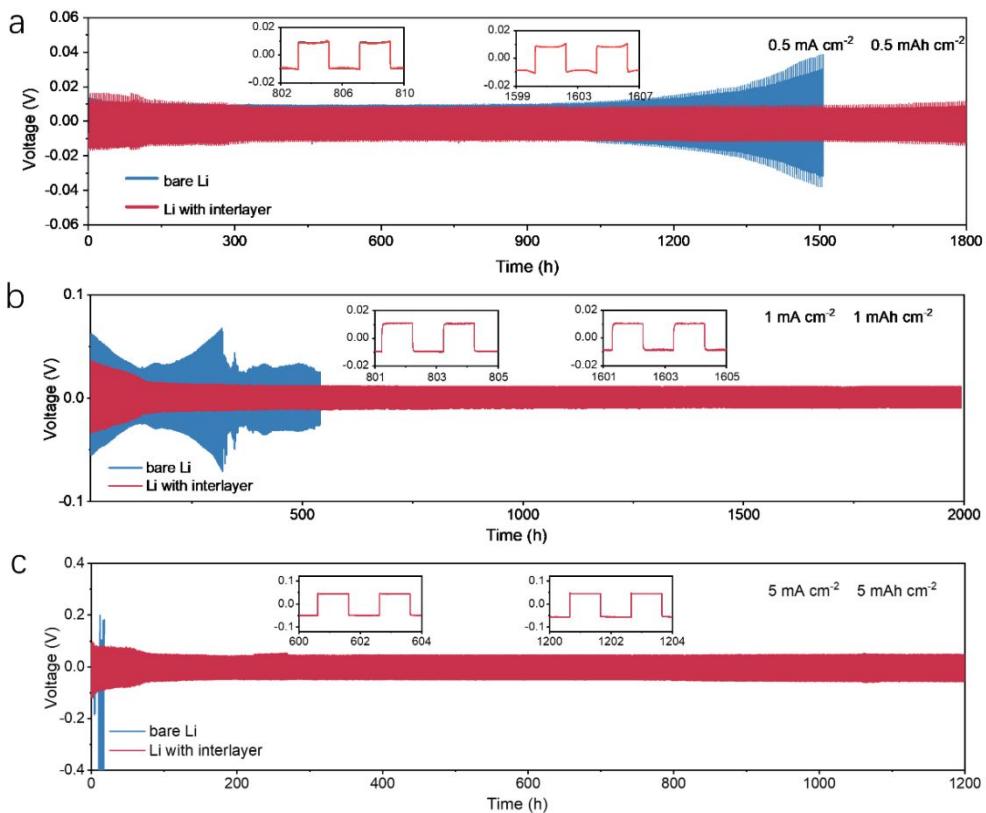


Figure S13. Voltage-time profiles of Li plating/stripping for bare Li and Co-PCNF/Li symmetric cells at different capacities and current densities.

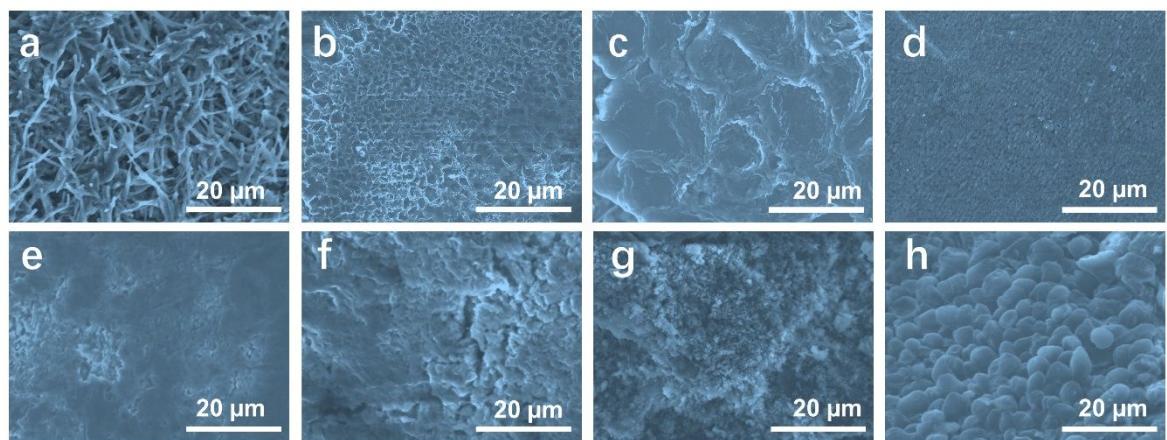


Figure S14. *Ex situ* SEM observations of the morphology evolution for (a-d) Co-PCNF/Li symmetric cell and (e-h) bare Li symmetric cell at a cycling time of (a,e) 10 h; (b,f) 100 h; (c,g) 200 h; (d,h) 400 h.

