

Recent Advances in Heterostructured Cathodic Electrocatalysts for Non-aqueous Li-O₂ Batteries

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Table S1 Comparison table of the synthesis methods and electrocatalytic performance based on reported typical heterostructures catalysts for Li-O₂ batteries.

Materials	Synthesis Method	1 st Discharge Capacity ^{a/} Current Density	Overall Potential Gap/ Current Density	Cycles or hours/Current Density-Fixed Capacity	Ref.
Mo ₂ C@CNTs	Ball Milling+Thermal Treatment	-	0.47 V/100 mA g ⁻¹	100/100 mA g ⁻¹ -500 mAh g ⁻¹	1
Co ₄ N@CNFs	Hydrothermal+Nitridation Process	~11000 mAh g ⁻¹ /00 mA g ⁻¹	1.23 V/700 mA g ⁻¹	177/200 mA g ⁻¹ -500 mAh g ⁻¹	2
CuGeO ₃ @Graphene	Hydrothermal+Thermal Treatment	10030 mAh g ⁻¹ /200 mA g ⁻¹	1.50 V/200 mA g ⁻¹	50/1000 mA g ⁻¹ -2000 mAh g ⁻¹	3
Co ₉ S ₈ @CFs	Hydrothermal+Thermal Treatment	6875 mAh g ⁻¹ /50 mA g ⁻¹	0.57 V/50 mA g ⁻¹	105/100 mA g ⁻¹ -500 mAh g ⁻¹	4
(Mn _{1/3} Co _{2/3})O@CNTs	One-pot Spray Pyrolysis Infiltrated+Thermal Treatment	20588 mAh g ⁻¹ /500 mA g ⁻¹ - 7646 mAh g ⁻¹ /200 mA g ⁻¹	0.64 V/200 mA g ⁻¹ ~1.25 V/200 mA g ⁻¹	245/200 mA g ⁻¹ -500 mAh g ⁻¹ 700h/500 mA g ⁻¹ -500 mAh g ⁻¹	5 6
N-Co@Graphene	Reaction+Thermal Treatment	-	~0.9 V/0.1 mA cm ⁻²	30/0.1 mA cm ⁻² -1 mAh cm ⁻²	7
Pd-C@CP	Electrophoretic+In-Situ Modification	5900 mAh g ⁻¹ /1500 mA g ⁻¹	~1 V/300 mA g ⁻¹	213/300 mA g ⁻¹ -1000 mAh g ⁻¹	8
Ag/La _{0.9} FeO _{3-δ}	Electrospinning+Thermal Treatment	8476 mAh g ⁻¹ /100 mA g ⁻¹	0.66 V/100 mA g ⁻¹	174/100 mA g ⁻¹ -1000 mAh g ⁻¹	9
Co ₃ O ₄ /Ag	Solution Reaction+Hydrothermal Thermal Treatment+In- Situ Modification+Atom Interdiffusion Solution	12000 mAh g ⁻¹ /200 mA g ⁻¹ 22551 mAh g ⁻¹ /1000 mA g ⁻¹	~1.2 V/200 mA g ⁻¹ 0.68 V/1000 mA g ⁻¹	80/200 mA g ⁻¹ -1000 mAh g ⁻¹ 268/1000 mA g ⁻¹ -3000 mAh g ⁻¹	10 11
Au/Cu@FCu	Immersion+Redox Replacement	27270 mAh g ⁻¹ /100 mA g ⁻¹	0.64 V/100 mA g ⁻¹	220/100 mA g ⁻¹ -500 mAh g ⁻¹	12
Pd/NiCo ₂ O ₄	Hydrothermal+Solution Immersion	4000 mAh g ⁻¹ /200 mA g ⁻¹	~1.3 V/100 mA g ⁻¹	100/200 mA g ⁻¹ -1000 mAh g ⁻¹	13
Ru/ZnIn ₂ S _{4-x}	Hydrothermal+Solution Immersion	3532mAh g ⁻¹ /500 mA g ⁻¹	0.77 V/500 mA g ⁻¹	1254h/500 mA g ⁻¹ -1000 mAh g ⁻¹	14
Pd/Pd ₄ S	Solution Reaction	8777 mAh g ⁻¹ /100 mA g ⁻¹	1.55 V/100 mA g ⁻¹	160/500 mA g ⁻¹ -500 mAh g ⁻¹	15
MnO ₂ /Co ₃ O ₄ @CP	Hydrothermal+Thermal Treatment	4850 mAh g ⁻¹ /103 mA g ⁻¹	0.95 V/103 mA g ⁻¹	53/103 mA g ⁻¹ -1030 mAh g ⁻¹	16
NiCo ₂ S ₄ /NiO	Hydrothermal+Solution	10050 mAh g ⁻¹ /200 mA g ⁻¹	0.88 V/200 mA g ⁻¹	300/200 mA g ⁻¹ -1000 mAh g ⁻¹	17

