Electronic Supplementary Information

MOF-Derived Hierarchical CoP Nanoflakes Anchored on Vertically-Erected Graphene Scaffolds as Self-Supported and Flexible Hosts for Lithium–Sulfur Batteries

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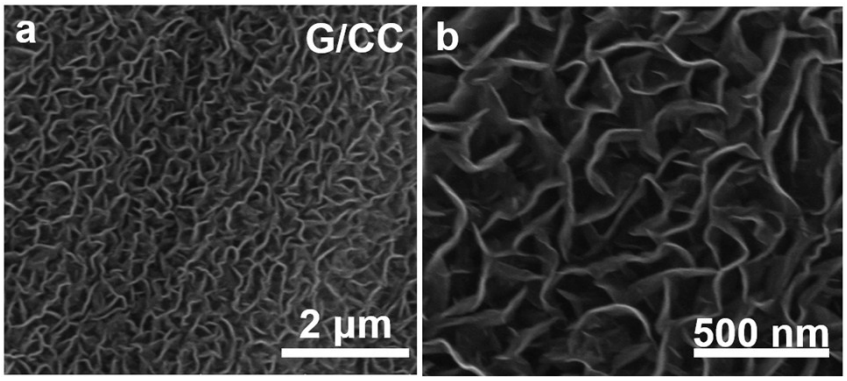

Fig. S1. SEM images of G/CC.

Element	Atomic %
O	18.38
P	24.89
Co	20.04

Fig. S2. The atomic ratio of Co, P, O elements studied by EDS.

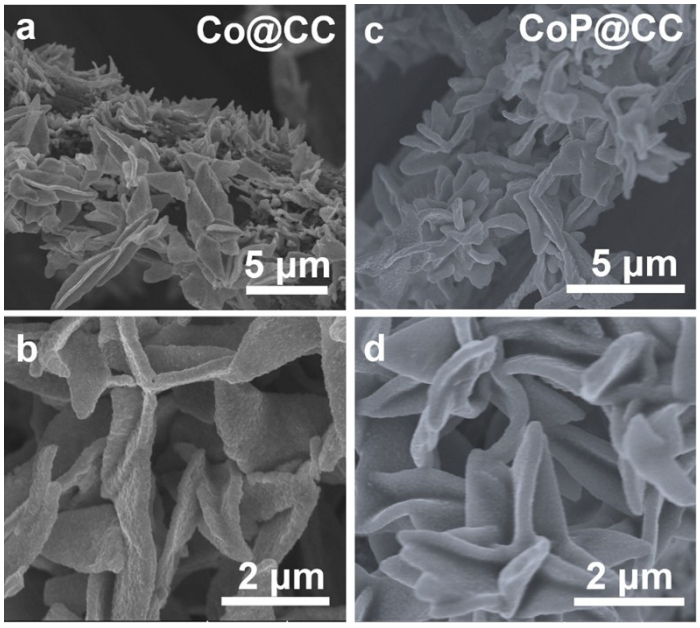

Fig. S3. SEM images of (a, b) Co@CC and (c, d) CoP@CC.

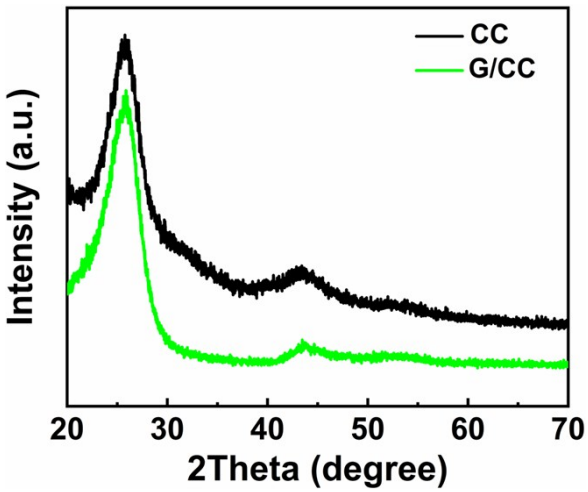

Fig. S4. XRD patterns of CC and G/CC.

