

Supporting Information

One-pot synthesis of N,S-doped pearl chain tube-loaded Ni₃S₂ composite materials for high-performance lithium-air batteries

Bohan An,^a Jiacheng Li,^a Xiaochao Wu,^a Wanqing Li,^{a,b,*} Yongliang Li,^a Lingna Sun,^a Hongwei Mi,^a Qianling Zhang,^a Chuanxin He,^a Xiangzhong Ren^{a,*}

^a College of Chemistry and Environmental Engineering, Shenzhen University,
Guangdong 518060, PR China

^b Key Laboratory of Optoelectronic Devices and Systems of Ministry of Education
and Guangdong Province, College of Physics and Optoelectronic Engineering,
Shenzhen University, Shenzhen 518060, China

*Corresponding author.

E-mail address: renxz@szu.edu.cn, , liwanqing136@foxmail.com

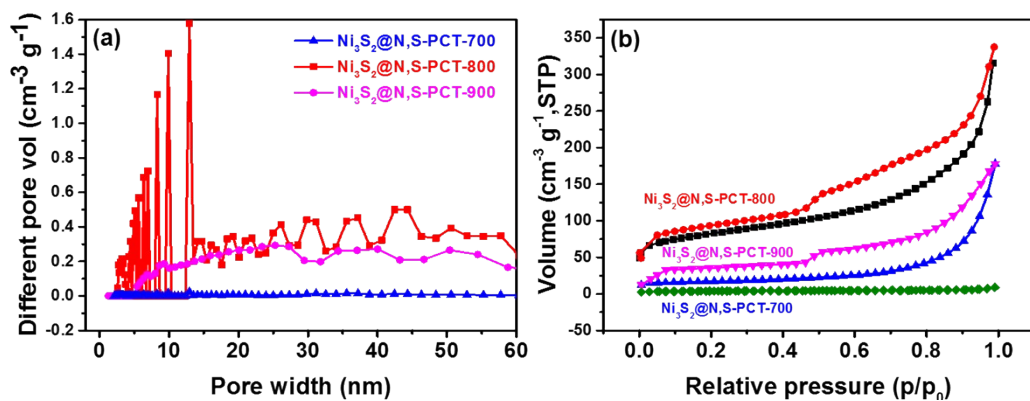


Figure S1 Under different temperature gradients (a) $\text{Ni}_3\text{S}_2@\text{N,S-PCT}$ of pore size distribution; (b) $\text{Ni}_3\text{S}_2@\text{N,S-PCT}$ of adsorption isotherm

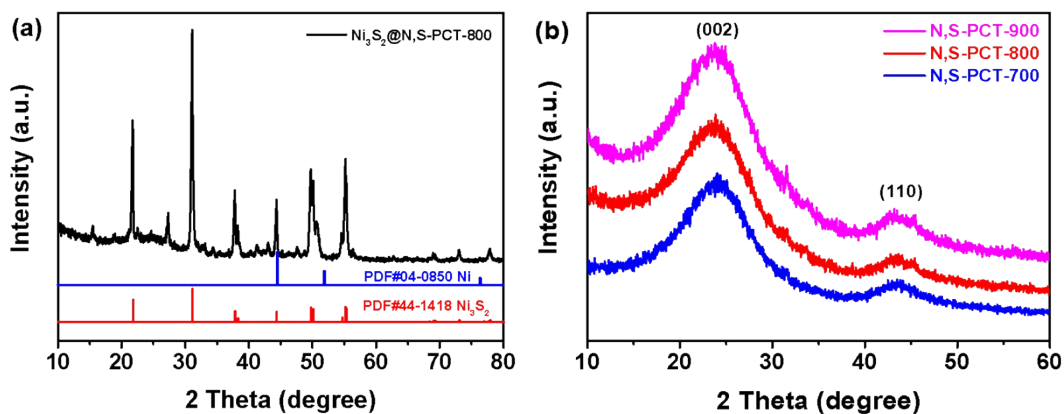


Figure S2 (a) XRD of material sample coated on carbon paper; (b) XRD of N,S-PCT at different temperatures

