Supporting Information

Hierarchical Ni_2P@NiFeAlO_x Nanosheet Arrays as Bifunctional Catalyst for Superior Overall Water Splitting

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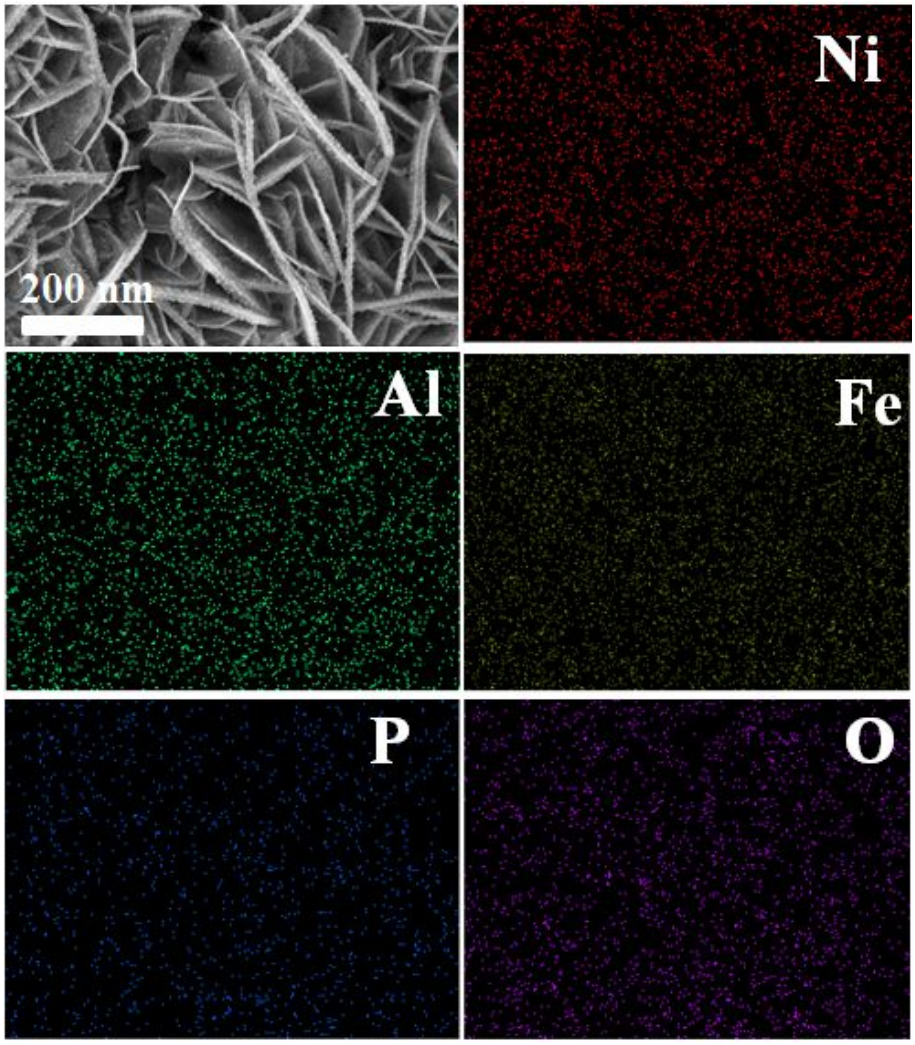

Figure S1 SEM image for Ni_2P@$NiFeAlO_x$ with elemental EDS mapping images.

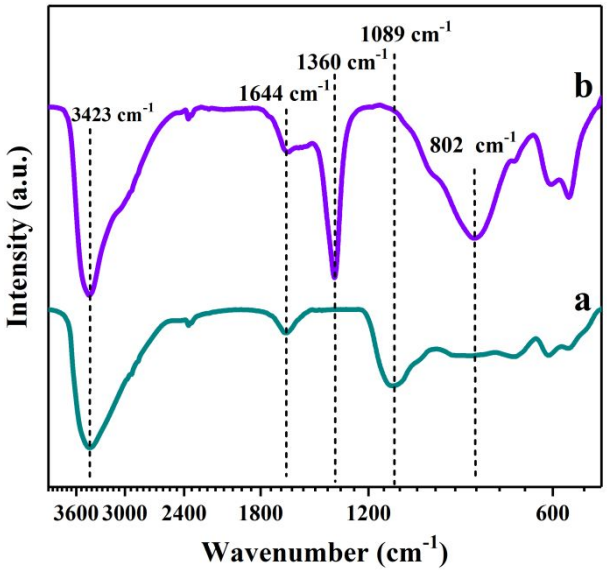

Figure S2 FT-IR spectra of NiFeAl-PO_4^{3-}-LDH (a) and NiFeAl-CO_3^{2-}-LDH (b).

