Supporting Information

P-Doped Nickel Sulfide Nanosheet Arrays for Highly Efficient Overall Water Splitting

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Fig. S1 The schematic illustration of the synthesis process for the P-Ni_3S_2 grown on Ni foam.

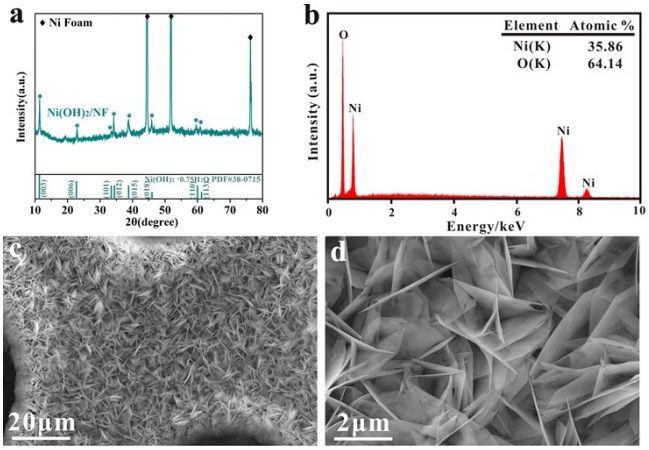

Fig. S2 (a) XRD pattern, (b) EDX spectrum and (c, d) SEM images of $Ni(OH)_2$/NF.

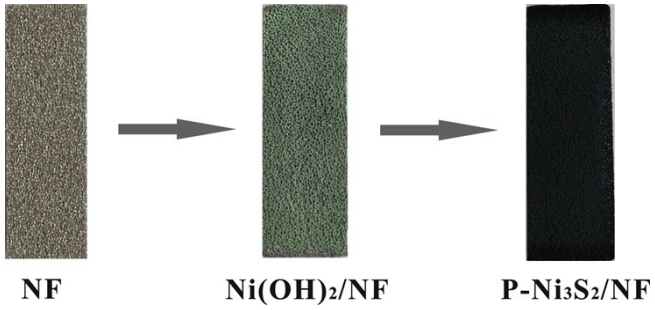

Fig. S3 Photographic images of Ni foam (NF), $Ni(OH)_2$/NF and P-Ni_3S_2/NF.

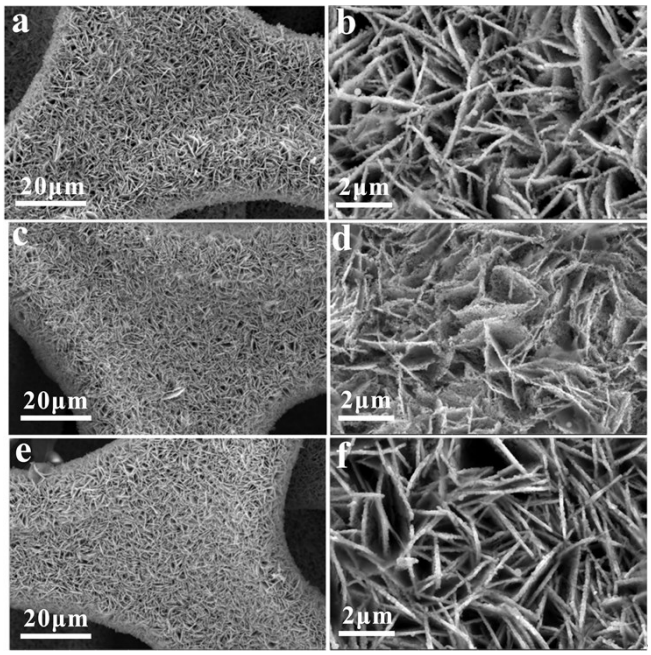

Fig. S4 SEM image of (a-b) $P_{3.0\%}$-Ni_3S_2/NF, (c-d) $P_{22.1\%}$-Ni_3S_2/NF and (e-f) Ni_3S_2/NF.

