

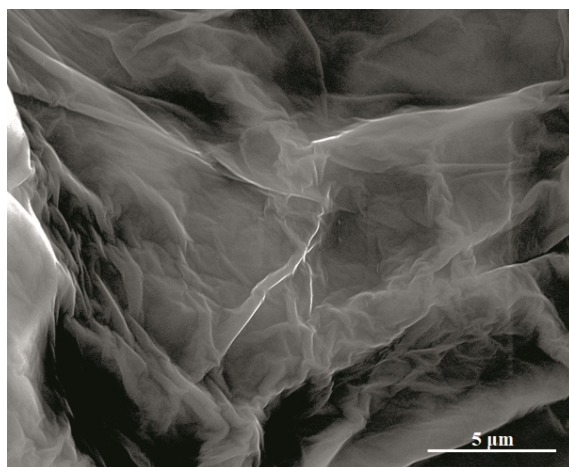
## Interfacial Engineering of NiFeP/NiFe-LDH Heterojunction for Efficient Overall Water Splitting

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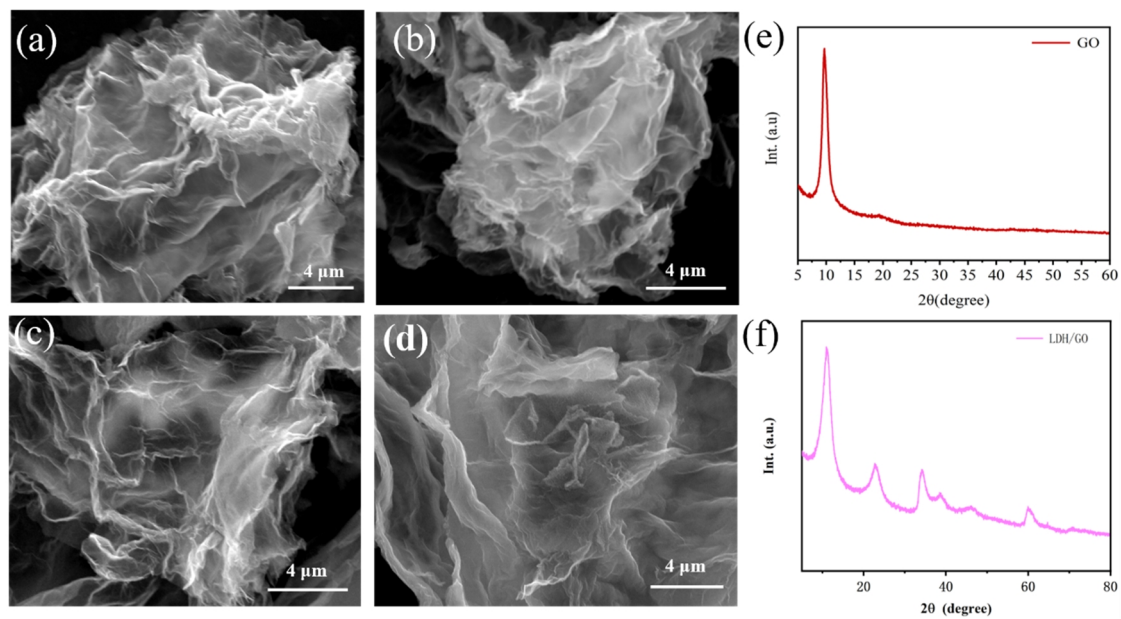
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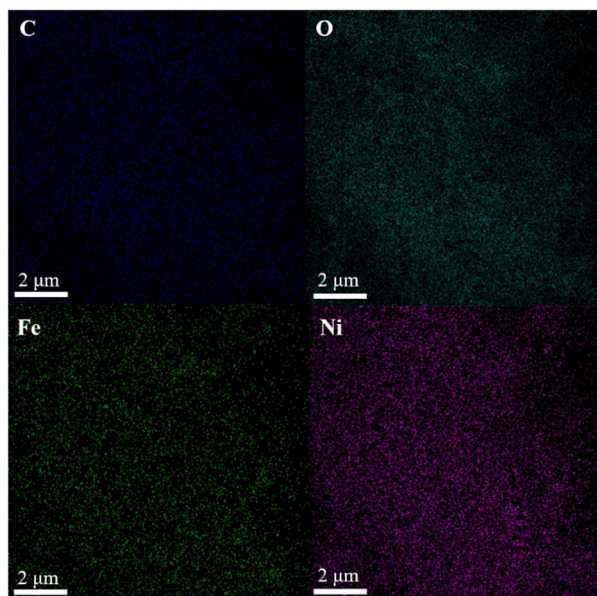
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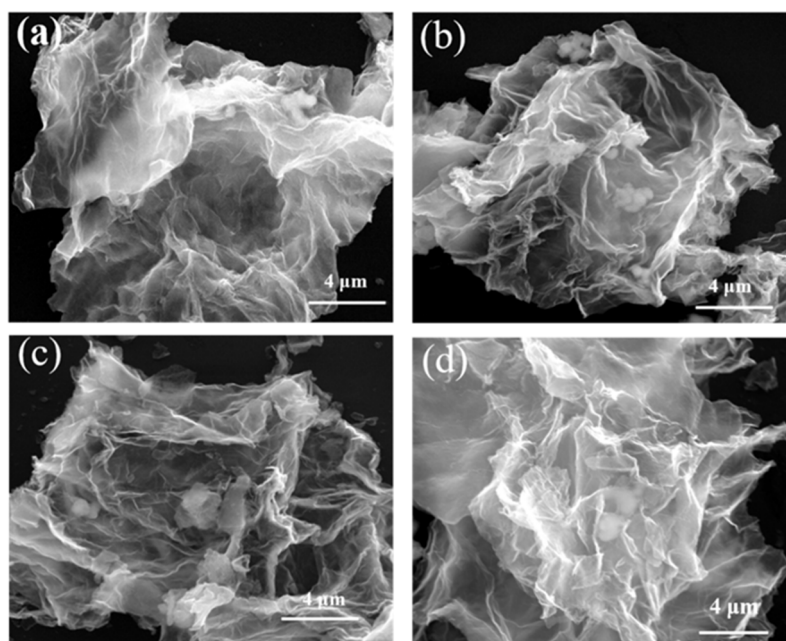
**Figure S1.** SEM image of graphene oxide (GO).



