Electronic Supplementary Information

Improving Hydrogen Evolution Performance of Self-Supported Hierarchical NiFe Layered Double Hydroxide via NH_3-Inducing at Room Temperature

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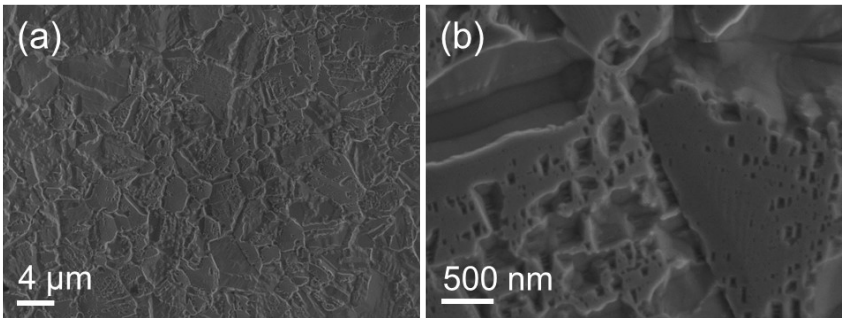

Fig. S1. SEM images of pristine nickel foam under different magnifications.

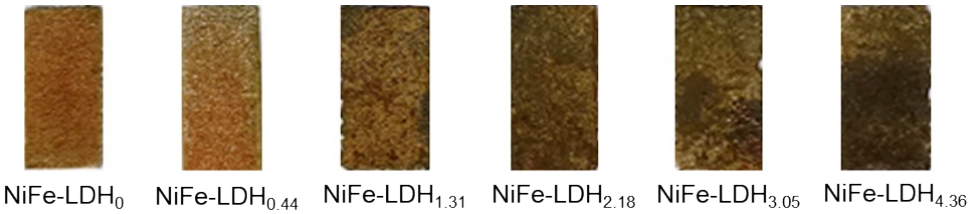

Fig. S2. The photographs of NiFe-LDH$_x$ samples obtained with the different concentrations of NH_3.