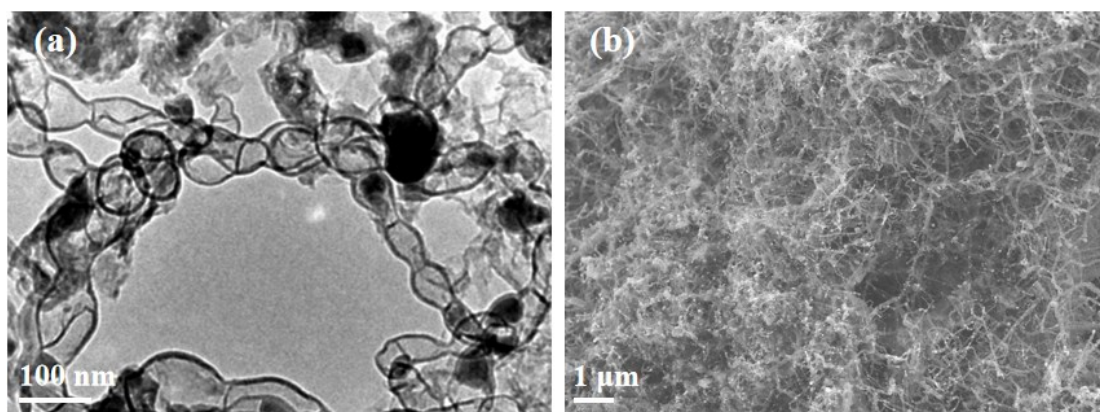


Figure S3 (a) Mapping the diameter of the carbon layer of N,S-PCTs; (b) the whole frame of $\text{Ni}_3\text{S}_2@\text{N,S-PCT-800}$

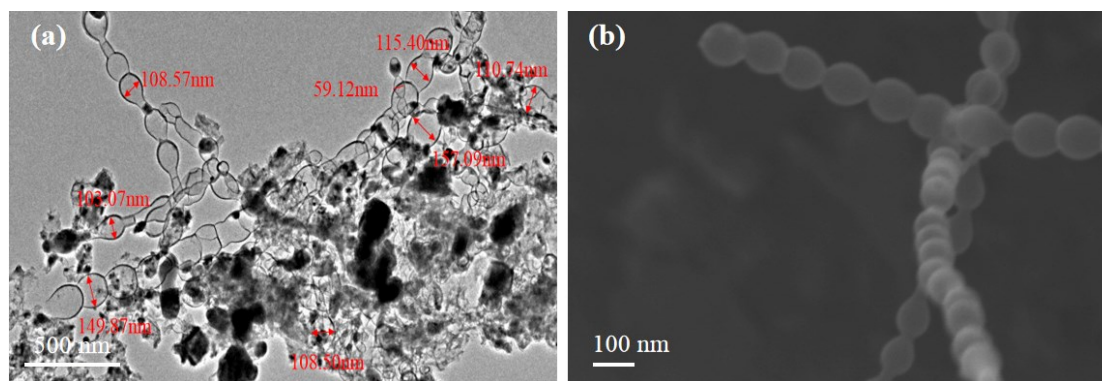


Figure S4 (a) Mapping the diameter of $\text{Ni}_3\text{S}_2@\text{N,S-PCT-800}$; (b) SEM images of N,S-PCT-800 . The N,S-PCT-800 obtained, the morphology of the carbon tube presents a pearl chain shape.