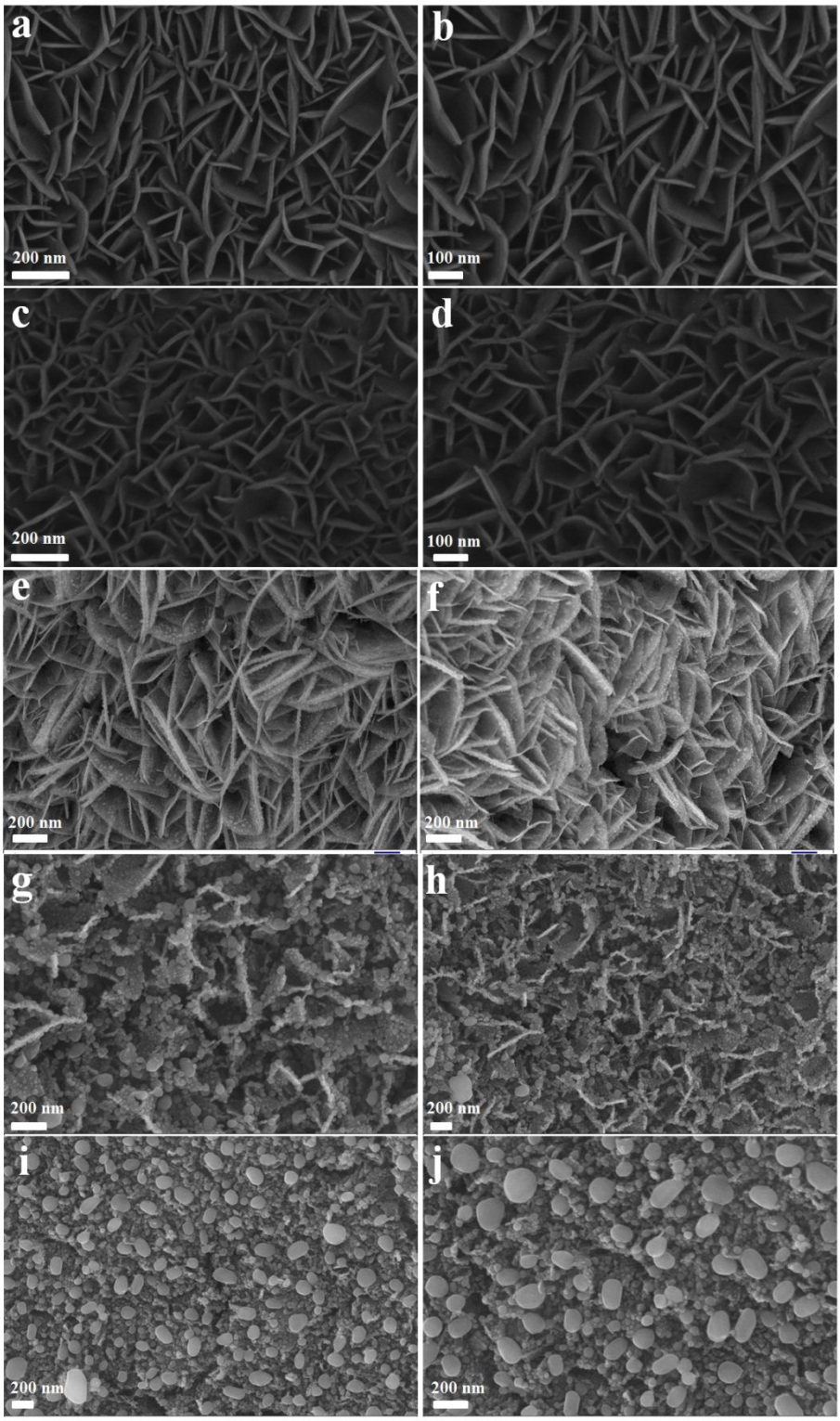

Figure S3 SEM images of Ni_2P@$NiFeAlO_x$ samples prepared at different reduction temperature of 300 (a,b), 400 (c,d), 500 (e,f), 600 (g,h) and 700 ℃ (i,j).

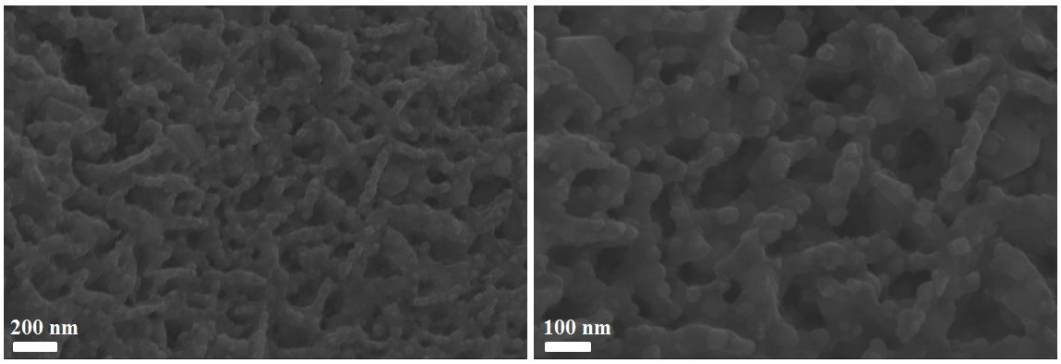

Figure S4 SEM images of $Ni_2P@NiAlO_x$ samples reduced at the temperature of 500 ℃.

