Electronic Supplementary Information (ESI) for New Journal of Chemistry

NiFe-CN catalysts derived from Solid-phase Exfoliation of NiFe-Layered Double Hydroxide for CO_2 Electroreduction

Yingke Fu[a], Lin Chen[a], Ying Xiong[a*], Hao Chen[a], Ruishi Xie[a], Bin Wang[a], Yaping Zhang[a], Tianxia Liu[b*], Ping Zhang[a*]

[a]*State Key Laboratory of Environment-friendly Energy Materials, School of Materials Science and Engineering, Southwest University of Science and Technology, Mianyang, 621010, PR China*

[b]*School of Chemistry and Chemical Engineering, North Minzu University, Yinchuan 750021, China*

Table of Contents

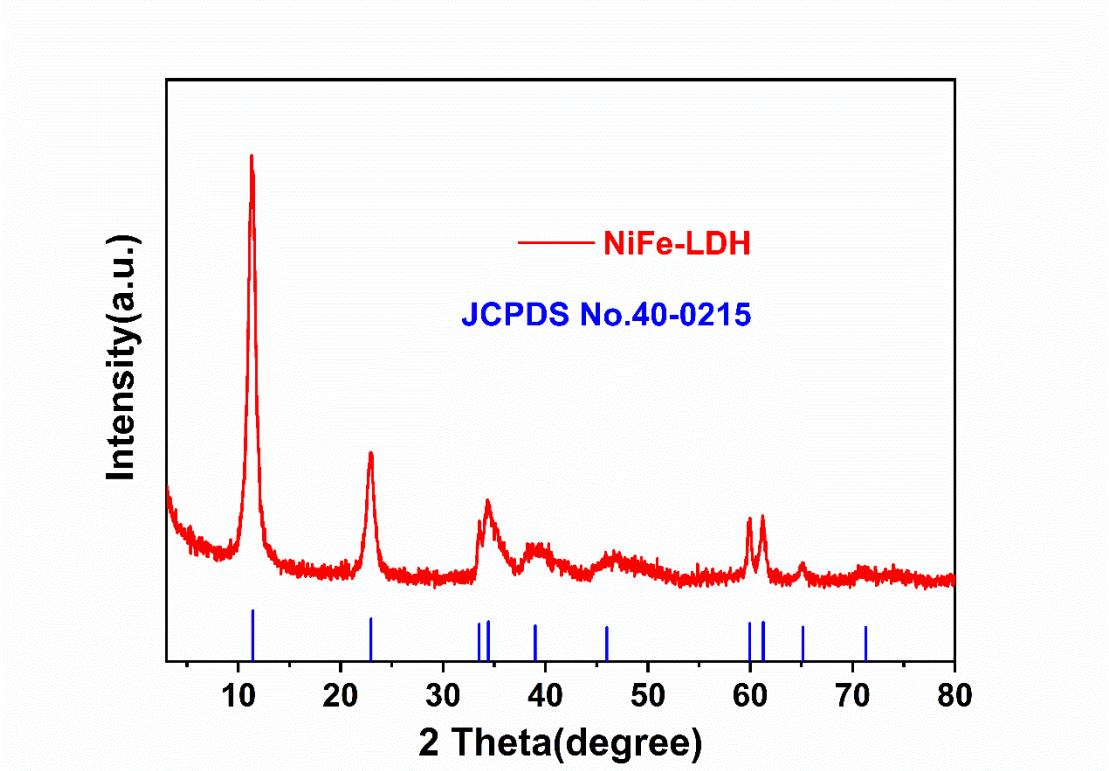

Figure S1. XRD pattern of NiFe-LDH

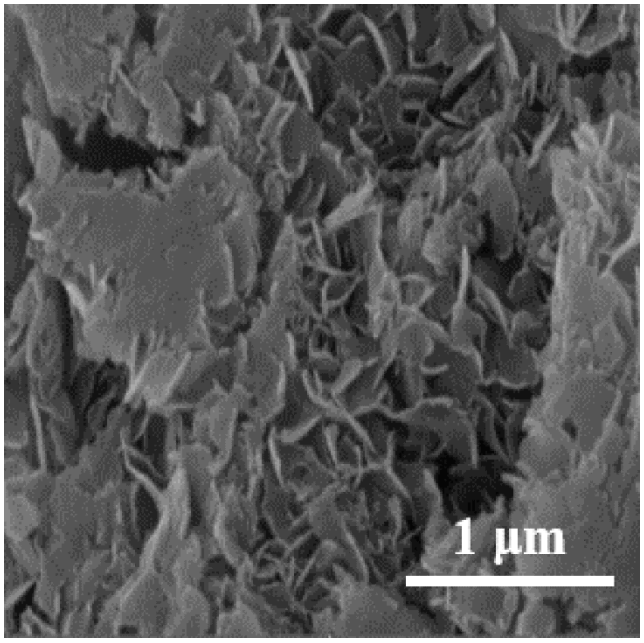

Figure S2. SEM image of NiFe-LDH.

