Supporting Information

1D/2D NiFeP/NiFe-OH heterostructure: roles of the unique nanostructure in stabilizing highly efficient oxygen evolution reaction

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Experimental section

Preparation of materials

Ferrous Sulfate ($FeSO_4 \cdot 7H_2O$), sodium citrate dihydrate ($Na_3C_6H_5O_7 \cdot 2H_2O$), sodium sulfate ($Na_2SO_4$), potassium hydroxide (KOH), boric acid (H_3BO_3), potassium carbonate (K_2CO_3) muriatic acid (HCl) and sulfuric acid (H_2SO_4) were purchased from the Beijing Chemical Works (Beijing, China). Sodium hypophosphite (NaH_2PO_2) were purchased from Innochem-Beijing. Nickel sulfate ($NiSO_4 \cdot 6H_2O$) and sodium hydroxide (NaOH) were purchased from Shanghai Aladdin Bio-Chem Technology Co., LTD. Trisodium phosphate anhydrous (Na_3PO_4**)** were purchased from Shanghai Macklin Bochemical Co., LTD. The nickel foam was purchased from the Tianyu factory of Shandong province.

Preparation of clean Ni Foam

The process include: (1) The Ni foam were washed and degreased in the solution containing 10 g L^{-1} NaOH, 5 g L^{-1} Na_2SO_4 and 15 g L^{-1} Na_3PO_4 for oil removal. (2) The oxide layer on the surface of the Ni foam was removed in the solution containing 10 g L^{-1} HCl and 20 g L^{-1} H_2SO_4.

Electrodeposition of single NiFe-OH

A 50 ml electrolytic bath was used for electrodeposition at a temperature of 25 °C. An aqueous nitrate solution with 0.1 M total metal ions (Ni^{2+} and Fe^{2+}) served as the electrolyte. The substrate was treated to a cathodic deposition at 20 mA cm^{-2} for 600 s in a conventional synthesis.

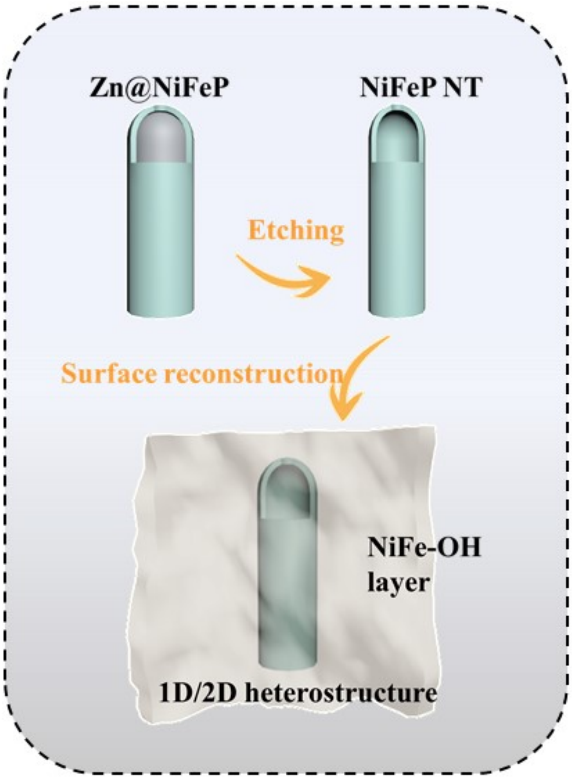

Figure S1. Schematic illustration of the synthesis processes.