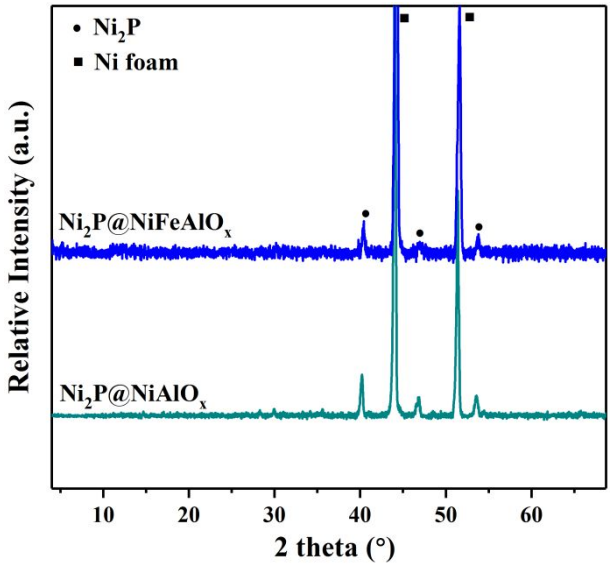

Figure S5 XRD patterns of $Ni_2P@NiAlO_x$ and $Ni_2P@NiFeAlO_x$.

Table S1 The actual atomic ratios of Ni, Fe and P in different catalysts determined by ICP-AES.

Catalysts	molar ratio of Ni/Fe/Al	
	feeding ratio	actual ratio
$Ni_2P@NiFeAlO_x$	3:1:1	2.72:0.63:1
$Ni_2P@NiAlO_x$	3:0:1	2.65:0:1
NiFeAl-PO_4^{3-}-LDH	3:1:1	2.88:0.74:1

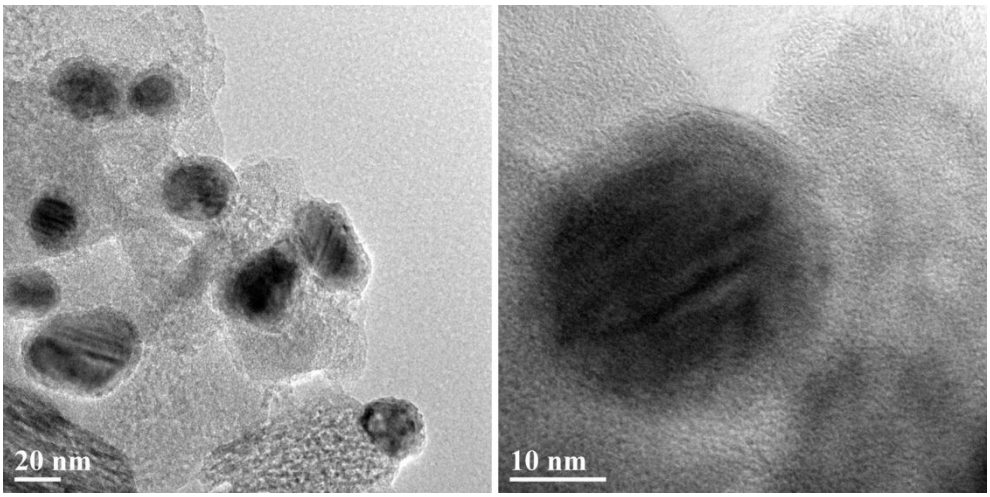

Figure S6 HRTEM images of Ni_2P@$NiFeO_x$.

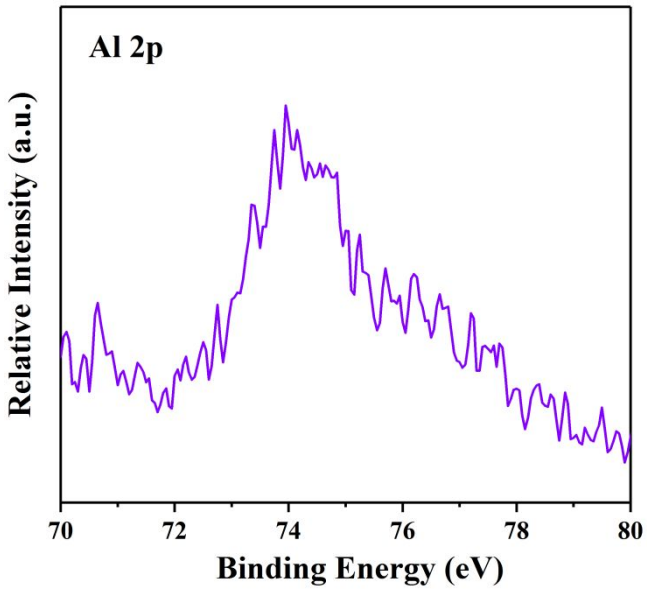

Figure S7 XPS spectra in the Al 2p region for Ni_2P@$NiFeAlO_x$ sample.

