Supporting information

Radially Aligned Hierarchical Nickel/Nickel-Iron (Oxy)hydroxide Nanotubes for Efficient Electrocatalytic Water Splitting

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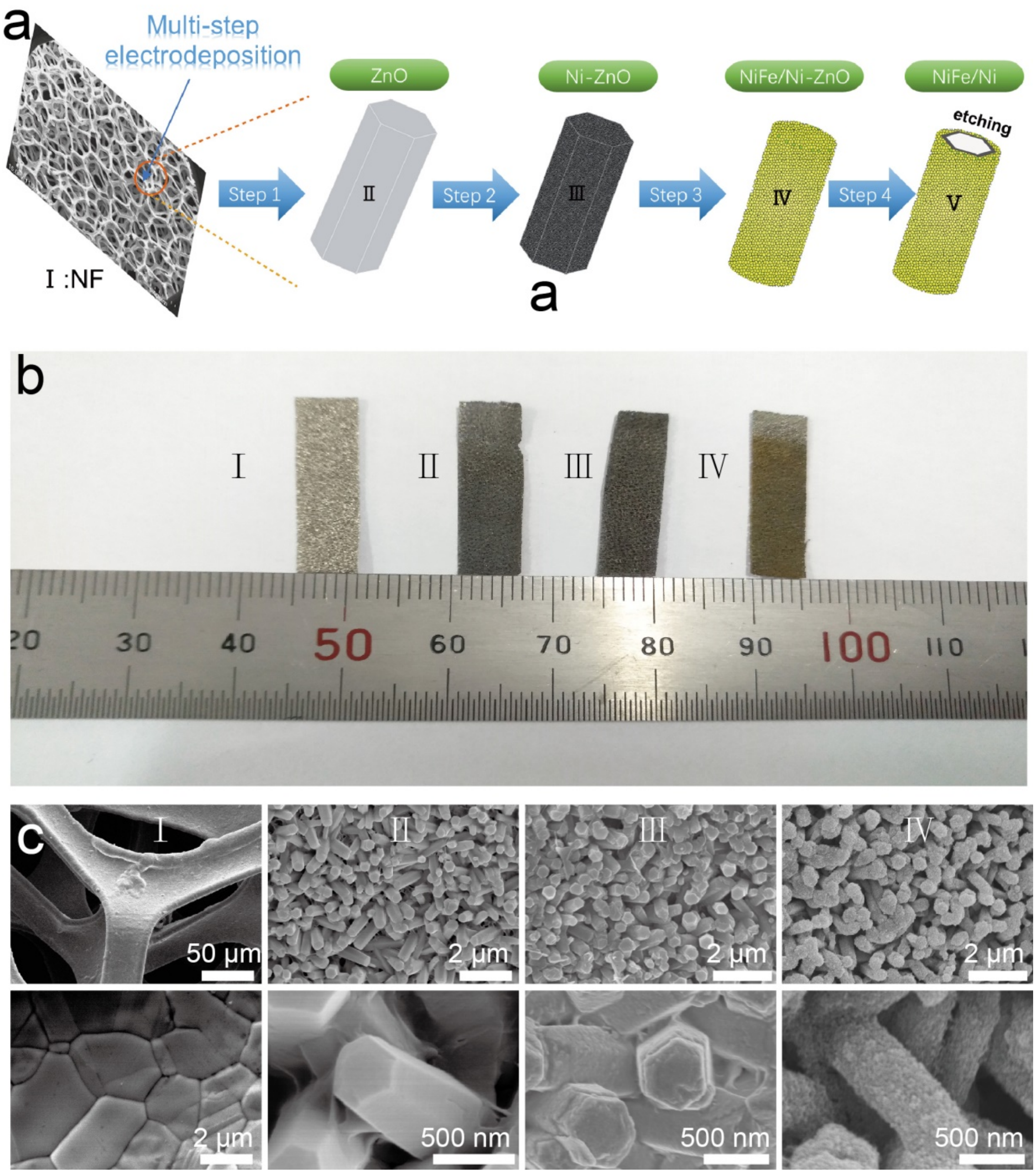

Figure S1. (a) Schematic diagram for sample growth, (b) digital photographs, and (c) low-magnification SEM images for samples at the different growth steps. The color change evidenced the successive growth of Zn, Ni, and NiFe (oxy)hydroxide.