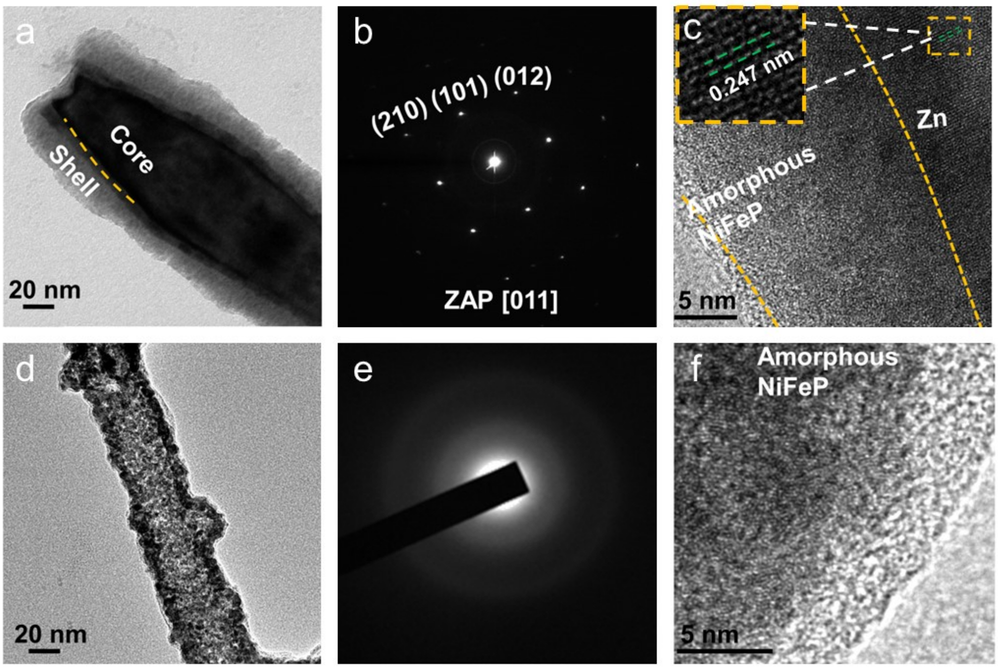

Figure S2. TEM results for the samples obtained at different steps. (a, d) TEM images, (b, e) SAED patterns, (c, f) HRTEM images of (a, b and c) Zn@NiFeP NWs and (d, e and f) 1D NTs.