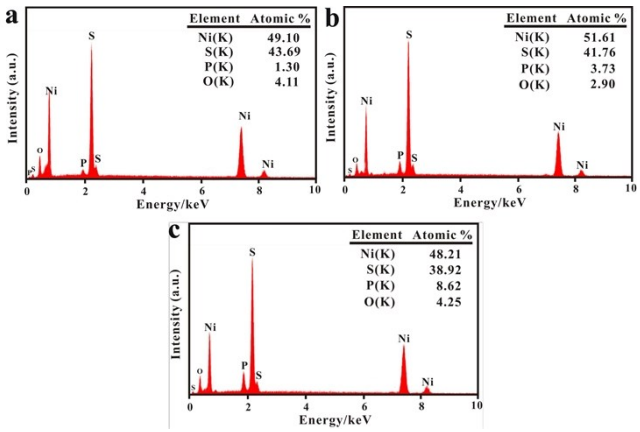

Fig. S5 EDX elemental mapping of (a) $P_{3.0\%}$-Ni_3S_2/NF, (b) $P_{8.9\%}$-Ni_3S_2/NF and (c) $P_{22.1\%}$-Ni_3S_2/NF.

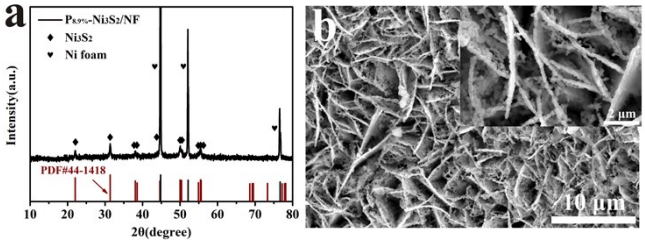

Fig. S6 (a) XRD pattern and (b) SEM of $P_{8.9\%}$-Ni_3S_2/NF after HER.

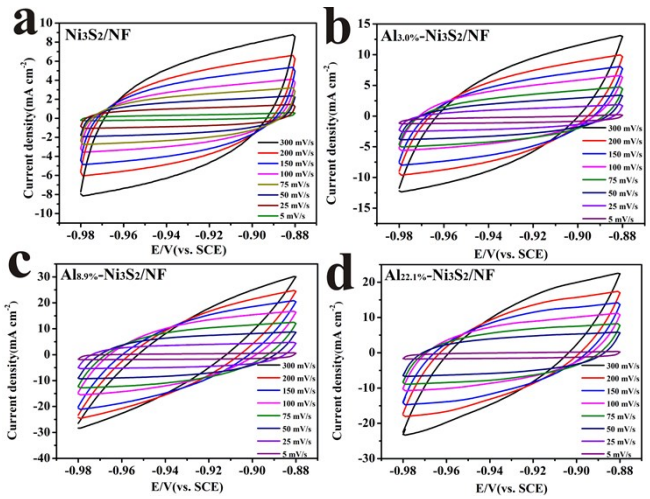

Fig. S7 Typical cyclic voltammetry curves of (a) Ni_3S_2/NF, (b) $P_{3.0\%}$-Ni_3S_2/NF, (c) $P_{8.9\%}$-Ni_3S_2/NF and (d) $P_{22.1\%}$-Ni_3S_2/NF with different scan rates in 1M KOH.

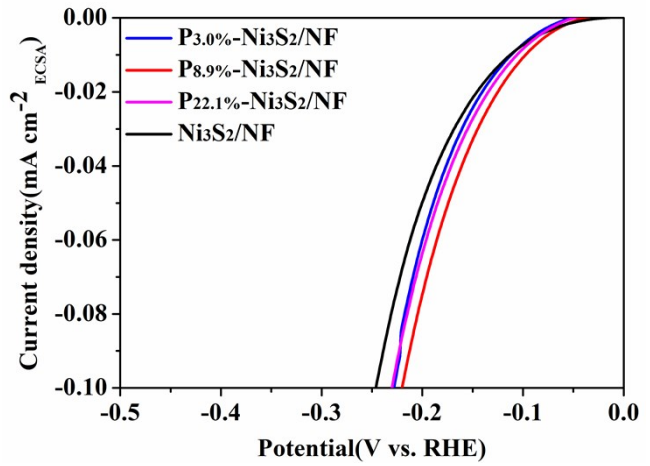

Fig. S8 Polarization curves from normalized to the electrochemical active surface area (ECSA) for HER.

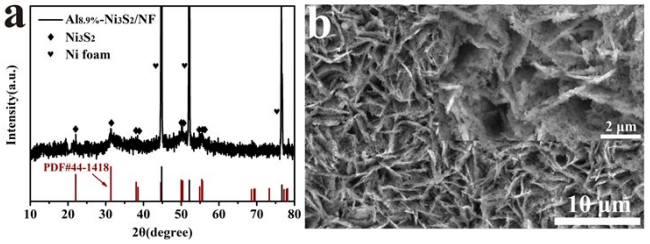

Fig. S9 D(a) XRD pattern and (b) SEM of $P_{8.9\%}$-Ni_3S_2/NF after OER.

