

Electronic Supplementary Information

**A novel strategy to boost the oxygen evolution reaction activity of
NiFe-LDHs with in situ synthesized 3D porous reduced graphene
oxide matrix as both substrate and electronic carrier**

Yanfang Gu, Yuanqiang Wang^{*}, Wei An, Yong Men, Yichuan Rui^{*}, Xinyi Fan, Bin Li

*College of Chemistry and Chemical Engineering, Shanghai University of Engineering
Science, Shanghai 201620, People's Republic of China*

**E-mail: yuanqiangw@sues.edu.cn (Y. Q. Wang), Tel: +86-021-67791216*

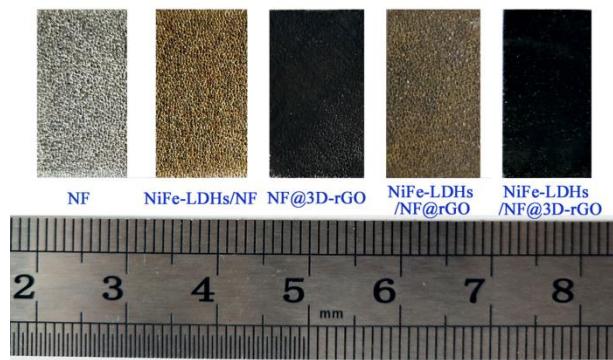


Fig. S1. Photograph images of the NF, and the synthesized NiFe-LDHs/NF, NF/3D-rGO, NiFe-LDHs/NF@rGO, NiFe-LDHs/NF@3D-rGO electrodes.

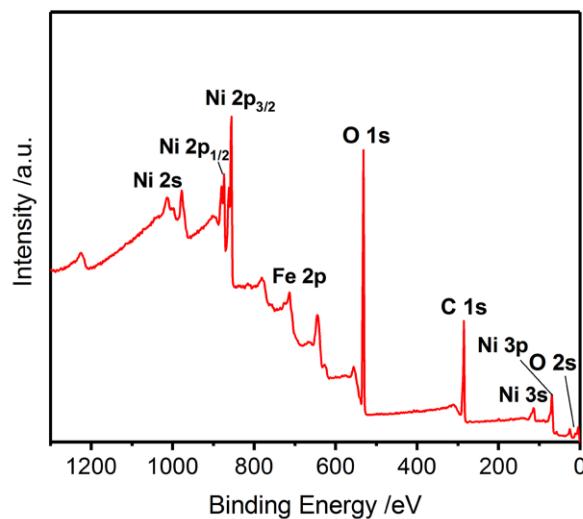


Fig. S2. XPS survey spectrum of the NiFe-LDHs/3D-rGO composite with the elements peaks of Ni, Fe, O and C.

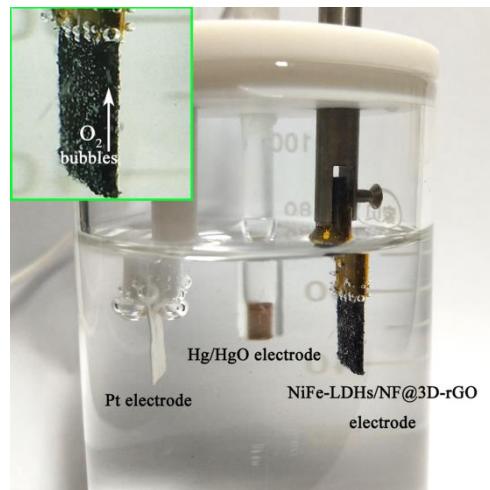


Fig. S3. Photograph of the device for the electrocatalytic oxygen evolution reaction system. The inset image is the enlarged view of the NiFe-LDHs/NF@3D-rGO electrode.

