Supporting Information

Rapid large-scale synthesis of ultrathin NiFe-layered double hydroxides nanosheets with tunable structures as robust oxygen evolution electrocatalysts

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Supporting information: Figures S1-S7 and Table S1-S3.

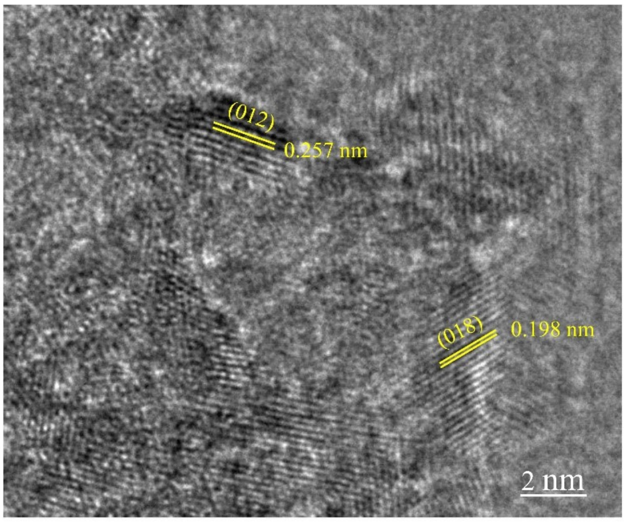

Fig. S1. The high-resolution transmission electron microscopy image of Ni_2Fe_1-LDHs sample.

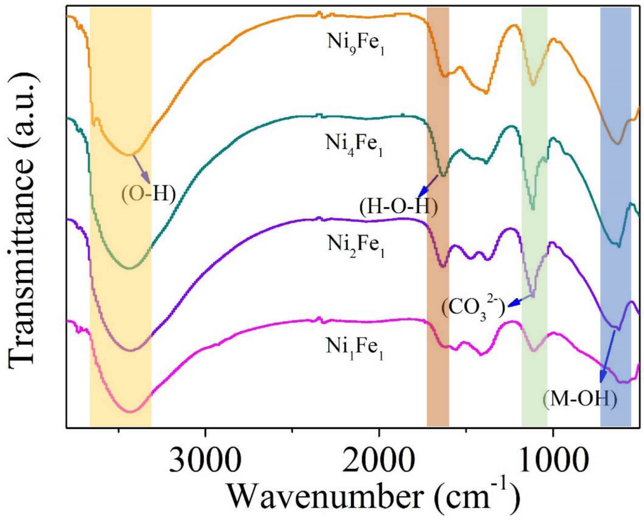

Fig. S2. Fourier transform infrared spectroscopy of NiFe-LDHs nanosheets.

For all NiFe-LDHs samples, the prominent and broad peak at 3445 cm^{-1} can be readily attributed to the vibration mode of *v* (H_2O) in the LDHs structure, while the band at 1630 cm^{-1} can be assigned to the vibration mode of δ (H_2O). The broad peak at 684 cm^{-1} is ascribed to the *v* (M–OH) vibration band of FeNi LDH, which is the feature for the NiFe-LDHs. In addition, the band at 1115 cm^{-1} demonstrates the presence of CO_3^{2-} anions.[1,2]

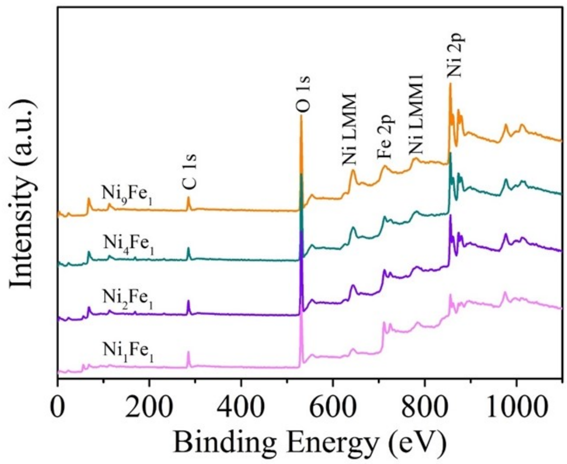

Fig. S3. XPS survey spectra of the as-obtained NiFe-LDHs ultrathin nanosheets.