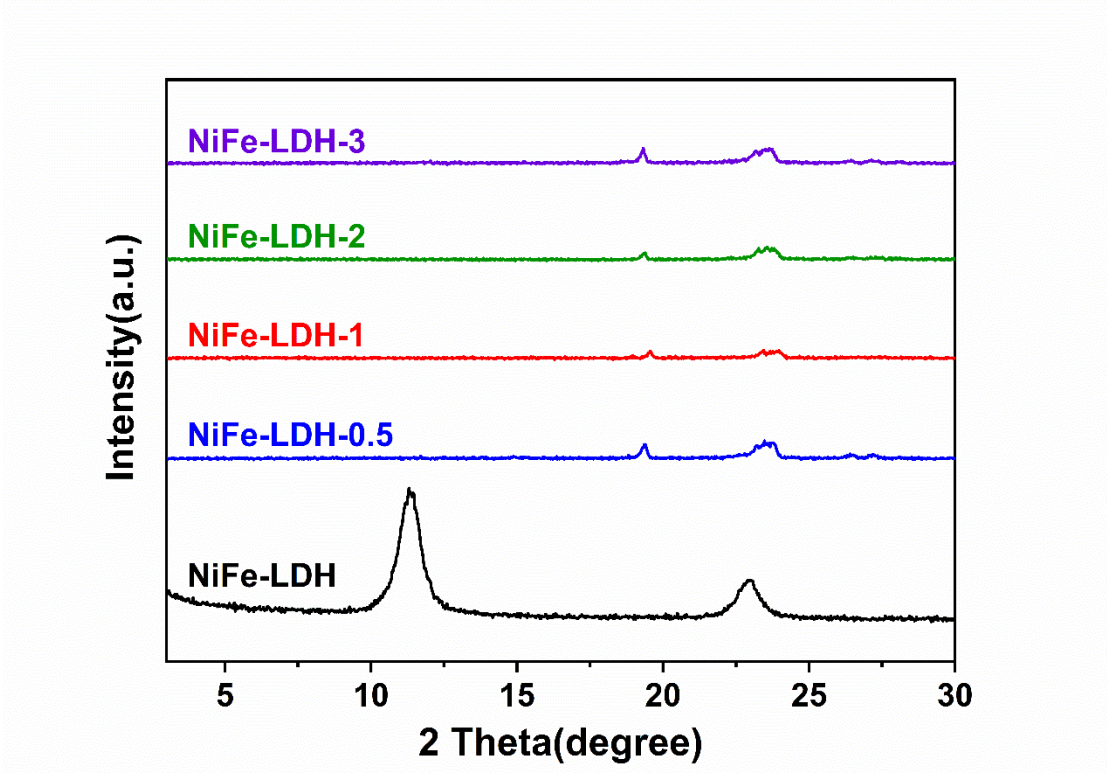

Figure S3. XRD pattern of exfoliated NiFe-LDH

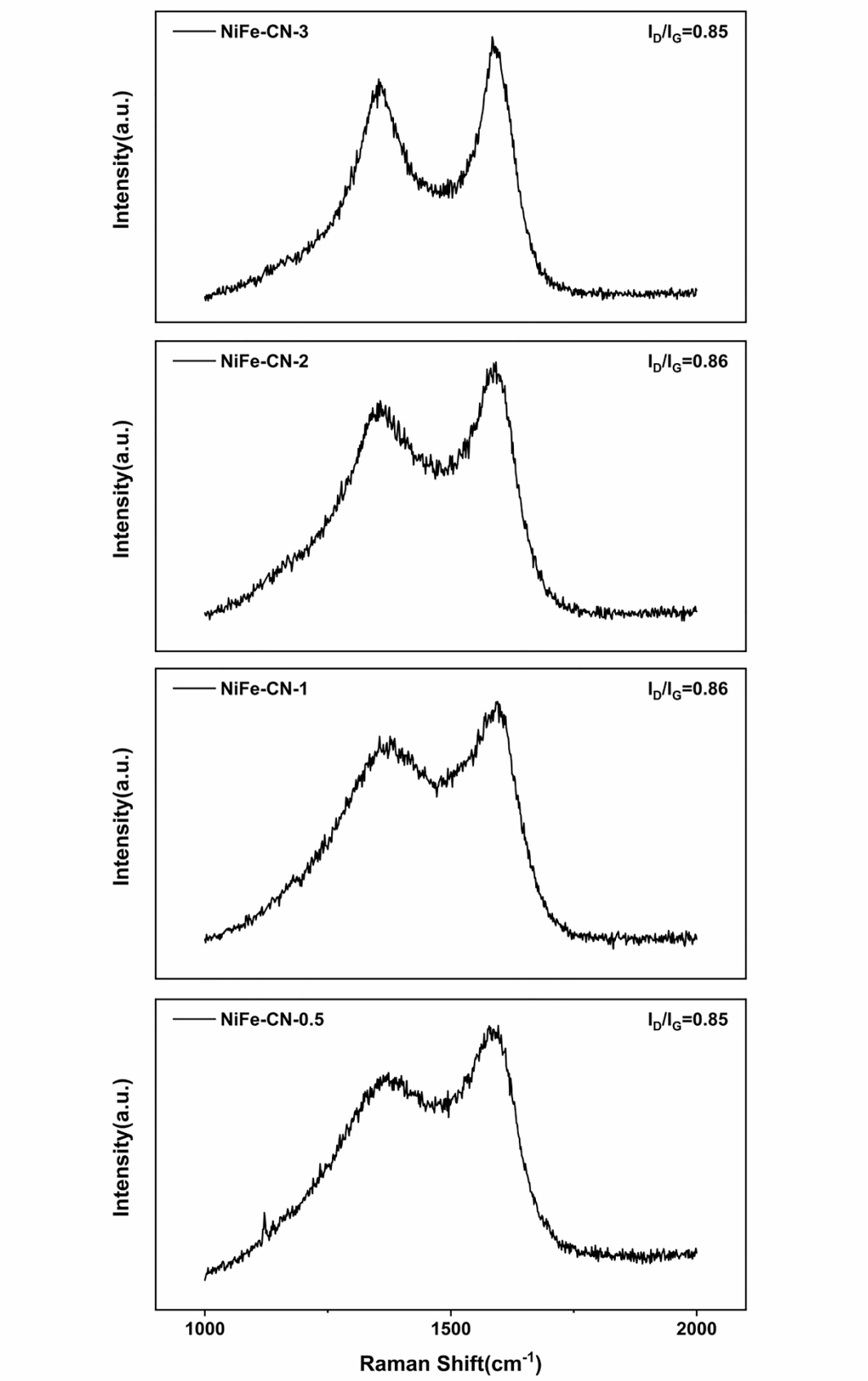

Figure S4. Raman spectrum of NiFe-NC-0.5, NiFe-NC-1, NiFe-NC-2, and NiFe-NC-3.