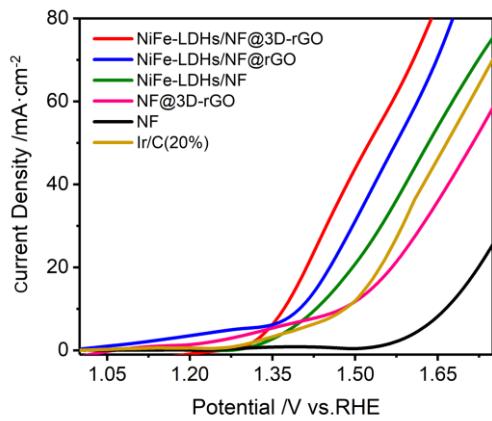


Fig. S4. OER Polarization curves of NF, NF@3D-rGO, NiFe-LDHs/NF, NiFe-LDHs/NF@rGO, NiFe-LDHs/NF@3D-rGO and Ir/C without IR-corrected.

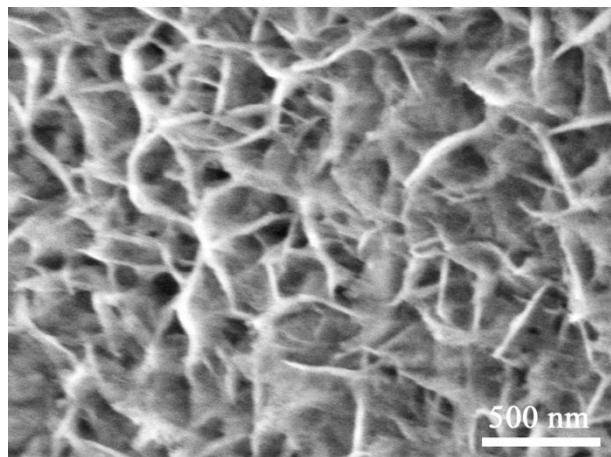


Fig. S5. SEM image of NiFe-LDHs/NF after the OER test.

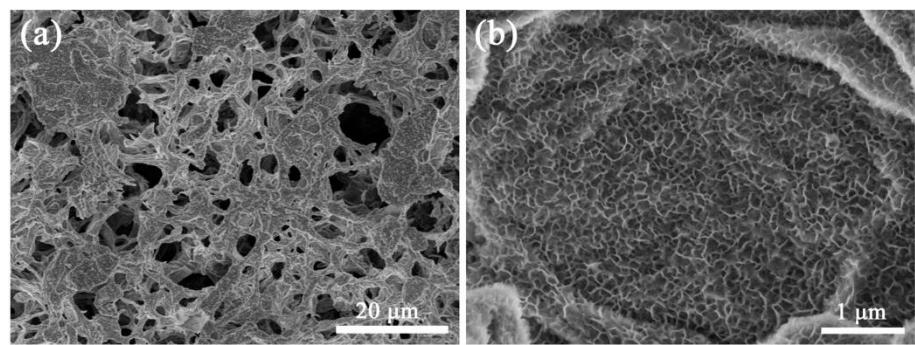


Fig. S6. (a, b) SEM images of NiFe-LDHs/NF@3D-rGO after the OER test.