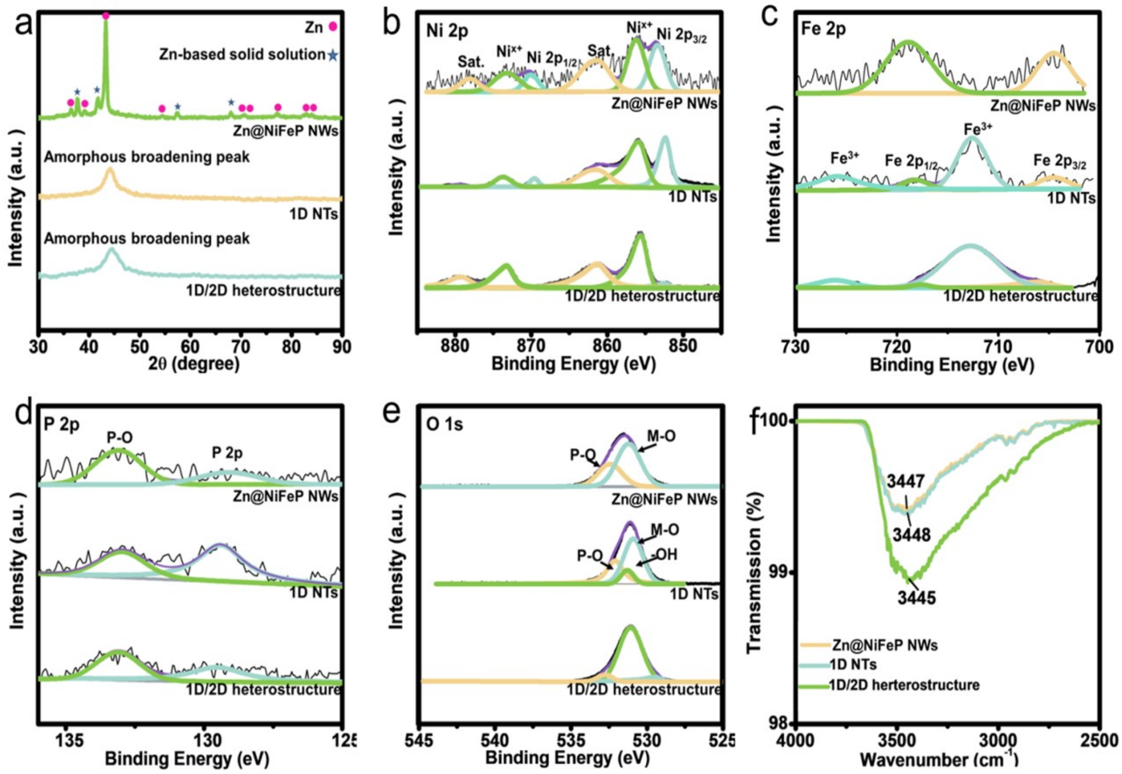

Figure S3. Changes of structure and composition of the sample obtained at different steps. (a) XRD patterns, (b) Ni 2p XPS spectra, (c) Fe 2p XPS spectra, (d) P 2p XPS spectra, (e) O 1s XPS spectra and (f) FTIR spectra of Zn@NiFeP NWs, 1D NTs and 1D/2D heterostructure.

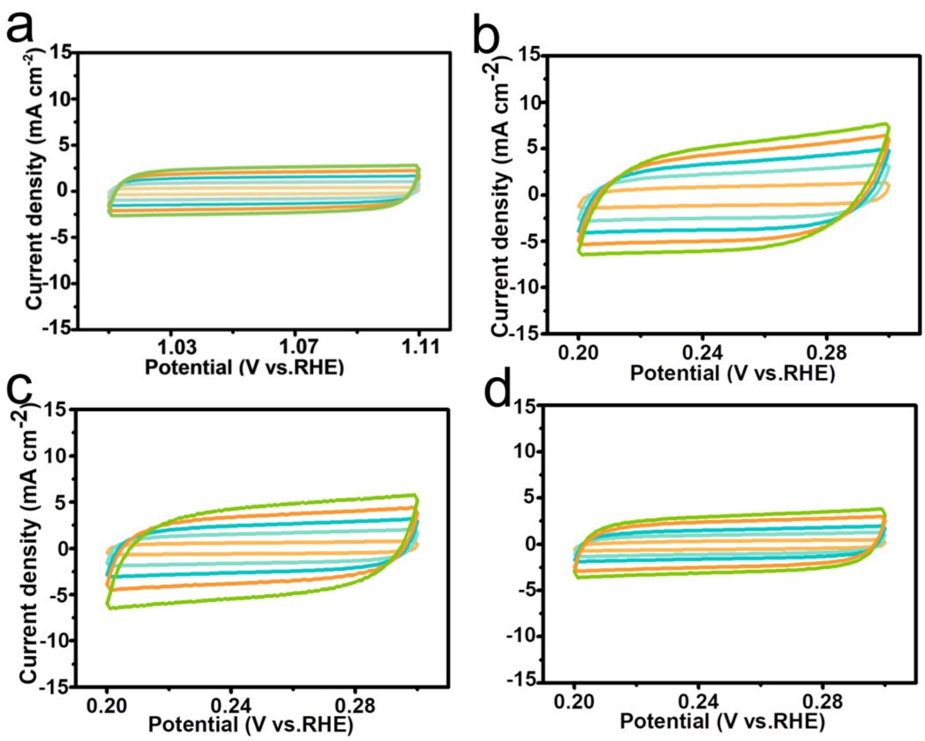

Figure S4. CV curves for ECSA measurements of IrO_2 and 1D/2D heterostructure with different Fe content. (a) IrO2. (b) Ni:Fe=96:4. (c) Ni:Fe=82:18. (d) Ni:Fe=64:36.

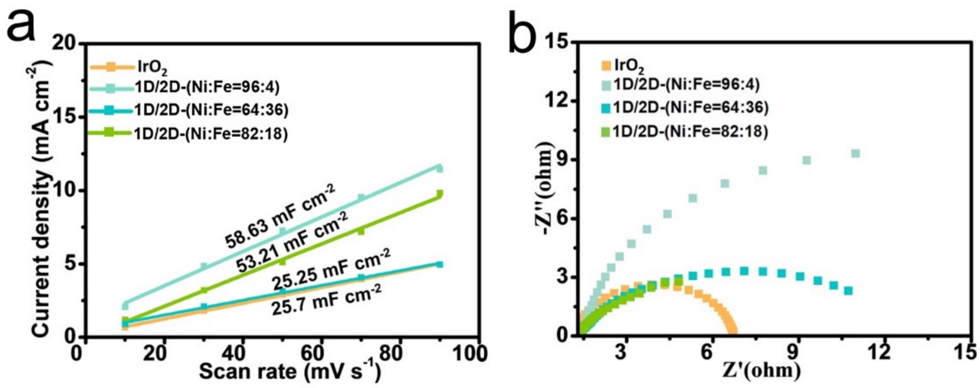

Figure S5. (a) Tafel plots of 1D/2D heterostructure with different Fe content. (b) EIS spectra of 1D/2D heterostructure with different Fe content