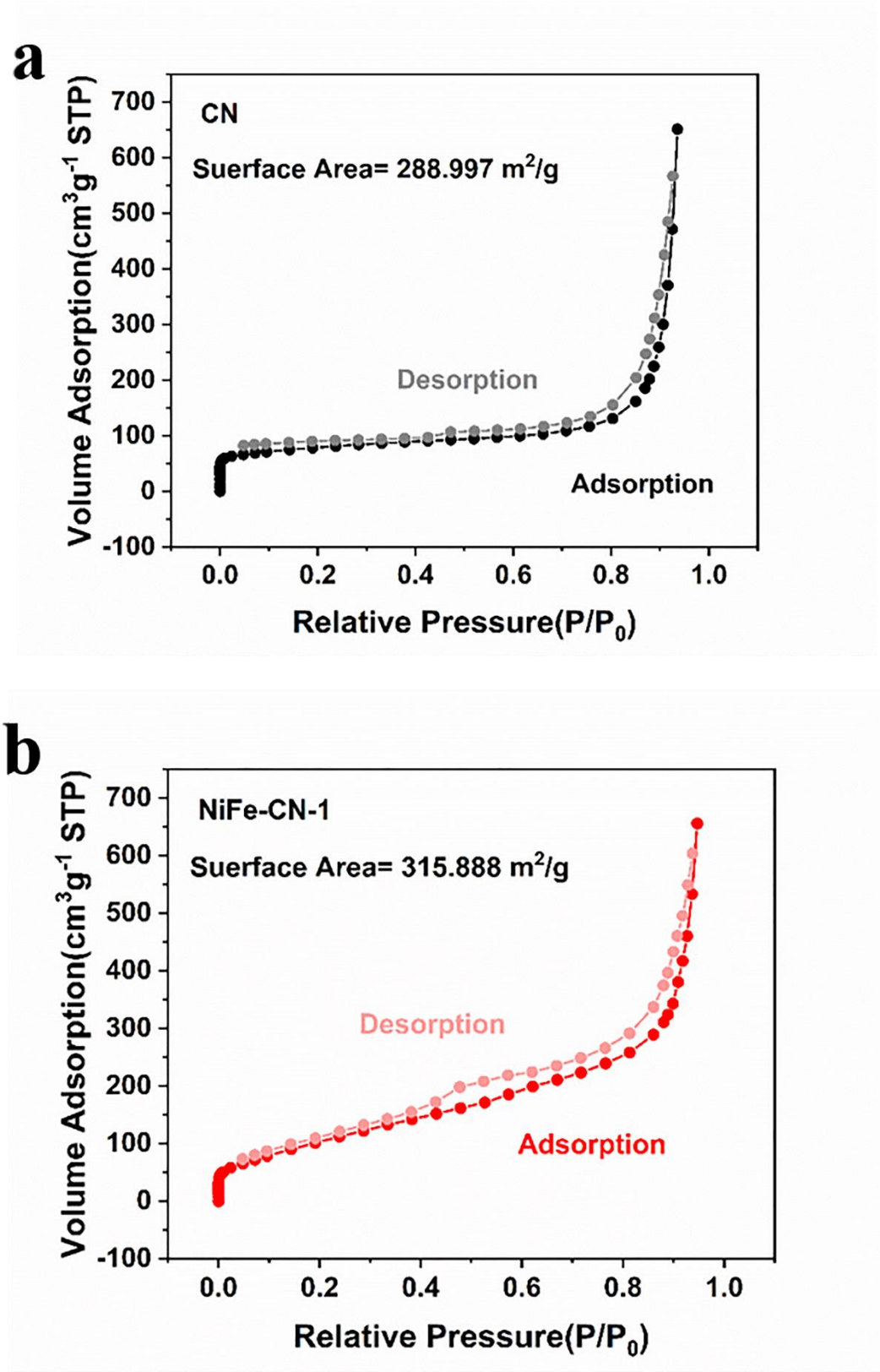

Figure S5. (a, b) Nitrogen adsorption-desorption isotherms of CN and NiFe-NC-1.

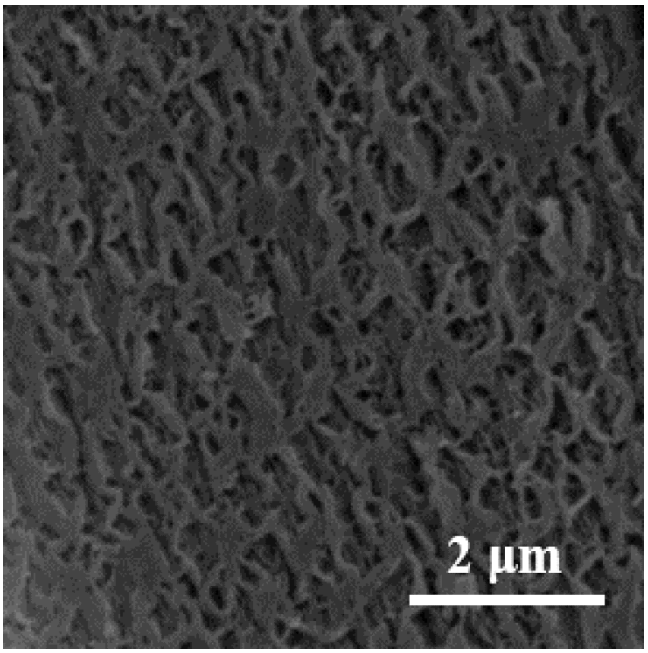

Figure S6. SEM image of CN.

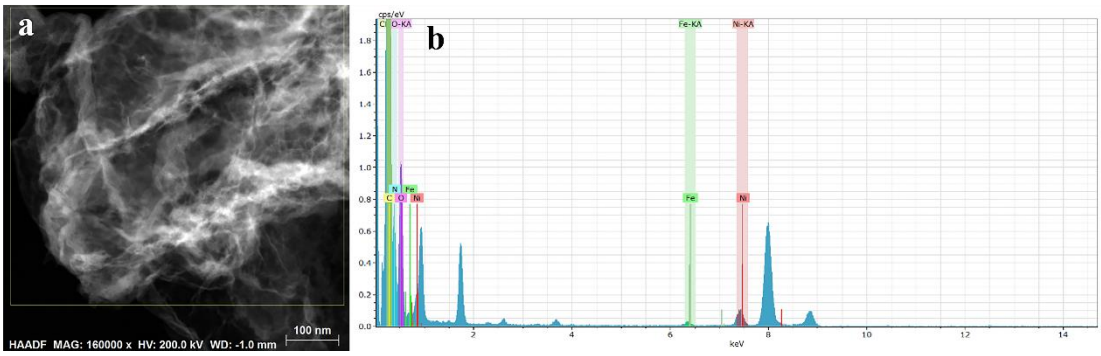

Figure S7. EDS of NiFe-NC-1.