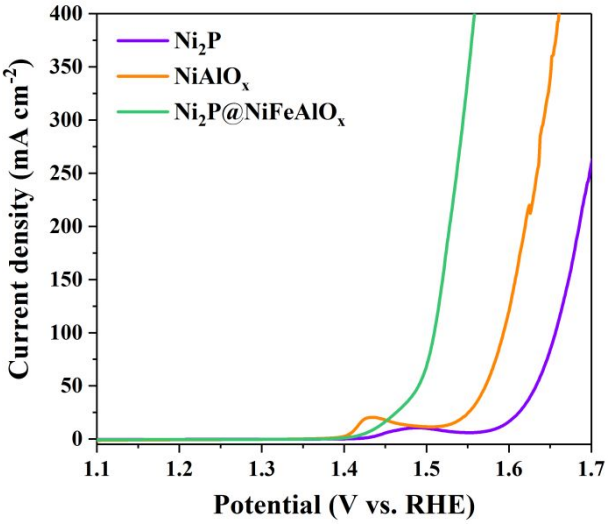

Figure S8 OER polarization curves of Ni_2P, $NiFeAlO_x$ and Ni_2P@$NiFeAlO_x$

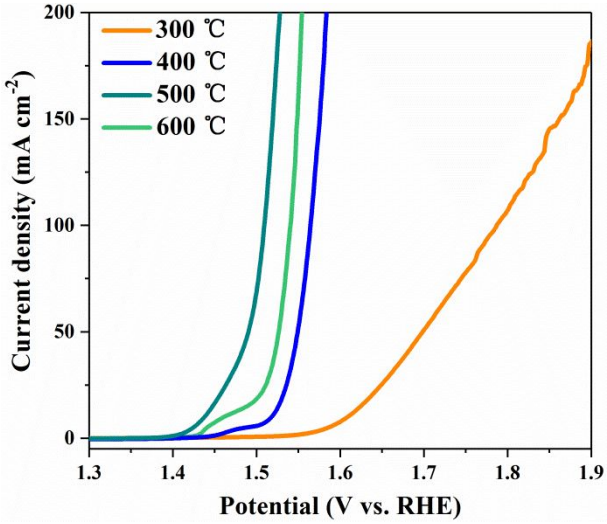

Figure S9 OER polarization curves of NiFeAl-PO_4^{3-}-LDH reduced at different temperatures.

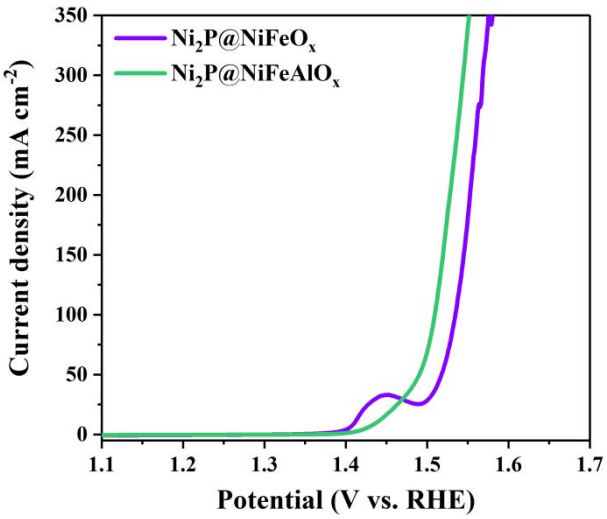

Figure S10 OER polarization curves of Ni_2P@$NiFeAlO_x$ and Ni_2P@$NiFeO_x$.

Table S2 Summary of different catalysts for OER.

Electrocatalysts	Current density ($mA\ cm^{-2}$)	Overpotential (mV)	Tafel slop ($mV\ dec^{-1}$)	Reference
Ni_2P@$NiFeAlO_x$	10	210	54	This work
IrO_2	10	325	155	This work
$NiFeO_x$	10	230	31.5	Nat. Commun. 2015, 6, 7261.
NiCoP@C	10	330	96	Angew. Chem. Int. Ed. 2017, 56, 3897.
NiFe LDH@NiCoP/NF	10	220	48.6	Adv. Funct. Mater. 2018, 1706847
$(Ni_{0.33}Fe_{0.67})_2P$	50	230	55.9	Adv. Funct. Mater. 2017,

				1702513.
NiCoP/NF	10	280	87	Nano Lett. 2016, 16, 7718.
Ni_2P nanoparticle	10	290	47	Energy Environ. Sci. 2015, 8, 2347.
NiFe-LDH@ DG10	10	210	110	Adv. Mater. 2017, 29, 1700017.
FeNi-rGO LDH	10	206	40	Angew. Chem. 2014, 126, 7714.
NiFe-LDH/CNT	10	228	31	J. Am. Chem. Soc. 2013, 135, 8452.
CoP@rGO	10	280	75	J. Am. Chem. Soc. 2016, 138, 14686.
$CoFe_2O_4$/C NRAs	10	240	45	Adv. Mater. 2017, 29, 1604437.