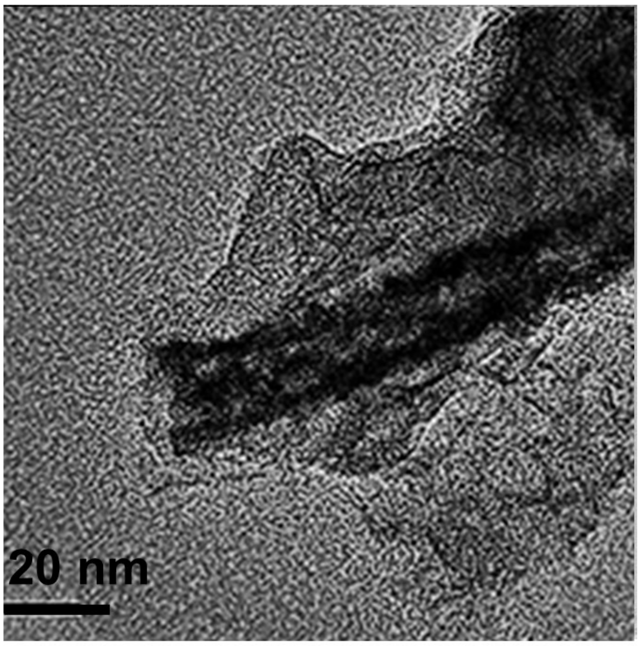

Figure S6. TEM image of 1D/2D heterostructure after 220 h OER measurement.

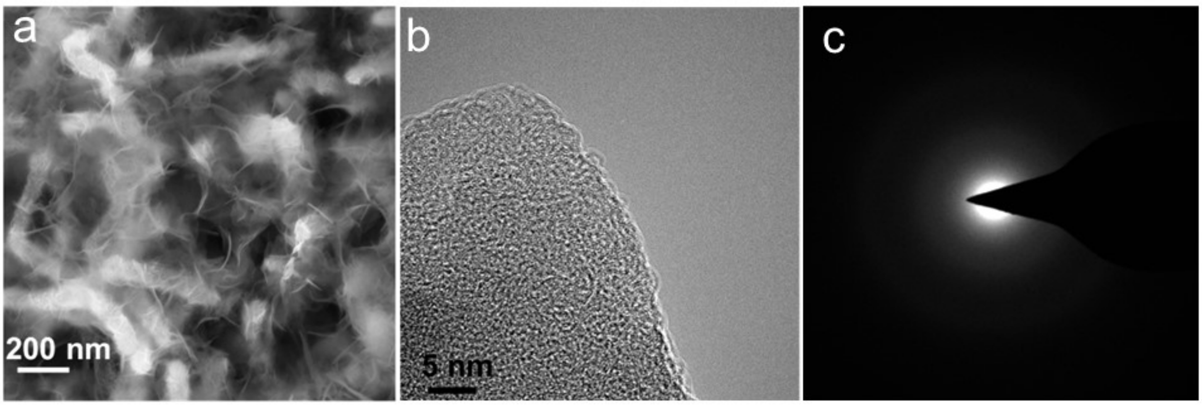

Figure S7. (a) SEM image, (b) HRTEM image and (c) SAED pattern of 1D/2D heterostructure after 220 h stability measurement.

