Supporting Information (SI):

Nanoporous NiAl-LDH nanosheet arrays with optimized Ni active sites for efficient electrocatalytic alkaline water splitting

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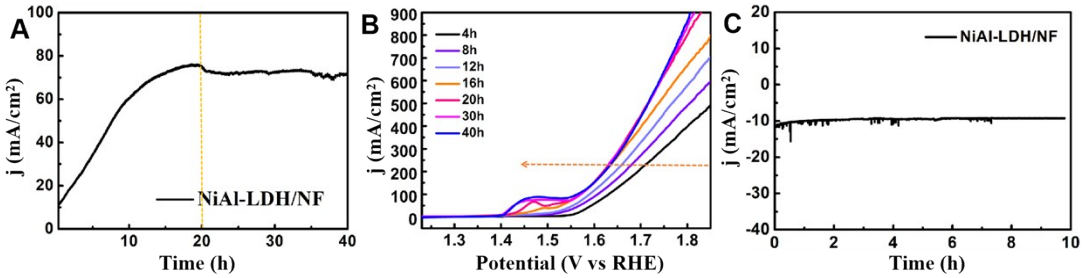

Fig. S1 (A) Electrochemical etching process; (B) OER polarization curves and (C) the long-term HER durability test of NiAl-LDH/NF in alkaline media (1 M KOH solution).

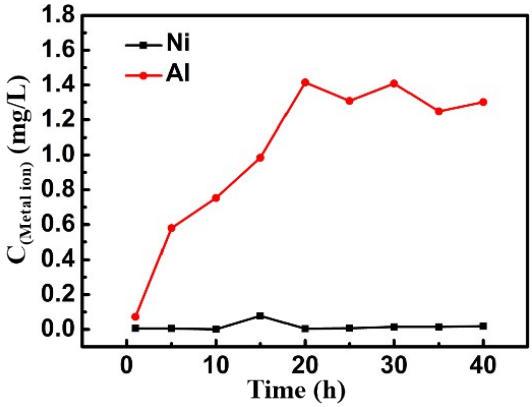

Fig. S2 The contents of Ni and Al species in the electrolyte as a function of electrocatalysis time at an applied potential of 1.56 V.

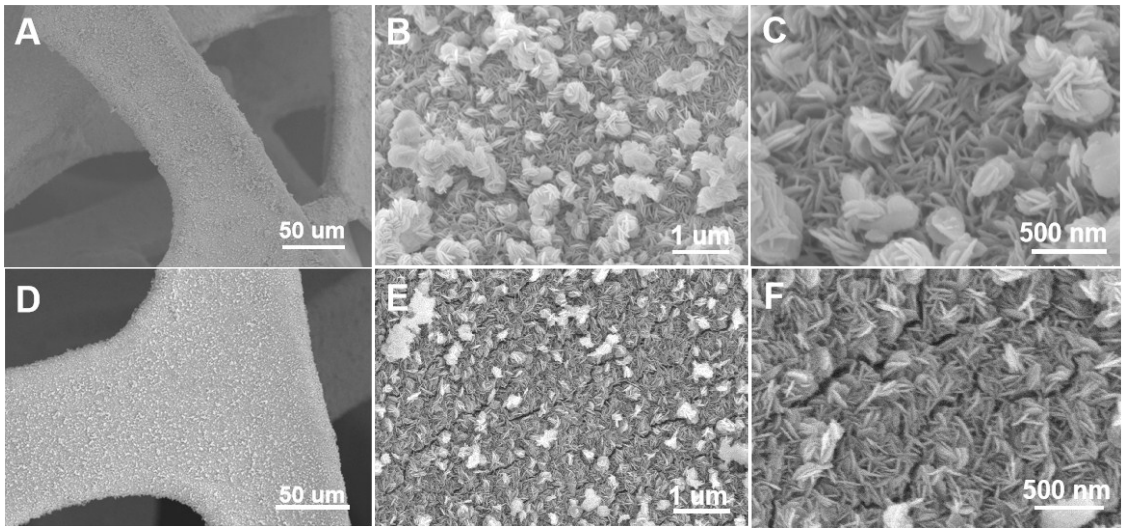

Fig. S3 (A-C) SEM images of np-NiAl-LDH/NF; (D-F) SEM images of NiAl-LDH/NF.