	Immersion+Thermal Treatment				
PdO/Co ₃ O ₄	Solution	-	0.22 V/200 mA g ⁻¹	90/200 mA g ⁻¹ -500 mAh g ⁻¹	18
	Immersion+Pyrolysis				
Co ₃ O ₄ /MnO ₂	Hydrothermal+Thermal Treatment	5738 mAh g ⁻¹ /100 mA g ⁻¹	0.82 V/100 mA g ⁻¹	60/200 mA g ⁻¹ -1000 mAh g ⁻¹	19
NiCo ₂ O ₄ /NiO	Hydrothermal	17463 mAh g ⁻¹ /500 mA g ⁻¹	0.98 V/500 mA g ⁻¹	500/100 mA g ⁻¹ -1000 mAh g ⁻¹	20
Mo ₂ C/MoO ₂	Hydrothermal+Thermal Treatment	~2000 mAh g ⁻¹ /800 mA g ⁻¹	0.56 V/200 mA g ⁻¹	100/200 mA g ⁻¹ -1000 mAh g ⁻¹	21
	Solution				
RuO ₂ /Mn ₂ O ₃	Reaction+Electrospinning	-	0.96 V/100 mA g ⁻¹	121/400 mA g ⁻¹ -1000 mAh g ⁻¹	22
	Hydrothermal+Thermal Treatment				
Urchin-NiO/NiCo ₂ O ₄	Hydrothermal	9231 mAh g ⁻¹ /100 mA g ⁻¹	1.48 V/100 mA g ⁻¹	80/100 mA g ⁻¹ -600 mAh g ⁻¹	23
NiS ₂ /ZnIn ₂ S ₄	Hydrothermal	3682 mAh g ⁻¹ /500 mA g ⁻¹	~1.2 V/500 mA g ⁻¹	490/500 mA g ⁻¹ -500 mAh g ⁻¹	24
CoSe ₂ /NiSe ₂	Hydrothermal+Thermal Treatment	3530 mAh g ⁻¹ /600 mA g ⁻¹	0.95 V/100 mA g ⁻¹	250/200 mA g ⁻¹ -1000 mAh g ⁻¹	25
Ni ₃ Se ₂ /NiSe ₂ @NF	Hydrothermal	23092 mAh g ⁻¹ /500 mA g ⁻¹	0.38 V/100 mA g ⁻¹	500/100 mA g ⁻¹ -1000 mAh g ⁻¹	26
	Hydrothermal+Solution Reaction				
CdSe/ZnS QD@CNT	Hydrothermal+Solution Reaction	-	~1.3 V/100 mA g ⁻¹	100/100 mA g ⁻¹ -1000 mAh g ⁻¹	27
	Co-precipitation +Self-assembling				
Co-Fe-(LDH) /RuO ₂	Co-precipitation +Self-assembling	~4300 mAh g ⁻¹ /10 mA cm ⁻²	0.64 V/100 mA g ⁻¹	100/10 mA cm ⁻² -800 mAh g ⁻¹	28

^aThe specific discharge capacities were calculated based on the amount of catalysts in the cathodes.

1. W. J. Kwak, K. C. Lau, C. D. Shin, K. Amine, L. A. Curtiss and Y. K. Sun, *ACS Nano*, 2015, **9**, 4129-4173.
2. K. R. Yoon, K. Shin, J. Park, S. H. Cho, C. Kim, J. W. Jung, J. Y. Cheong, H. R. Byon, H. M. Lee and I. D. Kim, *ACS Nano*, 2018, **12**, 128-139.
3. G. H. Lee, M. C. Sung, J. C. Kim, H. J. Song and D. W. Kim, *Adv. Energy Mater.*, 2018, **8**, 1801930.
4. X. D. Lin, R. M. Yuan, S. R. Cai, Y. H. Jiang, J. Lei, S. G. Liu, Q. H. Wu, H. G. Liao, M. S. Zheng and Q. F. Dong, *Adv. Energy Mater.*, 2018, **8**, 1800089.
5. J. H. Kim, Y. J. Oh and Y. C. Kang, *Carbon*, 2018, **128**, 125-133.
6. Y. Luo, C. Jin, Z. Wang, M. Wei, C. Yang, R. Yang, Y. Chen and M. Liu, *Journal of Materials Chemistry A*, 2017, **5**, 5690-5695.
7. G. Tan, L. Chong, R. Amine, J. Lu, C. Liu, Y. Yuan, J. Wen, K. He, X. Bi, Y. Guo, H. H. Wang, R. Shahbazian-Yassar, S. Al Hallaj, D. J. Miller, D. Liu and K. Amine, *Nano Lett.*, 2017, **17**, 2959-2966.
8. J. J. Xu, Z. L. Wang, D. Xu, L. L. Zhang and X. B. Zhang, *Nat. Commun.*, 2013, **4**, 2438.
9. Y. Cong, Q. Tang, X. Wang, M. Liu, J. Liu, Z. Geng, R. Cao, X. Zhang, W. Zhang, K. Huang and S. Feng, *ACS Catal.*, 2019, **9**, 11743-11752.
10. R. Gao, Z. Yang, L. Zheng, L. Gu, L. Liu, Y. Lee, Z. Hu and X. Liu, *ACS Catal.*, 2018, **8**, 1955-