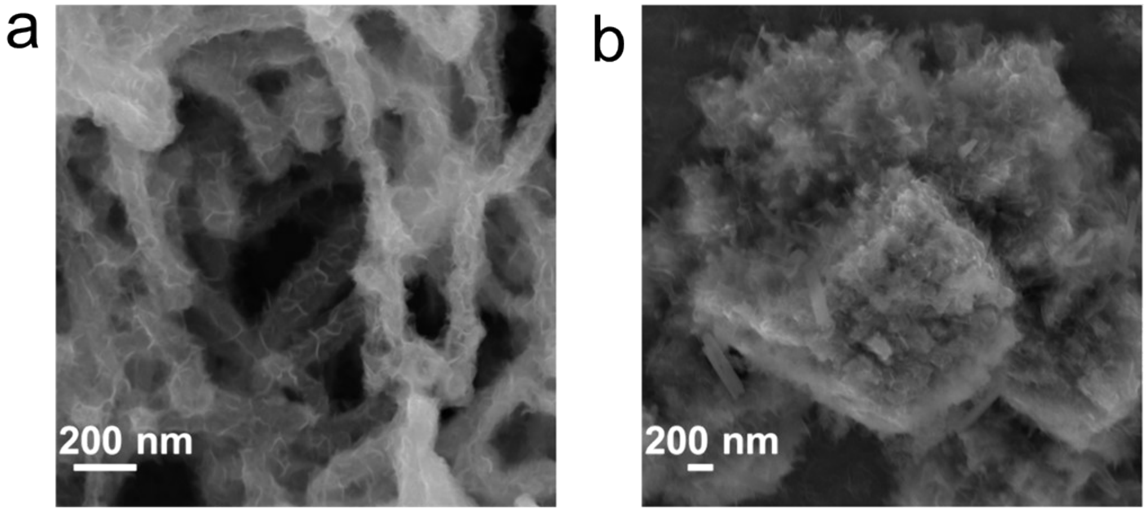

Figure S8. SEM images of sample with 4% Fe (a) and 36% Fe (b).

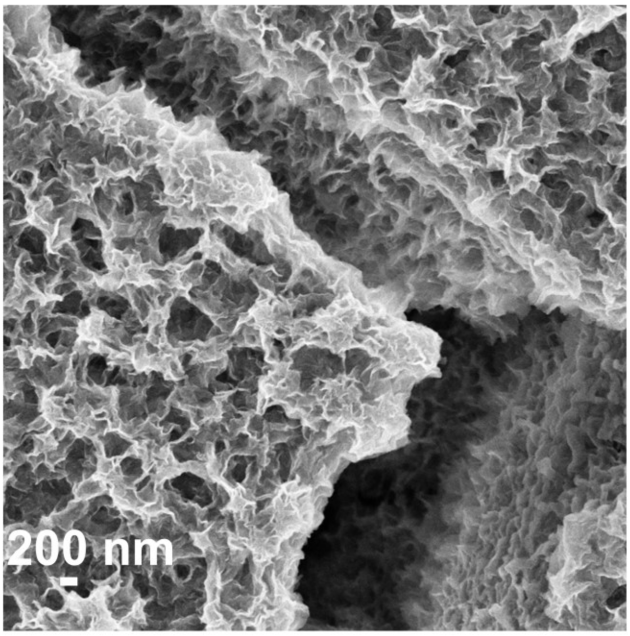

Figure S9. SEM image of measured single NiFe-OH.