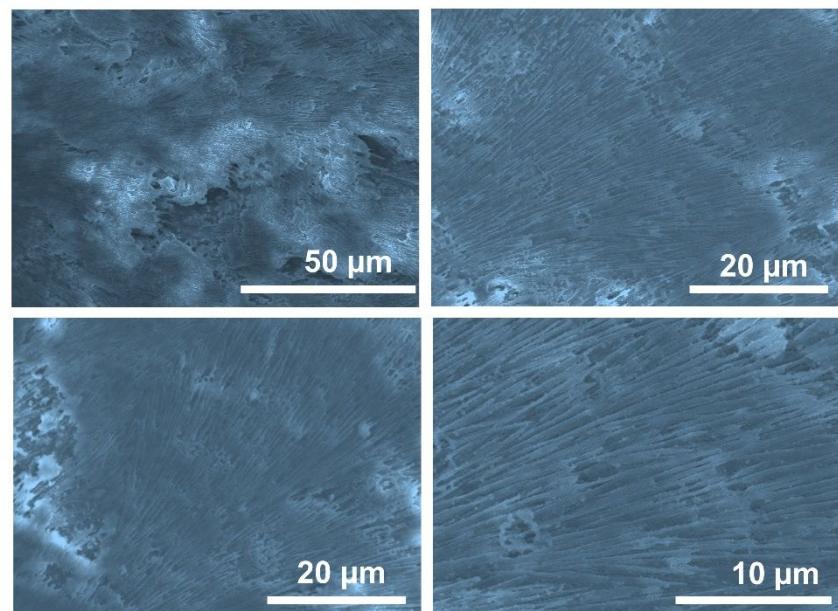


Figure S15. SEM images of Co-PCNF matrix loaded with sulfur.

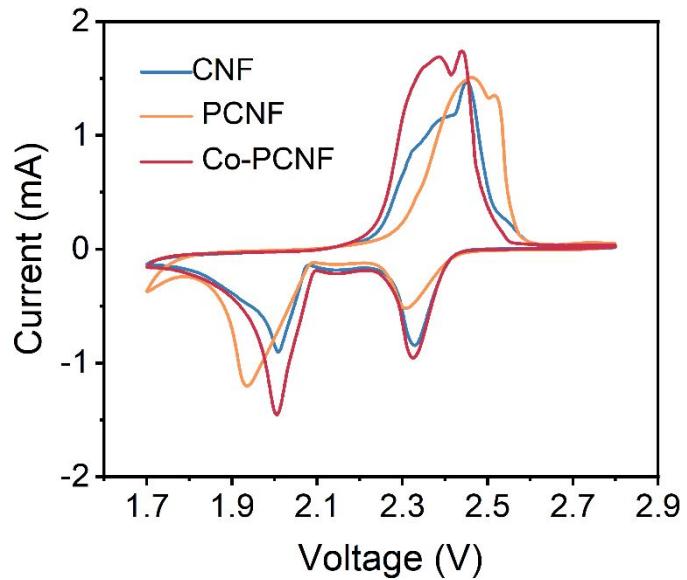


Figure S16. CV curves of S/Co-PCNF, S/PCNF and S/CNF cathodes at a scan rate of 0.05 mV s^{-1} .

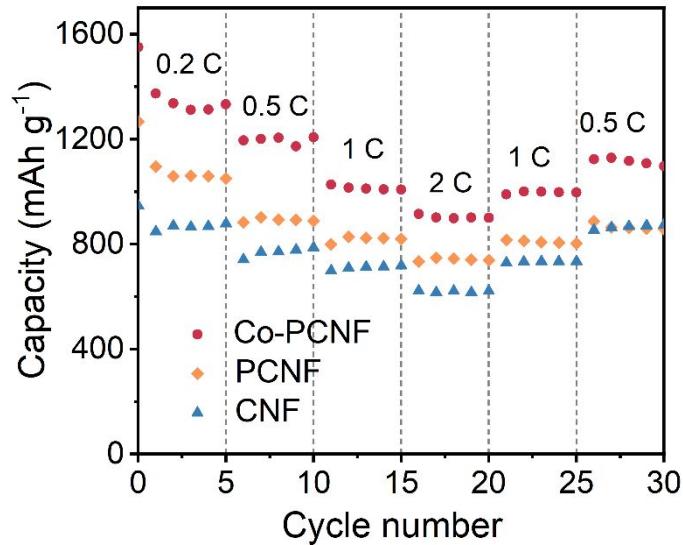


Figure S17. Rate performances of S/Co-PCNF, S/PCNF and S/CNF cathodes.

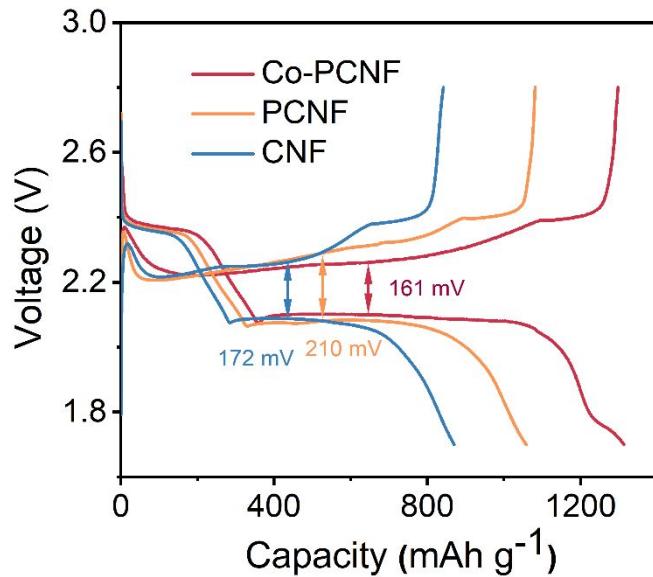


Figure S18. GCD curves of S/Co-PCNF, S/PCNF and S/CNF cathodes at 0.2 C.

