

## Cubic core-shell structure of $\text{NiCoS}_x/\text{CoS}_2$ as high-efficiency tri-functional catalyst for Zn-air battery and overall water splitting

Bingqian Wang, Jianyu Liu, Huaiyun Ge, Siwei Fan, Guanghui Zhang, Lingxue Zhao and Guangda Li\*

School of Materials Science and Engineering, Qilu University of Technology (Shandong Academy of Sciences), Shandong, Jinan 250353, China

Corresponding author.

E-mail address: ligd@qlu.edu.cn

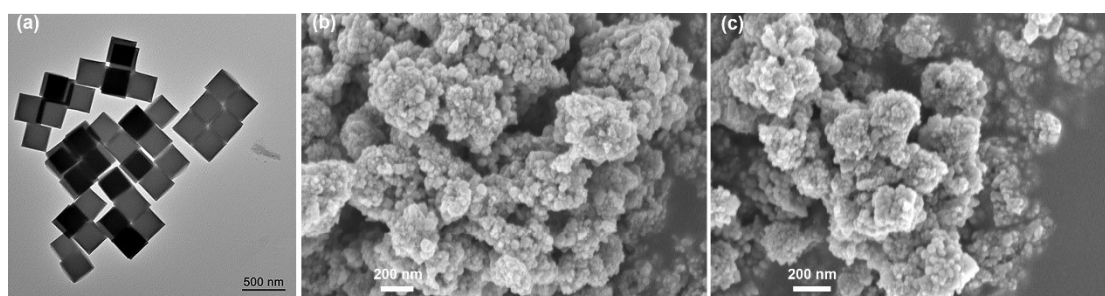


Fig. S1 (a) TEM images of the  $\text{Ni}_3[\text{Co}(\text{CN})_6]_2$ . (b~c) SEM images of the  $\text{NiCoS}_x$  without  $\text{Co}(\text{OH})_2$ .

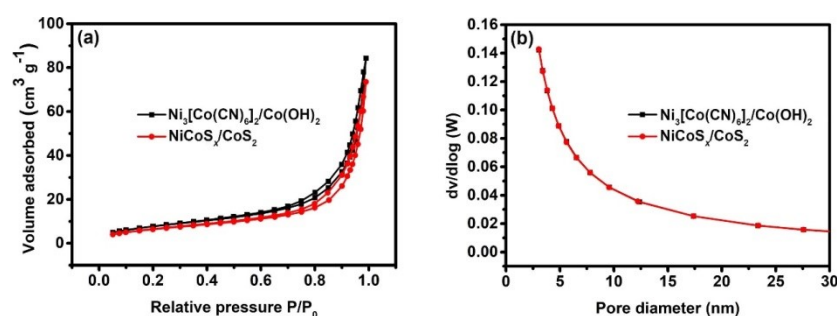


Fig. S2 The nitrogen adsorption-desorption isotherms (a) and the corresponding pore size distribution curves (b) of  $\text{Ni}_3[\text{Co}(\text{CN})_6]_2/\text{Co}(\text{OH})_2$  and  $\text{NiCoS}_x/\text{CoS}_2$ .

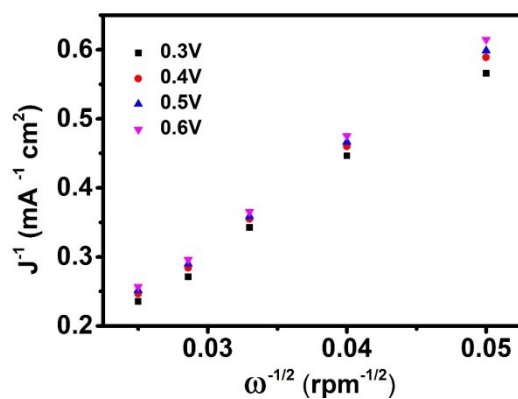


Fig. S3 K-L plots at 0.3 V, 0.4 V, 0.5 V, 0.6 V potentials for NiCoS<sub>x</sub>/CoS<sub>2</sub>.

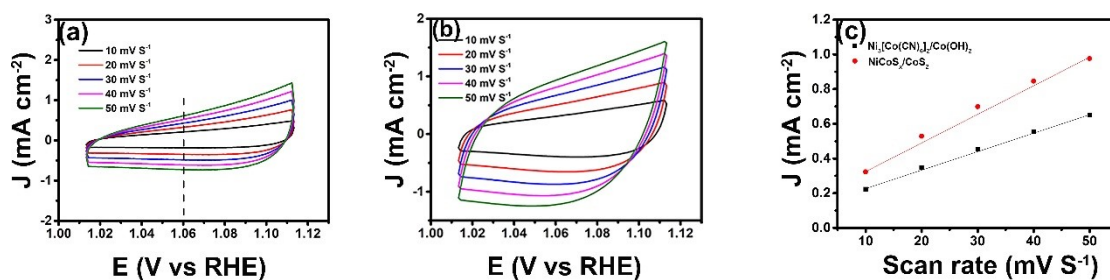


Fig. S4 CV curves of Ni<sub>3</sub>[Co(CN)<sub>6</sub>]<sub>2</sub>/Co(OH)<sub>2</sub> (a), NiCoS<sub>x</sub>/CoS<sub>2</sub> (b), in the potential range of 1.006 - 1.106 V (vs. RHE). (c) The corresponding linear fitting of the capacitive current densities vs. the scan rate.