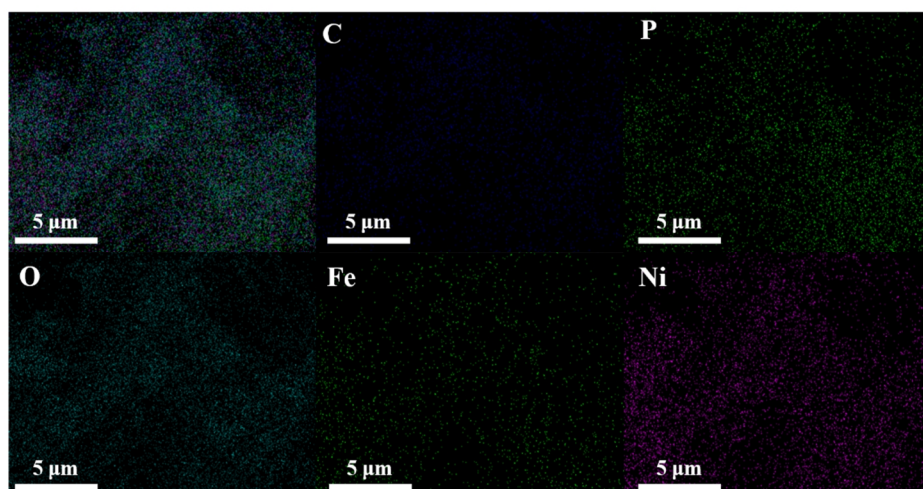
**Figure S2.** SEM images of the flower-like  $\text{Ni}_x\text{Fe}_y\text{-LDH/GO}$ : (a)  $x:y = 0.6:0.4$ ; (b)  $x:y = 0.8:0.2$ ; (c)  $x:y = 0.7:0.3$ ; (d)  $x:y = 0.5:0.5$ . (e) XRD of GO; (f) XRD of LDH/GO.