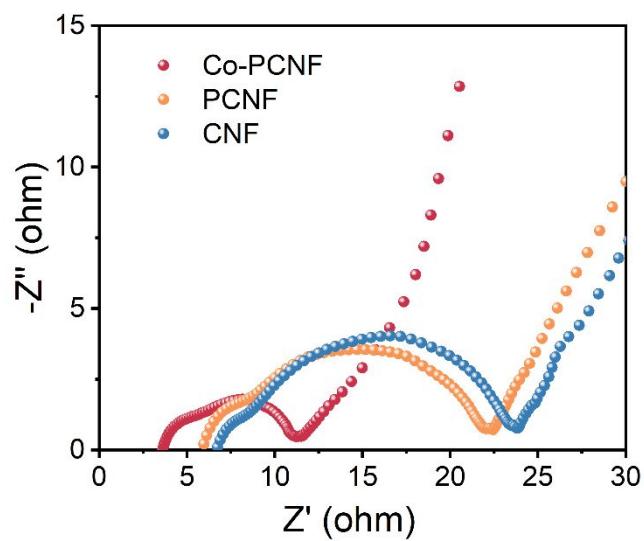


Figure S19. EIS profiles of S/Co-PCNF, S/PCNF and S/CNF cathodes.

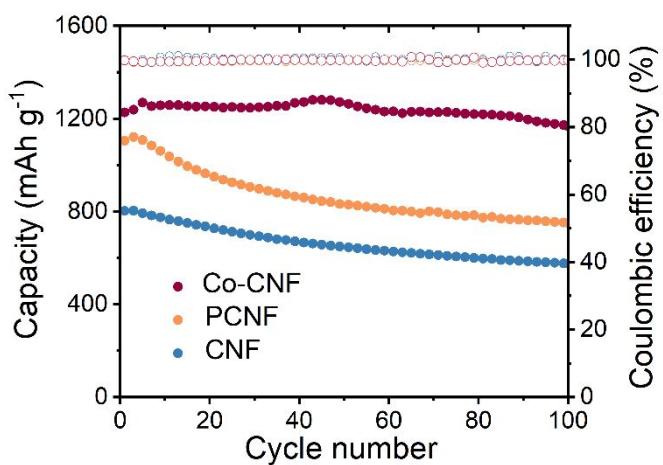


Figure S20. Cycling performances of S/Co-PCNF, S/PCNF and S/CNF cathodes at 0.5 C.

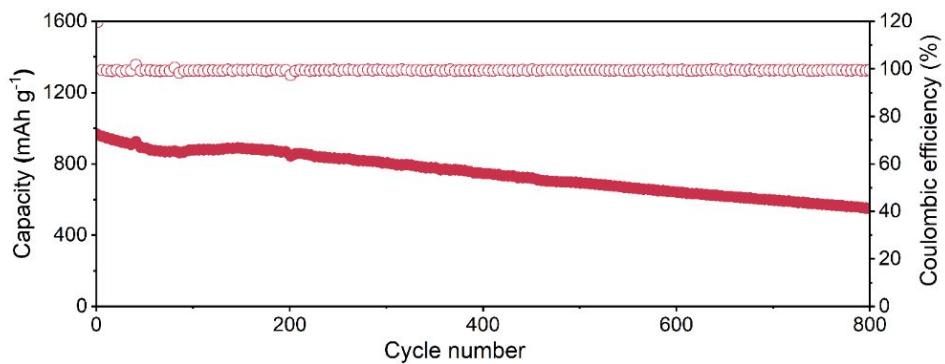


Figure S21. Cycling performance of S/Co-PCNF cathode at 1.0 C.

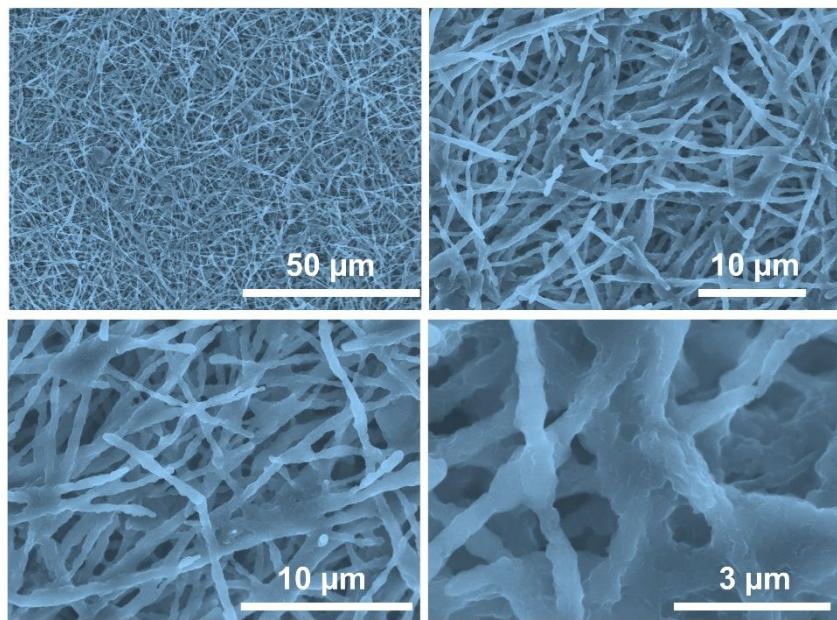


Figure S22. Post-mortem SEM images of S/Co-PCNF cathode at 1.0 C after 800 cycles.