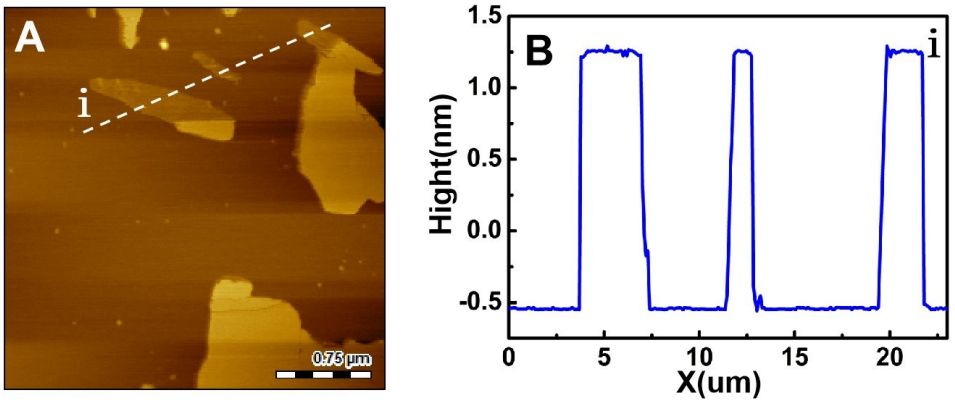

Fig. S4 (A) AFM image and (B) the corresponding height profile of np-NiAl-LDH/NF.

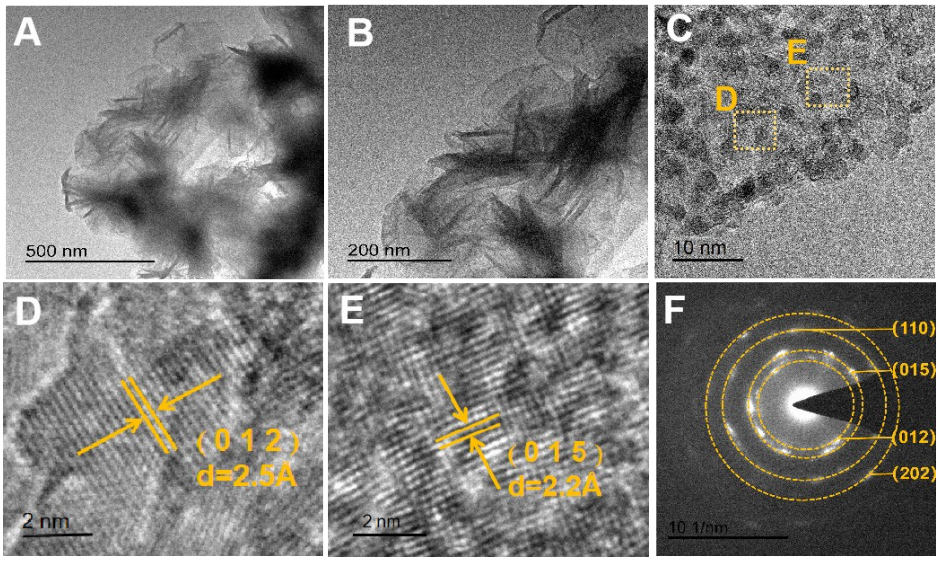

Fig. S5 (A-C) TEM images; (D,E) HRTEM images; and (F) the corresponding SAED pattern of NiAl-LDH/NF.

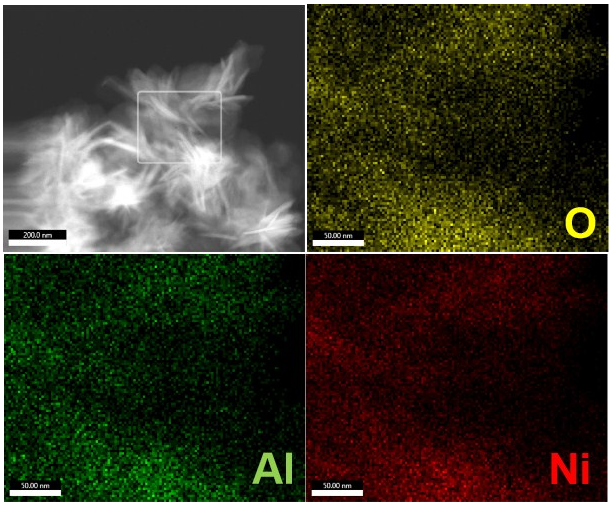

Fig. S6 The elemental mapping images of NiAl-LDH/NF.

