



Supplementary Materials

Pearson's Principle-Inspired Robust 2D Amorphous Ni-Fe-Co Ternary Hydroxides on Carbon Textile for High-Performance Electrocatalytic Water Splitting

Rong Hu ^{1,†}, Huiyu Jiang ^{1,†}, Jinglin Xian ^{1,†}, Shiyun Mi ¹, Liyun Wei ¹, Guangyu Fang ¹, Jiayue Guo ¹, Siqi Xu ¹, Ziyang Liu ¹, Huanyu Jin ^{2,*}, Huimin Yu ^{3,*} and Jun Wan ^{1,*}

¹ State Key Laboratory of New Textile Materials and Advanced Processing Technologies, Hubei Key Laboratory of Biomass Fibers and Eco-Dyeing & Finishing, Wuhan Textile University, Wuhan 430200, China; fulonlon@163.com (R.H.); huiyujiang@wtu.edu.cn (H.J.); xianjinglin1006@163.com (J.X.); mishiyan0426@163.com (S.M.); wly9520ft@163.com (L.W.); mmyz1031@163.com (G.F.); 2105250704@mail.wtu.edu.cn (J.G.); 1905250434@mail.wtu.edu.cn (S.X.); lzy1341969071@163.com (Z.L.)

² Institute for Sustainability, Energy and Resources, The University of Adelaide, Adelaide, SA 5005, Australia

³ Future Industries Institute, University of South Australia, Mawson Lakes Campus, Adelaide, SA 5095, Australia

* Correspondence: huanyu.jin@adelaide.edu.au (H.J.); huimin.yu@unisa.edu.au (H.Y.); wanj@wtu.edu.cn (J.W.)

† These authors contributed equally to the work.

1. Figures S1 to S2

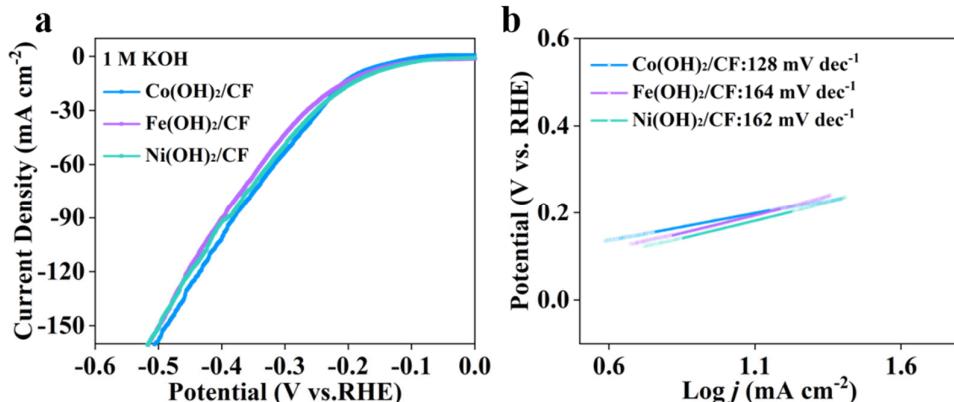


Figure S1. The LSV curves (a) and Tafel slope (b) of monohydroxide hydroxides.

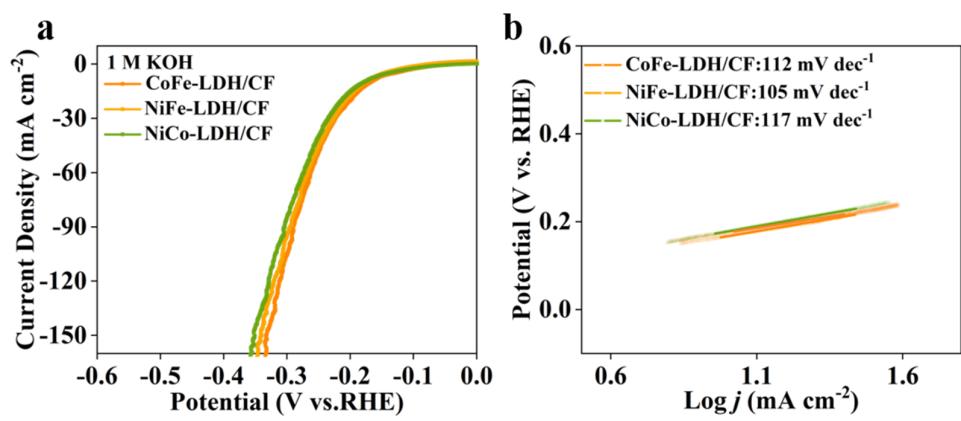


Figure S2. The LSV curves (a) and Tafel slope (b) of binary hydroxides.