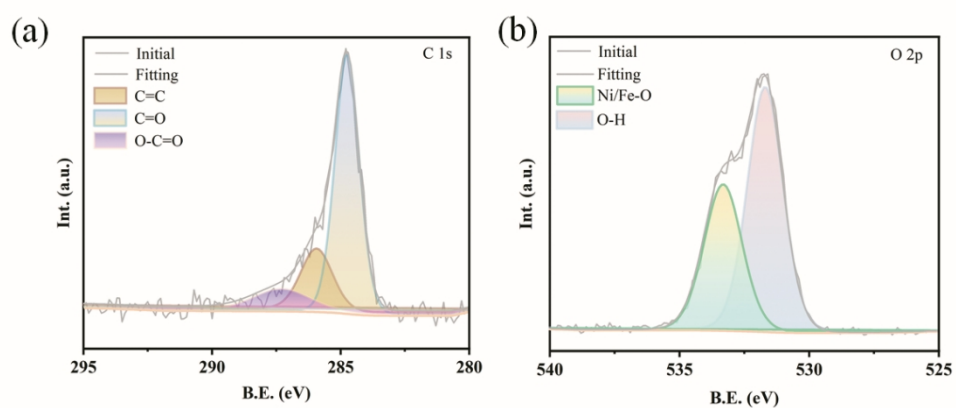
**Figure S3.** EDS mapping patterns of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{-LDH/GO}$ .



**Figure S4.** SEM images of the Ni<sub>x</sub>Fe<sub>y</sub>P/LDH/GO: (a) x:y = 0.6:0.4; (b) x:y = 0.8:0.2; (c) x:y = 0.7:0.3; (d) x:y = 0.5:0.5.



**Figure S5.** EDS mapping patterns of the flower-like  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$ .



**Figure S6.** The XPS measurement results for  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$ : (a) C 1s, (b) O 2p core levels.

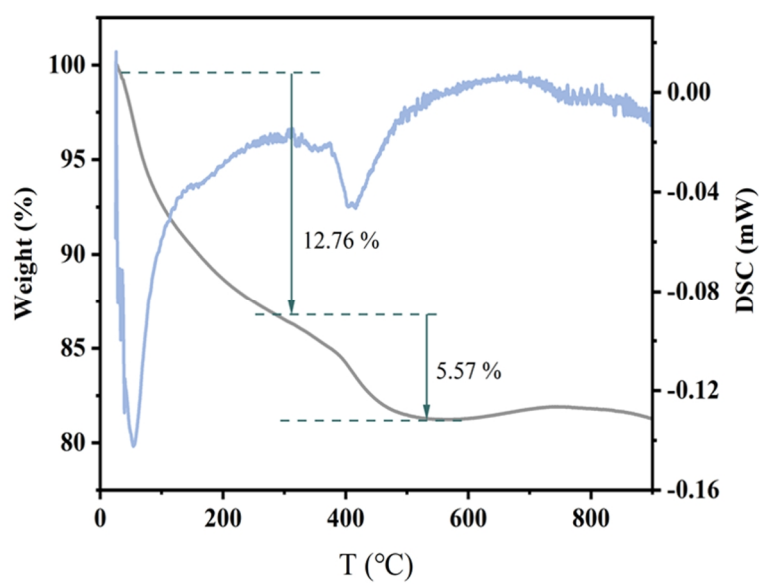
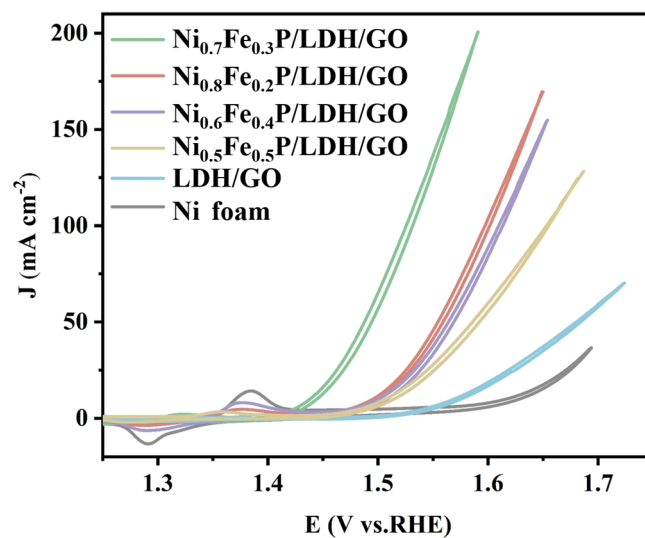
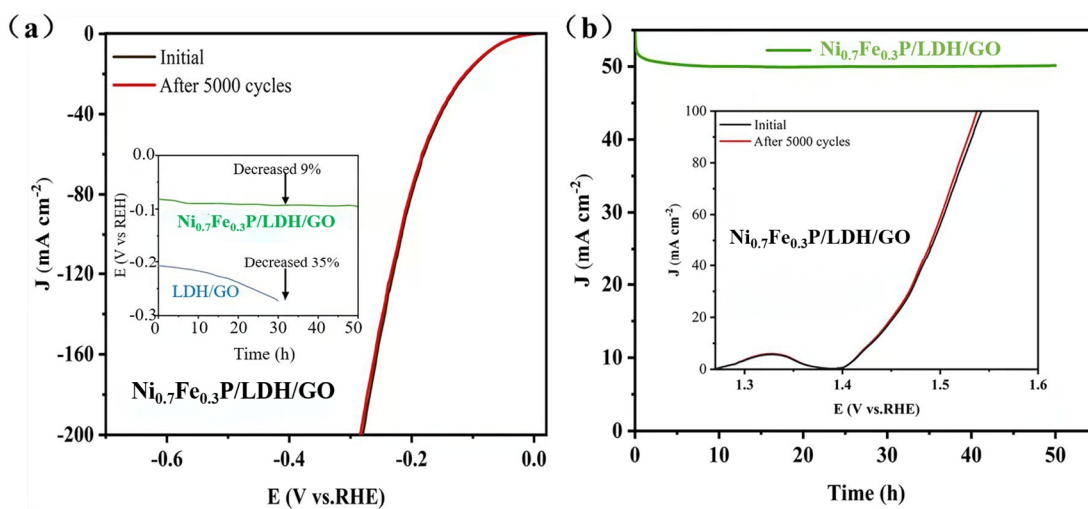