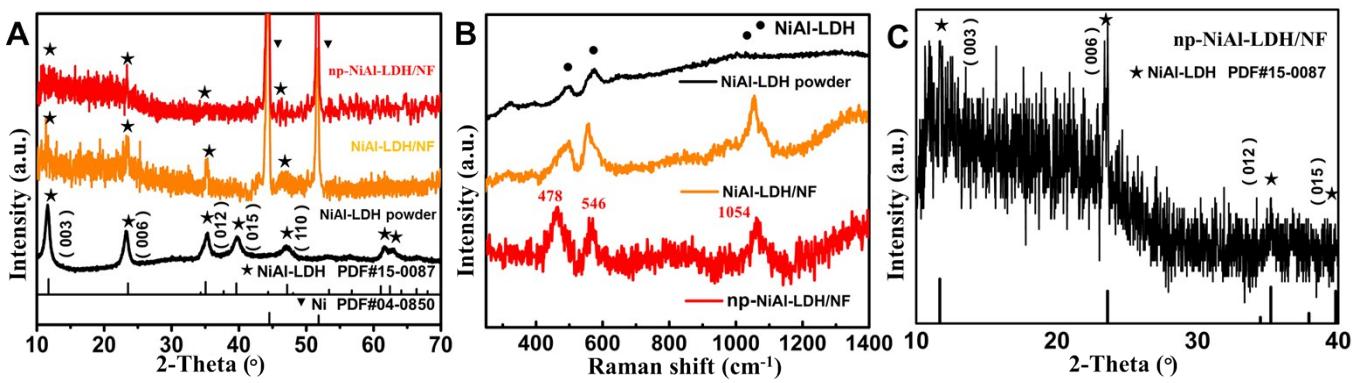

Fig. S7 (A) XRD patterns; (B) Raman spectra of NiAl-LDH powder, NiAl-LDH/NF and np-NiAl-LDH/NF and (C) the partially enlarged XRD pattern of np-NiAl-LDH/NF.

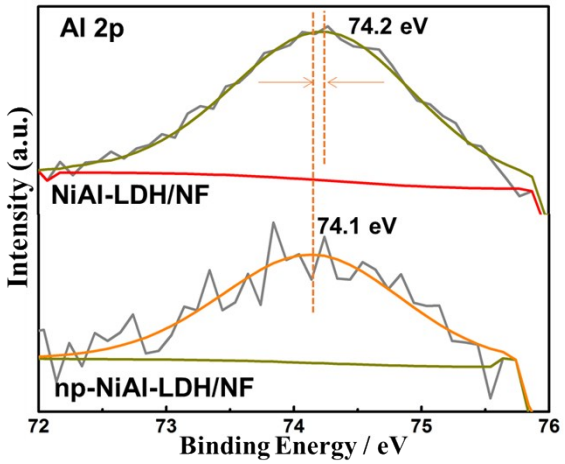

Fig. S8 Al 2p XPS high-resolution spectra of NiAl-LDH/NF and np-NiAl-LDH/NF.

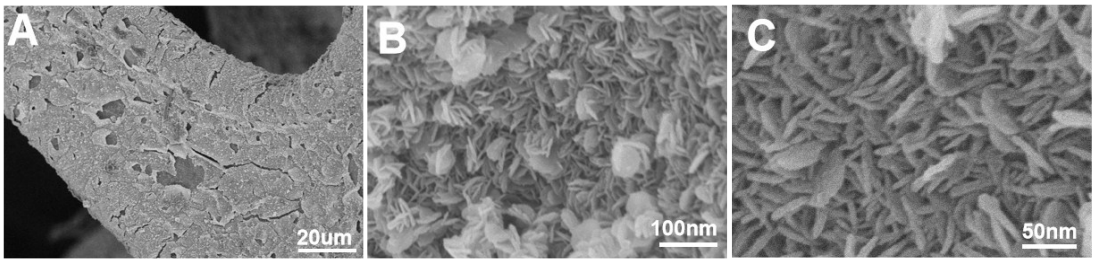

Fig. S9 (A-C) SEM images of np-NiAl-LDH/NF after 100 h-electrocatalytic HER test.