2. Table S1

Table S1. Comparison of HER performance in common LDH.

Reference	Material	Electrolyte	η^{10} (mV vs.RHE)	Tafel slope (mV dec ⁻¹)
This Work	NiFeCo-LDH/CF	1 M KOH	151	67
<i>Small</i> 2019, 15, e1902551 [1]	CoFe LDH	1 M KOH	120	134
<i>Electrochim. Acta</i> 2021, 395. [2]	NiFeW-LDHs	1 M KOH	117	112
<i>J. Mater. Chem. A</i> 2020, 8, 2490-2497.[3]	CoFe LDH	1 M KOH	194	127
<i>Adv Mater</i> 2018, 30, 1706279.[4]	NiFe-LDH	1 M KOH	269	153
<i>ACS Sustainable Chem. Eng.</i> 2019, 7, 10035-10043.[5]	NiFe-LDHs	1 M KOH	145	90
<i>Dalton Trans.</i> 2017, 46, 8372-8376.[6]	NiCo-LDH	1 M KOH	130	141
<i>J. Mater. Chem. A</i> 2016, 4, 7245-7250.[7]	Ni _{2.5} Co _{0.5} Fe-OH	1 M KOH	110	93
<i>ACS Appl. Energy Mater.</i> 2018, 1, 1200-1209.[8]	CoFe LDH@g-C ₃ N ₄	1 M KOH	210	79
<i>Adv. Mater.</i> 2017, 29, 1700017. [9]	NiFeLDH@DG	1 M KOH	270	110
<i>Nanoscale</i> 2018, 10, 19484-19491.[10]	NiFeLDH	1 M KOH	220	74
<i>Energy Environ. Sci.</i> 2019, 12, 572-581. [11]	NiFeLDH	1 M KOH	210	78
<i>Small</i> 2018, 14, 1702568.[12]	CoFe-OH/Nifoam	1 M KOH	110	72
<i>ACS Appl. Mater. Inter.</i> 2018, 10, 42453-42468.[13]	CoFe LDH with trace Fe	1 M KOH	170	83
<i>ACS Appl. Mater. Inter.</i> 2016, 8, 34474-34481.[14]	Defected CoFe LDH	1 M KOH	300	95
<i>Adv. Funct. Mater.</i> 2018, 28, 1804361.[15]	Au/ CoNi LDH	1 M KOH	210	92

3. Reference

- Meng, X.; Han, J.; Lu, L.; Qiu, G.; Wang, Z.L.; Sun, C. Fe(2+) -Doped Layered Double (Ni, Fe) Hydroxides as Efficient Electrocatalysts for Water Splitting and Self-Powered Electrochemical Systems. *Small* **2019**, 15, e1902551.
- Ding, L.; Li, K.; Xie, Z.; Yang, G.; Yu, S.; Wang, W.; Cullen, D.A.; Yu, H.; Zhang, F. W-Induced Morphological Modification of NiFe Layered Double Hydroxides as Efficient Electrocatalysts for Overall Water Splitting. *Electrochim. Acta* **2021**, 395, 139199.
- Liu, S.; Zhu, J.; Sun, M.; Ma, Z.; Hu, K.; Nakajima, T.; Liu, X.; Schmuki, P.; Wang, L. Promoting the Hydrogen Evolution Reaction through Oxygen Vacancies and Phase Transformation Engineering on Layered Double Hydroxide Nanosheets. *J. Mater. Chem. A* **2020**, 8, 2490-2497.
- Chen, G.; Wang, T.; Zhang, J.; Liu, P.; Sun, H.; Zhuang, X.; Chen, M.; Feng, X. Accelerated Hydrogen Evolution Kinetics on NiFe-Layered Double Hydroxide Electrocatalysts by Tailoring Water Dissociation Active Sites. *Adv Mater* **2018**, 30, 1706279.
- Babar, P.; Lokhande, A.; Karade, V.; Pawar, B.; Gang, M.G.; Pawar, S.; Kim, J.H. Bifunctional 2D Electrocatalysts of Transition Metal Hydroxide Nanosheet Arrays for Water Splitting and Urea Electrolysis. *ACS Sustainable Chem. Eng.* **2019**, 7, 10035-10043.