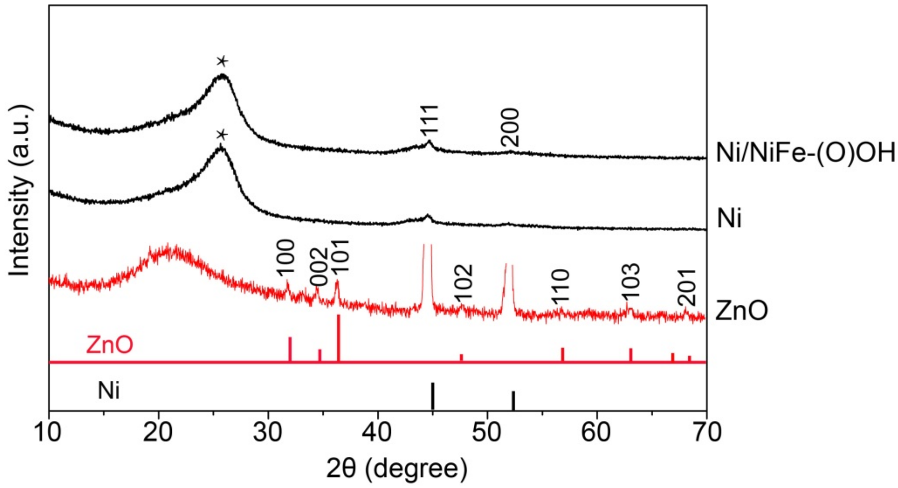

Figure S2. XRD patterns for the products at the different steps. For samples of ZnO and Ni, carbon cloth was used as substrate to exclude the possible strong diffracting interference of Ni foam.

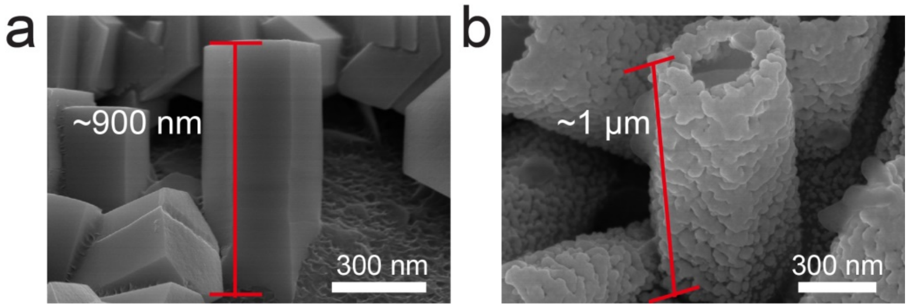

Figure S3. SEM images showing the length of (a) ZnO template and (b) NiFe (oxy)hydroxide tube, maintaining almost unchanged at ca. 1 μm.

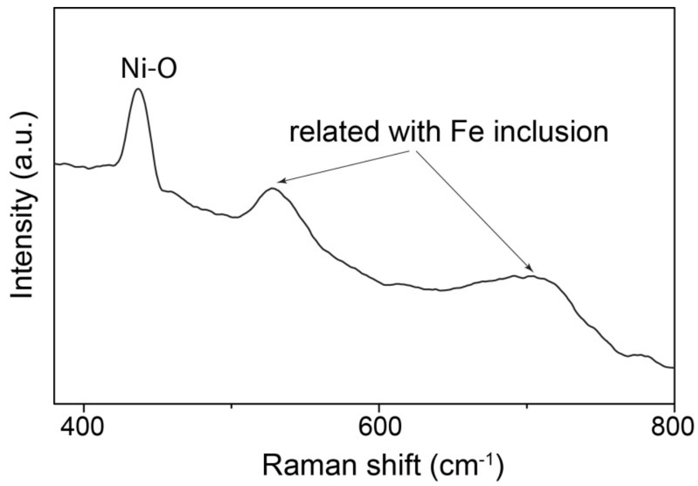

Figure S4. Raman spectrum for the Ni/NiFe nanotubes.