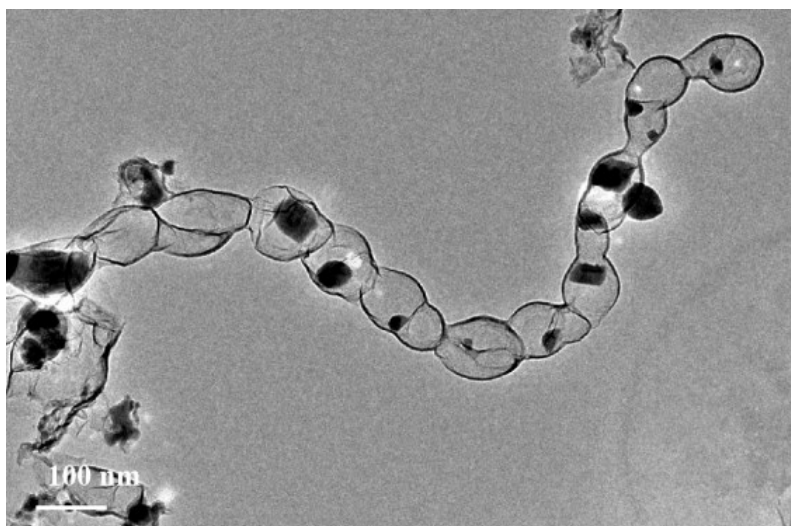


Figure S5. HRTEM images of $\text{Ni}_3\text{S}_2@\text{N,S-PCT-800}$

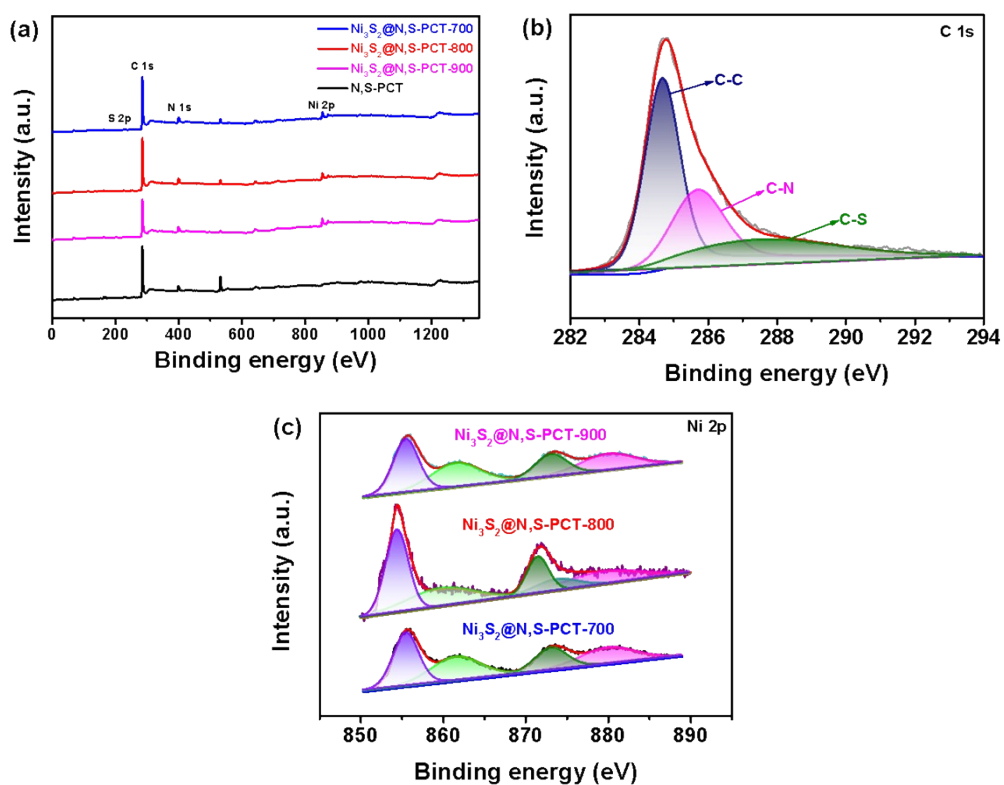


Figure S6 (a) XPS analysis of the samples in different temperatures; (b) XPS pattern of C 1s; (c) XPS pattern of Ni 2p from 700°C to 900°C