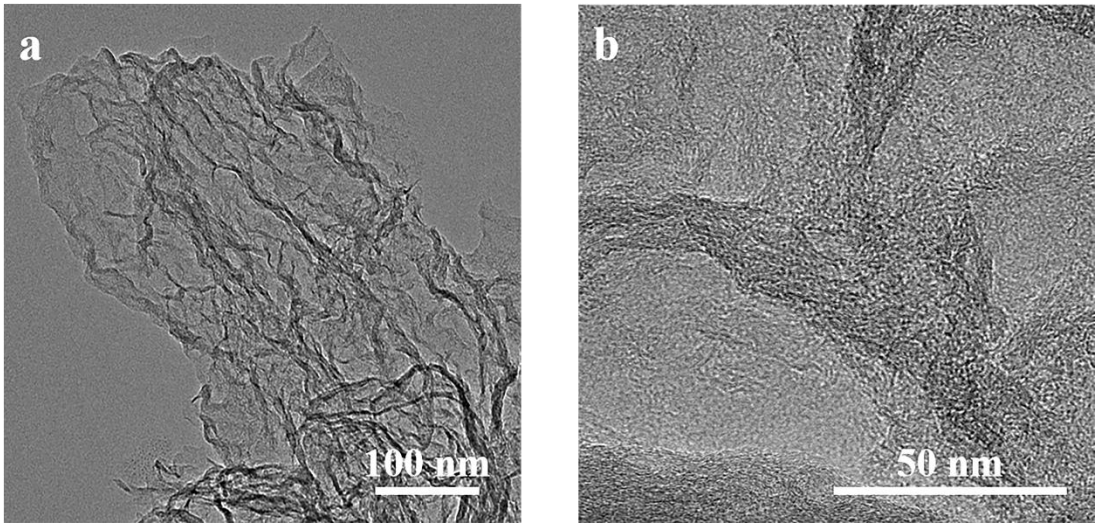

Figure S8. (a) TEM images, (b) HRTEM image of NiFe-NC-0.5.

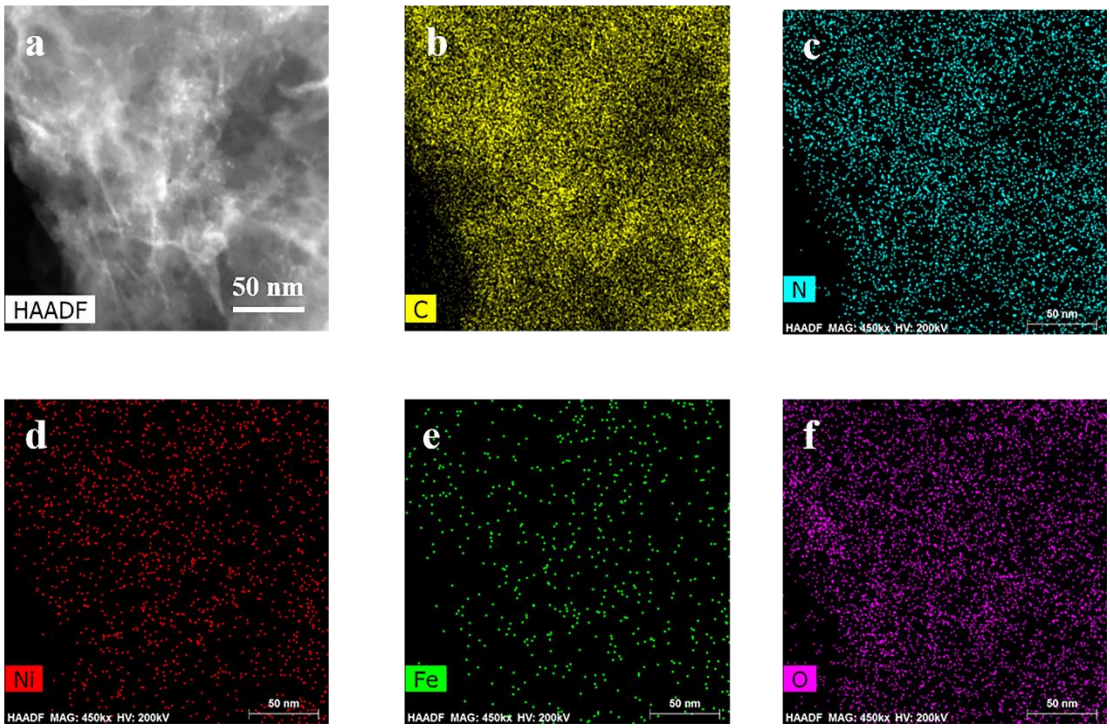

Figure S9. (a) STEM image, (b-f) the corresponding elemental mapping images of NiFe-NC-0.5.

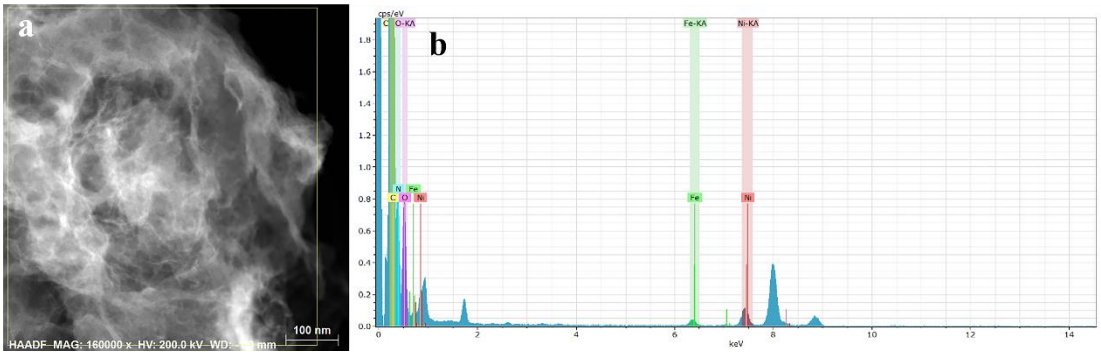

Figure S10. EDS of NiFe-NC-0.5.