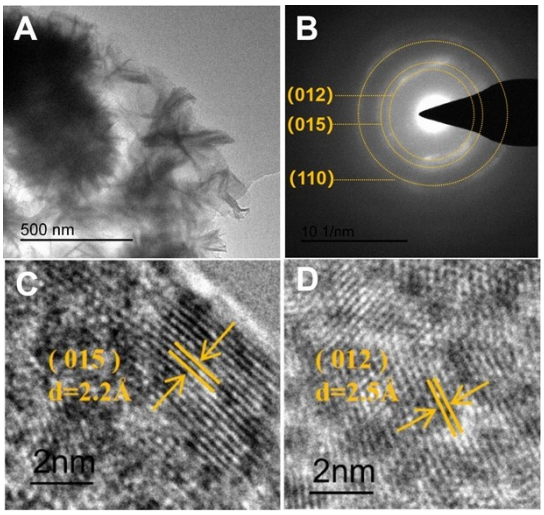

Fig. S10 (A) TEM image; (B) SAED pattern; and (C-D) High-resolution TEM images of np-NiAl-LDH/NF after 100 h-long electrocatalysis for HER.

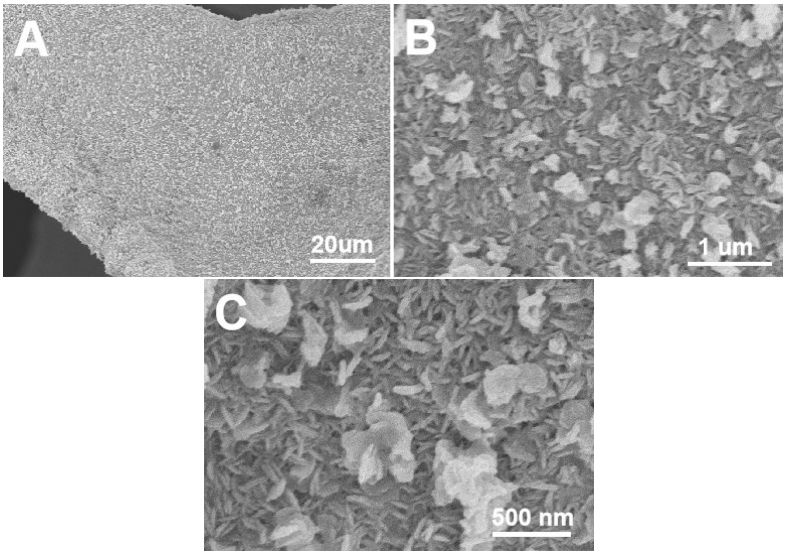

Fig. S11 (A-C) SEM images of np-NiAl-LDH/NF after 100 h-electrocatalytic OER test.

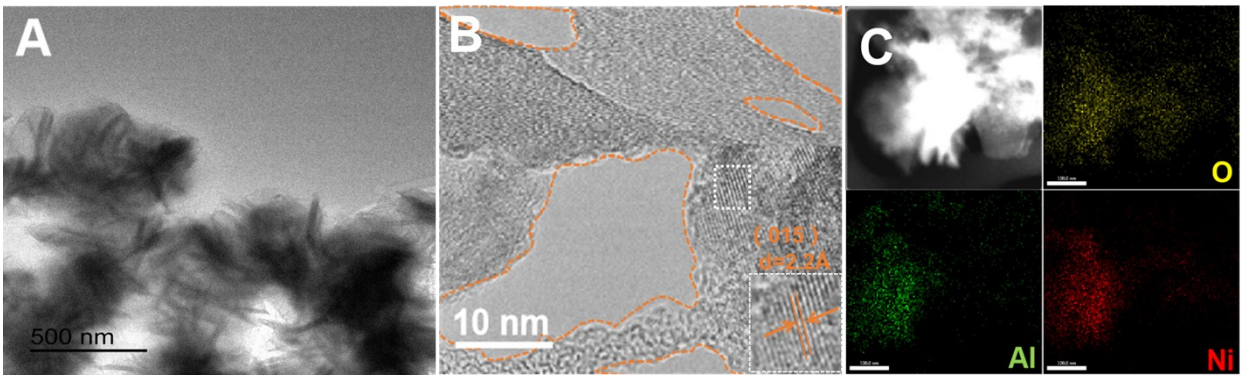

Fig. S12 (A,B) TEM images and (C) the corresponding elemental mapping images of np-NiAl-LDH/NF after 100 h-long electrocatalysis for OER.