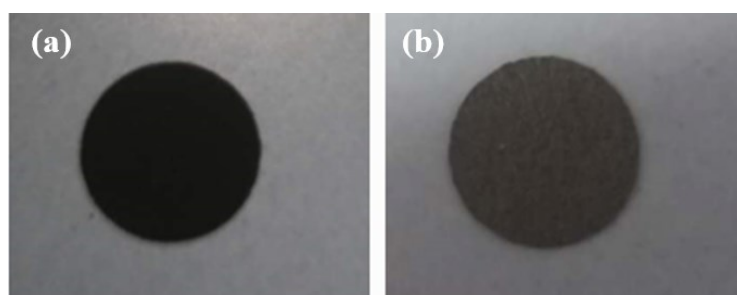


Figure S7. (a) loaded carbon paper (b) Original carbon paper

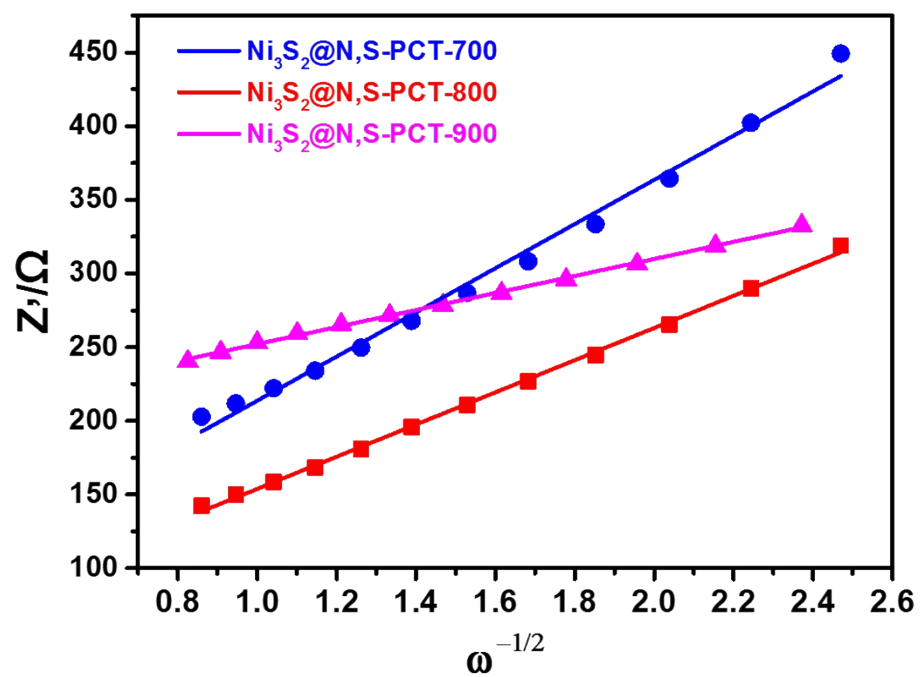


Figure S8. The relationship between Z' and $\omega^{-1/2}$ of O_2 electrode

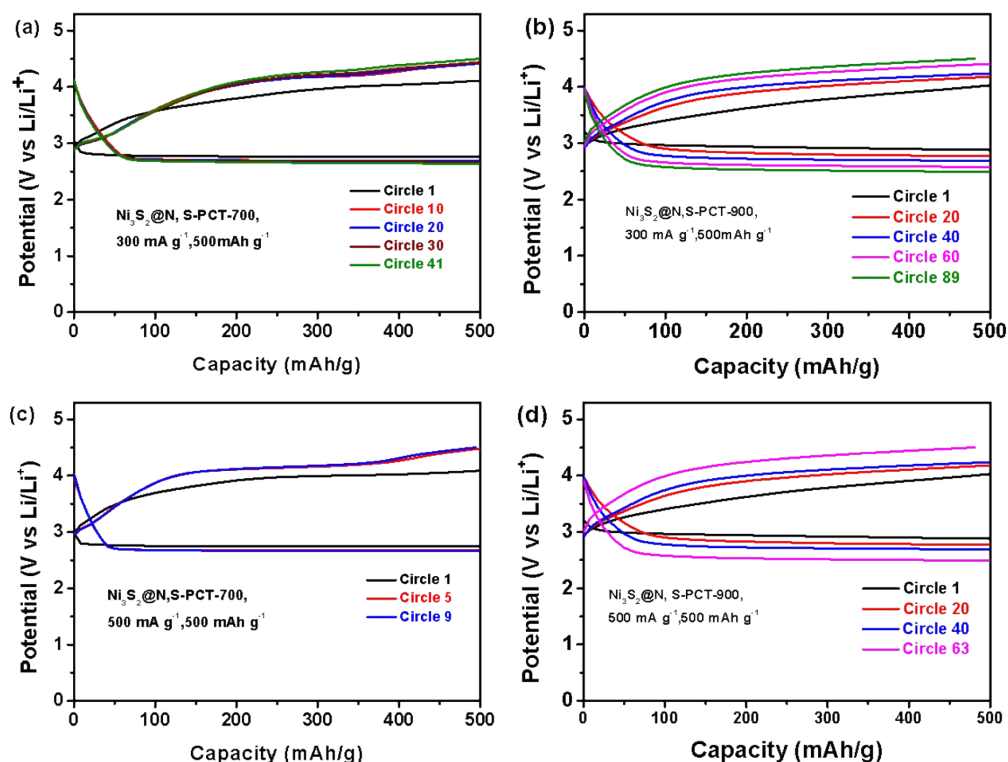


Figure S9. The discharge/charge profiles of (a) Ni₃S₂@N,S-PCT-700 and (b) Ni₃S₂@N,S-PCT-900, at a current density of 300 mA g⁻¹ with a limited capacity of 500 mAh g⁻¹. The