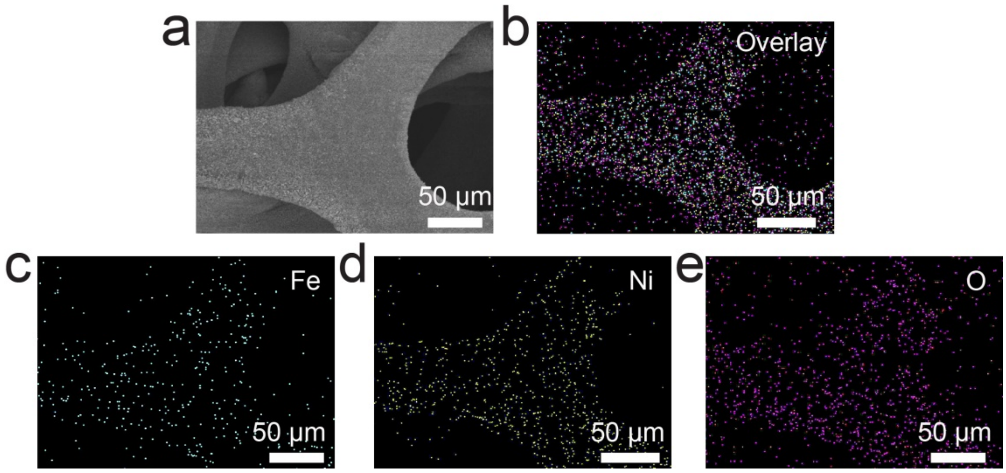

Figure S5. (a) Low-magnification SEM image for the final product of Ni/NiFe (oxy)hydroxide nanotube arrays and corresponding elemental mapping images of (b) overlaying, and individual (c) Fe, (d) Ni, (e) O elements, showing homogeneous distribution for all elements, which implied even growth of the product.

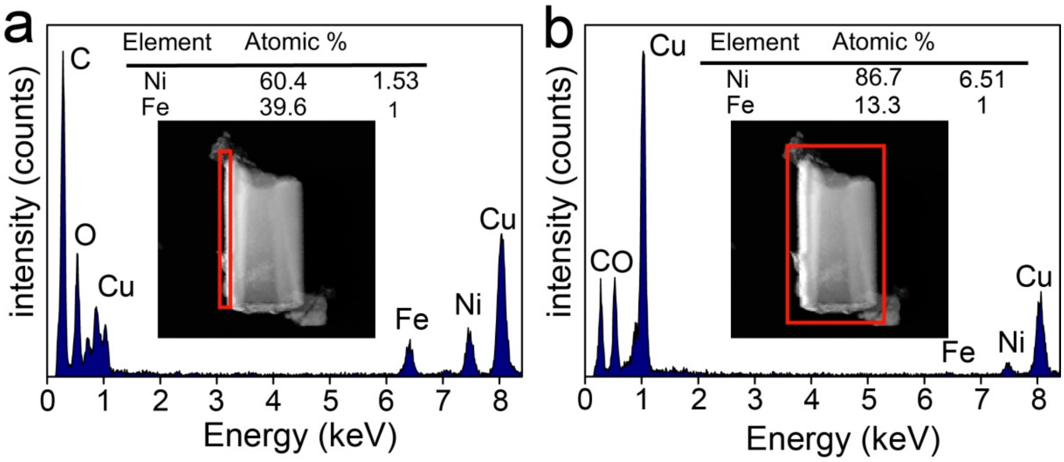

Figure S6. EDS spectrum for (a) outer NiFe (oxy)hydroxide and (b) the whole wall.

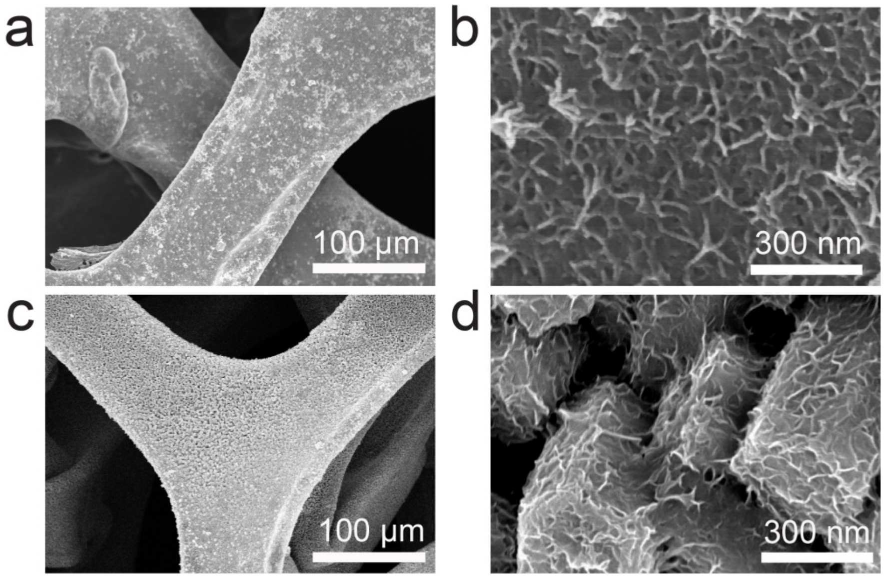

Figure S7. NiFe (oxy)hydroxide directly grown on (a-b) Ni foam and (c-d) ZnO. The morphology was sheet-like wrapping on Ni substrate or ZnO rods.