6. Liu, W.; Bao, J.; Guan, M.; Zhao, Y.; Lian, J.; Qiu, J.; Xu, L.; Huang, Y.; Qian, J.; Li, H. Nickel–Cobalt-Layered Double Hydroxide Nanosheet Arrays on Ni Foam as a Bifunctional Electrocatalyst for Overall Water Splitting. *Dalton Trans.* **2017**, *46*, 8372-8376.
7. Zhu, X.; Tang, C.; Wang, H. F.; Li, B. Q.; Zhang, Q.; Li, C.; Yang, C.; Wei, F. Monolithic-Structured Ternary Hydroxides as Free-standing Bifunctional Electrocatalysts for Overall Water Splitting. *J. Mater. Chem. A* **2016**, *4*, 7245-7250.
8. Bhowmik, T.; Kundu, M.K.; Barman, S. CoFe Layered Double Hydroxide Supported on Graphitic Carbon Nitrides: An Efficient and Durable Bifunctional Electrocatalyst for Oxygen Evolution and Hydrogen Evolution Reactions. *ACS Appl. Energy Mater.* **2018**, *1*, 1200-1209.
9. Jia, Y.; Zhang, L.; Gao, G.; Chen, H.; Wang, B.; Zhou, J.; Soo, M.T.; Hong, M.; Yan, X.; Qian, G.; Zou, J.; Du, A.; Yao, X. A Heterostructure Coupling of Exfoliated Ni–Fe Hydroxide Nanosheet and Defective Graphene as a Bifunctional Electrocatalyst for Overall Water Splitting. *Adv. Mater.* **2017**, *29*, 1700017.
10. Ye, W.; Fang, X.; Chen, X.; Yan, D. A Three-Dimensional Nickel–Chromium Layered Double Hydroxide Micro/Nanosheet Array as an Efficient and Stable Bifunctional Electrocatalyst for Overall Water Splitting. *Nanoscale* **2018**, *10*, 19484-19491.
11. Qiu, Z.; Tai, C. W.; Niklasson, G.A.; Edvinsson, T. Direct Observation of Active Catalyst Surface Phases and the Effect of Dynamic Self-Optimization in NiFe-Layered Double Hydroxides for Alkaline Water Splitting. *Energy Environ. Sci.* **2019**, *12*, 572-581.
12. Babar, P.; Lokhande, A.; Shin, H.H.; Pawar, B.; Gang, M.G.; Pawar, S.; Kim, J.H. Cobalt Iron Hydroxide as a Precious Metal-Free Bifunctional Electrocatalyst for Efficient Overall Water Splitting. *Small* **2018**, *14*, 1702568.
13. Rajeshkhanna, G.; Singh, T.I.; Kim, N.H.; Lee, J.H. Remarkable Bifunctional Oxygen and Hydrogen Evolution Electrocatalytic Activities with Trace-Level Fe Doping in Ni- and Co-Layered Double Hydroxides for Overall Water-Splitting. *ACS Appl. Mater. Inter.* **2018**, *10*, 42453-42468.
14. Liu, P.F.; Yang, S.; Zhang, B.; Yang, H.G. Defect-Rich Ultrathin Cobalt–Iron Layered Double Hydroxide for Electrochemical Overall Water Splitting. *ACS Appl. Mater. Inter.* **2016**, *8*, 34474-34481.
15. Sultana, U.K.; Riches, J.D.; O'Mullane, A.P. Gold Doping in a Layered Co-Ni Hydroxide System Via Galvanic Replacement for Overall Electrochemical Water Splitting. *Adv. Funct. Mater.* **2018**, *28*, 1804361.