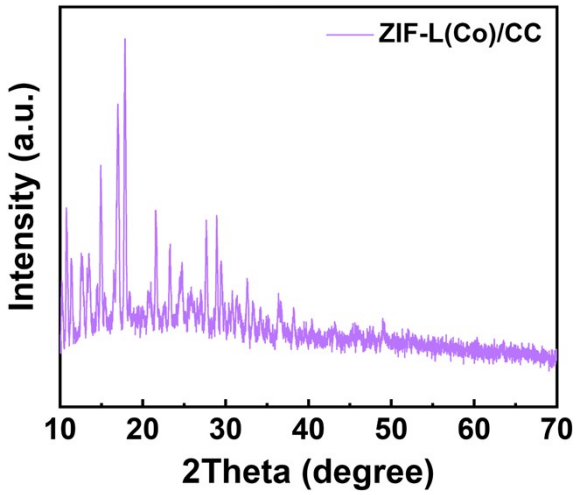

Fig. S5. XRD pattern of ZIF-L(Co)/CC.

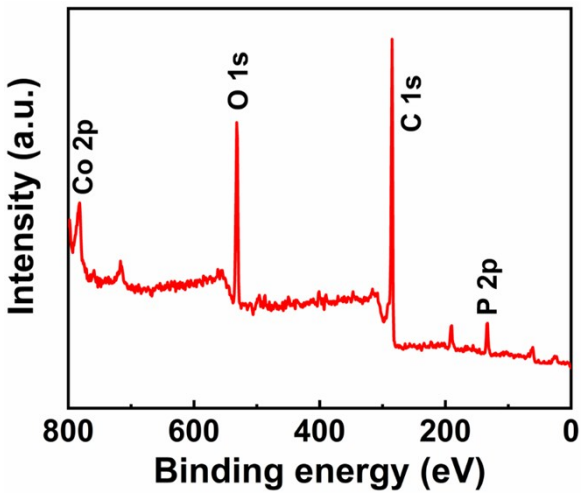

Fig. S6. XPS survey spectrum of CoP@G/CC.

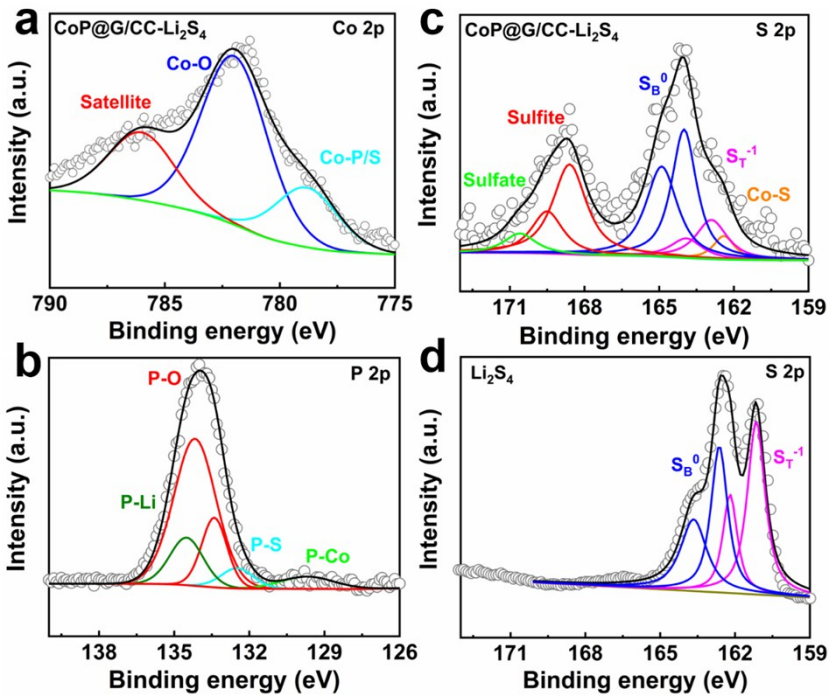

Fig. S7. XPS spectra of (a) Co 2p of CoP@G/CC after interacting with Li_2S_4, (b) P 2p of CoP@G/CC after interacting with Li_2S_4, (c) S 2p of CoP@G/CC after interacting with Li_2S_4 and (d) S 2p of bare Li_2S_4.