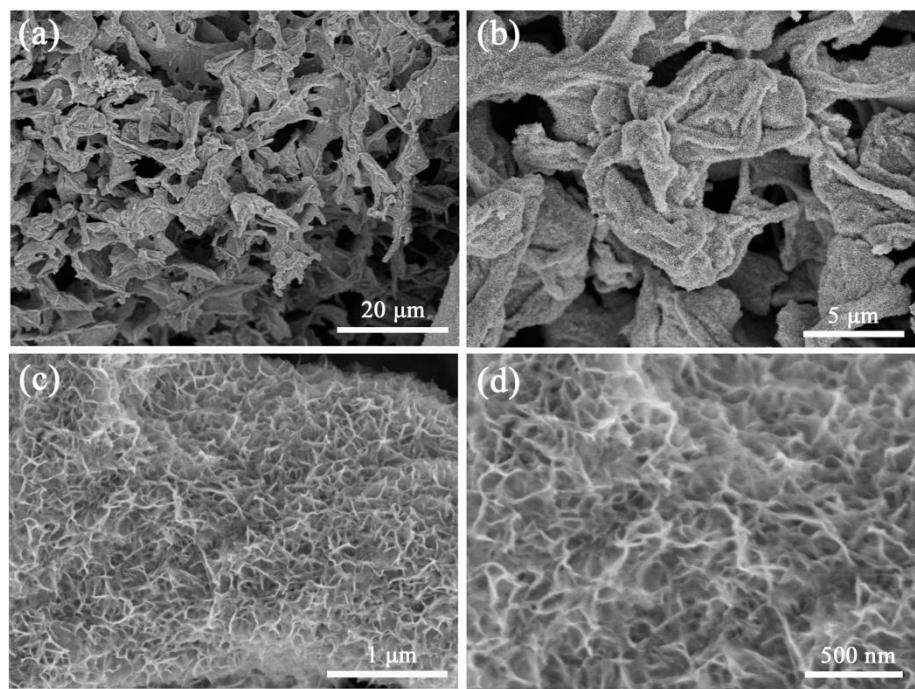


Fig. S7. (a, b, c, d) SEM images of NiFe-LDHs/NF@3D-rGO after the stability test.

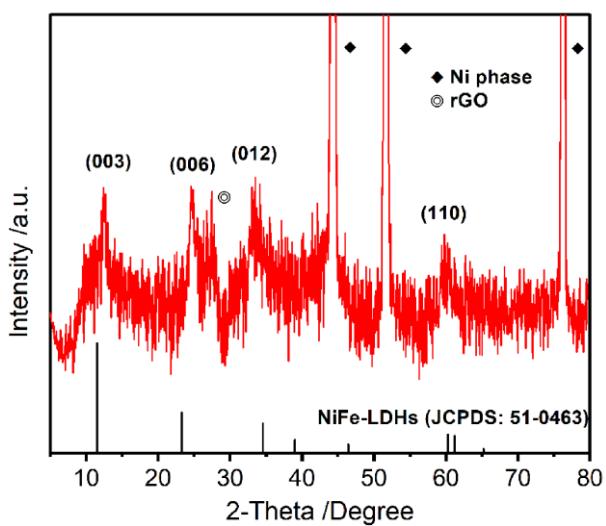


Fig. S8. XRD pattern of NiFe-LDHs/NF@3D-rGO after the OER test.