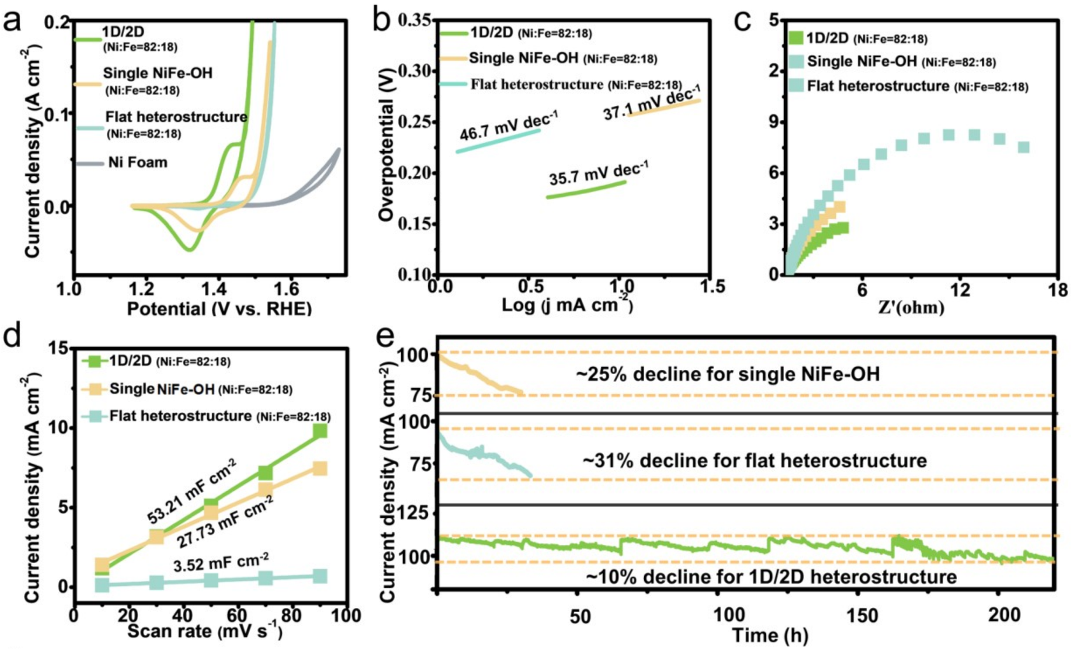

Figure S10. Electrochemical measurements. (a) CV curves, (b) Tafel slopes, (c) EIS spectra, (d) electrochemical double layer capacitance results for ECSA and (e) i-t curves of 1D/2D heterostructure, single NiFe-OH and flat heterostructure.

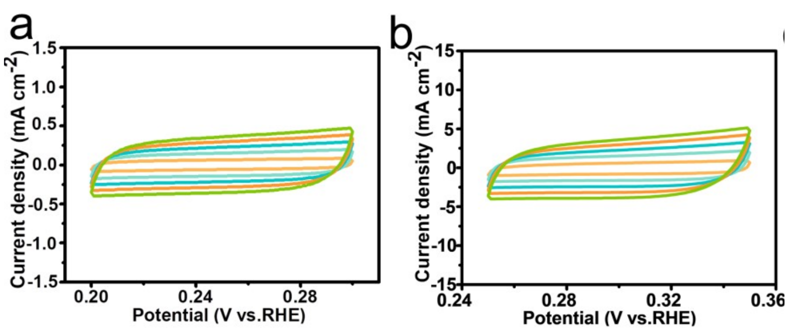

Figure S11. CV curves for ECSA measurements of (a) flat heterostructure and (b) single NiFe-OH.

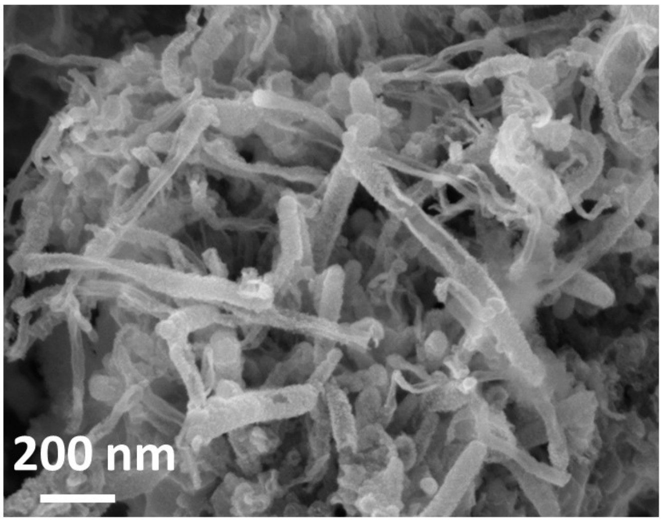

Figure S12. SEM image of measured 1D/2D heterostructure (220 h) after surface etching.