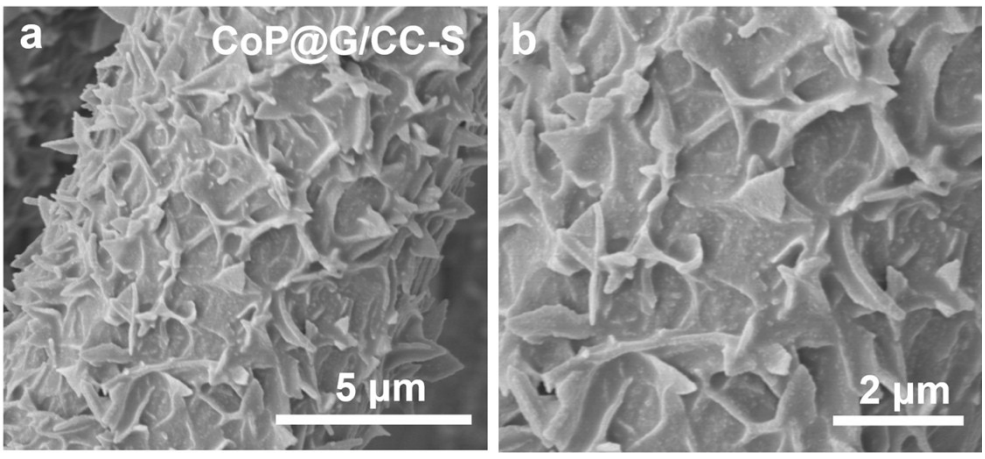

Fig. S8. SEM images of as-prepared CoP@G/CC-S.

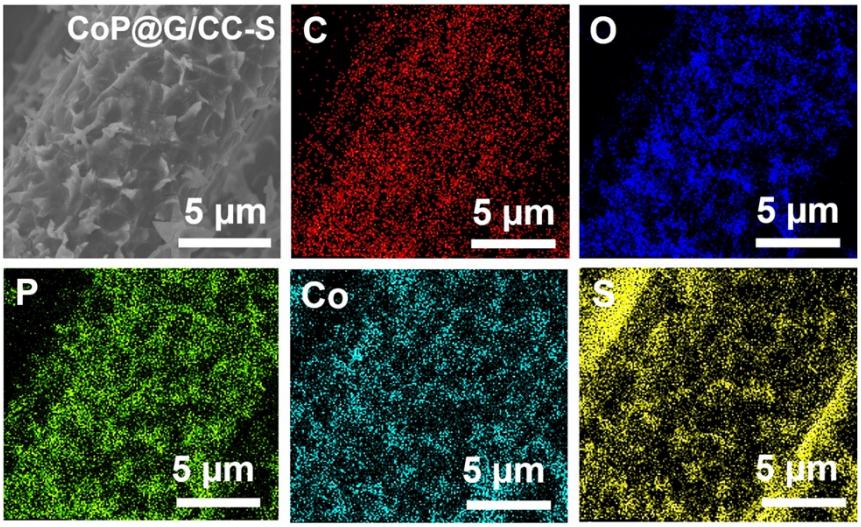

Fig. S9. Elemental maps of CoP@G/CC-S.

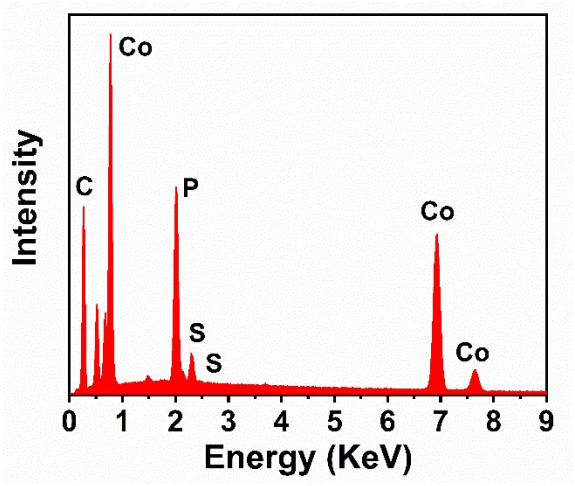

Fig. S10. EDS spectrum of CoP@G/CC-S cathode with a sulfur mass loading of 2.31 mg cm^{-2}.