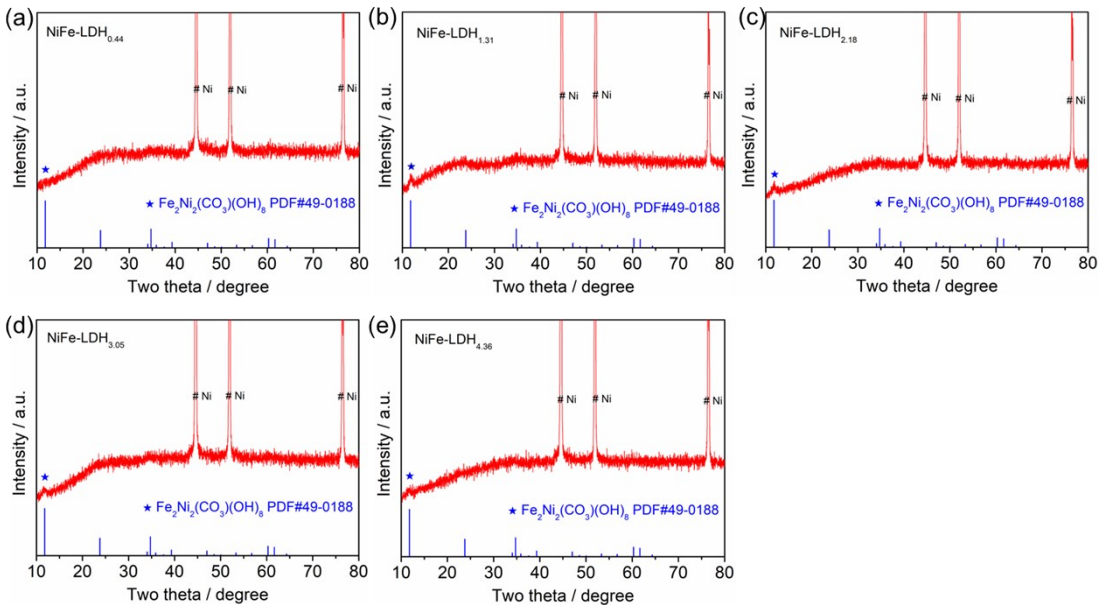

Fig. S3. XRD patterns of (a) NiFe-LDH$_{0.44}$, (b) NiFe-LDH$_{1.31}$, (c) NiFe-LDH$_{2.18}$, (d) NiFe-LDH$_{3.05}$ and (e) NiFe-LDH$_{4.36}$. The pattern at the bottom is the standard pattern of $Fe_2Ni_2(CO_3)(OH)_8$ (PDF#49-0188) for comparison.

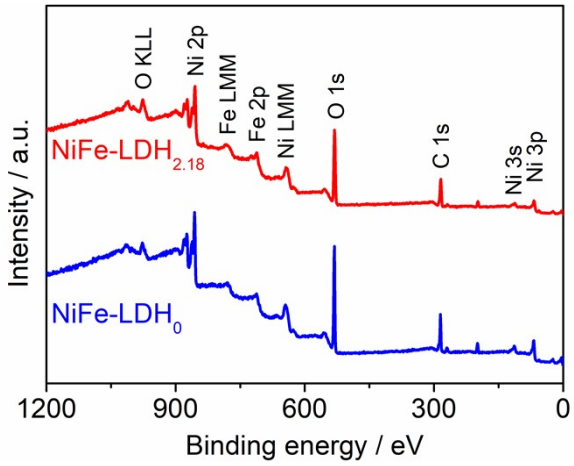

Fig. S4. XPS survey spectra of NiFe-LDH$_{2.18}$ and NiFe-LDH$_0$.

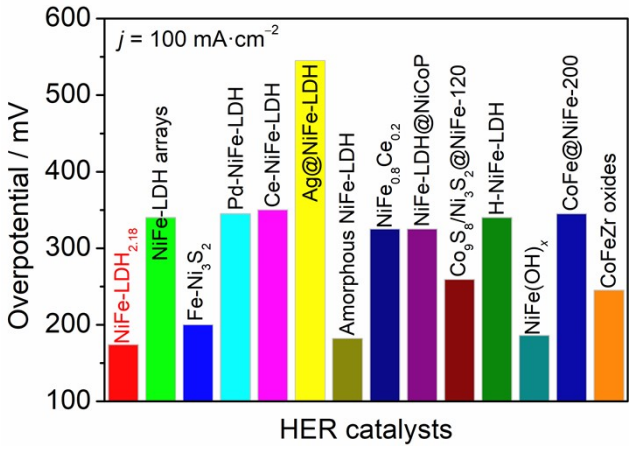

Fig. S5. Overpotentials required to achieve a current density of 100 mA·cm^{-2} on NiFeLDH$_{2.18}$ and other related HER catalysts.

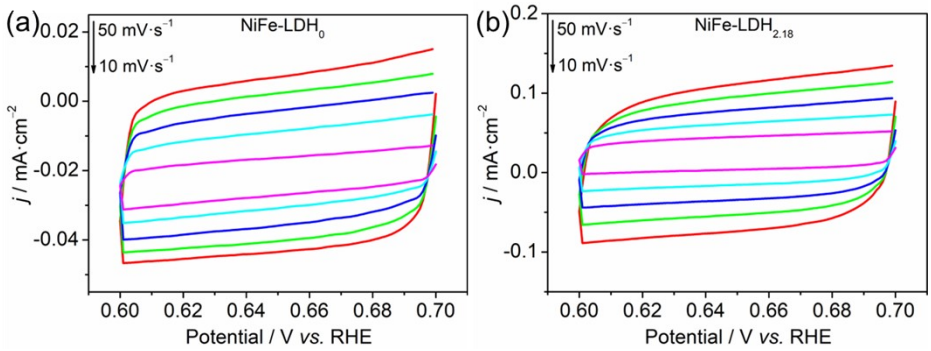

Fig. S6. CV curves of (a) NiFe-LDH$_0$ and (b) NiFe-LDH$_{2.18}$ in non-Faradaic region (0.6-0.7 V) at scan rates of 10, 20, 30, 40 and 50 mV·s^{-1} in 1.0 M KOH solution.