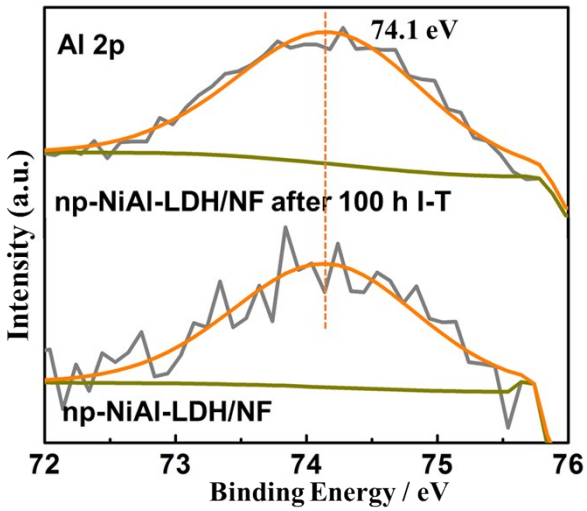

Fig. S13 High-resolution Al spectra of np-NiAl-LDH/NF and np-NiAl-LDH/NF after long-term catalytic OER stability test for 100 h.

Table S1. The Ni and Al contents were obtained from the EDX results of NiAl-LDH/NF and np-NiAl-LDH/NF.

Atomic %	NiAl-LDH/NF	np-NiAl-LDH/NF
Ni	43.28	51.04
Al	18.11	13.02
Ni/Al	2.39	3.92

Table S2. Comparison of the contents for four oxygen species from the fitting O 1s XPS spectra of NiAl-LDH/NF and np-NiAl-LDH/NF.

Oxygen species	NiAl-LDH/NF	np-NiAl-LDH/NF
adsorbed water (H_2O)	25.61%	32.09%
hydroxyl group (–OH)	25.85%	23.50%
oxygen vacancy (V_O)	17.34%	28.74%
metal–oxygen (M–O)	31.21%	15.67%

Table S3. Comparison of electrocatalytic performance of np-NiAl-LDH/NF with other recently reported Ni-based LDH catalysts for the HER in alkaline media.

Catalyst	Electrolyte	Overpotential at the corresponding *j*	Stability test (h)	Tafel slope	Reference
np-NiAl-LDH/NF	1 M KOH	~90@10 mA/cm^2 ~200@100 mA/cm^2	40	72	This work
NiAl-LDH/MoS_2	1 M KOH	220@10 mA/cm^2 ~335@100 mA/cm^2	10	82	ACS Energy Lett. 2018, 3, 952-960
Ni@NiFe LDH	1 M KOH	92@10 mA/cm^2 233@100 mA/cm^2	24	72.3	J Mater Chem A. 2019, 7(38), 21722-21729
CoFe@NiFe/NF	1 M KOH	240@10 mA/cm^2	30	46	Appl Catal B-Environ. 2019, 253, 131-139
Fe-Ni LDH/NF	1 M KOH	127@10 mA/cm^2 257@50 mA/cm^2	14	109.4	ACS Sustain Chem Eng. 2019, 7(17), 15073-15079
$NiCo_2O_4$@Ni_xCo_y-LDH/NF	1 M KOH	193@10 mA/cm^2	24	/	ACS Sustain Chem Eng. 2019, 7(5), 4784-4791
$Ni_{1-x}Fe_x$ LDH/NF	1 M KOH	170@10 mA/cm^2 ~260@50 mA/cm^2	36	83	ACS Appl. Mater. Interfaces. 2018, 10(49). 2245
Ni_1Cr_1 LDH/NF	1 M KOH	83@10 mA/cm^2	30	61	Nanoscale. 2018, 41(10), 19484-19491
CoSe/NiFe-LDH/NF	1 M KOH	260@10 mA/cm^2	10	/	Chem. Commun. 2016, 52, 908
NiAlCo-LDH	1 M KOH	475 @10 mA/cm^2	/	52.4	Appl Surf Sci. 2016, 370, 445-451
NiCo-LDH/NF	1 M KOH	162@10 mA/cm^2	20	141	Dalton T. 2017, 14, 8372-8376
NiFe LDH@ NiCoP/NF	1 M KOH	120@10 mA/cm^2	100	2	Adv. Funct. Mater. 2018, 28, 1706847
AlNi-LDH	1 M KOH	~200@10 mA/cm^2 ~290@100 mA/cm^2	7	130	ACS Energy Letter. 2018, 3(4), 9871-9876

Table S4. Comparison of the electrocatalytic performance of np-NiAl-LDH/NF with recently reported Ni-based electrocatalysts for the OER in alkaline media.