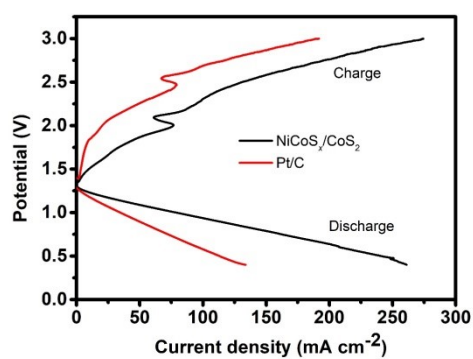


Fig. S5 Charge/discharge polarization curves of the rechargeable liquid ZABs based on the Pt/C catalyst and NiCoS<sub>x</sub>/CoS<sub>2</sub> catalyst, respectively.

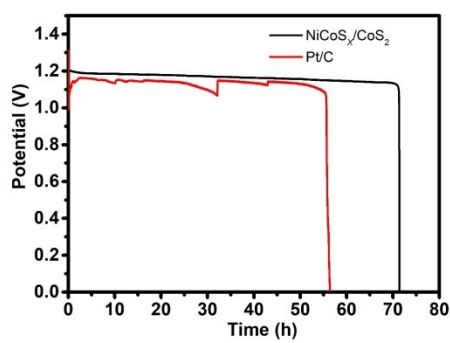


Fig. S6. Typical discharge curves of the rechargeable liquid ZABs based on the Pt/C catalyst and NiCoS<sub>x</sub>/CoS<sub>2</sub> catalyst, respectively.

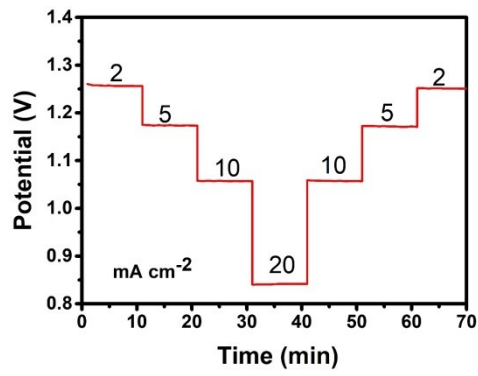


Fig. S7 Rate performance of NiCoS<sub>x</sub>/CoS<sub>2</sub> at different current densities.

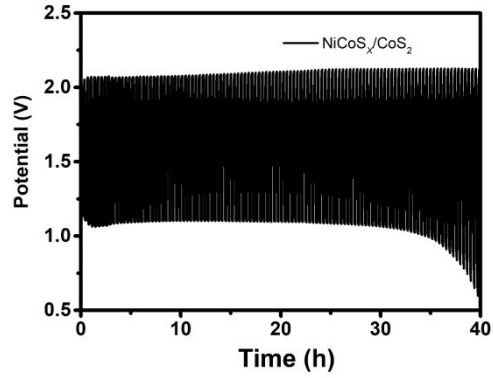


Fig. S8 Cycling stability of NiCoS<sub>x</sub>/CoS<sub>2</sub> at 10 mA cm<sup>-2</sup> charge/discharge.

Table S1 Comparison of the performance of the as-prepared other Co-based Ni-based sulfides catalysts ORR performance.

<i>Catalyst</i>	$E_{1/2}$	<i>Electrolyte</i>	<i>Ref.</i>
NiCoS <sub>x</sub> /CoS <sub>2</sub>	0.80	0.1 M KOH	This work
CoS <sub>2</sub> @MXene	0.80	0.1 M KOH	1
N, P/CoS <sub>2</sub> @TiO <sub>2</sub>	0.71	0.1 M KOH	2
CuS/NiS <sub>2</sub>	0.80	0.1 M KOH	3
Co <sub>9</sub> S <sub>8</sub> @NS-3DrGO	0.82	1 M KOH	4

Table S2 Comparison of the performance of the as-prepared other Co-based Ni-based sulfides catalysts OER performance.

<i>Catalyst</i>	$E_{j=20}$	<i>Electrolyte</i>	<i>Ref.</i>
NiCoS <sub>x</sub> /CoS <sub>2</sub>	1.54	0.1 M KOH	This work
NiCo <sub>2</sub> S <sub>4</sub>	1.56	1 M KOH	5
NiPS	1.63	1 M KOH	6
NiS <sub>2</sub>	1.48	1 M KOH	7
S-NiCoP/CC	1.55	1 M KOH	8

1. S. Han, Y. Chen, Y. Hao, Y. Xie, D. Xie, Y. Chen, Y. Xiong, Z. He, F. Hu and L. Li, *Science China Materials*, 2021, **64**, 1127-1138.
2. L. Guo, J. Deng, G. Wang, Y. Hao, K. Bi, X. Wang and Y. Yang, *Advanced Functional Materials*, 2018, **28**, 1804540.
3. L. An, Y. Li, M. Luo, J. Yin, Y. Q. Zhao, C. Xu, F. Cheng, Y. Yang, P. Xi and S. Guo, *Advanced Functional Materials*, 2017, **27**, 1703779.
4. Y. Li, Y. Zhou, H. Wen, J. Yang, C. Maouche, Q. Liu, Y. Wu, C. Cheng, J. Zhu and X. Cheng, *Dalton Transactions*, 2018, **47**, 14992-15001.
5. Y. Xue, Z. Zuo, Y. Li, H. Liu and Y. Li, *Small*, 2017, **13**, 1700936.
6. M. Wang, A. Saad, X. Li, T. Peng, Q.-T. Zhang, M. Kumar and W. Zhao, *Dalton Transactions*, 2021, **50**, 12870-12878.
7. S. Huang, Z. Jin, P. Ning, C. Gao, Y. Wu, X. Liu, P. Xin, Z. Chen, Y. Jiang and Z. Hu, *Chemical Engineering Journal*, 2021, **420**, 127630.
8. L. Mai-Thi, N. Hoang-Thy and Q. Bui, *Materials Chemistry and Physics*, 2021, **270**, 124746.