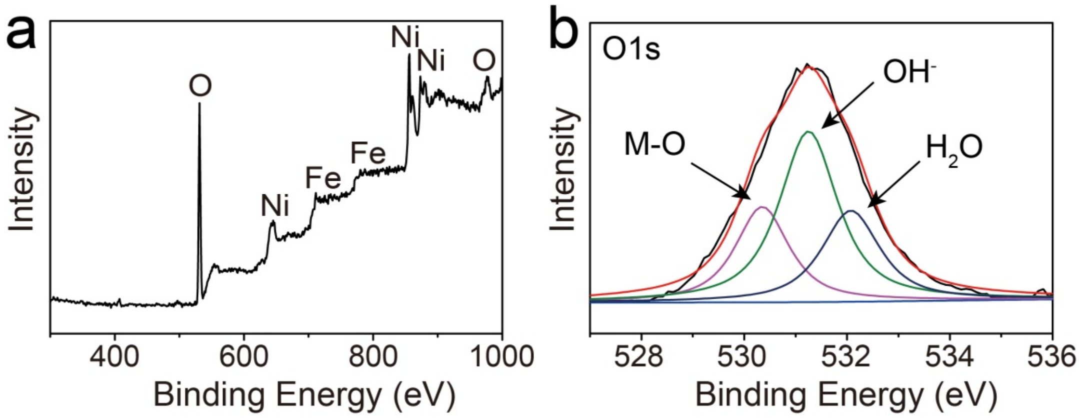

Figure S8. (a) XPS full-range and (b) O1s spectra for Ni/NiFe (oxy)hydroxide nanotubes.

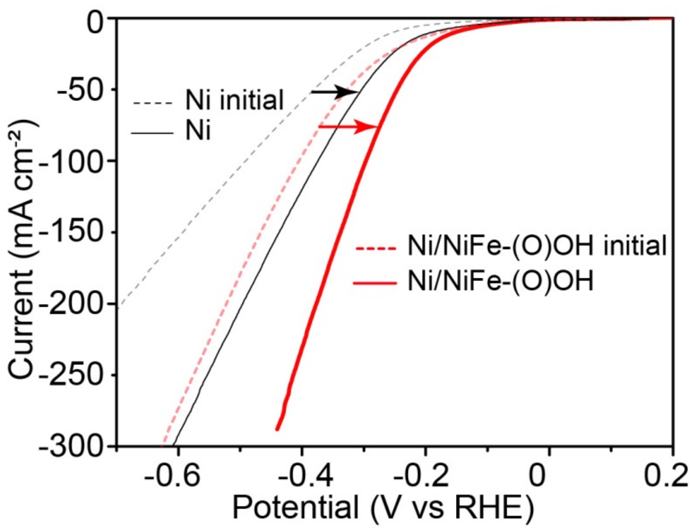

Figure S9. HER LSV curves of the Ni/NiFe (oxy)hydroxide before and after oxidation treatment, showing that the HER performance was to some extent optimized by the gentle oxidation treatment.

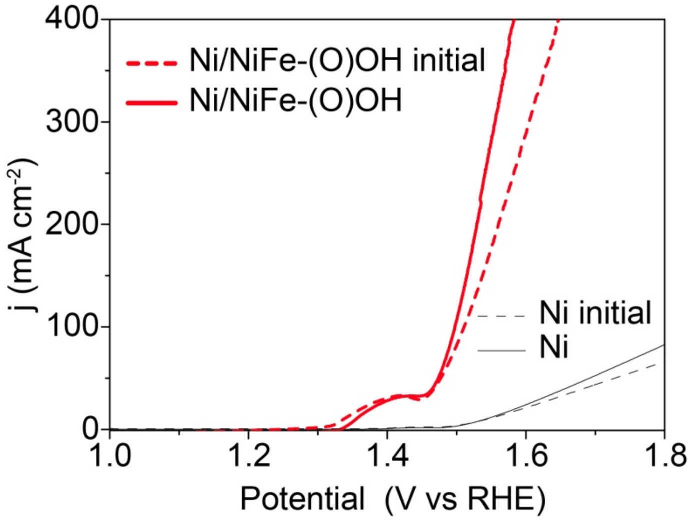

Figure S10. OER LSV curves of the Ni/NiFe (oxy)hydroxide before and after oxidation treatment, showing that the performance was little affected.