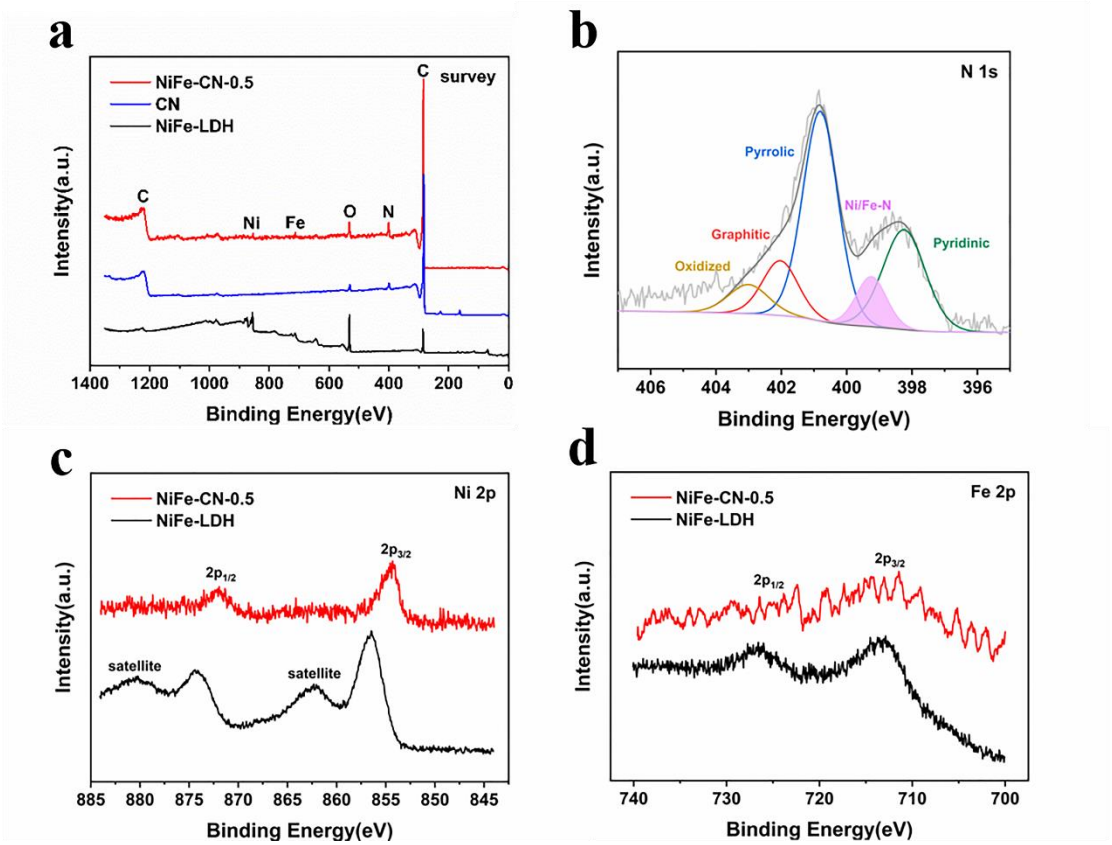

Figure S11. (a) XPS spectra of NiFe-LDH and NiFe-CN-0.5. (b) N 1s XPS spectra of NiFe-CN-0.5. (c) Ni 2p XPS spectra of NiFe-LDH and NiFe-CN-0.5. (d) Fe 2p XPS spectra of NiFe-LDH and NiFe-CN-0.5.

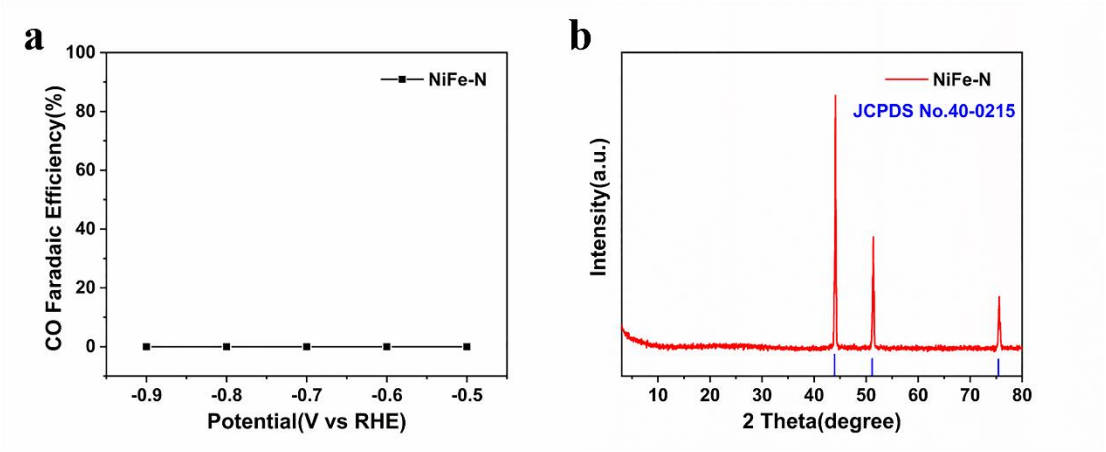

Figure S12. (a) Faradaic efficiency of CO of NiFe-N at different potentials in CO_2-saturated. (b) XRD of NiFe-N.