discharge/charge profiles of (c) Ni₃S₂@N,S-PCT-700 and (d) Ni₃S₂@N,S-PCT-900, at a current density of 500 mA g⁻¹ with a limited capacity of 500 mAh g⁻¹

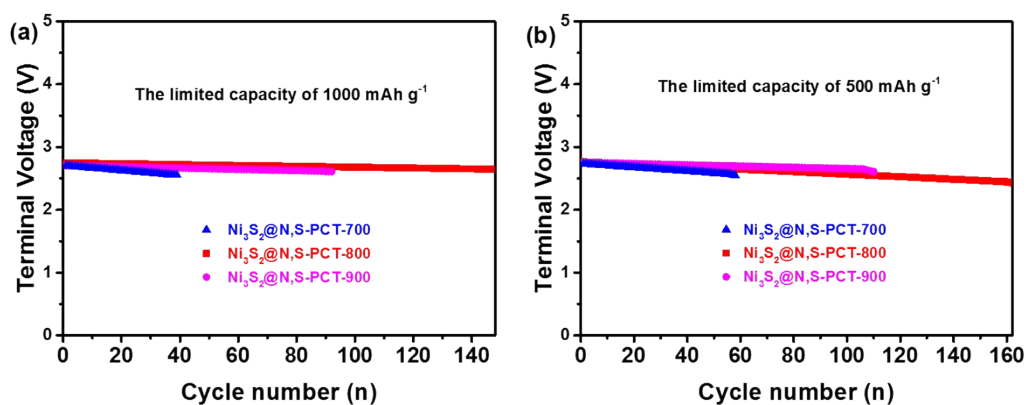


Figure S10. Cycle performance of the Ni₃S₂@N,S-PCT-900, Ni₃S₂@N,S-PCT-800 and Ni₃S₂@N,S-PCT-700 electrodes (a) at current density of 450 mA g⁻¹ with limited capacity of 1000 mAh g⁻¹ (b) at current density of 500 mA g⁻¹ with limited capacity of 500 mAh g⁻¹