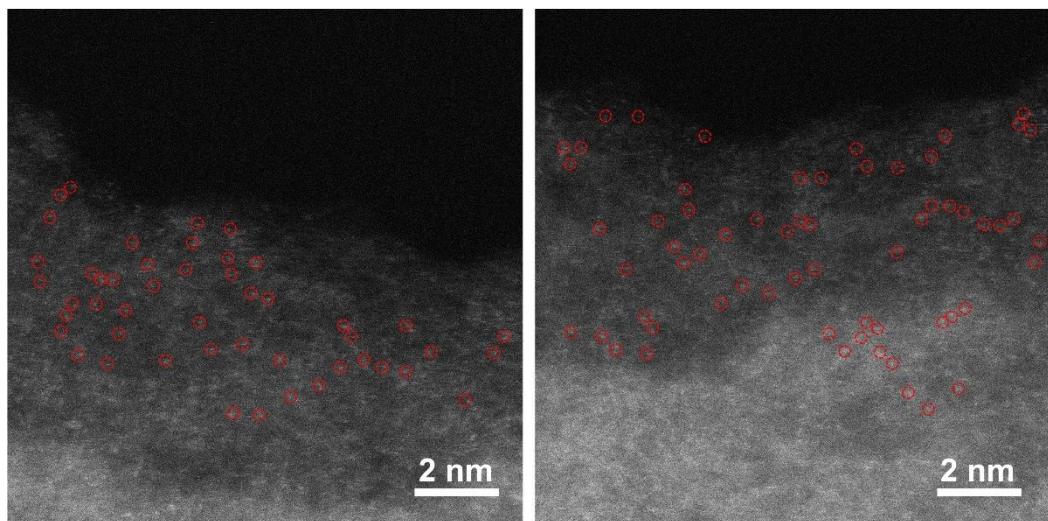


Figure S23. Atomic-resolution HAADF-STEM images of the cycled Co-PCNF after 100 cycles at 0.5 C, showing the preservation of single atoms.

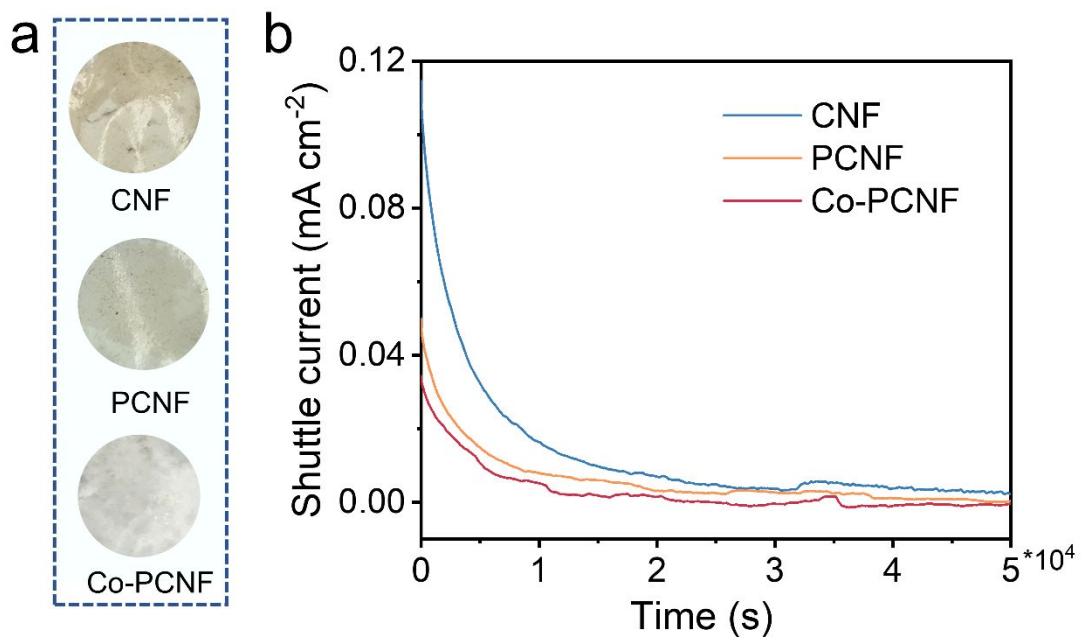


Figure S24. (a) Photographs of the separators of the S/Co-PCNF, S/PCNF and S/CNF paired cells after 100 cycles at 0.5 C; (b) Shuttle current curves of Li–S batteries with S/Co-PCNF, S/PCNF and S/CNF cathodes.

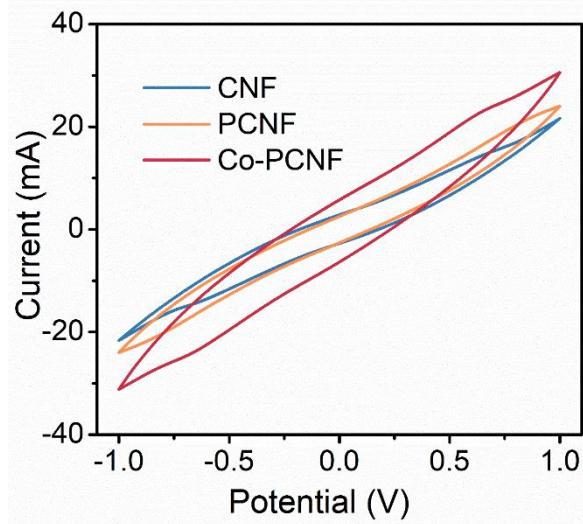


Figure S25. CV profiles of symmetric cells of Co-PCNF, PCNF and CNF at a scan rate of 50 mV s^{-1} .