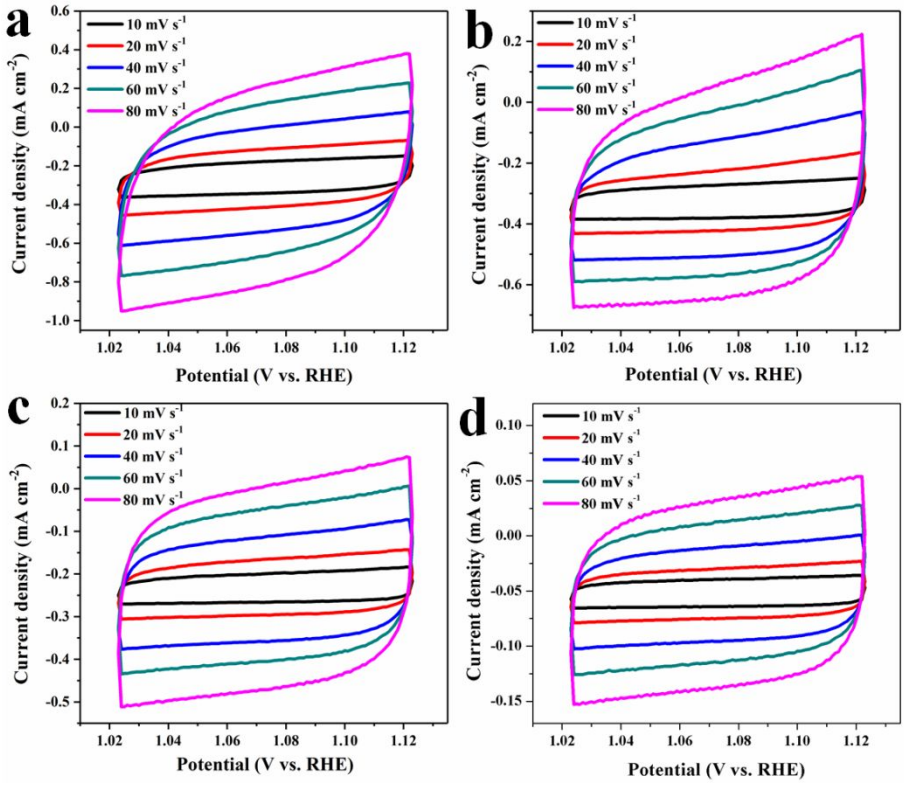

Figure S11 CV plots of (a) Ni_2P@$NiFeAlO_x$, (b) Ni_2P@$NiAlO_x$, (c) NiFeAl-PO_4^{3-}-LDH and (d) Ni foam tested at various scan rates from 10 to 80 mV s^{-1}.

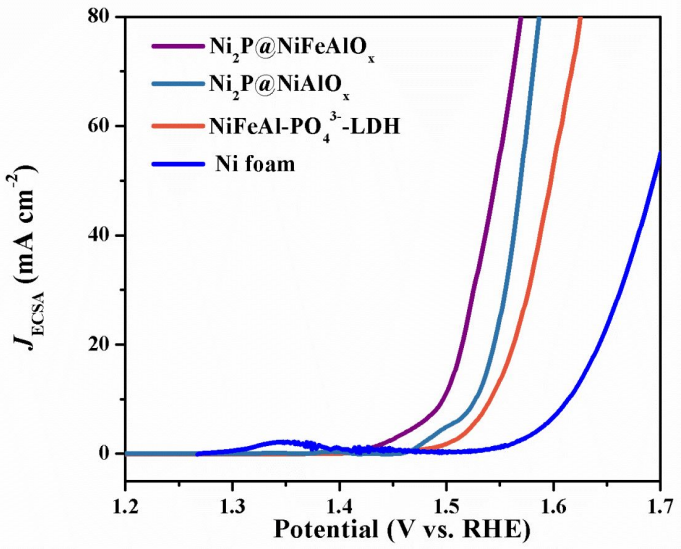

Figure S12 ECSA normalized polarization curves in different samples.