Figure S7. TG analysis of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$ .

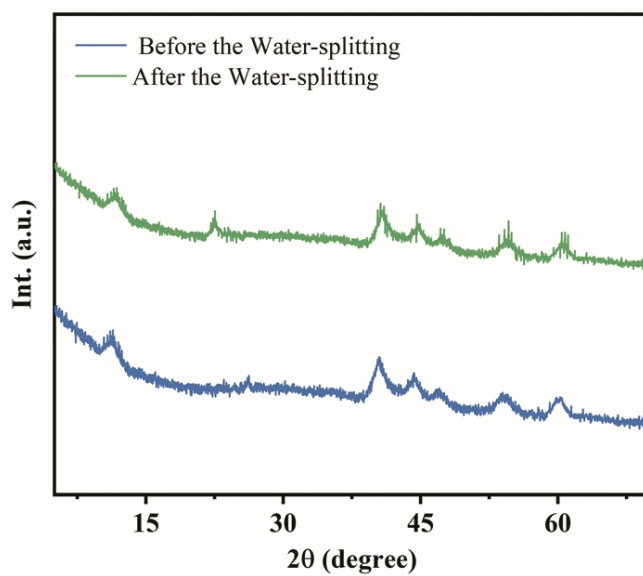




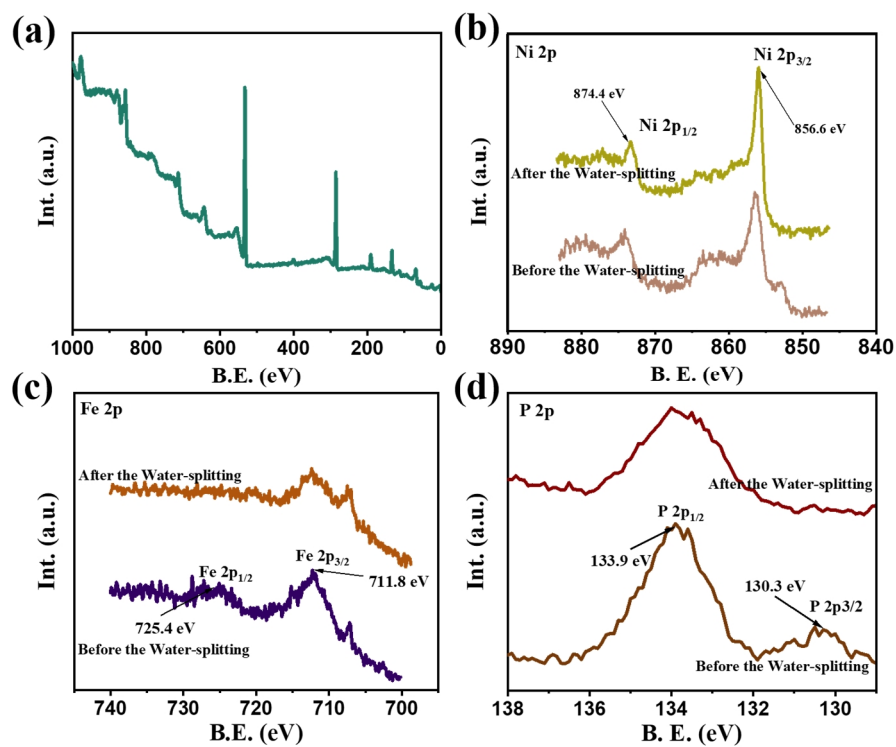
**Figure S8.** Cyclic voltammetry curves of samples for OER in 1 M KOH at a scan rate of 1 mV·s<sup>-1</sup> without stirring.