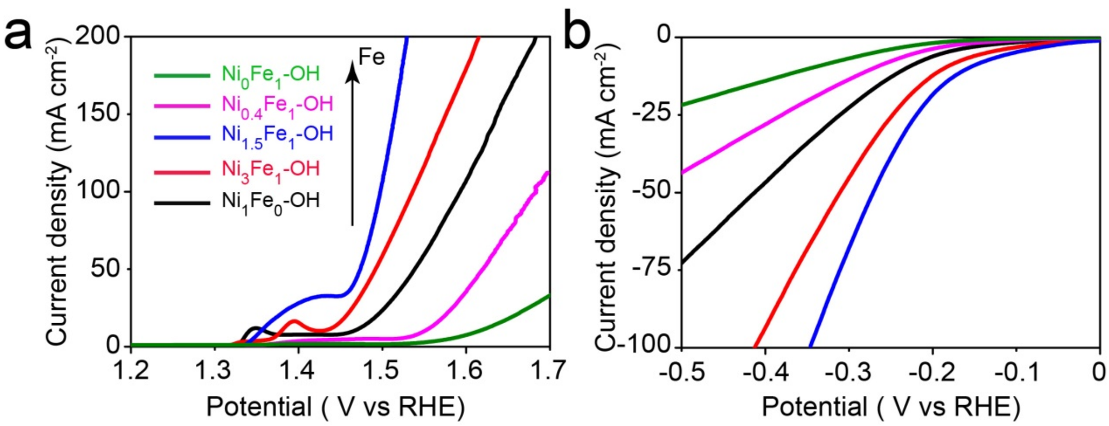

Figure S11. LSV curves for samples with varying Fe content toward (a) OER and (b) HER reactions, from which it could be drawn that $Ni_{1.5}Fe$ was optimal.

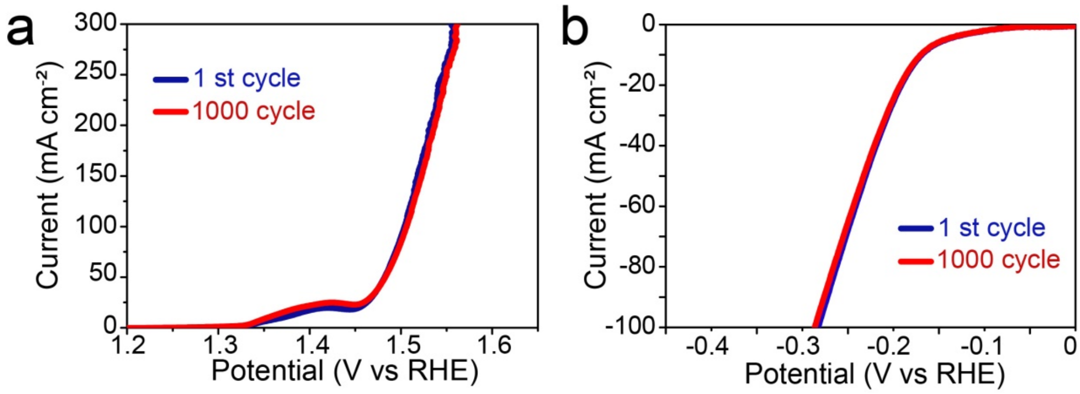

Figure S12. LSV curve comparison for (a) OER and (b) HER cyclic reactions.

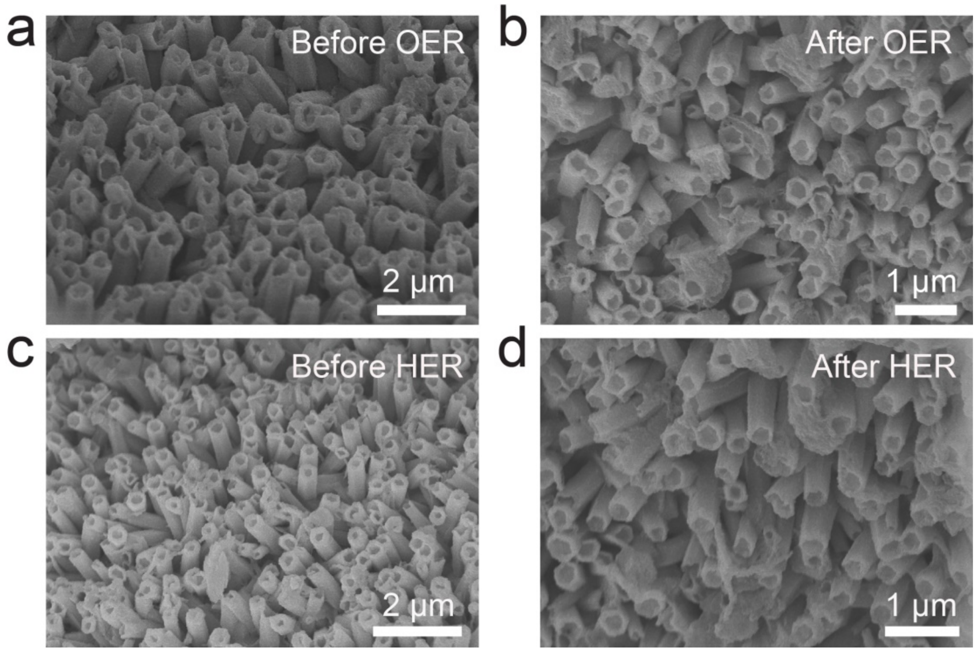

Figure S13. SEM morphology comparison before and after (a-b) OER and (c-d) HER cyclic reactions.