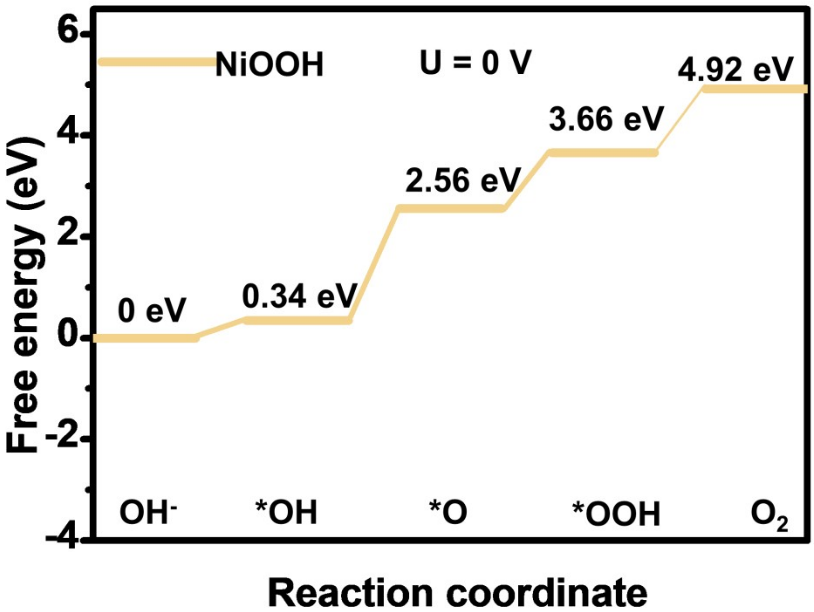

Figure S13. TEM image of 1D/2D heterostructure after 220 h OER measurement.

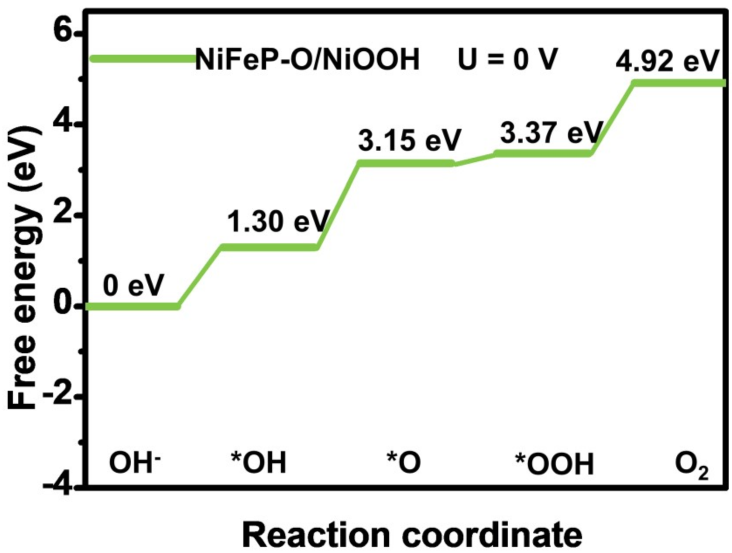

Figure S14. The calculated Gibbs free energy of NiFeP-O/NiOOH.

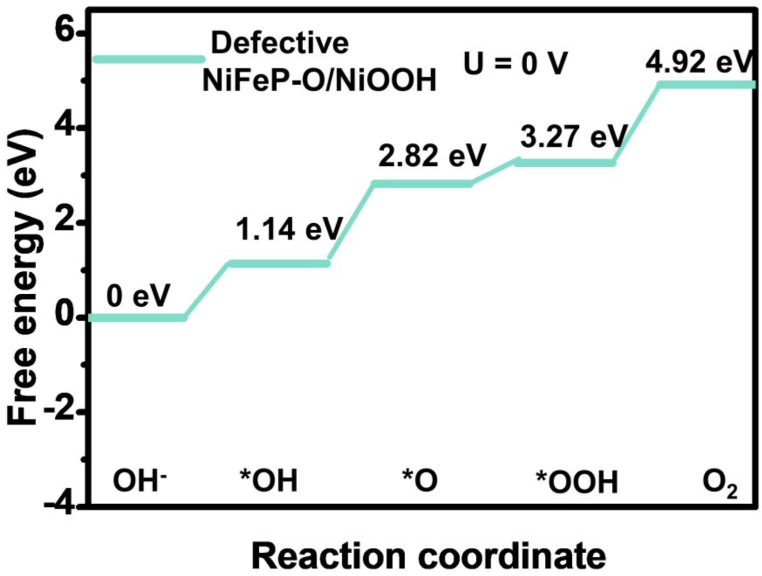

Figure S15. The calculated Gibbs free energy of defective NiFeP-O/NiOOH.