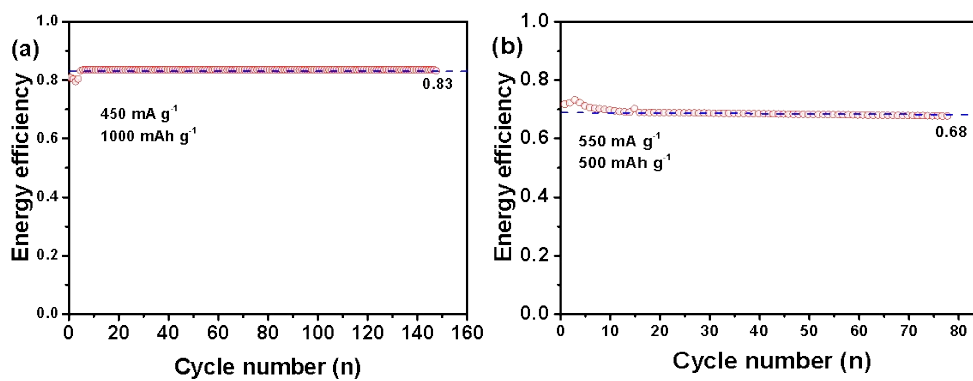


Figure S11. The cycle efficiency of Ni₃S₂@N,S-PCT-800 (a) the current density is 450 mA g⁻¹ and the limited capacity is 1000 mA g⁻¹ (b) the current density is 550 mA g⁻¹ and the limited capacity is 500 mA g⁻¹

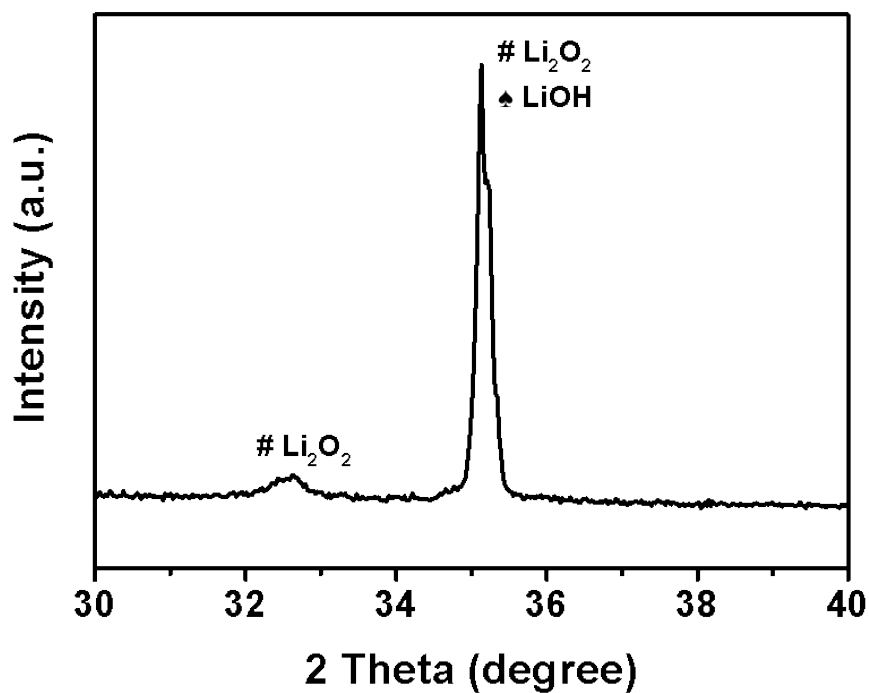


Figure S12. XRD analysis chart of discharge products in a certain range.