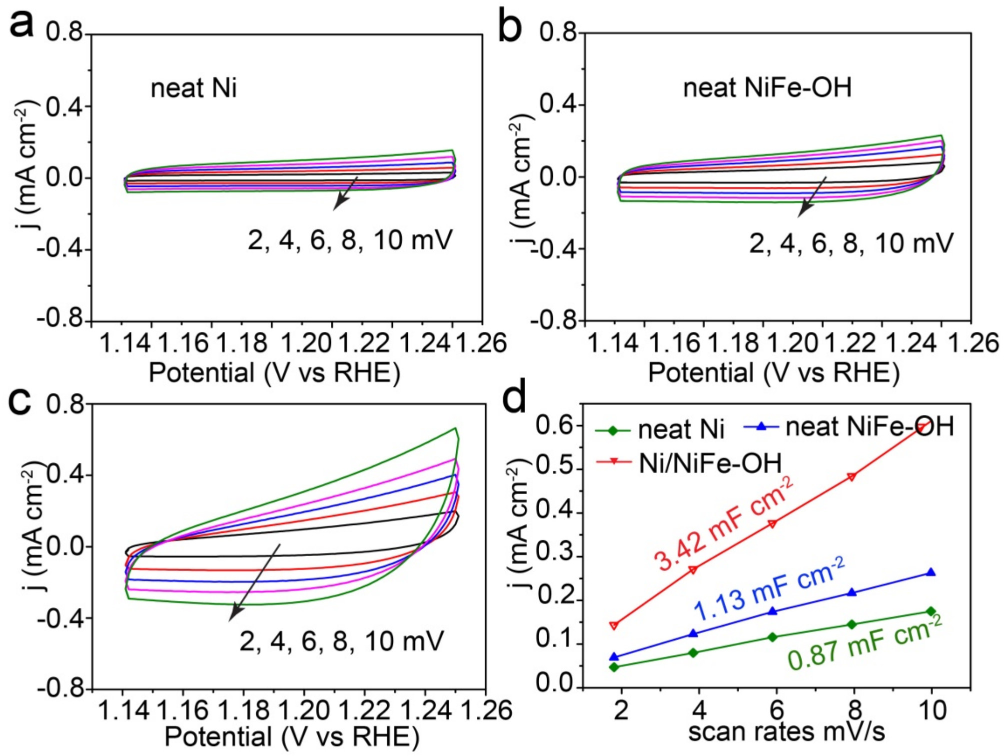

Figure S14. (a-c) Cyclic voltammograms curves measured in a non-Faradaic region (1.14-1.25 V) at varying scan rates, 2, 4, 6, 8 10 mV s^{-1} for the Ni/NiFe heterostructure nanotube, neat Ni, and neat NiFe (oxy)hydroxide. (d) Estimation of electrochemical double-layer capacitances for the samples. The designed Ni/NiFe nanotube manifested the largest value, underlining the advantage of the designed structure.

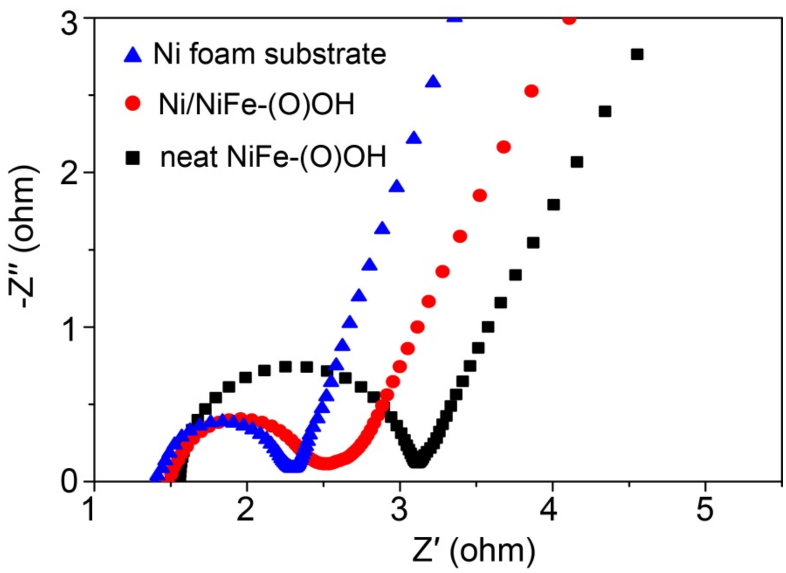

Figure S15. Nyquist plots of the samples, which indicated that the Ni seeds could work as effective electron highways for Ni/NiFe heterostructure, displaying electron conductivity even comparable to Ni foam substrate