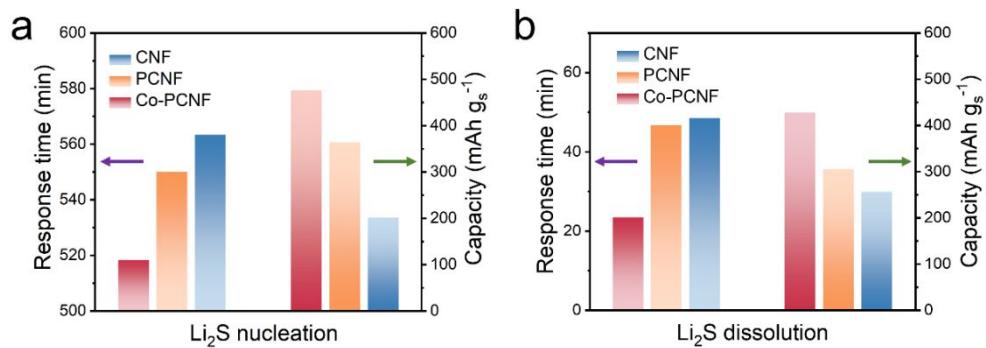


Figure S26. Response time and calculated specific capacity during Li_2S nucleation and dissolution of CNF, PCNF and Co-PCNF paired cells.

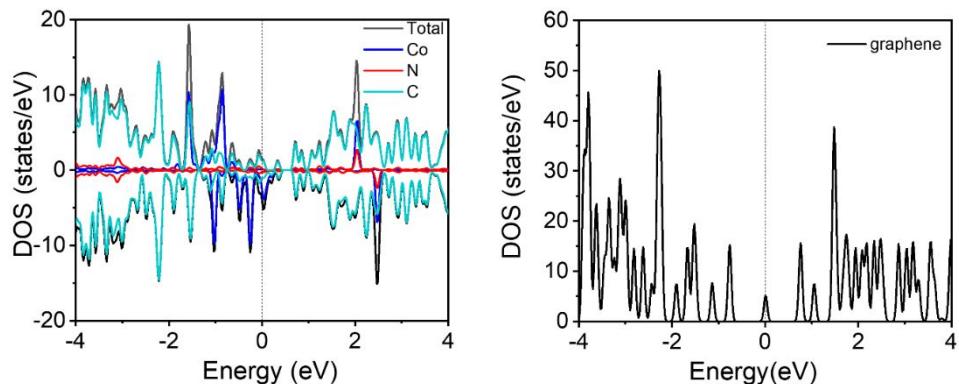


Figure S27. pDOS spectra of Co-PCNF and CNF monolayer.

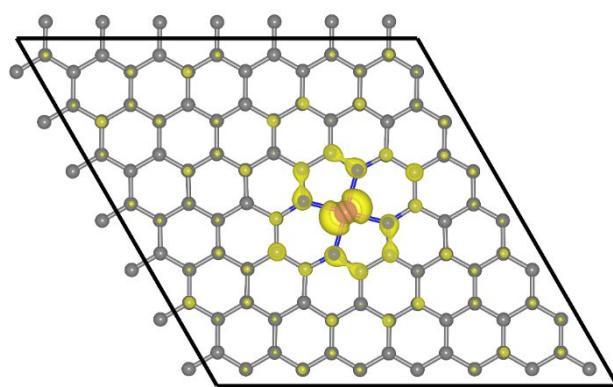


Figure S28. Partial charge density around the Fermi level.

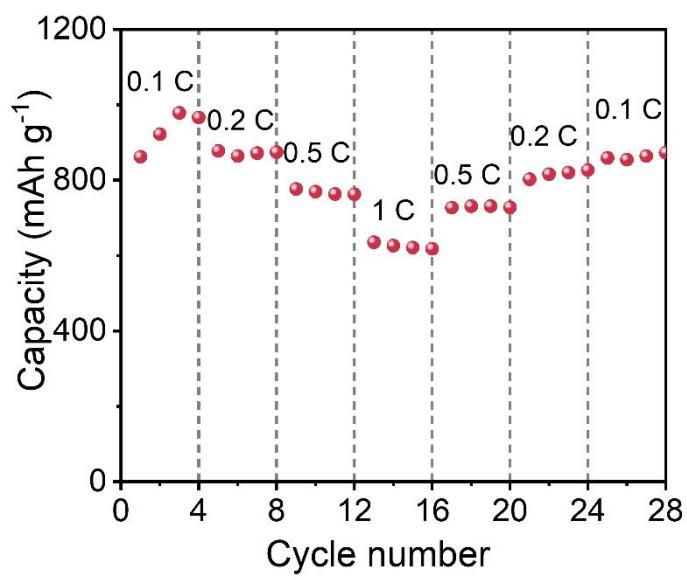


Figure S29. Rate performance of full battery with a sulfur loading of 3.5 mg cm^{-2} .