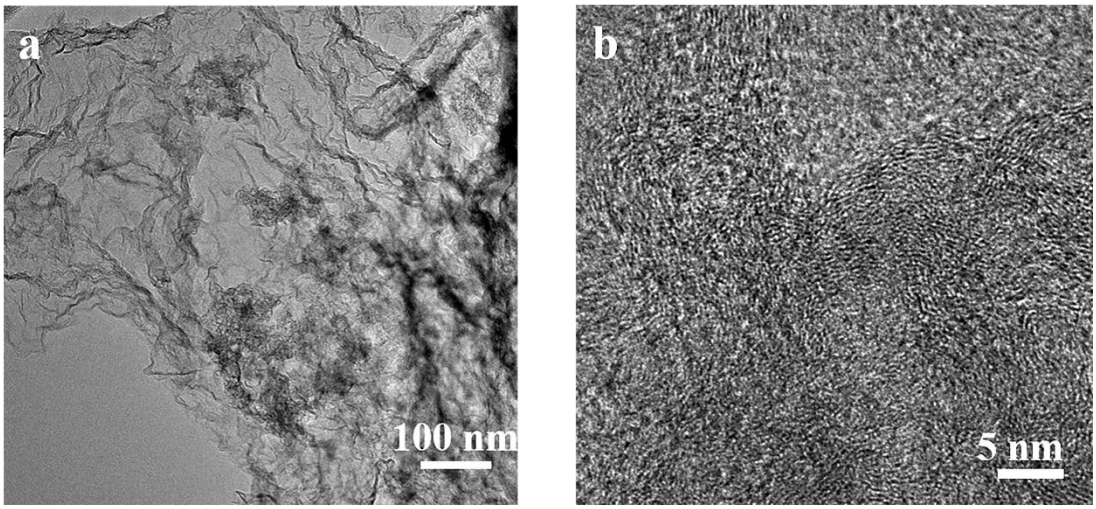

Figure S13. (a) TEM images, (b) HRTEM image of NiFe-NC-1 after electrolysis.

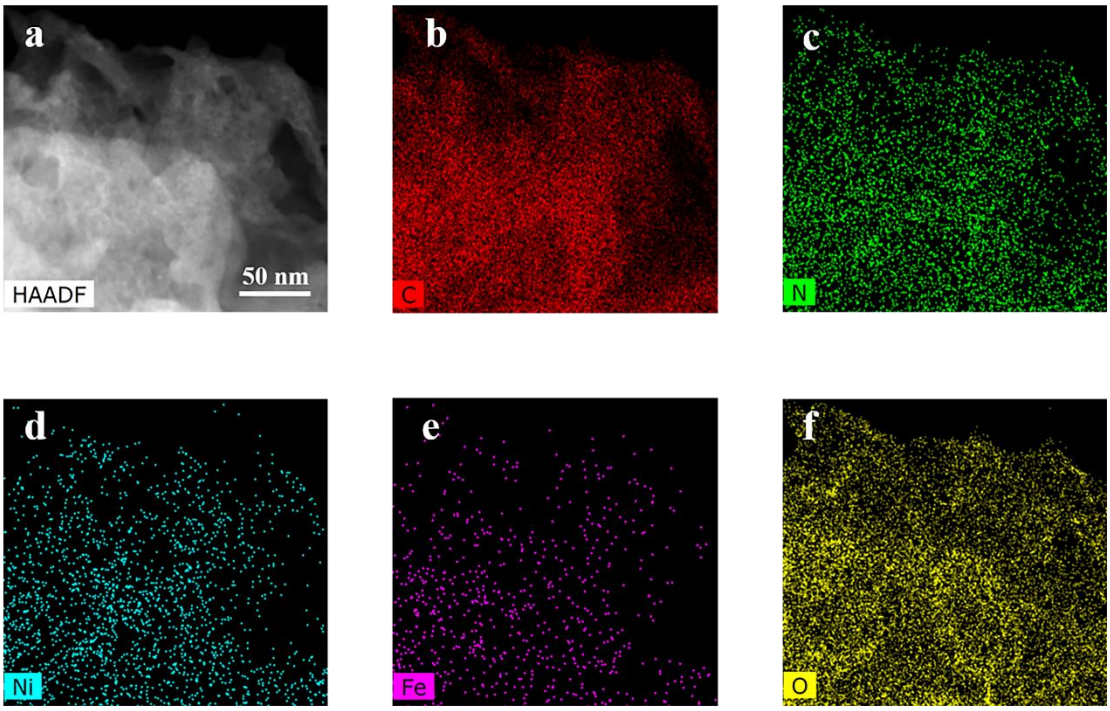

Figure S14. (a) STEM image, (b-f) the corresponding elemental mapping images of NiFe-NC-1 after electrolysis.

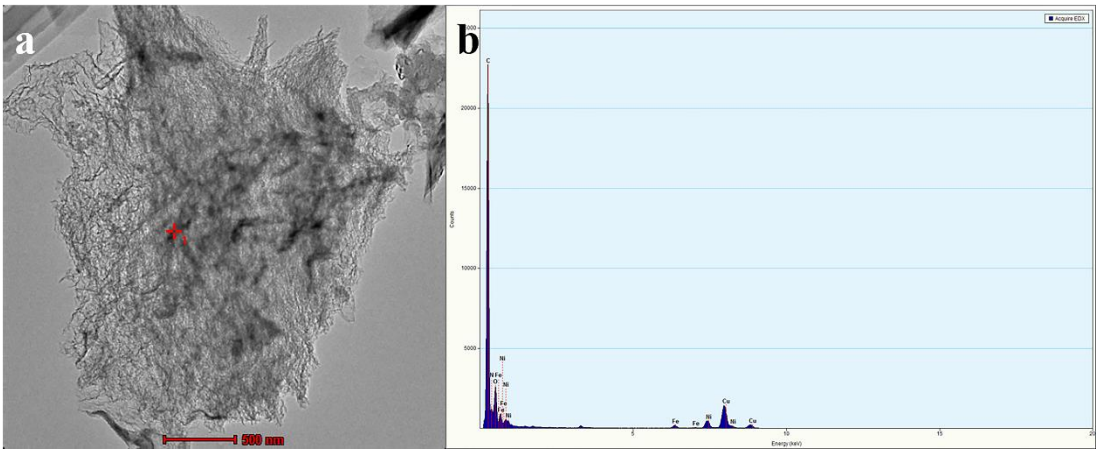

Figure S15. EDS of NiFe-NC-1 after electrolysis.