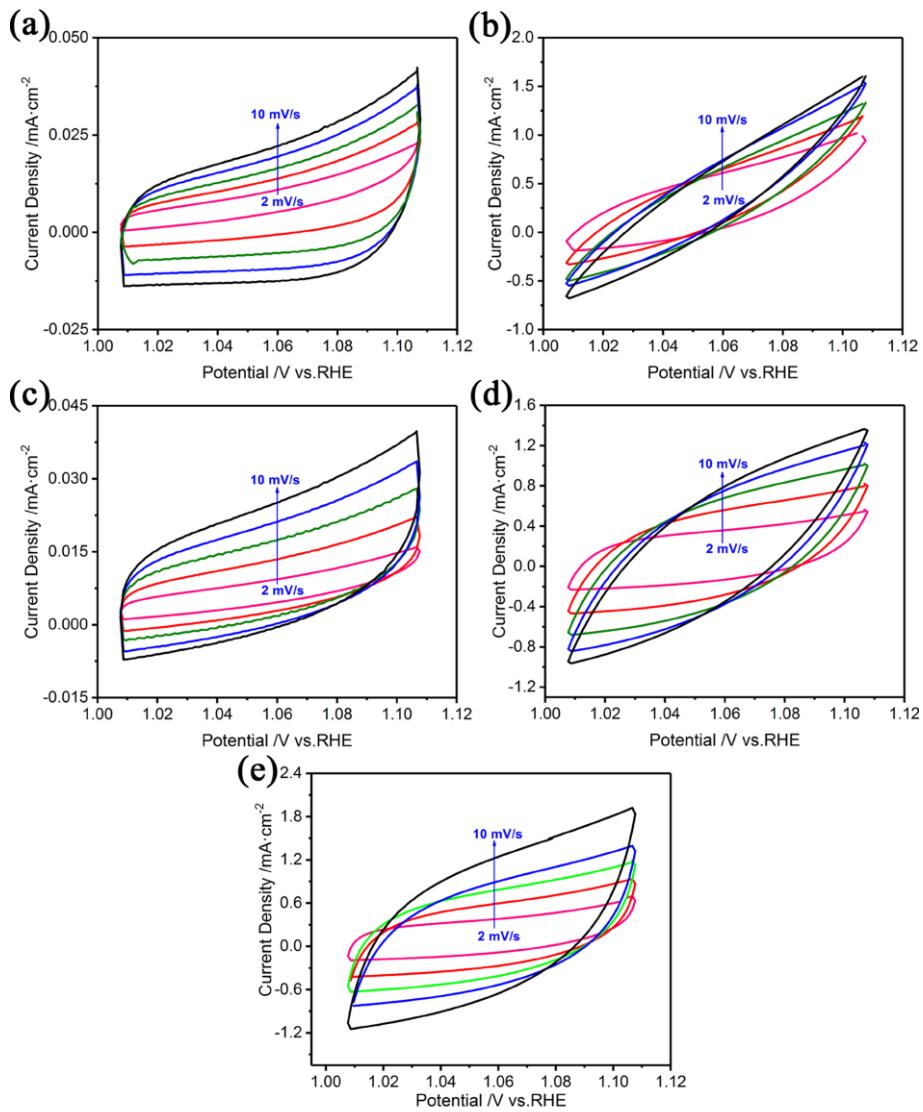


Fig. S9. Cyclic voltammograms of (a) NF, (b) NF/3D-rGO, (c) NiFe-LDHs/NF, (d) NiFe-LDHs/NF@rGO and (e) NiFe-LDHs/NF@3D-rGO in a potential window (1.01 - 1.11 V) at various scan rates of 2, 4, 6, 8, and 10 mV s^{-1} in 1.0 M KOH solution.

Table S1 The comparison of OER activity of composite catalysts associated with NiFe-LDHs (overpotential η was calculated by the formula of $\eta = E(\text{RHE}) - 1.23 \text{ V}$).

Electrocatalyst	Electrolyte	η (mV) @ $j(\text{mA} \cdot \text{cm}^{-2})$	Tafel Slope (mV dec $^{-1}$)	Electrode	Reference
Single-layer NiFe-LDH	1 M KOH	~300@10	40	Ni Foam	S1
Ni–Fe LDH	1 M KOH	210@10	40.4	Fe Foam	S2
NiFeCr LDH	1 M KOH	225@25	69	Carbon paper	S3
NiFe LDH-NS@DG10	1 M KOH	210@10	52	Ni Foam	S4
$\text{Ni}_{0.75}\text{Fe}_{0.25}$ LDHs	1 M KOH	140@10	31	Ni Foam	S5
FeOOH/NiFe	1 M KOH	290@100	-	Ni Foam	S6
LDHs@CCH NAs					
NiFe/RGO	1 M KOH	245@10	-	-	S7
NiFe-LDH/CNT	1 M KOH	300@10	31	GC	S8
(NiFe)S-2	1 M KOH	320@10	61	GN	S9
FeNi-P	1 M KOH	310@10	-	Ni Foam	S10
NiFe-NiCoO ₂	1M KOH	286@10	49.3	Carbon paper	S11
Ir/C	1 M KOH	345@10	107	Ni Foam	S12
Ir/C	1M KOH	327@10	-	GC	S13
NiFe-LDHs/NF@3D-rGO	1 M KOH	170@20	57	Ni Foam	This work