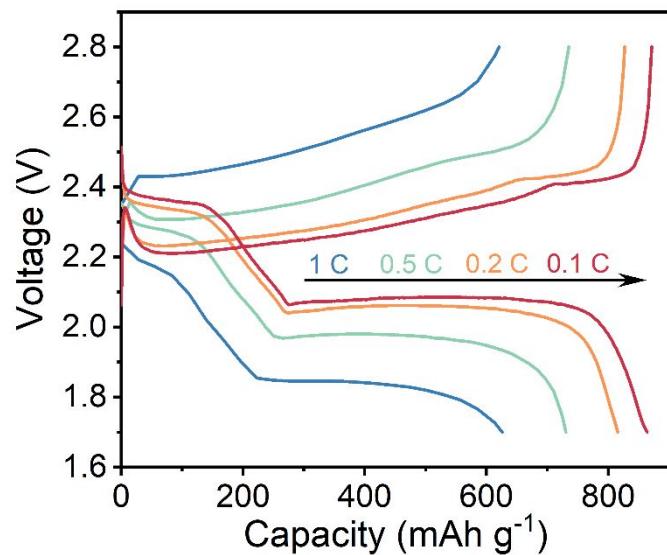


Figure S30. GCD curves of full battery at different current densities.

Table S1. Comparison of single-atom based sulfur hosts between this work and reported studies.

Materials	Sulfur loading	Rate capability	Rate/cycles/capacity retention	Ref
S-SAV@NG	2 mg cm ⁻²	1230 mAh g ⁻¹ (0.2 C)/645 mAh g ⁻¹ (3 C)	0.5 C/400/70.64%	[1]
Co-SAs@NC	2 mg cm ⁻²	1425 mAh g ⁻¹ (0.1 C)/680 mAh g ⁻¹ (10 C)	1 C/600/72%	[2]
S@Co-N/G	2 mg cm ⁻²	1210 mAh g ⁻¹ (0.2 C)/618 mAh g ⁻¹ (4 C)	1 C/500/71.6%	[3]
HFeNG-S	3 mg cm ⁻²	1298 mAh g ⁻¹ (0.2 C)/810 mAh g ⁻¹ (5 C)	3 C/600/85.7%	[4]
S-Ni@NG	1.5 mg cm ⁻²	1598 mAh g ⁻¹ (0.1 C)/612 mAh g ⁻¹ (10 C)	1 C/500/78%	[5]
Fe-PNC	1.3 mg cm ⁻²	1138.6 mAh g ⁻¹ (0.1 C)/280 mAh g ⁻¹ (2 C)	0.5 C/300/80.5%	[6]
Li2S@NC:SAFe	2.0-2.3 mg cm ⁻²	989 mAh g ⁻¹ (0.5 C)/589 mAh g ⁻¹ (12 C)	2 C/1000/54.3%	[7]
SC-Co	1.2 mg cm ⁻²	1130 mAh g ⁻¹ at 0.5 C	0.5 C/300/74.1%	[8]
CoSA-N-C	-	1574 mAh g ⁻¹ (0.05 C)/624 mAh g ⁻¹ (5 C)	1 C/1000/65%	[9]
NC@SA-Co	1.0 mg cm ⁻²	-	0.1 C/700/59.4%	[10]
Fe/C ₂ N	1 mg cm ⁻²	1480 mAh g ⁻¹ (0.1 C)/683 mAh g ⁻¹ (5 C)	1 C/900/75.14%	[11]
CNT@SACo	1 mg cm ⁻²	1496 mAh g ⁻¹ (0.1 C)/641 mAh g ⁻¹ (2 C)	1 C/500/67.6%	[12]
Mo-N ₂ /C	2 mg cm ⁻²	1360.2 mAh g ⁻¹ (0.1 C)/743.9 mAh g ⁻¹ (5 C)	2 C/550/90.1%	[13]
Fe-N ₂ /CN	1.3– 1.5 mg cm ⁻²	1301 mAh g ⁻¹ (0.2 C)/ 607 mAh g ⁻¹ (5 C)	2 C/2000/78%	[14]
Fe–N/MHCS	2 mg cm ⁻²	1110 mAh g ⁻¹ (0.2 C)/ 949 mAh g ⁻¹ (2 C)	0.1 C/200/69.2%	[15]
SA-ZnMXene	1.7 mg cm ⁻²	1136 mAh g ⁻¹ (0.2 C)/ 517 mAh g ⁻¹ (6 C)	4 C/400/88%	[16]
Mn/C-(N, O)	1.3 mg cm ⁻²	1330 mAh g ⁻¹ (0.2 C)/900 mAh g ⁻¹ (1 C)	0.5 C/600/52%	[17]
Co-PCNF	1.7 mg cm⁻²	1373.5 mAh g⁻¹(0.2 C)/ 914.3 mAh g⁻¹ (2 C)	1 C/800/57%	This work

Table S2. Comparison of device performances of Li–S batteries employing “two-in-one” hosts.

Materials	Current density (mA cm ⁻²), capacity (mAh cm ⁻²)	Anode cycle length (h)	Sulfur loading (mg cm ⁻²)	Rate (C)	Cycle number	Areal capacity (mAh cm ⁻²)	Ref.
Co/N-PCNSs	0.5,0.5	350	0.8-1.0	0.2	60	-	[18]
Cu/Ni CF	1,2	500	3.2	~0.4	260	2.4	[19]
VN	2,2	1200	4.0	0.5	~400	4.8	[20]
TiN-VN	2,1	1000	5.6	0.1	100	5.5	[21]
CoNi@P NCFs	1,2	1000	7.0	0.2	50	5.8	[22]
V ₈ C ₇ -VO ₂	3,1	400	9.2	0.1	100	7.3	[23]
Co-PCNF	3,3	1400	6.9	0.1	50	7.1	This work

Table S3. Comparison of electrochemical performances of Li anode employing selected conductive materials.