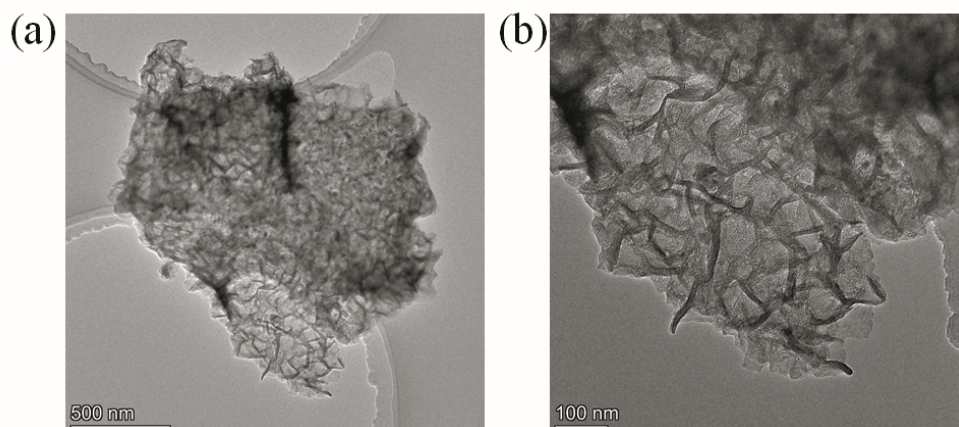
**Figure S9.** (a) Polarization curves of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$  before and after 5000 cycles for HER (Inset: The galvanostatic tests of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$  and LDH/GO are conducted for HER in 1 M KOH at  $j = 10 \text{ mA cm}^{-2}$ ); (b) The constant voltage test of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$  for OER in 1 M KOH at  $E = 1.49 \text{ V vs. RHE}$  (Inset: polarization curves of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$  before and after 5000 cycles for OER).



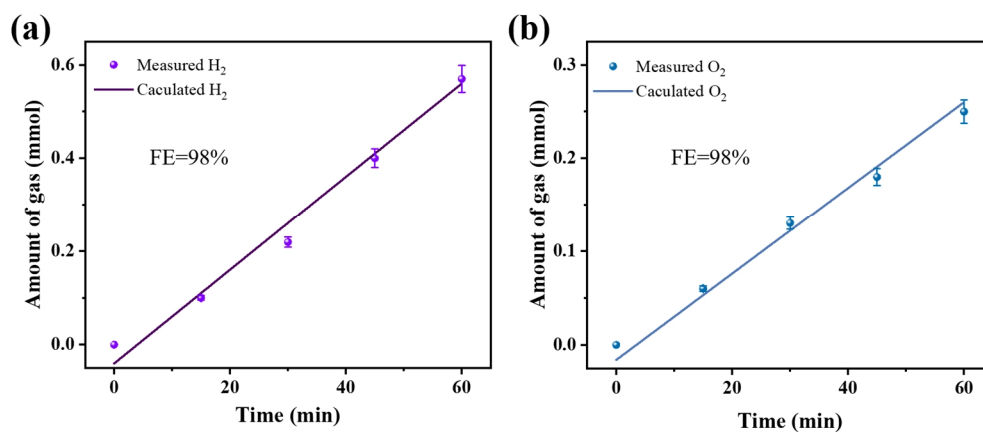
**Figure S10.** XRD patterns of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$  before and after overall water splitting.