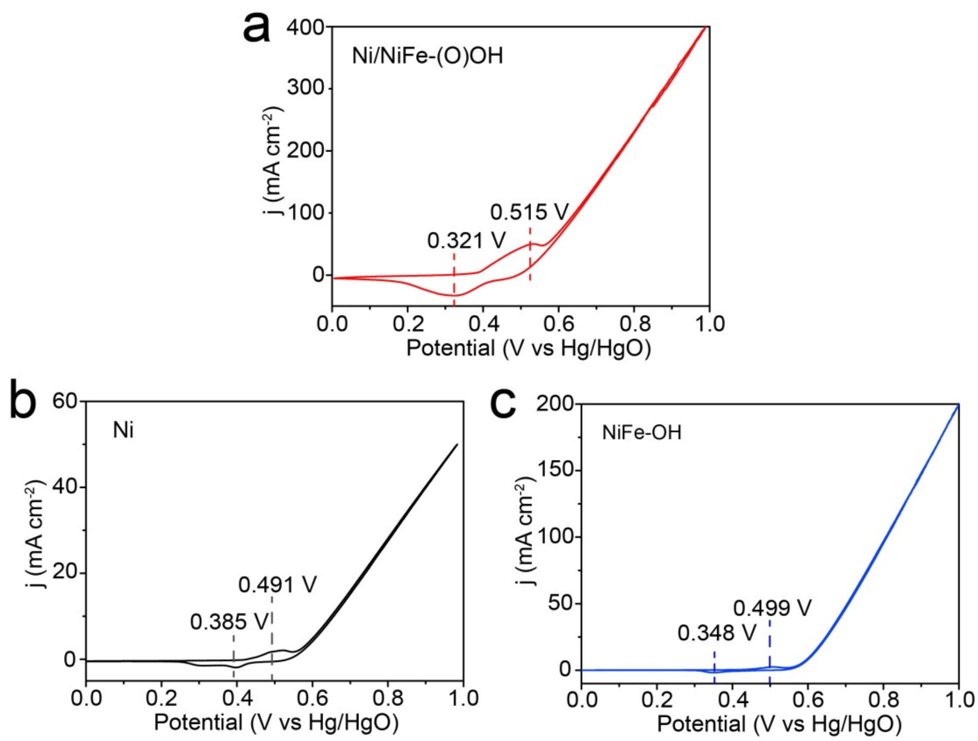

Figure S16. CV curves for the (a) Ni/NiFe (oxy)hydroxide heterostructure compared with (b,c) the individual components.

Table S1. A summarizing table comparing OER performance of our designed electrocatalyst with that of representative documented references.

Catalyst	Electrolyte	J (mA cm^{-2})	Overpotential (mV)	Tafel slope (mV dec^{-1})
NiFe-Ni/NF	**1 M KOH**	**10** **100**	**205** **265**	**53**
NiFe LDH/NF[1]	1 M NaOH	10	240	/
NiFe/ NF[2]	1 M KOH	10	250	28
NiFe-LDH/graphene oxide[3]	1 M KOH	10	222	40
Ni_3S_2/NF[4]	1 M KOH	10	260	/
FeOOH/Co/FeOOH nanotube arrays[5]	1 M NaOH	91	300	/
CeO_2/FeOOH heterolayered nanotube[6]	1 M NaOH	70.9	300	92.3
$W_{0.5}Co_{0.4}Fe_{0.1}$/NF[7]	1 M KOH	10	250	32
$CoFeO_x$/NF[8]	1 M KOH	10	270	/
NiFe-LDH/$NiCo_2O_4$[9]	1 M KOH	50	290	53
Hierarchical $NiCo_2O_4$ Hollow microcuboids[10]	1 M NaOH	10	290	53
$NiCo_2O_4$ nanowires[11]	1 M KOH	10	460	90
NiFeOH@Ni_2S_3/NF[12]	1 M KOH	10	165	93
N-doped NiFe LDH/NF[13]	1 M KOH	10	230	35
NiFe/NiC_2O_4/NF[14]	1 M KOH	10	＞240	38.8
MoS_2/Ni_3S_2/NF[15]	1 M KOH	10	218	88
$Ni_xFe_{1-x}Se_2$-derived oxide[16]	1 M KOH	10	195	28
Carbon quantum dots/NiFe-LDH[17]	1 M KOH	10	236	35
$NiFeO_x$[18]	1 M KOH	10	310	28
Exfoilated NiFe LDH[19]	1 M KOH	10	302	40
Fe,O-doped Co_2P[20]	1 M KOH	10	274.5	51.7
3D graphene network/CoAl-LDH[21]	1 M KOH	10	301	36
$Ni_{0.5}Co_{0.5}O_x$[22]	1 M KOH	10	355	37
NiCo-LDH[23]	1 M KOH	10	367	40
Co-Bi nanosheet /graphene[24]	1 M KOH	10	290	53

