Supporting Information

Layer-structured FeCo bihydroxide as ultra-stable bifunctional

electrocatalyst for water splitting at high current densities

Chaoyang Sun,^a Hui Wang,^a* Shan Ji,^{b**} Xuyun Wang,^a Vladimir Linkov,^c Xinlong Tian,^d Long Yao,^a Jiarui Zhao,^a Rongfang Wang ^a*

a. State Key Laboratory Base for Eco-Chemical Engineering, College of Chemical Engineering, Qingdao University of Science and Technology, Qingdao, 266042, China

b. College of Biological, Chemical Science and Engineering, Jiaxing University, Jiaxing, 314001, China

c. South African Institute for Advanced Materials Chemistry, University of the Western Cape, Cape Town, 7535, South Africa.

d. State Key Laboratory of Marine Resource Utilization in South China Sea, Hainan Provincial Key Lab of Fine Chemistry, School of Chemical Engineering and Technology, Hainan University, Haikou, 570228, China

Physical characterizations

The X-ray diffraction (XRD) was done on Shimadzu XD-3A Instrument, which was fitted with filtered Cu-K α radiation ($\lambda = 0.15418$ nm) and operated at 30 mA and 40 kV. The 2θ scan rate for XRD analysis was set at 5° min⁻¹. JEOL (JEM-2000 FX) microscope operated at 200 kV was used for transmission electron microscopy (TEM), high angle annular dark field scanning transmission electron microscopy (STEM) images and selected area electron diffraction (SAED) analysis. X-ray photoelectron spectra (XPS) were carried out on VG Escalab210 Spectrometer with Mg 300 W X-ray source.

Electrochemical measurements

A three-electrode electrochemical cell linked with a potentiostat/galvanostat (CHI 760, CH Instruments) was applied to evaluate the HER and OER electrocatalytic properties. In this three-electrode cell, Hg/HgO and graphite rod were used as reference electrode (RE) and counter electrode (CE) respectively. 1.0 M KOH aqueous solution was used as electrolyte for all electrocatalytic testing. Electrochemical impedance spectroscopy (EIS) spectra were measured at corresponding OER and HER electrode potentials from 0.01 to 1,000,000 Hz with an amplitude of 5 mV. Linear sweep voltammetry (LSV) was done with a scan rate of 5 mV s⁻¹. All potentials stated in this work were normalized to the reversible hydrogen electrode (RHE). *iR* compensation was applied for all electrochemical experiments.



Figure S1. SEM images of NM(a,b,c); SEM pictures of different time of (d,e,f) 10 s,

and (g,h,i) 60 s.



Figure S2. Survey XPS spectrum of FeCo-LDH/NM



Figure S3. (a) HER LSV curves of the FeCo-LDH/NM samples prepared with different time, (b) OER LSV curves of the FeCo-LDH / NM samples prepared with different time.



Figure S4. long-term overall water splitting chronopotentiometry of FeCo-LDH/NM at 1000 mA cm⁻² for (a) 10 h; (b) 30 h; (c) 60 h; (d) 90 h; (e) 120 h; (f) 150 h.



Figure S5. Cyclic voltammetry plots of (a) FeCo-LDH/NM; (b) NM for HER; (c) FeCo-LDH/NM; and (e) NM for OER.



Figure S6. Picture of size measurement of electrode material used in electrochemical

test

Table S1. Comparison of the stability of Fe-based electrocatalysts in overall water

Catalysts	Morphology	η _{50,overall} (V)	stability(h)	Ref.
FeCo-LDH/NM	Nanosheets	1.69	Over 150 h	This work
FeCoP/C NF	Nanosheet	1.68	3 h	1
FeCo/Co ₂ P	Nanoparticles	1.81	12 h	2
Fe ₂ P/CoP/Ni ₅ P ₄	Core-Shell	1.71	24 h	3
FeCoOOH	Nanosheets	1.72	15 h	4
Co-FeNiO _X	Nanosheets	1.70	20 h	5
Ni ₃ Se ₄ @NiFeLDH	Nanosheets	1.69	100 h	6
NiFeOH/CoSx	Nanosheets	1.71	24 h	7
CoNi/CoFe ₂ O ₄	Nanosheets	1.65	48 h	8
Fe _{13.7%} -Ni ₃ S ₂	Nanowire	1.6	14 h	9
Ni ₂ P and FeP	nanoparticles	1.76	100 h	10

hydrolysis.

Table S2. Comparison of the stability of different substrates in water splitting.

Substrate material	Stability	Ref.	
stainless steel wires (SSW)	24 h	11	
Carbon substrate	6-30 h	12, 13	
Nickel foam (NF)	24-120 h	14, 15	
Cu mesh	70 h	16	
Nickel mesh (NM)	Over 150 h	This work	

Reference

- R. Xiang, Y. Duan, C. Tong, L. Peng, J. Wang, S. S. A. Shah, T. Najam, X. Huang and Z. Wei, Electrochimica Acta, 2019, 302, 45-55.
- Q. Shi, Q. Liu, Y. Ma, Z. Fang, Z. Liang, G. Shao, B. Tang, W. Yang, L. Qin and X. Fang, Advanced Energy Materials, 2020, 10, 1903854.
- 3. Y. Du, J. Chen, L. Li, H. Shi, K. Shao and M. Zhu, *ACS Sustainable Chemistry & Engineering*, 2019, 7, 13523-13531.
- 4. T. H. Nguyen, J. Lee, J. Bae and B. Lim, *Chemistry*, 2018, 24, 4724-4728.
- 5. Z. Wu, X. Wang, J. Huang and F. Gao, *Journal of Materials Chemistry A*, 2018, 6, 167-178.
- T. Zhang, L. Hang, Y. Sun, D. Men, X. Li, L. Wen, X. Lyu and Y. Li, *Nanoscale Horizons*, 2019, 4, 1132-1138.
- R. Bose, V. R. Jothi, K. Karuppasamy, A. Alfantazi and S. C. Yi, *Journal of Materials Chemistry A*, 2020, 8, 13795-13805.
- S. Li, S. Sirisomboonchai, A. Yoshida, X. An, X. Hao, A. Abudula and G. Guan, *Journal of Materials Chemistry A*, 2018, 6, 19221-19230.
- 9. X. Wang, W. Zhang, J. Zhang and Z. Wu, *ChemElectroChem*, 2019, 6, 4550-4559.
- 10. H. J. Shin, S. W. Park and D. W. Kim, *International Journal of Energy Research*, 2020, 44, 11894-11907.
- 11. V. R. Jothi, K. Karuppasamy, T. Maiyalagan, H. Rajan, C. Y. Jung and S. C. Yi, *Advanced Energy Materials*, 2020, **10**, 1904020.
- 12. S. Cui, Y. He and X. Bo, Journal of Electroanalytical Chemistry, 2019, 853, 113557.
- H.-W. Lin, D. Senthil Raja, X.-F. Chuah, C.-T. Hsieh, Y.-A. Chen and S.-Y. Lu, *Applied Catalysis B: Environmental*, 2019, 258, 118023.
- 14. X. Yi, X. He, F. Yin, B. Chen, G. Li and H. Yin, *Dalton Trans*, 2020, 49, 6764-6775.
- 15. Q. Xu, W. Gao, M. Wang, G. Yuan, X. Ren, R. Zhao, S. Zhao and Q. Wang, *International Journal of Hydrogen Energy*, 2020, **45**, 2546-2556.
- J. Zhao, W. Wu, X. Jia, Z. Zhao and X. Hu, *Catalysis Science & Technology*, 2020, 10, 1803-1808.