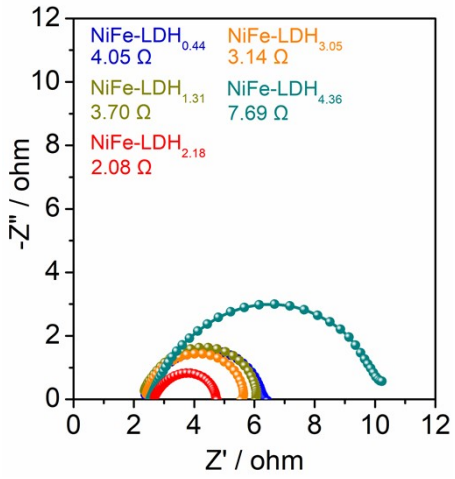

Fig. S7. Nyquist plots of NiFe-LDH$_x$ measured at –0.3 V.

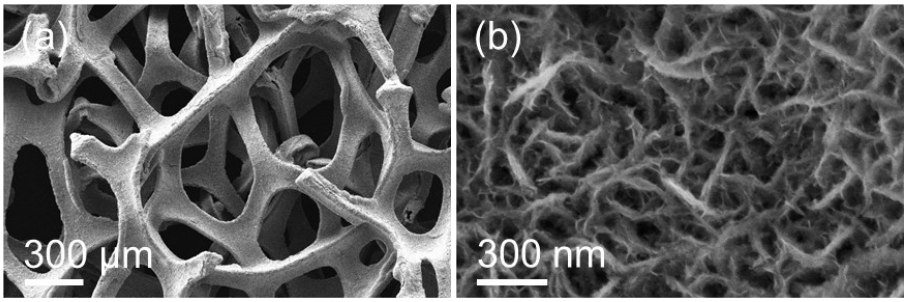

Fig. S8. SEM images of NiFe-LDH$_{2.18}$ after the stability test.

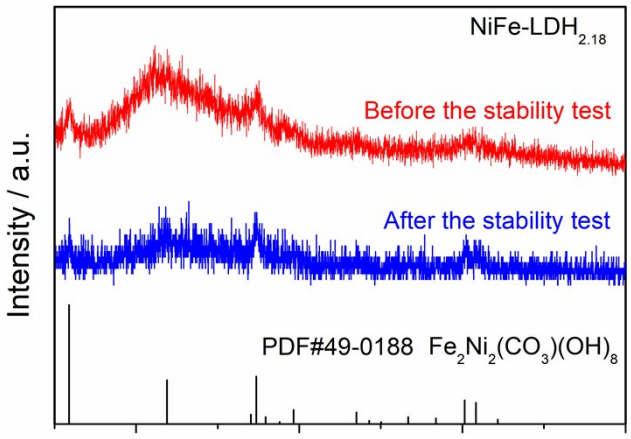

Fig. S9. XRD patterns of NiFe-LDH$_{2.18}$ before and after the stability test.

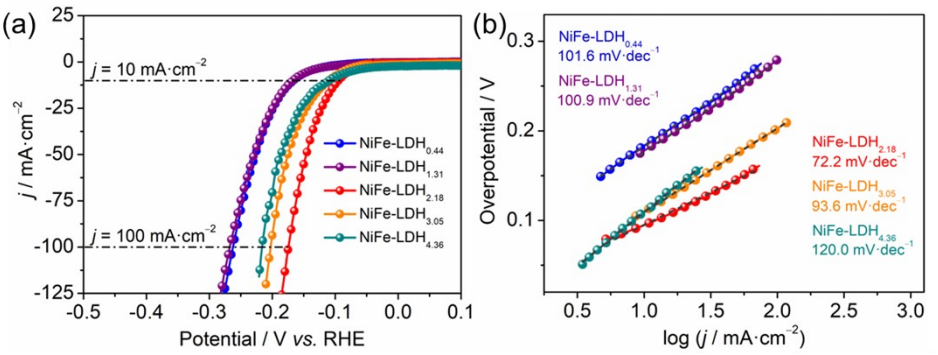

Fig. S10. The influence of the concentration of NH_3 on the HER performances of NiFe-LDH nanosheets: (a) *iR*-corrected HER polarization curves, (b) corresponding Tafel plots.