	Materials	Current density (mA cm ⁻²), capacity (mAh cm ⁻²)	Life span	Overpotential	Ref
Carbon cloth (CC)	CC	4, 4	350 h	18 mV	[24]
	Zn@NC@CC	3, 1	400 h	9.6 mV	[25]
	CC@CN-Co	5, 5	1000 h	20 mV	[26]
	<i>N,P</i> -codoped CC (NPCC)	5, 1	240 h	140 mV	[27]
	Au-CC	3, 1	350 h	40 mV	[28]
	CC/Co ₃ O ₄ -NC	1, 1	1000 h	18 mV	[29]
	NRA-CC	2, 4	1000 h	35 mV	[30]
	CC@ZnO	0.5, 1	1500 h	42 mV	[31]

CNF	3D hollow carbon fiber (3D-HCF)	2, 1	600 h	20 mV	[32]
	CNF/Ag	1, 1	400 h	6.5 mV	[33]
	Ag–CNFs	0.5, 1	500 h	25 mV	[34]
	CFs@Au	1, 2	750 h	60 mV	[35]
	F-rich macroporous CNFs	3, 3	400 h	50 mV	[36]
	NCH@CFs	3, 3	500 h	23 mV	[37]
	Co/CNFs interlayers	0.5, 0.5	1500 h	38.1 mV	[38]
Graphene	CNF/TiN	1, 1	600 h	14.1 mV	[39]
	MnO ₂ /graphene foam	2, 1	300 h	96 mV	[40]
	rGO	3, 1	70 h	80 mV	[41]
	Ag/GO	40, 1	50 h	120 mV	[42]
	crumpled graphene ball	0.5, 1	750 h	-	[43]
	Au-GA	4, 4	1200 h	87 mV	[44]
	3D rGO Foam	1, 1	500 h	31.2 mV	[45]
	3D N-doped graphene	1, 1	1400 h	15 mV	[46]
Cu nanowire	G/Li	5, 2.5	1000	-	[47]
	CuNW network	1, 2	550 h	40 mV	[48]
	Cu-CNF	3, 3	400 h	20 mV	[49]
Cu foam	CuNW-P	1, 1	1000 h	18 mV	[50]
	3D Cu@Ag 3D foam host	2, 2	300h	6.5 mV	[51]
	3D Porous Cu	1, 0.2	1000h	-	[52]
	Cu mesh/Li	2, 1	120 h	-	[53]
	3D HPC/CF	0.5, 1	600 h	27 mV	[54]
	porous Cu current collector porous Cu	1, 1	200 h	20 mV	[55]
	3D Cu ₂ S NWs/Cu foam	1, 1	140 h	20 mV	[56]
	3D Cu	0.2, 0.5	600 h	50 mV	[57]
	CuO-Cu mesh	5, 1	1000 h	100 mV	[58]
Ni foam	Ni foam	1, 5	40 h	200 mV	[59]
	3D porous Ni	1,1	600 h	-	[60]
	NiCo ₂ O ₄ @ Ni foam	1, 1	1000 h	16 mV	[61]

	V ₂ O ₅ @ Ni foam	1, 1	1600 h	18 mV	[62]
	ZnO@ Ni foam	5, 1	250 cycles	45 mV	[63]
	Ni ₂ P@nickel foam	2, 1	2000 h	-	[64]
	Ni ₃ N@ nickel foam	5, 1	160 cycles	34 mV	[65]
	NCNT/NF	3, 3	400 h	62 mV	[66]
	Co-PCNF	5, 5	1200 h	40 mV	This work

Table S4. Comparison of electrochemical performances of sulfur cathode employing selected conductive materials.

	Material	Sulfur loading	Rate capability	Rate/cycles/capacity decay per cycle	Ref
Carbon cloth (CC)	CoP@G/CC	2 mg cm ⁻²	1371.9 mAh g ⁻¹ (0.2 C)/ 930.1 mAh g ⁻¹ (3 C)	2C/500/0.03%	[67]
	VSe ₂ -VG@ CC	1.4–1.7 mg cm ⁻²	1480 mAh g ⁻¹ (0.2 C)/ 450 mAh g ⁻¹ (5.0 C)	5 C/800/0.039%	[68]
	CC@WS ₂	1.0–1.2 mg cm ⁻²	1581 mAh g ⁻¹ (0.1 C)/ 448 mA h g ⁻¹ (3 C)	0.5 C/500/0.0313%	[69]
	CC-CoS ₂	1.2 mg cm ⁻²	1481 mAh g ⁻¹ (0.1 C)/ 823 mAh g ⁻¹ (4 C)	4C/1000/0.021%	[70]
	ZnS _{1-x} /CC	1.7 mg cm ⁻²	1043 mAh g ⁻¹ (0.2 C)/ 374 mAh g ⁻¹ (5 C)	1 C/500/0.04%	[71]
	CC@CoP/C	1.81 mg cm ⁻²	1201 mAh g ⁻¹ (0.1 C)/ 708 mA h g ⁻¹ (2 C)	2C/600/0.016%	[72]
	CC@SnO ₂ @TMS	2.75 mg cm ⁻²	1500 mAh g ⁻¹ (0.2 C)/ 860 mAh g ⁻¹ (5 C)	5 C/4000/0.009%	[73]
	CC/VN	8.1 mg cm ⁻²	1310.8 mAh g ⁻¹ (0.1 C)/ 591.6 mAh g ⁻¹ (5 C)	0.1C/250/0.078%	[74]
Carbon nanotube (CNT)	CNT paper	2.0 mg cm ⁻²	1093 mAh g ⁻¹ (0.38 mA cm ⁻²)/ 814 mA·h g ⁻¹ (1.51 mA cm ⁻²)	0.05 C/150/0.2%	[75]
	CNT-UGF	2.4 mg cm ⁻²	-	0.5 C/400/0.063%	[76]
	HKUST-1/CNT	1mg cm ⁻²	1,263 mAh g ⁻¹ at (0.2 C)/449 mAh g ⁻¹ (10 C)	0.2 C/500/0.08%	[77]
	porous CNT microsphere s	~2.0 mg cm ⁻²	1200 mAh g ⁻¹ (0.2 C)/785 mAh g ⁻¹ (3.0 C)	1 C/500/0.046%	[78]
	CNT@MPC	1 mg cm ⁻²	1200 mAh g ⁻¹ (0.1 C)/800	0.1 C/200/0.05%	[79]