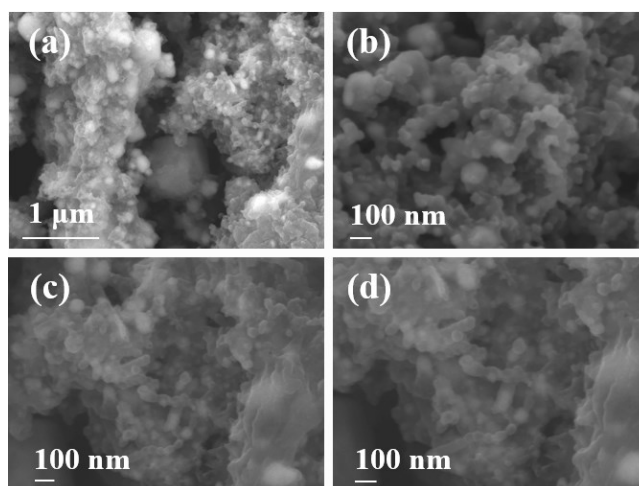


Figure S13. SEM image of materials after circulation in different positions (a) Magnification is 20,000; (b) Magnification is 50,000; (c) Magnification is 40,000; (d) Magnification is 50,000

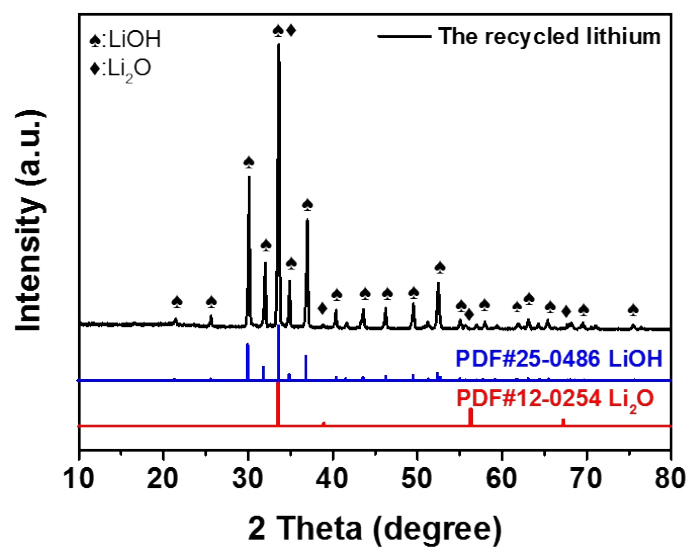


Figure S14. XRD patterns of the recycled lithium

Table S1. Elemental content percentage measured by XPS

Sample name	Ni (at. %)	O (at. %)	C (at. %)	N (at. %)	S (at. %)
Ni ₃ S ₂ @N,S-PCT-900	1.8	11.06	76.51	7.73	2.91
Ni ₃ S ₂ @N,S-PCT-800	3.24	2.98	81.95	10.63	1.34
Ni ₃ S ₂ @N,S-PCT-700	1.88	4	83.96	9.13	1.03
N,S-PCT	0.53	2.7	85.67	10.07	1.17

Table S2. Comparison of Li-O₂ battery properties of Ni₃S₂@N,S-PCT-800 cathode with those of representative state-of-the-art cathodes reported in literature.

Catalysts	Current Density (mA g ⁻¹)	Overpotention (V)	Cycling Performance (Cycles/Limited Capacity)	First Discharge Capacity (mAh g ⁻¹)	Ref.
2D Co ₃ S ₄ nanosheets	100	0.92	25/500	5917	1
Ni ₃ S ₂	200	1.29	50/500	7478	2
MoS ₂ nanoflakes	100	≥1.0	50/500	1250	3
Ni ₃ S ₂ /PBSC NFs	100	0.68	120/500	12874	4
Co ₉ S ₈	100	1.37	100/500	3500	5
MoS _x /HRG	0.05	1.5	30/500	6678	6
TiC/MWNTs-Ru	250	0.49	90/1000	3841	7
GDP-Mo ₂ C@NCF	100	1.2	100/1000	7437	8
NCS/S-3DPG	150	1.38	102/1000	14,173	9
Co ₂ P/Ru/CNT	100	1.22	120/1000	12 800	10
Ni₃S₂@N,S-PCT-800	450	1.45	148/1000	16733.7	This work