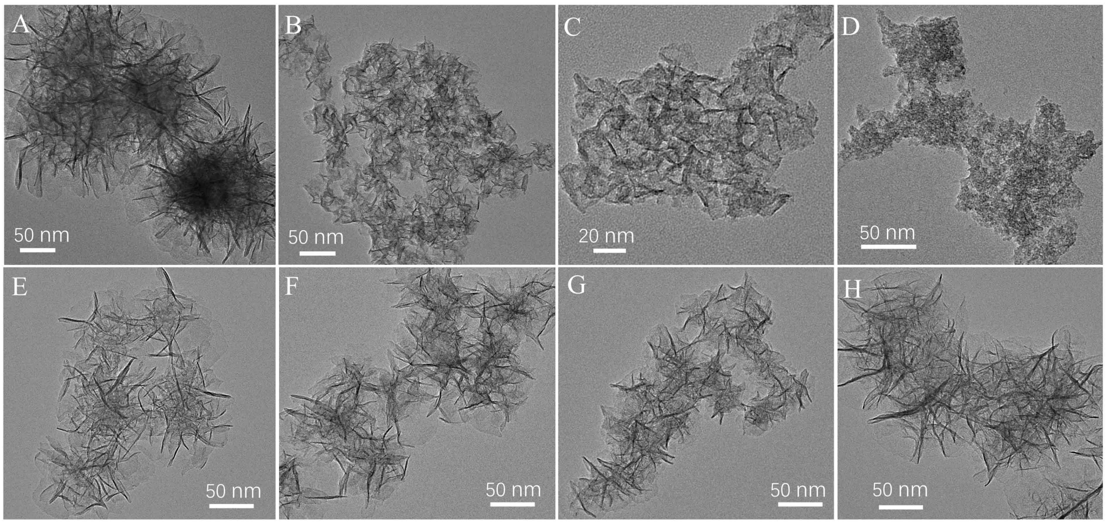

Fig. S4. (A-D) TEM images of the as-obtained CoFe-based ultrathin nanosheets with different Co/Fe atomic ratios: (A) 9:1, (B) 4:1, (C) 2:1, (D) 1:1. (E-H) TEM images of the as-obtained NiCo-based ultrathin nanosheets with different Ni/Co atomic ratios: (E) 9:1, (F) 4:1, (G) 2:1, (H) 1:1. All TEM images reveal their sheet-like morphology and ultrathin thickness.

Fig. S5. Illustration for the large-scale preparation of Ni_2Fe_1-LDHs sample and its weight.

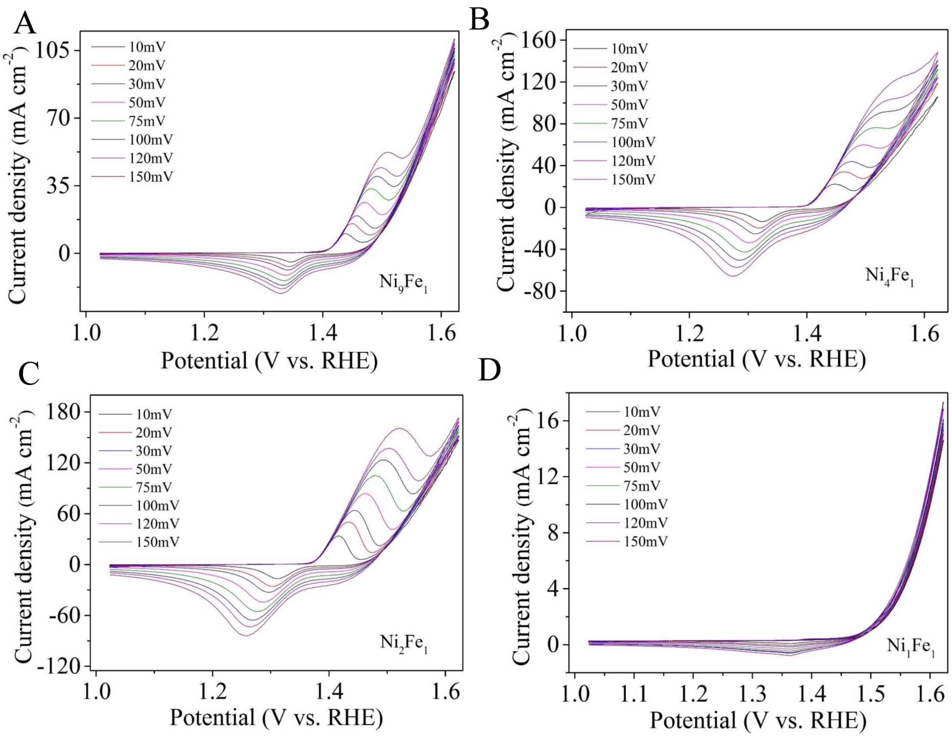

Fig. S6. Cyclic voltammetry curves of different NiFe-LDHs samples in 1 M KOH solution at potential sweep rates of 10, 20, 50, 70, 100, 120 and 150 mV s^{-1}.