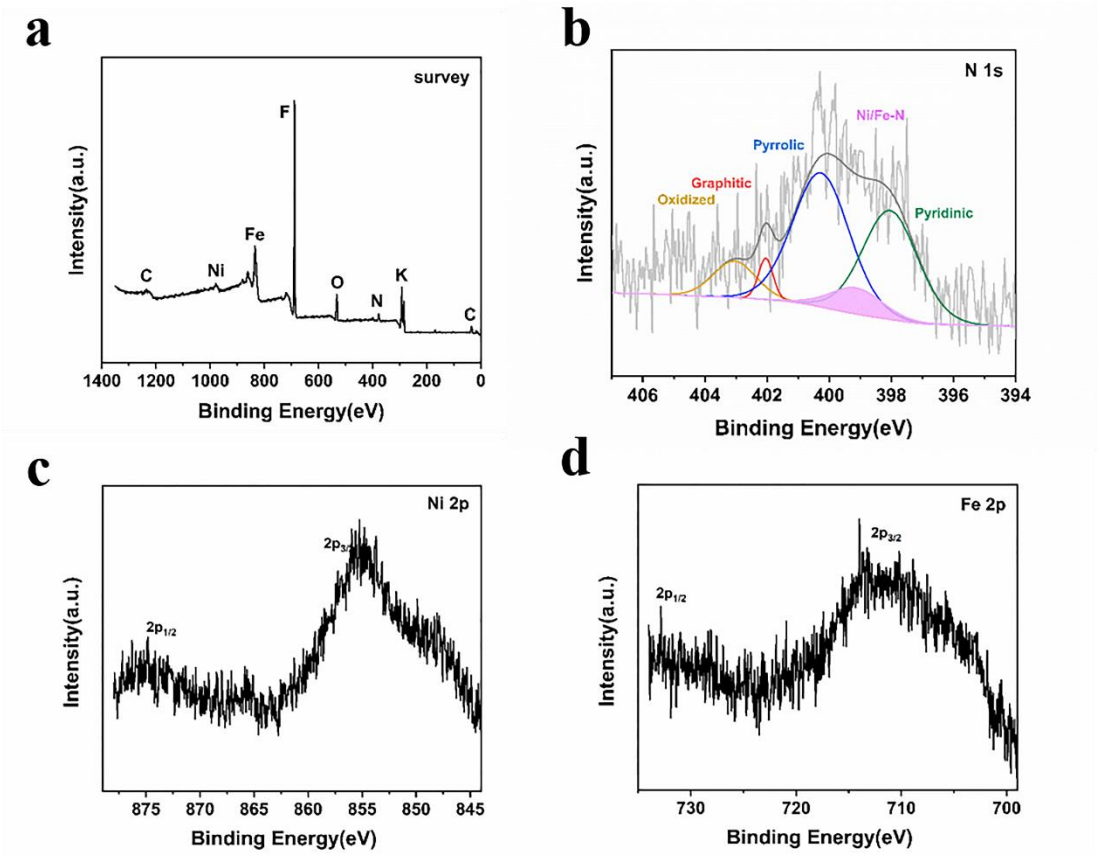

Figure S16. (a) XPS spectra of NiFe-CN-1 after electrolysis. (b) N 1s XPS spectra of NiFe-CN-1 after electrolysis. (c) Ni 2p XPS spectra of NiFe-CN-1 after electrolysis. (d) Fe 2p XPS spectra of NiFe-CN-1 after electrolysis.

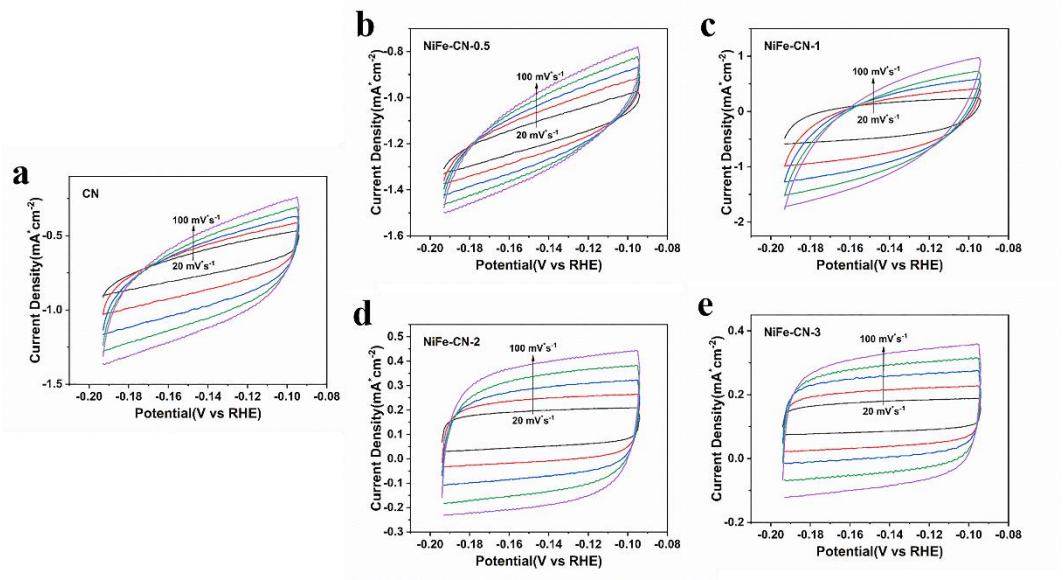

Figure S17. The cyclic voltammetry curves at different scan rates (20, 40, 60, 80, and 100 mV s^{-1}) of (a) CN, (b) NiFe-NC-0.5, (c) NiFe-NC-1, (d) NiFe-NC-2, and (e) NiFe-NC-3.