**Figure S11.** XPS patterns of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$  before and after overall water: (a) survey, (b) Ni 2p, (c) Fe 2p, (d) P 2p core levels.



**Figure S12.** TEM of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$  after overall water splitting.



**Figure S13.** The amount of gas theoretically calculated and experimentally measured vs. time for  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$ : (a) for HER, (b) for OER in 1 M KOH at  $\eta = 296$  mV vs. theoretically calculated quantities assuming 100% FE for both HER and OER.

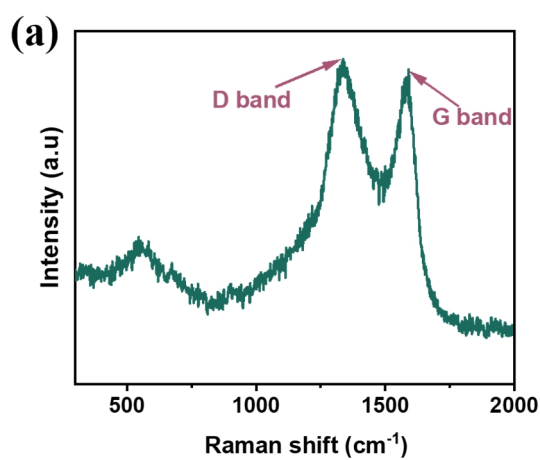
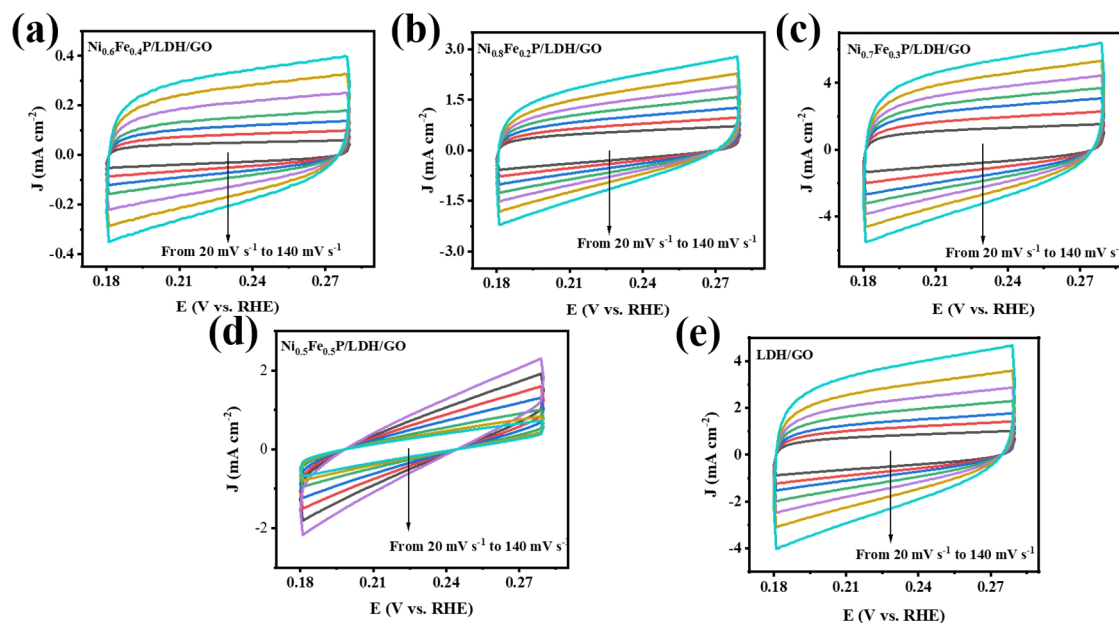
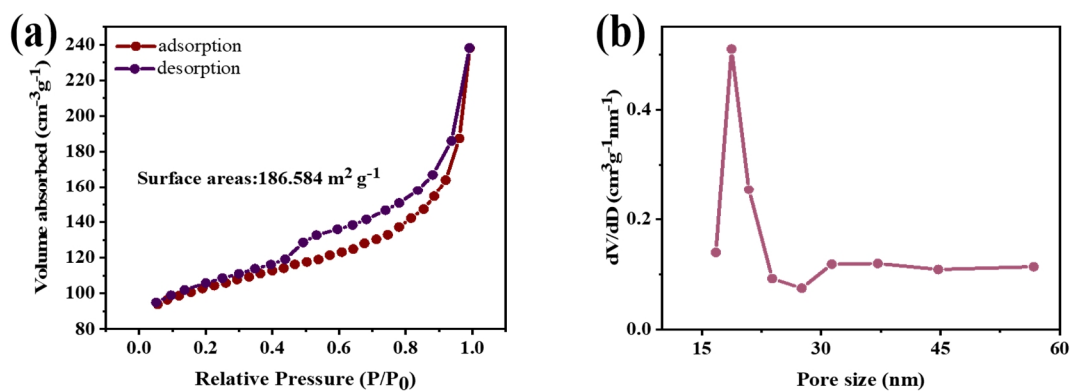