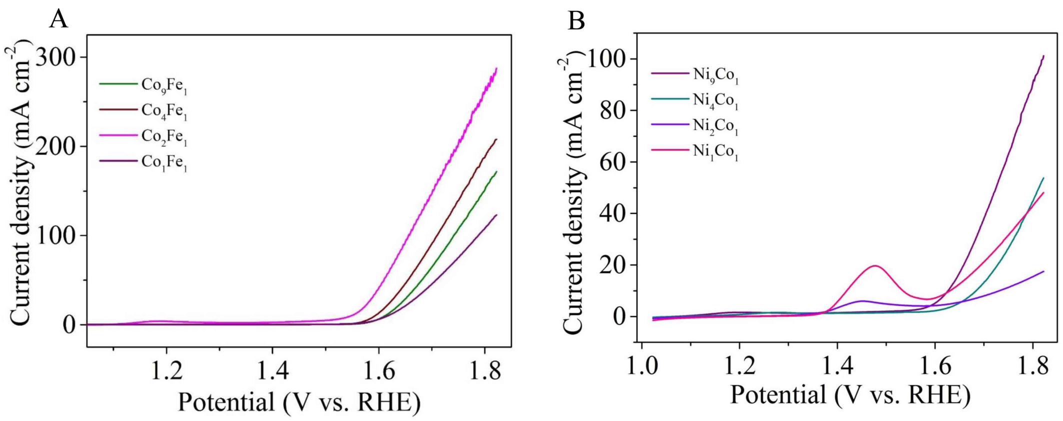

Fig. S7. Polarization curves of various samples measured at a scan rate of 5 mV s^{-1} in 1 M KOH solution. (A) CoFe-based ultrathin nanosheets. (B) NiCo-based ultrathin nanosheets.

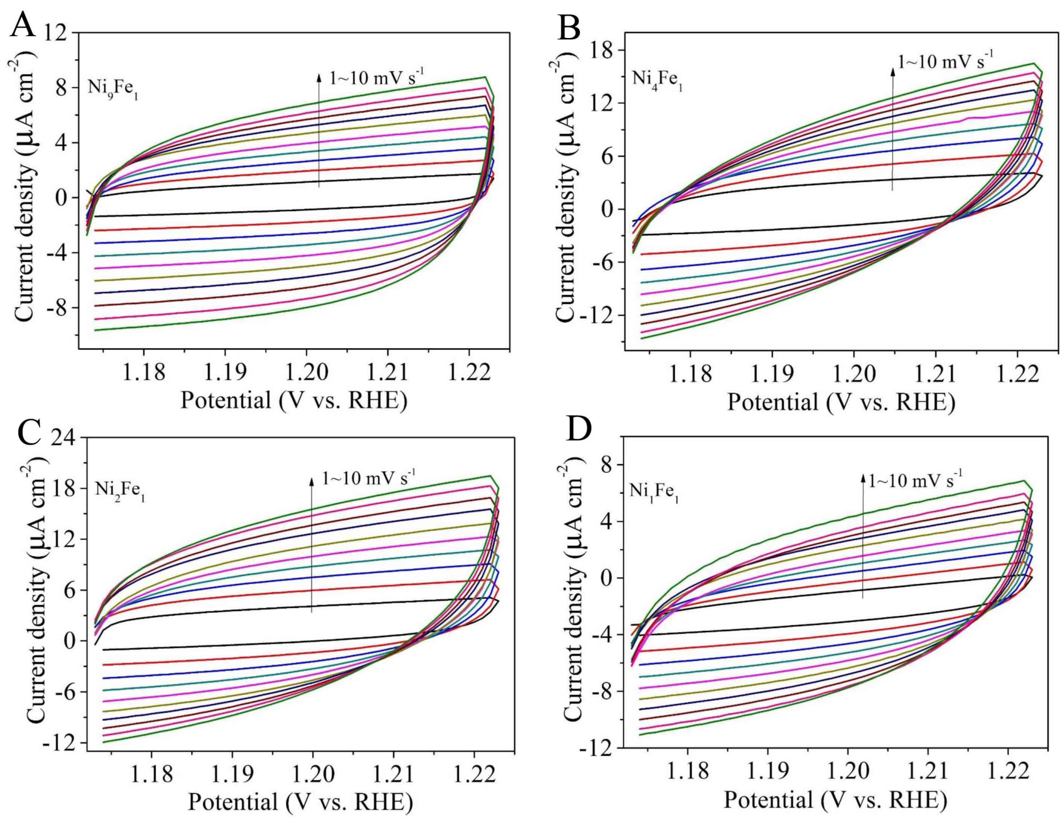

Fig. S8. Cyclic voltammetry curves of various NiFe-LDHs samples measured in 1 M KOH solution at scan rates from 1 to 10 mVs^{-1}.