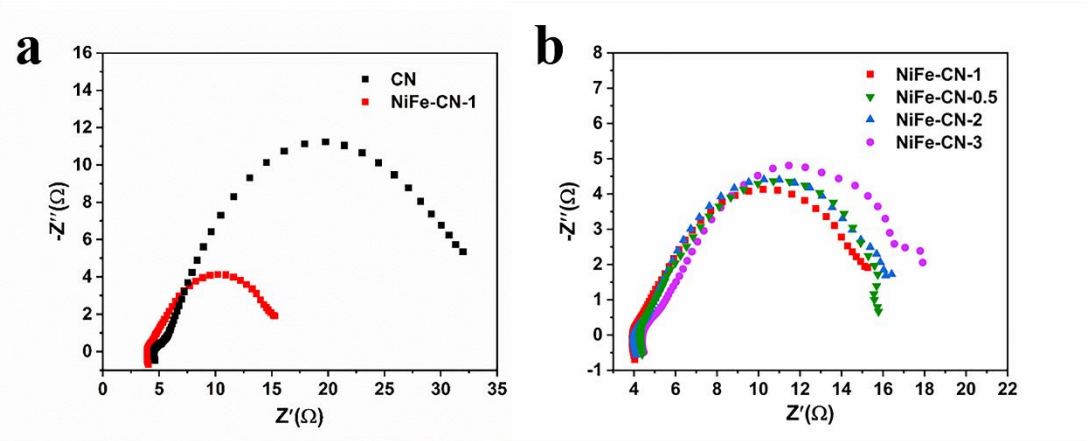

Figure S18. Nyquist plots at a potential of -0.8 V (vs RHE) of the samples.

Table S1. Evolution of the binding energies of XPS peaks.

Sample	Ni $2p_{3/2}$	Ni $2p_{1/2}$	Fe $2p_{3/2}$	Fe $2p_{1/2}$
NiFe-LDH	856.45	874.21	713.14	726.64
NiFe-NC-0.5	854.27	871.97	713.18	726.65
NiFe-NC-1	854.46	871.90	713.25	762.81

Table S2. Raman spectra parameters of CN and NiFe-CN.

Sample	D band (cm^{-1})	G band (cm^{-1})	I_D/I_G
CN	1166.83	1398.24	0.83
NiFe-NC-0.5	1777.41	2076.52	0.85
NiFe-NC-1	1141.42	1328.24	0.86
NiFe-NC-2	1188.51	1370.98	0.86
NiFe-NC-3	1235.43	1449.03	0.85

Table S3. Comparison of CO_2 reduction performance on various catalysts of the NiFe-NC with recently reported electrocatalysts.

Catalysts	Electrolyte	Potential V (vs RHE)	FE CO (%)	Reference
NiFe-NC	0.5 M $KHCO_3$	-0.8	94.4	This work
FeMn-N-C	0.1 M $KHCO_3$	-0.65	80	[1]
Fe-N-C/Graphene	0.1 M $KHCO_3$	-0.5	80	[2]
NFe-CNT/CNS	0.5 M $KHCO_3$	-0.6	60	[3]
NiSAs/N-C	0.5 M $KHCO_3$	-0.89	71.9	[4]
CoNi-NC	0.1 M $KHCO_3$	-0.5	55	[5]
CNT-N-NiFe	0.5 M $KHCO_3$	-0.7	82	[6]
Cu-Sn NWs	0.1 M $KHCO_3$	-0.7	82	[7]
CoPc-CN/CNT(3.5%)	0.1 M $KHCO_3$	-0.97	95	[8]
CuPd NP/C	0.1 M $KHCO_3$	-0.9	87	[9]
Au-Cu NP/C	0.5 M $KHCO_3$	-0.7	50	[10]

REFERENCES

1. A. S. Varela, N. R. Sahraie, J. Steinberg, W. Ju, H. S. Oh and P. Strasser, *Angew Chem Int Edit*, 2015, **54**, 10758-10762.
2. C. H. Zhang, S. Z Yang, J. J. Wu, M. J. Liu, S. Yazdi, M. Q. Ren, J. W. Sha, J. Zhong, K. Q. Nie, A. S. Jalilov, Z. Y. Li, H. M. Li, B. I. Yakobson, Q. Wu, E. L. Ringe, H. Xu, P. M. Ajayan and J. M. Tour, *Adv Energy Mater*, 2018, **8**, 1703487.
3. F. P. Pan, H. L. Zhao, W. Deng, X. H. Feng and Y. Li, *Electrochim Acta*, 2018, **273**, 154-161.
4. C. M. Zhao, X. Y. Dai, Y. Tao, W. X. Chen, X. Q. Wang, J. Wang, J. Yang, S. Q. Wei, Y. Wu and Y. D. Li, *J Am Chem Soc* **2017,** 139, 8078.
5. Q. He, D. Liu, J. H. Lee, Y. Liu, Z. Xie, S. Hwang, S. Kattel, L. Song and J. G.

Chen, *Angew Chem Int Edit*, 2020, **59**, 3033-3037.
6. H. Chen, P. Zhang, R. S. Xie, Y. Xiong, C. H. Jia, Y. K. Fu, P. A Song, L. Chen, Y. P. Zhang and T. Liao, *Adv Mater Interfaces*, 2021, **8**, 2101165..
7. W. B. Ju, F. Z. Jiang, H. Ma, Z. Y. Pan, Y. B. Zhao, F. Pagani, D. Rentsch, J. Wang and C. Battaglia, *Adv Energy Mater*, 2019, **9**, 1901514.
8. X. Zhang, Z. S. Wu, X. Zhang, L. W. Li, Y. Y. Li, H. M. Xu, X. X. Li, X. L. Yu, Z. S. Zhang, Y. Y. Liang and H. L. Wang, *Nat Commun*, 2017, **8**, 14675.
9. Y. Mun, S. Lee, A. Cho, S. Kim, J. W. Han and J. Lee, *Appl Catat B-Environ*, 2019, **246**, 82-88.
10. J. H. Zhou, D.W. Lan, S. S. Yang, Y. Guo, K. Yuan, L. X. Dai and Y. W. Zhang, Inorg Chem Front, 2018, **5**, 1524-1532.