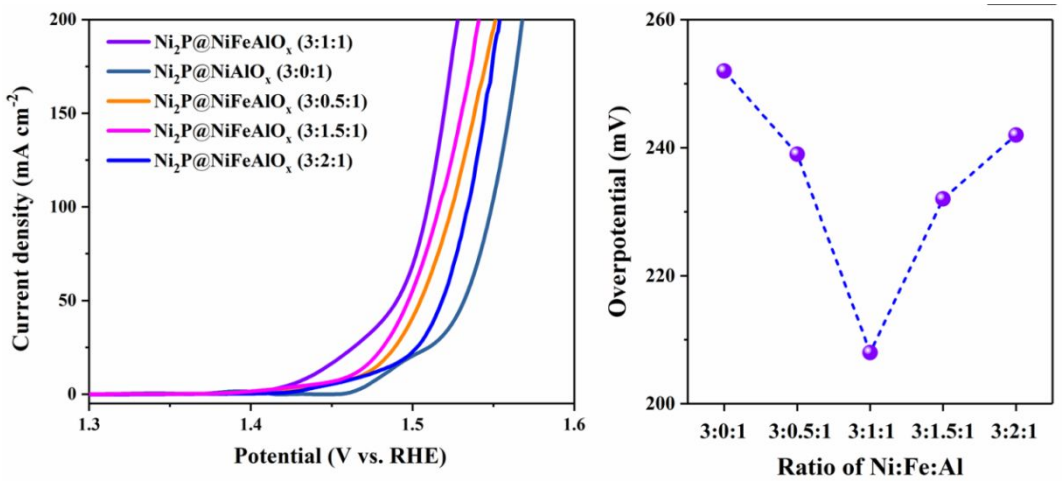

Figure S13 OER polarization curves of Ni_2P@NiFeAlO$_x$(3:n:1) samples with different Ni/Fe/Al molar ratios (n = 0, 0.5, 1, 1.5, 2) and overpotentials required to reach 10 mA cm^{-2} *versus* Ni/Fe/Al ratios.

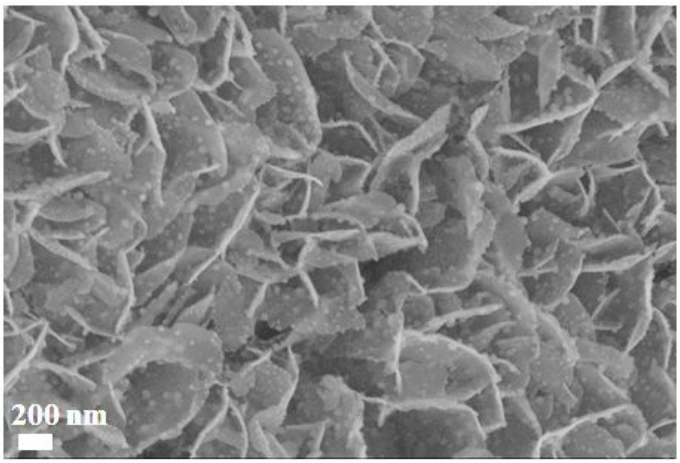

Figure S14 SEM image of Ni_2P@$NiFeAlO_x$ after the stability test of OER.

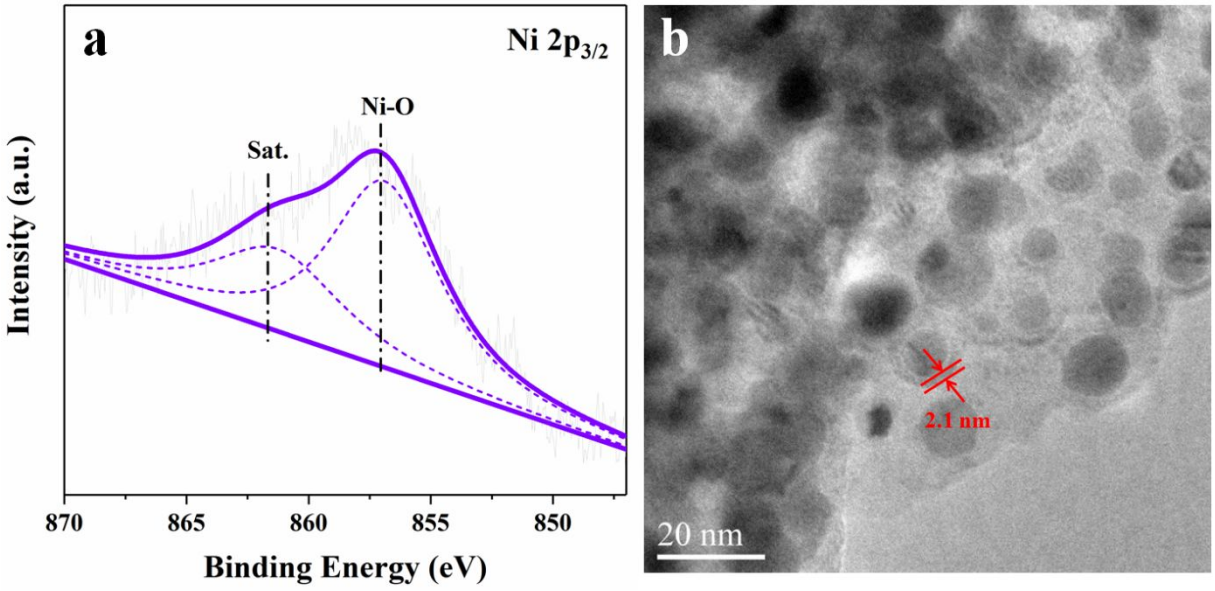

Figure S15 XPS spectra in Ni $2p_{3/2}$ region and TEM image for Ni_2P@$NiFeAlO_x$ after the stability test of OER for 24 h.