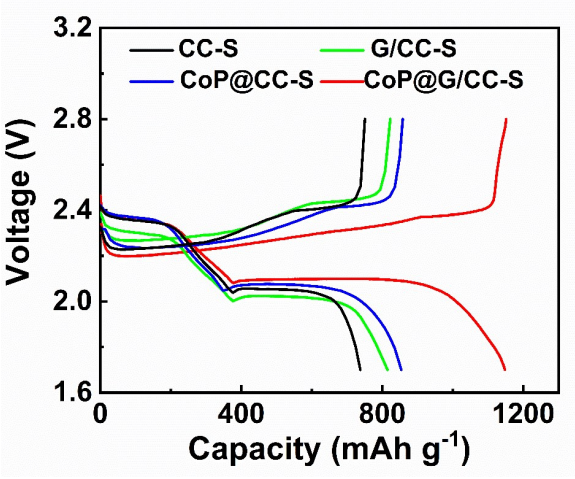

Fig. S11. Galvanostatic charge/discharge profiles of different cathodes at 0.5 C.

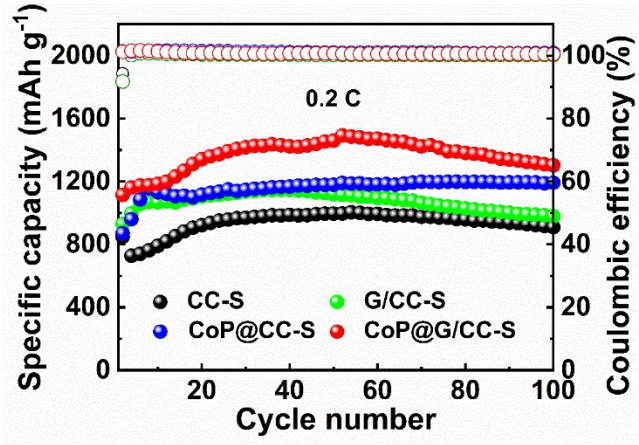

Fig. S12. Cycling performances of different cathodes at 0.2 C.

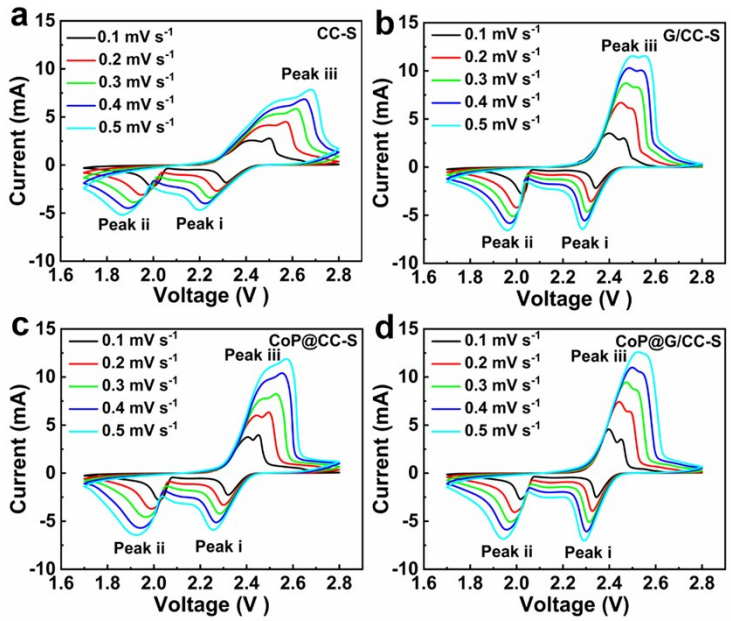

Fig. S13. CV profiles of different electrodes at different scan rates.