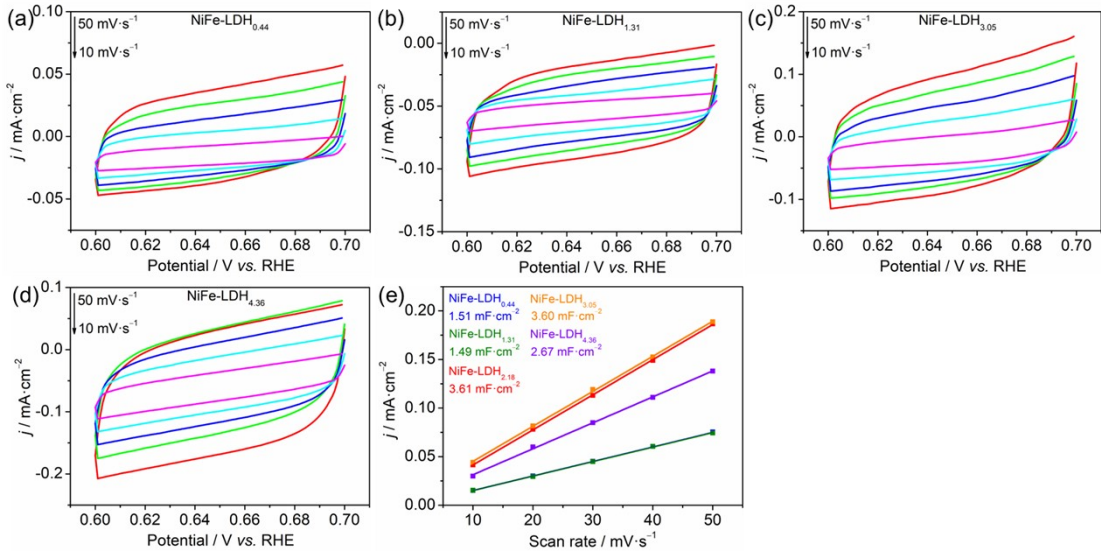

Fig. S11. CV curves of (a) NiFe-$LDH_{0.44}$, (b) NiFe-$LDH_{1.31}$, (c) NiFe-$LDH_{3.05}$ and (d) NiFe-$LDH_{4.36}$ in non-Faradaic region at scan rates of 10, 20, 30, 40 and 50 $mV \cdot s^{-1}$ in 1 M KOH solution, (e) current density at 0.65 V as a function of scan rate.

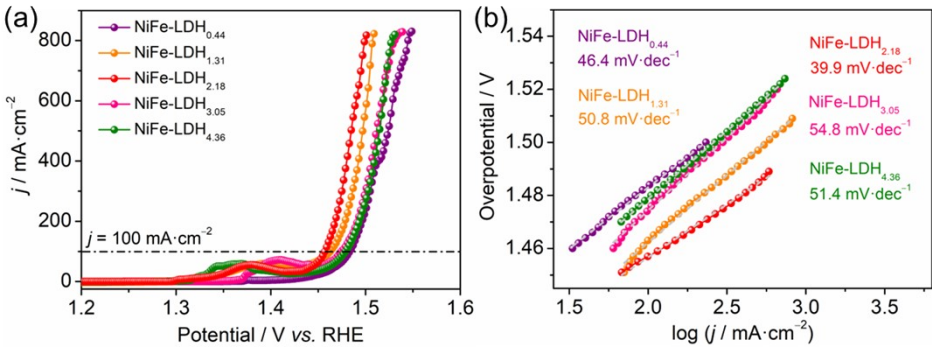

Fig. S12. (a) *iR*-corrected OER polarization curves, (b) corresponding Tafel plots of NiFe-LDH$_x$ series samples obtained with different concentrations of NH_3.