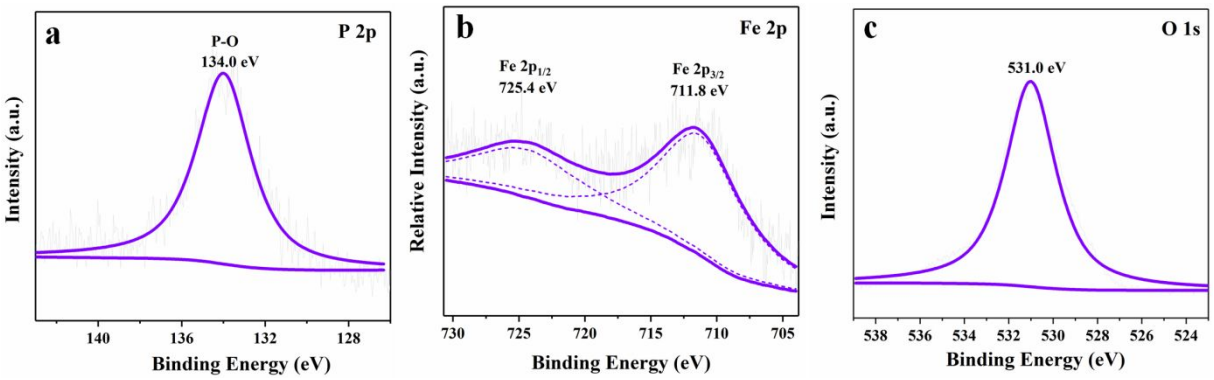

Figure S16 XPS spectra in of Ni_2P@$NiFeAlO_x$ after the stability test of OER for 24 h.

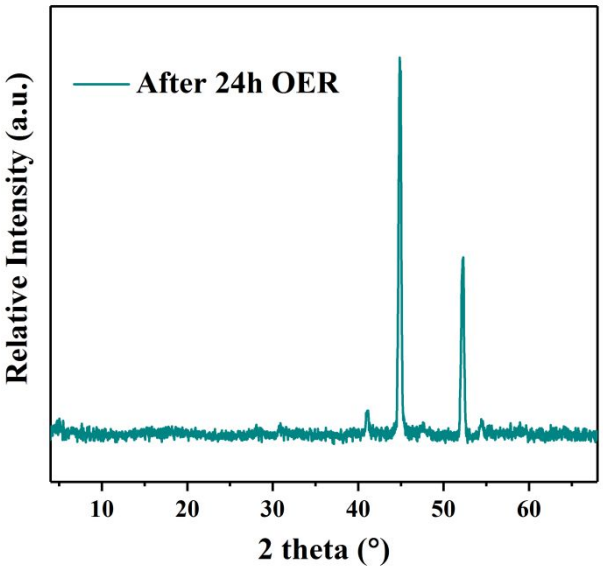

Figure S17 XRD pattern of Ni_2P@$NiFeAlO_x$ after the stability test of OER

Table S3 Summary of different catalyst for HER.

Electrocatalysts	Current density (mA cm^{-2})	Overpotential (mV)	Tafel slop (mV/dec)	Reference
Ni_2P@$NiFeAlO_x$	10	105	106	This work
Pt/C	10	54	58	This work
NiFe LDH/$NiCo_2O_4$/NF	10	192	59	ACS Appl. Mater. Interfaces 2017, 9, 1488.
Ni_5P_4/Ni foil	10	150	53	Angew. Chem. Int. Ed. 2015, 54, 12361.
NiFe/$NiCo_2O_4$/NF	10	105	88	Adv. Funct. Mater. 2016, 26, 3515.
NiFe LDH/NF	10	210	58.9	Science 2014, 345, 1593.
Ni_2P/Ni/NF	10	98	72	ACS Catal. 2016, 6, 714-721

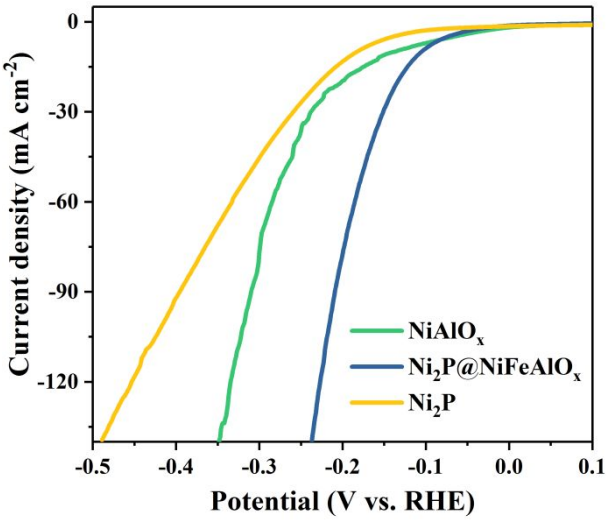

Figure S18 HER polarization curves of Ni_2P@$NiFeAlO_x$, Ni_2P and $NiFeAlO_x$.