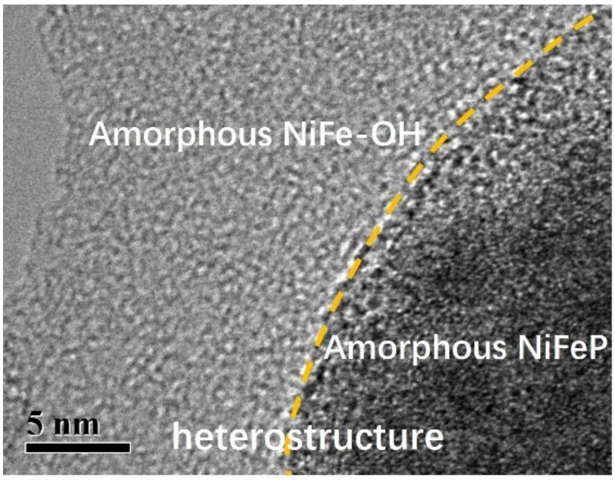

Figure S16. HRTEM image of 1D/2D heterostructure

Table S1. The results of EDS mapping

	Zn@NiFeP NWs	**NiFeP NTs**	**1D/2D heterostructure**	**1D/2D heterostructure after 220 h**	**1D/2D heterostructure (low content of Fe)**	**1D/2D heterostructure (low content of Fe)**
	Atomic percent (%)	Atomic percent (%)	Atomic percent (%)	Atomic percent (%)	Atomic percent (%)	Atomic percent (%)
P	5.45	9.14	10.6	8.90	7.10	6.09
Fe	5.38	14.03	16.29	12.60	3.60	29.94
Ni	19.23	76.83	73.11	78.50	89.30	62.97
Zn	69.94	-	-	-	-	-

Table S2. Activity and stability of various reported NiFe-LDH and related NiFe hydroxide catalysts for OER in 1M KOH.

OER electrocatalysts	η_{10}(mV)	Dur(h)	References
This work	190	220	
NiFe LDH@defective	210	10	1
NiFe LDH-rGO	250	9	2
NiFe LDH@HPGG	265	50	3
NiFe LDH @NGH	290	12	4
NiFe LDH QD@GH	220	10	5
NiFe LDH /C	210	28	6
NiFeCr LDH@CP	225	6	7
NiFeCe LDH-CNT	227	8.3	8
S–NiCoFe LDH NA	206	30	9
NiFe LDH@Cu NA	199	48	10
NiFe LDH-Cu_3P NA	235	11	11
Ni NP-NiFe LDH	328	18	12
NiFe LDH@NiCoP NA	220	100	13
NiFe LDH@Ni_3S_2	190	40	14
$NiFe_2O_4$/NiFe LDH	213	20	15
NiFe LDH@Co_3O_4	269	40	16
FeOOH/NiFe LDH NA	207	20	17
Fe–$Ni(OH)_2$	270	40	18
Ni/$NiFe(OH)_x$ NA	240	38	19
NiFe–Cu_2O NA	215	50	20
R–NiFeOOH	270	11	21

Table S3. Calculation results of ZPE-TS for Gibbs free energy.

	H_2O	H_2	sur-OH*	sur-O*	sur-OOH*
ZPE – TS (eV)	0.084	0.05	0.34	0.08	0.28

Table S4. Thermodynamic data used in the calculations of Gibbs free energy

	E(eV)	G
H_2O	-14.218518	-14.134807
H_2	-6.77	-6.817005
O_2	-	-9.71559
OH^-	-	-10.726305
NiFeP-O/NiOOH	-654.77046	-683.04
NiFeP-O/NiOOH-OH*	-664.54105	-681.744
NiFeP-O/NiOOH-O*	-659.01612	-679.888
NiFeP-O/NiOOH-OOH*	-669.72842	-679.674
NiOOH	-387.82304	-416.093
NiOOH-OH*	-398.54586	-415.749
NiOOH-O*	-392.65726	-413.529
NiOOH-OOH*	-402.48702	-412.433
Defective NiFeP-O/NiOOH	-618.90051	-647.170
Defective NiFeP-O/NiOOH-OH*	-623.47677	-646.036
Defective NiFeP-O/NiOOH-O*	-633.96012	-644.349
Defective NiFeP-O/NiOOH-OOH*	-628.83232	-643.906

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