Figure S14. Raman spectra analysis of Ni<sub>0.7</sub>Fe<sub>0.3</sub>P/LDH/GO.



**Figure S15.** (a–e) cyclic voltammetry (CV) measurement with varied scan rates (20, 40, 60 etc. mV·s<sup>-1</sup>) for Ni<sub>x</sub>Fe<sub>y</sub>P/LDH/GO under different scan rates in the potential region from 0.18 to 0.28 V vs. RHE





**Figure S16.** Structural characterization patterns of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$ : (a) The nitrogen adsorption-desorption isotherm, (b) The pore size distribution result.

**Table S1.** Inductive Coupled Plasma Emission Spectrometer (ICP-MS) Analysis of Ni<sub>x</sub>Fe<sub>y</sub>P/LDH/GO Heterojunction with Different Ni/Fe Molar Ratios.

Materials	Fe (g)	Ni (g)	Ni/Fe (molar ratios)
Ni <sub>0.6</sub> Fe <sub>0.4</sub> P/LDH/GO	0.358	0.562	0.592:0.378
Ni <sub>0.8</sub> Fe <sub>0.2</sub> P/LDH/GO	0.129	0.518	0.883:0.231
Ni <sub>0.7</sub> Fe <sub>0.3</sub> P/LDH/GO	0.182	0.425	0.724:0.325
Ni <sub>0.5</sub> Fe <sub>0.5</sub> P/LDH/GO	0.287	0.291	0.496:0.512

**Table S2.** Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) Analysis of  
Ni<sub>0.7</sub>Fe<sub>0.3</sub>P/LDH/GO Heterojunction before and after Etching for 20 s.

Materials	Fe (atomic fraction %)	Ni (atomic fraction %)	P (atomic fraction %)
Pristine	23.4	51.8	14.6
Etched	23.7	52.1	1.7

**Table S3.** Comparison of the Water Splitting Property of  $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$  to Other Catalysis in 1 M KOH ( $j_{10} = 10 \text{ mA}\cdot\text{cm}^{-2}$ )

Catalyst	Overpotential for HER (mV)	Overpotential for OER (mV)	Cell voltage (V)	Stability (h)	References
$\text{Ni}_{0.7}\text{Fe}_{0.3}\text{P/LDH/GO}$	79	198	1.526	25	This work
$\text{Co}_3\text{S}_4@\text{MoS}_2$	136	280	1.58	10	2
$\text{MoO}_2@\text{MoS}_2@\text{Co}_9\text{S}_8$	160	310	1.62	20	3
NG-NiFe@ $\text{MoC}_2$	150	320	1.53	10	4
Co-P films	94	345	1.56	24	5
Ni/NiP	130	270	1.61	24	6
Al, Fe codoped CoP/RGO	145	280	1.66	10	7
$\text{NiCo}_2\text{O}_4@\text{Ni}_2\text{P/NAs}$	141	350	1.52	6	8
NP-RuO <sub>2</sub>	87	250	1.62	12	9
NiCoP/CNTs	82	250	1.558	20	10
$\text{NiCo}_2\text{O}_4@\text{C}$	109	220	1.608	48	11
FeCo/CFPs	163	283	1.68	10	12
CoNi-OOH	210	279	1.76	60	13
NiFe@C	195	274	1.63	20	14
$\text{Ni}_{1-x}\text{Fe}_x\text{-HP}$	215	280	1.57	24	15
NiO	424	370	1.6	6	16

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