			mAh g ⁻¹ (5 C)		
CNTs/Co ₃ S ₄ -NBs	1.2 mg cm ⁻²	1330 mAh g ⁻¹ (0.2 C)/ 702 mAh g ⁻¹ (5 C)	1 C/500/0.042%	[80]	
CoP-CNT	3 mg cm ⁻²	-	1 C/200/0.018%	[81]	
Macropore CNT particles (M-CNTPs)	1 mg cm ⁻²	1343 mA h g ⁻¹ (0.2 C)/ 992 mA h g ⁻¹ (2 C)	2 C/100/0.29%	[82]	
Carbon nanofiber (CNF)	nitrogen-doped porous carbon nanofibers (NPCN)	6.1 mg cm ⁻²	1248 mA h g ⁻¹ (0.05 C)/ 702 mA h g ⁻¹ (2 C)	0.5 C/400/0.04%	[83]
	CPZC	1.5 mg cm ⁻²	1200 mAh g ⁻¹ (0.2 C)/622.6 mAh g ⁻¹ (10 C)	1 C/2000/0.016%	[84]
	porous CNFs	2.0 mg	954 mAh g ⁻¹ (0.5 C)/ 602.2 mAh g ⁻¹ (2 C)	0.5 C/350/ 0.068%	[85]
	activated multichannel CNF	2.2 mg cm ⁻²	1351 mAh g ⁻¹ (0.2 C)/ 847 mA h g ⁻¹ (5 C)	0.2 C/300/0.07%	[86]
	activated PCNT	2.2 mg cm ⁻²	1270 mAh g ⁻¹ (0.2 C)/ 857 mAh g ⁻¹ (5 C)	0.2 C/300/0.09%	[87]
	LRC/S@EFG	3.6 mg cm ⁻²	1,300 mAh g ⁻¹ (0.1 C)/ 363 mAh g ⁻¹ (2 C)	0.2 C/200/0.1%	[88]
	CNF@Co ₃ S ₄	1.7 mg cm ⁻²	1079 mA h g ⁻¹ (0.1 C)/ 850 mA h g ⁻¹ (1.5 C)	1 C/200/0	[89]
	GC-TiO@CHF	5 mg cm ⁻²	1,000 mAh g ⁻¹ (0.1 C)/ 620 mAh g ⁻¹ (2 C)	0.2 C/400/0.085%	[90]
Graphene	<i>N</i> -doped graphene sheet	1.5 mg cm ⁻²	1486 mAh g ⁻¹ (0.1 C)/ 950 mAh g ⁻¹ (1 C)	1.17 mA cm ⁻² /300/ 0.08%	[91]
	rNGO	1.2 mg cm ⁻²	803 mAh g ⁻¹ (0.1 C)/ 592 mAh g ⁻¹ (1 C)	1C/1000/-	[92]
	hydrothermally reduced graphene oxide (HrGO)	1.3-1.6 mg cm ⁻²	1033 mAh g ⁻¹ (0.1 C)/ 588 mAh g ⁻¹ (1 C)	1.5 A g ⁻¹ / 500 / 0.07%	[93]
	3D <i>N</i> -doped graphene foam	2.05 mg cm ⁻²	1145 mAh g ⁻¹ (0.1 C)/ 625 mAh g ⁻¹ (2 C)	2 C/500/0.061%	[94]
	GA composite	2.5 mg cm ⁻²	1100 mAh g ⁻¹ (0.1 C)/300 mAh g ⁻¹ (1 C)	0.5 C/200/-	[95]

	hierarchical porous graphene	-	1068 mAh g ⁻¹ (0.5 C)/ 543 mAh g ⁻¹ (10 C)	1C/80/-	[96]
<i>N,S</i> co-doped porous graphene matrix	1.5 mg cm ⁻²		1203 mAh g ⁻¹ (0.1 C)/ 855 mAh g ⁻¹ (2 C)	0.2 C/600/0.056%	[97]
	rGO	2.2 mg cm ⁻²	1200 mAh g ⁻¹ (0.1 C)/ 924 mAh g ⁻¹ (2 C)	0.1 C/200/0.12%	[98]
	Co-PCNF	1.7 mg cm⁻²	1373.5 mAh g⁻¹(0.2 C)/ 914.3 mAh g⁻¹ (2.0 C)	1 C/800/0.05%	This work

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