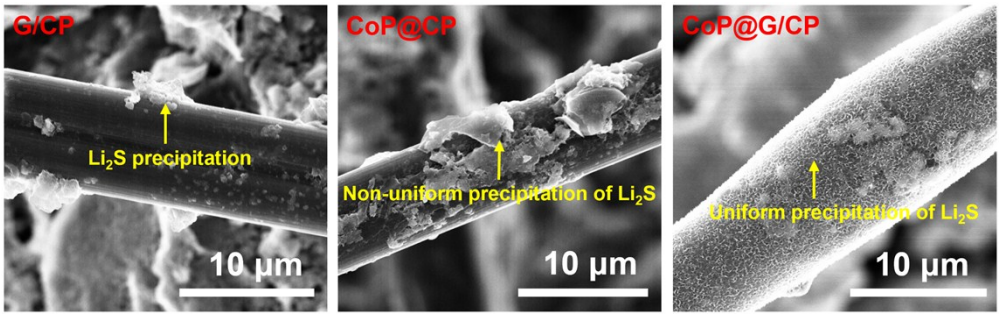

Fig. S14. SEM observations of Li_2S nucleation on the surface of G/CP, CoP@CP and CoP@G/CP, respectively.

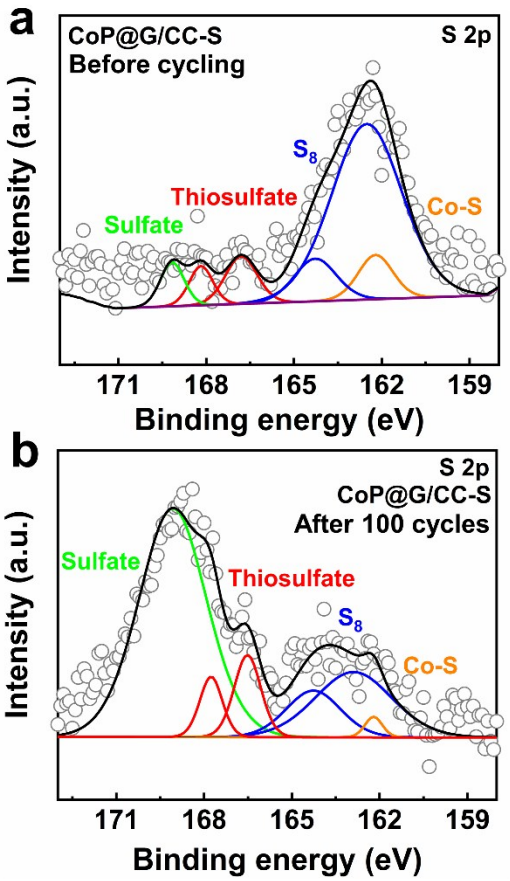

Fig. S15. XPS S 2p analysis of CoP@G/CC-S cathode before and after 100 cycles.

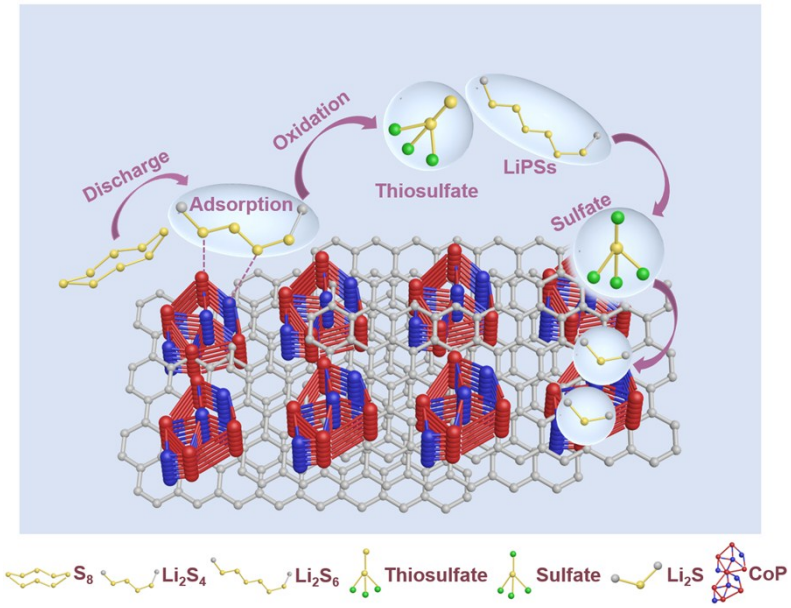

Fig. S16. Schematic illustration of S_8 evolution on CoP@G/CC hybrid host.