Catalyst	Electrolyte	Overpotential at the corresponding *j*	Stability test (h)	Reference
np-NiAl-LDH/NF	**1 M KOH**	**~180@10 mA/cm^2 ~320@100 mA/cm^2**	**60**	**This work**
Ni@NiFe LDH	**1 M KOH**	**218@10 mA/cm^2 269@100 mA/cm^2**	**24**	**J Mater Chem A. 2019, 7(38), 21722-21729**
CoFe@NiFe/NF	**1 M KOH**	**190@10 mA/cm^2**	**30**	**Appl Catal B-Environ. 2019, 253, 131-139**
NiAl-LDH/MoS_2	**1 M KOH**	**~310@10 mA/cm^2**	**15**	**ACS Energy Lett. 2018, 3, 952-960**
NiFeAl LDHs	**1 M KOH**	**260@10 mA/cm^2**	**10**	**Nano Res. 2018, 11, 4524-4534**
FeNi LDH/NF	**1 M KOH**	**193@10 mA/cm^2 306@50 mA/cm^2**	**25**	**ACS Sustain Chem Eng. 2019, 7(17), 15073-15079**
$NiCo_2O_4$@Ni_xCo_yLDH /NF	**1 M KOH**	**115@10 mA/cm^2**	**24**	**ACS Sustain Chem Eng. 2019, 7(5), 4784-4791**
Ni_2Cr_1 LDH/NF	**1 M KOH**	**390@100 mA/cm^2**	**20**	**Nanoscale. 2018, 41(10), 19484-19491**
AlNi-LDH	**1 M KOH**	**~320@100 mA/cm^2**	**7**	**ACS Energy Letter. 2018, 3(4), 9871-9876**
N-NiAl-LDH	**1 M KOH**	**~400@100mA/cm^2**	**6**	**J Mater Chem A. 2018, 6, 23283-23288**
$Ni_{1-x}Fe_x$ LDH/NF	**1 M KOH**	**106@10mA/cm^2**	**36**	**ACS Appl. Mater. Interfaces. 2018, 10(49), 2245**

Table S5. Comparison of the electrocatalytic performance of np-NiAl-LDH/NF with recently reported Ni-based LDH catalysts for overall water splitting in alkaline media.

Catalyst	Electrolyte	Cell voltage (mV)	Stability test (h)	Reference
np-NiAl-LDH/NF	1 M KOH	1.5@10 mA/cm^2 1.75@100 mA/cm^2	55	This work
Ni@NiFe LDH	1 M KOH	1.53@10 mA/cm2 1.78@100 mA/cm2	24	J Mater Chem A. 2019, 7(38), 21722-21729
CoFe@NiFe/NF	1 M KOH	1.59@10 mA/cm2	24	Appl Catal B-Environ. 2019, 253, 131-139
NiAl-LDH/MoS_2	1 M KOH	1.5@10 mA/cm2	10	ACS Energy Lett. 2018, 3, 952-960
AlNi-LDH	1 M KOH	1.58@100 mA/cm^2	7	ACS Energy Letter. 2018, 3(4), 9871-9876
Co_9S_8@NiCo LDH/NF	1 M KOH	1.6@10 mA/cm^2	10	Sci Bull. 2019, 60–66
Ni_3S_2@NiV-LDH	1 M KOH	1.53@10 mA/cm^2	160	Nanoscale. 2019, 7585-7591
Fe-Ni LDH/NF	1 M KOH	1.65@10 mA/cm2 1.73@100 mA/cm2	20	ACS Sustain Chem Eng. 2019, 7(17), 15073-15079
$NiCo_2O_4$@Ni_xCo_y LDH/NF	1 M KOH	~1.63@10 mA/cm^2	24	ACS Sustain Chem Eng. 2019, 7(5), 4784-4791
$Ni_{1-x}Fe_x$ LDH	1 M KOH	1.59@10 mA/cm^2	24	ACS Appl. Mater. Interfaces. 2018, 10(49), 2245
Ni_2Cr_1 LDH/NF	1 M KOH	1.55@10 mA/cm^2	30	Nanoscale. 2018, 41(10), 19484-19491