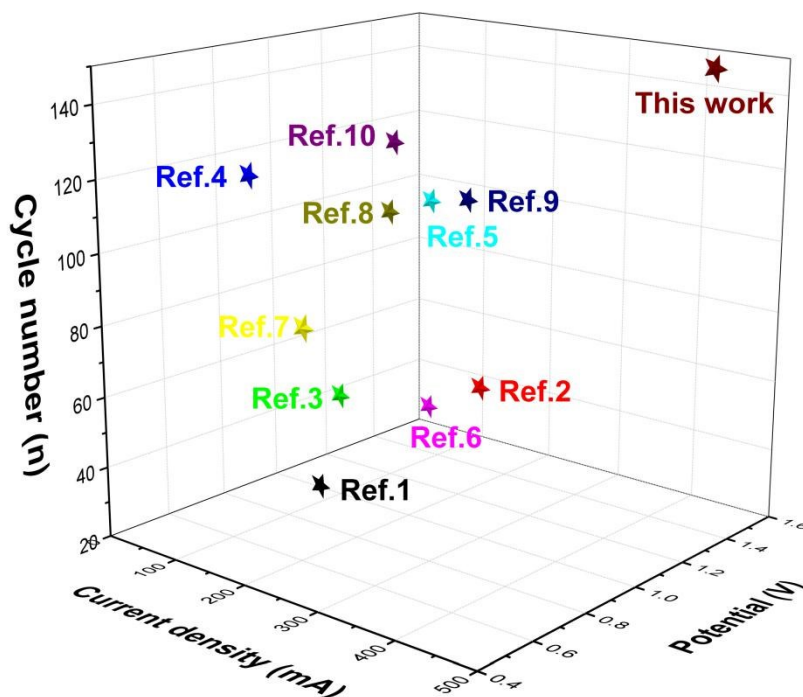


Figure S12 Lithium-air battery performance of different types of cathode materials

REFERENCE

1. P. Sennu, M. Christy, V. Aravindan, Y.-G. Lee, K. S. Nahm and Y.-S. Lee, *Chem. Mater.*, 2015, **27**, 5726-5735.
2. Q. Wang, X. Wang and H. He, *J. Power Sources*, 2020, **448**, 227397.
3. M. Asadi, B. Kumar, C. Liu, P. Phillips, P. Yasaei, A. Behranginia, P. Zapol, R. F. Klie, L. A. Curtiss and A. Salehi-Khojin, *ACS Nano*, 2016, **10**, 2167-2175.
4. Z. Zhang, K. Tan, Y. Gong, H. Wang, R. Wang, L. Zhao and B. He, *J. Power Sources*, 2019, **437**, 226908.
5. G. Wang, Y. Li, L. Shi, R. Qian and Z. Wen, *Chem. Eng. J.*, 2020, **396**, 125228.
6. L. Li, C. Chen, J. Su, P. Kuang, C. Zhang, Y. Yao, T. Huang and A. Yu, *J. Mater. Chem. A*, 2016, **4**, 10986-10991.
7. C.-S. Yang, Z. Sun, Z. Cui, F.-L. Jianga, J.-W. Denga and T. Zhang, *Energy Storage Mater.*, 2020, **30**, 59-66.
8. Z. Sun, B. Li, C. Feng, X. Cao, X. Zheng, K. Zeng, C. Jin, X. Wu, D. Dai and R. Yang, *J. Mater. Chem. A*, 2020, **8**, 14815-14821.
9. S. Hyun, B. Son, H. Kim, J. Sanetuntikul and S. Shanmugam, *Appl. Catal. B*, 2020, **263**, 118283.
10. P. Wang, C. Li, S. Dong, X. Ge, P. Zhang, X. Miao, Z. Zhang, C. Wang and L. Yin, *Small*, 2019, **15**, 1900001.