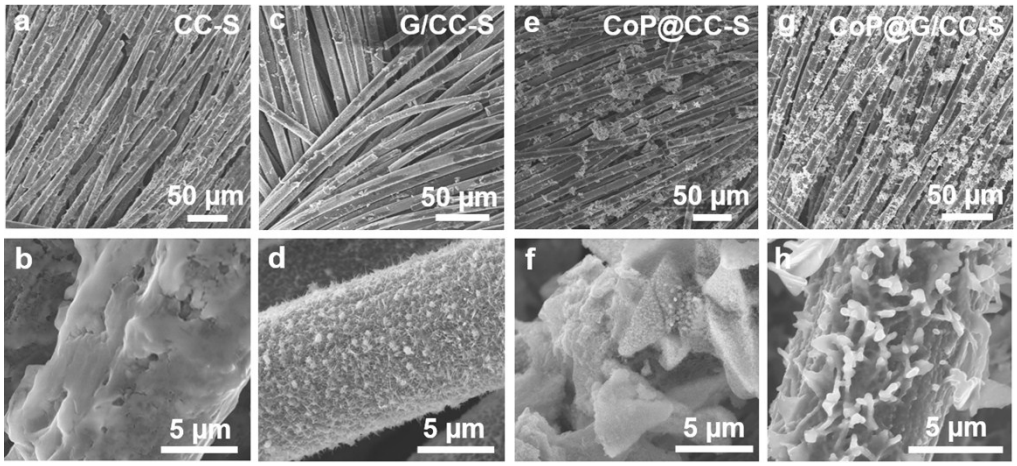

Fig. S17. SEM images of CC-S (a, b), G/CC-S (c, d), CoP@CC-S (e, f) and CoP@G/CC-S (g, h) cathodes before (a, c, e, g) and after (b, d, f, h) 100 cycles.

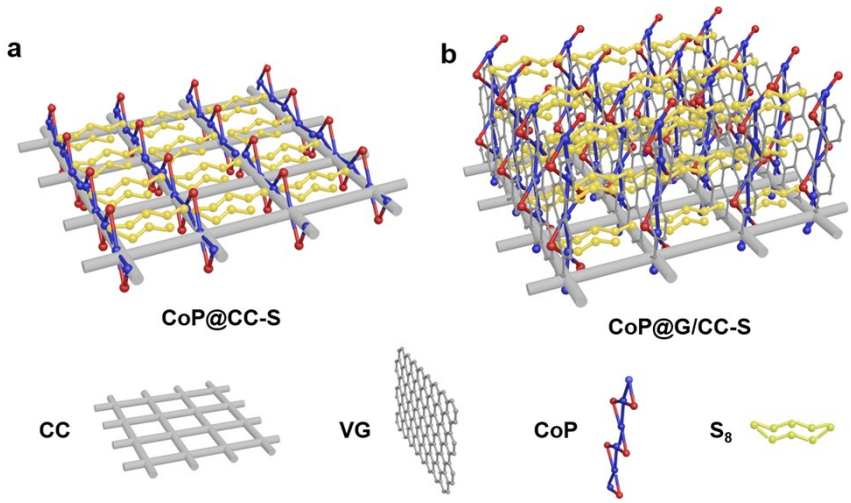

Fig. S18. Schematic illustration of the sulfur distribution at the CoP@CC and CoP@G/CC.