References

- [S1] F. Song and X. Hu, Exfoliation of layered double hydroxides for enhanced oxygen evolution catalysis, *Nat. Commun.* 5 (2014) 4477.
- [S2] X. Yang, C.-J. Wang, C.-C. Hou, W. -F. Fu, and Y. Chen, Self-Assembly of Ni-Fe layered double hydroxide on Fe foam as 3D integrated electrocatalysts for oxygen evolution: dependence of the catalytic performance on anions under in situ condition, *ACS Sustainable Chem. Eng.* 6 (2018) 2893–2897.
- [S3] Y. Yang, L. N. Dang, M. J. Shearer, H. Y. Sheng, W. J. Li, J. Chen, P. Xiao, Y. H. Zhang, R. J. Hamers, S. Jin, Highly active trimetallic NiFeCr layered double hydroxide electrocatalysts for oxygen evolution reaction, *Adv. Energy Mater.* 8 (2018).
- [S4] Y. Jia, L. Z. Zhang, G. P. Gao, H. Chen, B. Wang, J. Z. Zhou, M. T. Soo, M. Hong, X. C. Yan, G. R. Qian, J. Zou, X. D. Yao, A heterostructure coupling of exfoliated Ni-Fe hydroxide nanosheet and defective graphene as a bifunctional electrocatalyst for Overall Water Splitting, *Adv. Mater.* 29 (2017) 1700017.
- [S5] Q. Y. Liu, H. Wang, X. N. Wang, R. Tong, X. L. Zhou, X. N. Peng, H. B. Wang, H. L. Tao, Z. H. Zhang, Bifunctional $\text{Ni}_{1-x}\text{Fe}_x$ layered double hydroxides/Ni foam electrodes for high-efficient overall water splitting: A study on compositional tuning and valence state evolution, *Int. J. Hydrogen Energ.* 42 (2017) 5560-5568.
- [S6] J. Chi, H. M. Yu, G. Jiang, J. Jia, B. W. Qin, B. L. Yi, Z. G. Shao, Construction of orderly hierarchical FeOOH/NiFe layered double hydroxides supported on cobaltous carbonate hydroxide nanowire arrays for a highly efficient oxygen evolution reaction, *J. Mater. Chem. A* 6 (2018) 3397-3401.
- [S7] D. H. Youn, Y. Bin Park, J. Y. Kim, G. Magesh, Y. J. Jang, J. S. Lee, One-pot synthesis of NiFe layered double hydroxide/reduced graphene oxide composite as an

efficient electrocatalyst for electrochemical and photoelectrochemical water oxidation, Journal of Power Sources 294 (2015) 437-443.

[S8] M. Gong, Y. Li, H. Wang, Y. Liang, J. Z. Wu, J. Zhou, J. Wang, T. Regier, F. Wei and H. Dai , An advanced Ni-Fe layered double hydroxide electrocatalyst for water oxidation, J. Am. Chem. Soc. 135 (2013) 8452-8455.

[S9] C. Y. Liu, H. Ma, M. W. Yuan, Z. H. Yu, J. Li, K. R. Shi, Z. P. Liang, Y. Yang, T. J. Zhu, G. B. Sun, H. F. Li, S. L. Ma,(NiFe)S-2 nanoparticles grown on graphene as an efficient electrocatalyst for oxygen evolution reaction, Electrochim Acta 286 (2018) 195-204.

[S10] Q. Yan, T. Wei, J. Wu, X. Y. Yang, M. Zhu, K. Cheng, K. Ye, K. Zhu, J. Yan, D. X. Cao, G. L. Wang, Y. Pan, Self-supported FeNi-P nanosheets with thin amorphous layers for efficient electrocatalytic water splitting, Acs. Sustain Chem. Eng. 6 (2018) 9640-9648.

[S11] R. Shi, J. Wang, Z. Wang, T. Li, Y.-F. Song, Unique NiFe-NiCoO₂ hollow polyhedron as bifunctional electrocatalysts for water splitting, J. Energy Chem. 33 (2019) 78-80.

[S12] S. Oh, H. Kim, Y. Kwon, M. Kim, E. Cho, H. Kwon, Porous Co-P foam as an efficient bifunctional electrocatalyst for hydrogen and oxygen evolution reactions, J. Mater. Chem. A. 4 (2016) 18272-18277.

[S13] J. Bao, X. Zhang, B. Fan, J. Zhang, M. Zhou, W. Yang, Ultrathin Spine-Structured Nanosheets Rich in Oxygen Deficiencies for Enhanced Electrocatalytic Water Oxidation. Angew. Chem. 127(2015), 7507–7512.