1963.

11. J. J. Xu, Z. W. Chang, Y. B. Yin and X. B. Zhang, *ACS Cent. Sci.*, 2017, **3**, 598-604.
12. N. Luo, G. J. Ji, H. F. Wang, F. Li, Q. C. Liu and J. J. Xu, *ACS Nano*, 2020, **14**, 3281-3289.
13. D. A. Agyeman, M. Park and Y. M. Kang, *J. Mater. Chem. A*, 2017, **5**, 22234-22241.
14. R. X. Liang, C. Z. Shu, A. J. Hu, C. X. Xu, R. X. Zheng, M. L. Li, Y. W. Guo, M. He, Y. Yan and J. P. Long, *J. Mater. Chem. A*, 2020, **8**, 11337-11345.
15. Q. S. Huang, F. Dang, H. T. Zhu, L. L. Zhao, B. He, Y. Wang, J. Wang and X. M. Mai, *J. Power Sources*, 2020, **451**, 227738.
16. P. Zhang, S. Zhang, M. He, J. Lang, A. Ren, S. Xu and X. Yan, *Adv. Sci.*, 2017, **4**, 1700172.
17. P. Wang, C. Li, S. Dong, X. Ge, P. Zhang, X. F. Miao, R. Wang, Z. Zhang and L. Yin, *Adv. Energy Mater.*, 2019, **9**, 1900788.
18. Y. Zhang, J. Ma, M. W. Yuan, Y. Li, R. A. Shen, W. C. Cheong, T. Han, G. B. Sun, C. Chen and C. Y. Nan, *Chem. Commun.*, 2019, **55**, 12683-12686.
19. Y. J. Lee, D. H. Kim, T.-G. Kang, Y. Ko, K. Kang and Y. J. Lee, *Chemistry of Materials*, 2017, **29**, 10542-10550.
20. R. X. Liang, A. J. Hu, M. L. Li, Z. Q. Ran, C. Z. Shu and J. Long, *Electrochim Acta*, 2019, **321**, 134716.
21. C. Wu, Y. Y. Hou, J. C. Jiang, H. P. Guo, H. K. Liu, J. Chen and J. Z. Wang, *J. Power Sources*, 2020, **470**, 228317.
22. K. R. Yoon, G. Y. Lee, J. W. Jung, N. H. Kim, S. O. Kim and I. D. Kim, *Nano Lett.*, 2016, **16**, 2076-2083.
23. W. Zhao, X. Li, R. Yin, L. Qian, X. Huang, H. Liu, J. Zhang, J. Wang, T. Ding and Z. Guo, *Nanoscale*, 2018, **11**, 50-59.
24. A. Hu, W. Lv, T. Lei, W. Chen, Y. Hu, C. Shu, X. Wang, L. Xue, J. Huang, X. Du, H. Wang, K. Tang, C. Gong, J. Zhu, W. He, J. Long and J. Xiong, *ACS Nano*, 2020, **14**, 3490-3499.
25. R. X. Liang, C. Z. Shu, A. J. Hu, M. L. Li, Z. Q. Ran, R. X. Zheng and J. P. Long, *Chem. Eng. J.*, 2020, **393**, 124592.
26. Z. Q. Ran, C. Z. Shu, Z. Q. Hou, L. J. Cao, R. X. Liang, J. B. Li, P. Hei, T. S. Yang and J. P. Long, *J. Power Sources*, 2020, **468**, 228308.
27. V. Veeramani, Y. H. Chen, H. C. Wang, T. F. Hung, W. S. Chang, D. H. Wei, S. F. Hu and R. S. Liu, *Chem. Eng. J.*, 2018, **349**, 235-240.
28. X. Y. Jin, D. A. Agyeman, S. Kim, Y. H. Kim, M. G. Kim, Y. M. Kang and S. J. Hwang, *Nano Energy*, 2020, **67**, 104192.