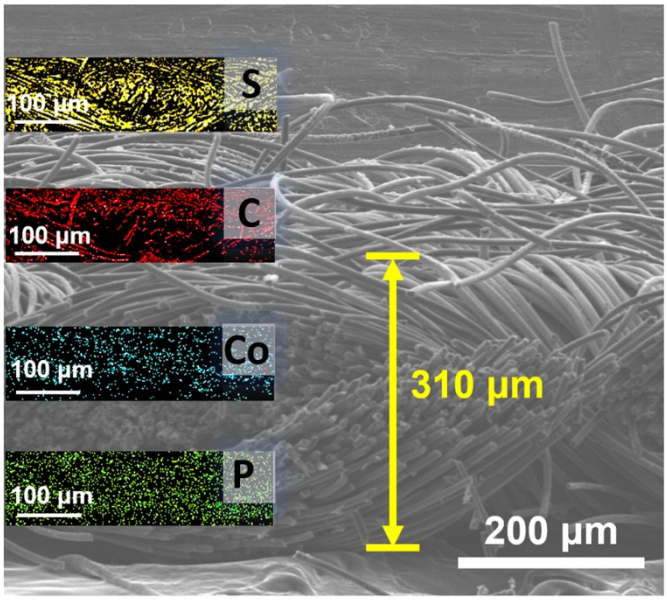

Fig. S19. Cross-sectional SEM/EDS characterization of the CoP@G/CC-S cathode with a sulfur loading of 10.83 mg cm^{-2}.

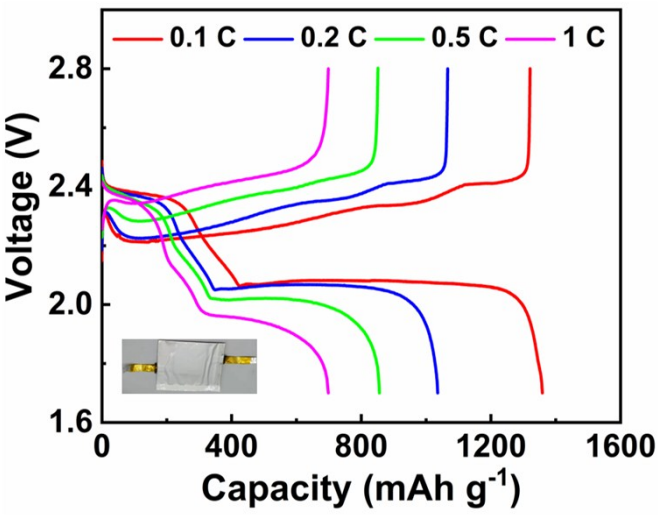

Fig. S20. Galvanostatic charge/discharge profiles of assembled pouch cell with CoP@G/CC-S cathode at different current densities.

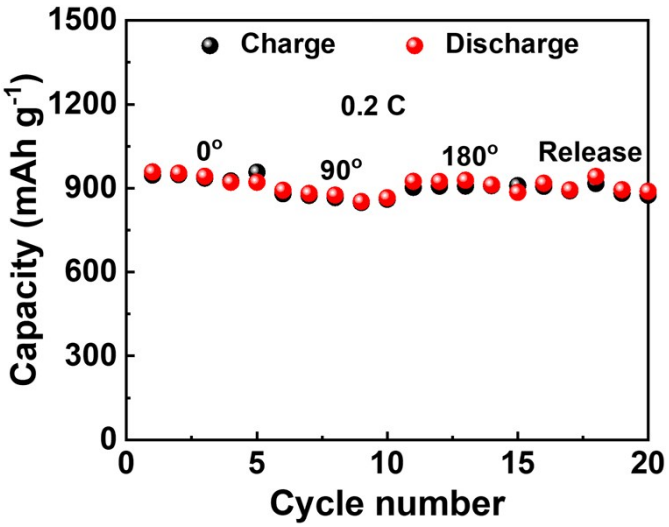

Fig. S21. Cycling performance of the pouch cell under bent-release states.

Table S1. Comparison of the discharge specific capacities of CoP@G/CC-S cathodes at various current densities with those of other reported materials.