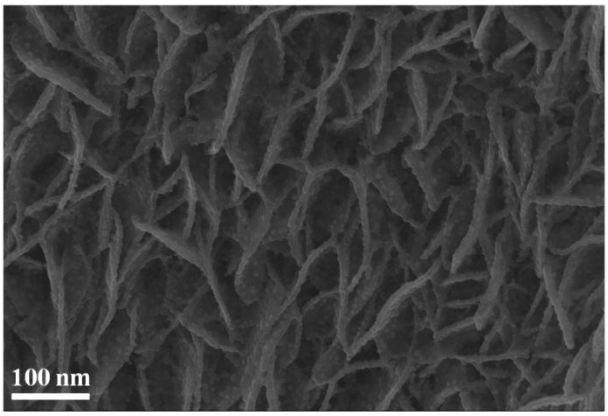

Figure S19 SEM image Ni_2P@$NiFeAlO_x$ after the stability test of HER.

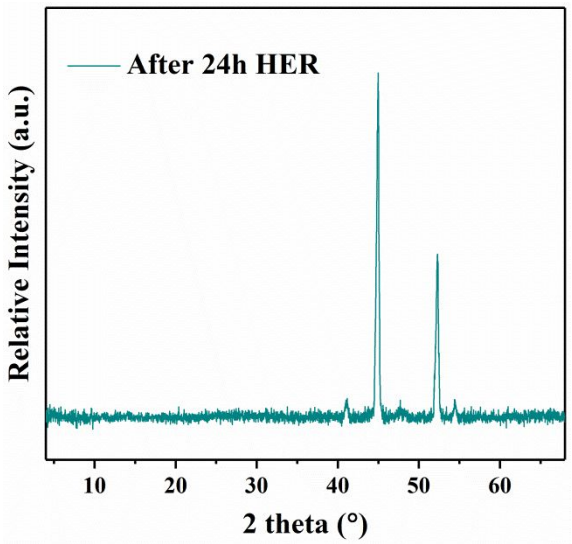

Figure S20 XRD pattern of Ni_2P@$NiFeAlO_x$ after the stability test of HER

Table S4 Comparison of cell voltage to reach 10 mA cm^{-2} for overall water splitting of Ni_2P@$NiFeAlO_x$ and some other reported bifunctional catalysts in alkaline electrolyte.

Electrocatalysts	Electrolyte (KOH)	Voltage (V)	Reference
Ni_2P@$NiFeAlO_x$	1 M	1.52	This work
IrO_2 // Pt/C	1 M	1.55	This work
Ni_2P nanoparticle	1 M	1.63	Energy Environ. Sci. 2015, 8, 2347.
NiCoP/NF	1 M	1.58	Nano Lett. 2016, 16, 7718.
$(Ni_{0.33}Fe_{0.67})_2P$	1 M	1.49	Adv. Funct. Mater. 2017, 1702513
Ni_5P_4/NF	1 M	1.70	Angew. Chem. Int. Ed. 2015, 54, 12361.
NiP/NF	1 M	1.61	Adv. Funct. Mater. 2016, 26, 3314.
NiFe LDH@NiCoP/NF	1 M	1.57	Adv. Funct. Mater. 2018, 1706847
NiFe LDH/$NiCo_2O_4$/NF	1 M	1.60	ACS Appl. Mater. Interfaces 2017, 9, 1488.
NiFe LDHNS@DG10	1 M	1.5 @20 mA cm^{-2}	Adv. Mater. 2017, 29, 1700017.
NiFe LDH@$NiCo_2S_4$/NF	1 M	1.60	ACS Appl. Mater. Interfaces 2017, 9, 15364.
CoP@a-CoO_x	1 M	1.66	Adv. Sci. 2018, 5, 1800514
Fe doped CoP	1 M	1.60	Nano Energy, 2017, 41, 583.
$Ni_{0.9}Fe_{0.1}$/nanocarbon	1 M	1.58	ACS Catal. 2016, 6, 580.
NiSe NWs	1 M	1.63	Angew. Chem. Int. Ed. 2015, 127 9483.
Ni@NC-800	1 M	1.60	Adv. Mater. 2017, 29, 1605957.
$NiCo_2O_4$ hollow microcuboids	1 M	1.65	Angew. Chem. Int. Ed. 2016, 55, 6290.
MoS_2/Ni_3S_2	1 M	1.56	Angew. Chem. Int. Ed. 2016, 128, 6814.
CoO_x@CN	1 M	1.55	J. Am. Chem. Soc. 2015, 137, 2688–2694.
Porous MoO_2 Nanosheets	1 M	1.53	Angew. Chem. Int. Ed. 2015, 127, 9483.