Table S1. Inductively coupled plasma atomic emission spectrometry results of different NiFe-LDHs samples.

Sample	Ni (g/kg)	Fe (g/kg)	Ni/Fe ratio
Ni_9Fe_1	462.4	50.1	8.81
Ni_4Fe_1	430.5	99.2	4.14
Ni_2Fe_1	302.0	141.3	2.03
Ni_1Fe_1	275.9	250.0	1.05

Table S2. The DFT data for the adsorption of water molecular on the catalyst surface.

Species	Energy (eV)	Adsorption energy (eV)
Ni_9Fe_1 Ni_9Fe_1-H_2O	-66291.8524 -66758.2781	-2.70872
Ni_4Fe_1 Ni_4Fe_1-H_2O	-65314.132 -65780.7648	-2.91582
Ni_2Fe_1 Ni_2Fe_1-H_2O	-62869.9265 -63336.9353	-3.29182
Ni_1Fe_1 Ni_1Fe_1-H_2O	-60394.0037 -60860.3526	-2.63192
H_2O	-463.716979	--

Table S3. The DFT data for the formation energy of active NiOOH species.

Species	Energy (eV)	Formation energy (eV)
Ni_9Fe_1 Ni_9Fe_1-OH	-66291.6545 -66735.2784	4.65589
Ni_4Fe_1 Ni_4Fe_1-OH	-65313.8976 -65758.4921	3.68529
Ni_2Fe_1 Ni_2Fe_1-OH	-62869.7586 -63314.9711	3.06729
Ni_1Fe_1 Ni_1Fe_1-OH	-60393.8358 -60837.8957	4.21989
OH	-448.27979	--

Table S4. Comparison of the electrocatalytic performance between the Ni_2Fe_1-LDHs sample and other recently reported catalysts.

Samples	Overpotential (mV) at 10 mA/cm^2	Current density (mA cm^{-2}) at 1.6 V (vs RHE)	References
Ni_2Fe_1-LDHs	263	102.8	This work
$Ni_{0.8}Fe_{0.2}$-LDH	235	~90	Nanoscale, 2020, 12, 10751.
$(Co,Ni)Se_2$@NiFe	277	~58	ACS Appl. Mater. Interfaces 2019, 11, 8, 8106.
IrO_x/U-NiFe-LDH	236	~160	Chem. Commun., 2020, 56, 11465.
$Ni_{0.83}Fe_{0.17}(OH)_2$	245	Over 100	ACS Catal. 2018, 8, 6, 5382.
NiFe LDH nanomesh	--	~125	Nano Energy 2018, 53, 74.
V_{Ni}-rich α-$Ni(OH)_2$	~300	~100	ACS Energy Letters 2018, 3,1373.
β-$Ni(OH)_2$ nanomeshes	--	~105	Adv. Mater. 2017, 29, 1604765.
Exfoliated NiFe LDH	302	--	Nat. Commun. 2014, 5, 4477
NiFe-LDH with tensile strain	270	--	Angew. Chem. Int. Ed. 2019, 58,736.
$Ni_{2/3}Fe_{1/3}$-rGO	230	--	ACS Nano 2015, 9, 1977.
NiFe/C	220	--	ACS Appl. Mater. Interfaces 2015, 7, 9203.
Commercial IrO_2	330	23	Angew. Chem. Int. Ed. 2015, 54, 8722.

Reference

[1] X. X. Jiang, J. Y. Xue, Z. Y. Zhao, C. Li, F. L. Li, C. Cao, Z. Niu, H. W. Gu and J. P. Lang, *RSC Adv.*, **2020**, *10*,12145.

[2] A. Jawad, Z. W. Liao, Z. H. Zhou, A. Khan, T. Wang, J. Ifthikar, A. Shahzad, Z. L. Chen and Z. Q. Chen, *ACS Appl. Mater. Interfaces*, **2017**, *9*, 28451.