Materials	Capacity (mAh g^{-1})					Ref.
	0.2 C	0.5 C	1 C	2 C	3 C	
VO_2(P)-NCNT	~1200.0	-	864.0	760.0	-	[1]
CoO/Co@PCF-S	946.7	828.5	753.9	684.3	-	[2]
$Ti_3C_2T_x$/S paper	-	~1290.0	~1180.0	~1080.0	-	[3]
CC@CoP/C-S	1088.0	895.0	821.0	708.0	-	[4]
TiO_2 NW/G	1270.0	~1100.0	~1020.0	850.0	-	[5]
VN/G	1447.0	1241.0	1131.0	953.0	701.0	[6]
CoP@G/CC-S	**1371.9**	**1265.5**	**1189.6**	**1053.1**	**930.1**	**This work**

Table S2. Comparison of battery performances based on pouch cell between this work and other reported studies.

Materials	S loadings ($mg\ cm^{-2}$)	Initial capacity (Rate) ($mAh\ g^{-1}$)	Cycle number (cycles) / Decay rate (% per cycle)	References
Co_9S_8–Celgard separator	2.0	1185 (0.1 C)	30 / 0.86%	[7]
P_2S_5/S@THF	3.8-4.2	(~1200) (0.2 C)	20 / 1.6%	[8]
B_4C@CNF	3.3	1008 (0.1 C)	100 / 0.50%	[9]
CC@CoP/C-S	3.4	1100 (0.1 C)	50 / 0.54%	[4]
S-G@PP separator cathode	1.5-2.8	985 (~0.45 C)	30 / 0.89%	[10]
S-CNTs/CoNCNFs/PVDF membrane	~2.0	(~900) (0.2 C)	350 / 0.06%	[11]
CoP@G/CC-S	**2.0**	**1151.6 (0.2 C)**	**100 / 0.25%**	**This work**

Supporting references

1 S. Wang, J. Liao, X. Yang, J. Liang, Q. Sun, J. Liang, F. Zhao, A. Koo, F. Kong, Y. Yao, X. Gao, M. Wu, S.-Z. Yang, R. Li and X. Sun, *Nano Energy*, 2019, **57**, 230-240.

2 W. Ren, W. Ma, M. M. Umair, S. Zhang and B. Tang, *ChemSusChem*, 2018, **11**, 2695-2702.

3 H. Tang, W. Li, L. Pan, K. Tu, F. Du, T. Qiu, J. Yang, C. P. Cullen, N. McEvoy and C. Zhang, *Adv. Funct. Mater.*, 2019, **29**, 1901907.

4 Z. Wang, J. Shen, J. Liu, X. Xu, Z. Liu, R. Hu, L. Yang, Y. Feng, J. Liu, Z. Shi, L. Ouyang, Y. Yu and M. Zhu, *Adv. Mater.*, 2019, **31**, 1902228.

5 G. Zhou, Y. Zhao, C. Zu and A. Manthiram, *Nano Energy*, 2015, **12**, 240-249.

6 Z. Sun, J. Zhang, L. Yin, G. Hu, R. Fang, H. M. Cheng and F. Li, *Nat. Commun.*, 2017, **8**, 14627.

7 J. He, Y. Chen and A. Manthiram, *Energy Environ. Sci.*, 2018, **11**, 2560-2568.

8 Y. Lu, S. Gu, X. Hong, K. Rui, X. Huang, J. Jin, C. Chen, J. Yang and Z. Wen, *Energy Storage Mater.*, 2018, **11**, 16-23.

9 L. Luo, S. H. Chung, H. Y. Asl and A. Manthiram, *Adv. Mater.*, 2018, **30**, 1804149.

10 G. Zhou, L. Li, D. W. Wang, X. Y. Shan, S. Pei, F. Li and H. M. Cheng, *Adv. Mater.*, 2015, **27**, 641-647.

11 J. Wang, G. Yang, J. Chen, Y. Liu, Y. Wang, C. Y. Lao, K. Xi, D. Yang, C. J. Harris, W. Yan, S. Ding and R. V. Kumar, *Adv. Energy Mater.*, 2019, **9**, 1902001.