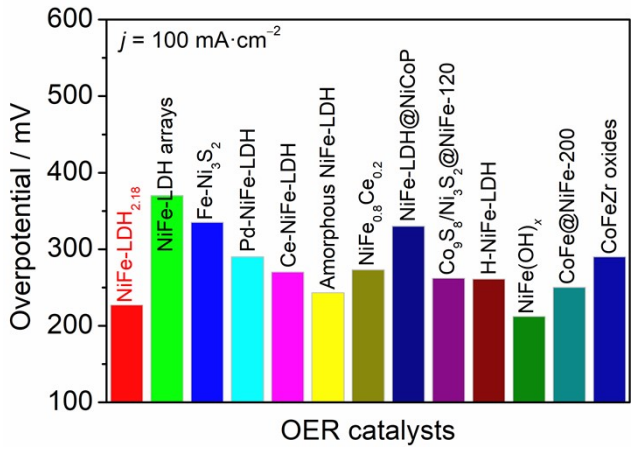

Fig. S13. Overpotentials required to achieve a current density of 100 mA·cm^{-2} on NiFeLDH$_{2.18}$ and other related OER catalysts.

Table S1 Comparison of electrochemical HER/OER performance between NiFe-$LDH_{2.18}$ in this work and other representative self-supported transition-metal-based catalysts reported in literatures (electrolyte is 1.0 M KOH).

Catalysts/Substrate	HER			OER			Water splitting	Ref.
	$\eta_{j=10}$[a] (mV)	$\eta_{j=100}$[b] (mV)	Tafel slope (mV·dec^{-1})	$\eta_{j=10}$ (mV)	$\eta_{j=100}$ (mV)	Tafel slope (mV·dec^{-1})	$E(j$=10 mA·cm^{-2}) (V)	
NiFe-$LDH_{2.18}$/Ni foam	95	174	72.2	NA	227	39.9	1.61	This work
NiFe-LDH nanosheets/Ni nanowire	116	NA	74	295*	NA	68	1.69 (30 mA·cm^{-2})	1
NiFe-LDH arrays	127	340*	109.4	193	370	143.1	1.652	2
NiFe-LDH/Ni foam	59	NA	62.3	184	NA	28.83	NA	3
Fe-Ni_3S_2/Fe-Ni alloy foam	75	200*	103	267	335	36	1.52*	4
Pd-NiFe-LDH/Ni foam	130	345*	46	156	290	47	1.514	5
Ce-NiFe-LDH/Ni foam	147	350*	112	175	270*	59	1.59	6
Ag@NiFe-LDH/carbon paper	200	545*	87	330	NA	89	1.7	7
Amorphous NiFe-LDH/Ni foam	95	182	NA	197	243	46.6	1.52	8
$NiFe_{0.8}Ce_{0.2}$/Ni foam	147	325	112	175	273	59	1.59	9
NiFe-LDH@NiCoP/Ni foam	120	325	88.2	220	330*	48.6	1.57	10
Co_9S_8/Ni_3S_2@NiFe-120/Ni foam	152	259	99.1	229	262	47.4	1.62	11
H-NiFe-LDH/Ni foam	189	340*	108	NA	261	72.2	1.575	12
$NiFe(OH)_x$/Fe foam	124 (20 mA·cm^{-2})	186	94.7	150 (20 mA·cm^{-2})	212	26.1	1.71 (100 mA·cm^{-2})	13
CoFe@NiFe-200/Ni foam	240	345*	88.88	190	250*	45.71	1.59	14
CoFeZr oxides/Ni foam	104	245*	119.3	264 (20 mA·cm^{-2})	290*	54.2	1.63	15

[a] The overpotential at the current density of 10 mA·cm^{-2},

[b] The overpotential at the current density of 100 mA·cm^{-2},

* The value is reckoned from the curves shown in the literature.

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