$Au@Co_3O_4$[25]	1 M KOH	10	378	60
Ni-Co oxides layers[26]	1 M NaOH	10	325	39
$Co_3O_4/NiCo_2O_4$[27]	1 M KOH	10	340	88
IrO_x[22]	1 M KOH	10	370	50
IrO_2[28]	1 M KOH	10	330	52
RuO_2[29]	0.1 M KOH	10	387	90

Table S2. A summarizing table comparing HER performance of our designed electrocatalyst with that of representative documented references.

Catalyst	Electrolyte	J (mA cm^{-2})	Overpotential (mV)	Tafel slope (mV dec^{-1})
NiFe-Ni/NF	**1 M KOH**	**10** **100**	**154** **297**	**71**
Hierarchical $NiCo_2O_4$ Hollow microcuboids[10]	1 M NaOH	10	110	49.7
MoS_2/Ni_3S_2/NF[15]	1 M KOH	10	110	83.1
NiFe-LDH/$NiCo_2O_4$/NF[9]	1 M KOH	10	192	59
NiS_2/NF[4]	1 M KOH	10	223	/
CoP nanowire /carbon cloth[30]	1 M KOH	10	209	51
Ni wire[31]	1 M KOH	10	350	/
CoS film/fluorine-doped tinoxide[32]	1 M KOH	10	480	93
CoOx@N-doped carbon[33]	1 M KOH	10	232	115
N, S-doped/CNT[34]	1 M KOH	5	400	56
A- $PrBa_{0.8}Ca_{0.2}Co_{1.5}Fe_{0.5}O_{5+\delta}$-H[35]	1 M KOH	20	224	42
$Pr_{0.5}(Ba_{0.5}Sr_{0.5})_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$[36]	1 M KOH	20	250	45
Porous N-rich carbon/Co nanopatricles[37]	1 M KOH	10	298	131
NiFe LDH/NF[1]	1 M NaOH	10	210	/
MoB nanoparticles[38]	1 M KOH	10	212	140-210
Cobalt-embedded nitrogen-rich carbon nanotubes[39]	1 M KOH	10	380	80
$NiSe_2$ nanosheets[40]	1 M KOH	10	184	37
NiFeLDH-NS@DG_{10}[41]	1 M KOH	10	300	110
$NiCo_2S_4$/carbon cloth[42]	1 M KOH	10	263	141
NiCoP 300[43]	1 M KOH	10	150	60.6

Table S3. A summarizing table comparing overall water splitting performance of our designed electrocatalyst with that of representative documented references.

Catalyst	Electrolyte	J (mA cm^{-2})	Potential (V)	Electrode
NiFe-Ni/NF	**1 M KOH**	**5** **10**	**1.5** **1.56**	**Ni foam**
Porous N-rich carbon/Co nanopatricles[37]	1 M KOH	10	1.64	Ni foam
Co-P film[44]	1 M KOH	10	1.65	Cu foil
Ni_2P[45]	1 M KOH	10	1.63	Ni foam
CoOx@N-doped carbon[33]	1 M KOH	20	1.55	Ni foam
NiFe LDH/NF[1]	1 M KOH	10	1.7	Ni foam
NiSe/NF[46]	1 M KOH	10	1.63	Ni foam
NiFe-LDH/$NiCo_2O_4$/NF[9]	1 M KOH	10	1.6	Ni foam
NiFe/NiC_2O_4/NF[14]	1 M KOH	10	1.67	Ni foam
Ni_3S_2/NF[4]	1 M KOH	10	1.76	Ni foam
Ni/NiP[47]	1 M KOH	10	1.61	Ni foam
Porous MoO_2 Nanosheets[48]	1 M KOH	10	1.53	Ni foam
$NiCo_2O_4$ nanowires Array[42]	1 M KOH	20	>1.9	Carbon cloth
EG/$Co_{0.85}Se$/NiFe-LDH[49]	1 M KOH	10	1.67	Graphite foil
CoOx-CoSe films[50]	1 M KOH	20	1.66	Ni foam
Nanoporous CoFe phosphides[51]	1 M KOH	10	1.53	/

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