Table S1. S/P Feed Ratio and Atomic Ratio of the synthesized $P_{X\%}$-Ni_3S_2/NF.

Sample	Precursor ratio (S: P) (mmol : mmol)	S: P atomic ratio (determined by EDS)
$P_{3.0\%}$-Ni_3S_2	34.17 : 0.342 = 100 : 1	39.32 : 1.65 = 23.83 : 1
$P_{8.9\%}$-Ni_3S_2	34.17 : 1.026 = 33 : 1	37.94 : 3.73 = 10.17 : 1
$P_{22.1\%}$-Ni_3S_2	34.17 : 1.710 = 20 : 1	34.22 : 5.31 = 6.45 : 1

Table S2. The chemical composition of P-Ni_3S_2 and Ni_3S_2 measured by XPS.

Samples	Atomic Percentage (At %)				P Doping concentration (At %)
	Ni	P	S	O	$\frac{n(P)}{n(P)+n(S)}$
Ni_3S_2	15.75	--	10.80	28.07	--
$P_{3.0\%}$-Ni_3S_2	16.28	0.35	10.62	29.46	3.19
$P_{8.9\%}$-Ni_3S_2	16.14	1.02	10.44	30.54	8.90
$P_{22.1\%}$-Ni_3S_2	16.07	2.90	10.25	31.77	22.05

Table S3. Comparison of the HER performance of $P_{8.9\%}$-Ni_3S_2/NF with other well-performed electrocatalysts.

Catalysts	η@10 mA cm^{-2} mV	η@20 mA cm^{-2} mV	η@100 mA cm^{-2} mV	Tafel slope mV dec^{-1}	Ref.
$P_{8.9\%}$-Ni_3S_2/NF	101	127	224	85	This work
Ni_3S_2/NF	137	159	230	96	[1]
N-Ni_3S_2/NF	110	~160	~230	--	[2]
Sn-Ni_3S_2/NF	137	~200	~320	148	[3]
N-Ni_3S_2@C/NF	113	--	--	90	[4]
$Fe_{11.1\%}$- Ni_3S_2/NF	126	--	--	89	[5]
$Fe_{17.5\%}$- Ni_3S_2/NF	47	142	232	95	[6]

$Ni_{1.5}Fe_{0.5}P$/CF	282	--	--	125	[7]
H-$Co_{0.85}Se$|P	150	180	--	83	[8]
Ni_2P-$NiSe_2$/CC	66	--	--	72.6	[9]
$Ni_xCo_{3-x}S_4$/Ni_3S_2/NF	136	--	258	107	[10]
NiFe LDH@NiCoP/NF	120	--	--	88.2	[11]
$P_{9.03\%}$-(Ni, Fe)$_3S_2$ /NF	98	126	218	88	[12]

Table S4. EIS and C_{dl} results of $P_{X\%}$-Ni_3S_2/NF, Ni_3S_2/NF. (x=3.0, 8.9 and 22.1)

Sample	Ni_3S_2	$P_{4.2\%}$-(Ni, Fe)$_3S_2$/NF	$P_{8.9\%}$-(Ni, Fe)$_3S_2$/NF	$P_{15.5\%}$-(Ni, Fe)$_3S_2$/NF
Rct [Ω]	4.05	2.03	1.83	2.30
C_{dl} [mF cm^{-2}]	18.4	36.8	42.6	32.5

Table S5. Comparison of the OER performance of $P_{8.9\%}$-Ni_3S_2/NF with other well-performed electrocatalysts.

Catalysts	η@10 mA cm^{-2} mV	η@20 mA cm^{-2} mV	η@100 mA cm^{-2} mV	Tafel slope mV dec^{-1}	Ref.
$P_{8.9\%}$-Ni_3S_2/NF	256	267	330	30	This work
N-Ni_3S_2/NF	--	--	330	79	[2]
Fe-Ni_3S_2/FeNi	282	--	--	54	[13]
Ni/NiS	--	~320	~390	109.8	[14]
MoS_2/Ni_3S_2	218	--	~290	88	[15]
NiFe LDH@NiCoP/NF	220	--	--	48.6	[11]
$Ni_{0.36}Fe_{2.64}O_4$/Ni	225	--	--	44	[16]
$P_{9.03\%}$-(Ni, Fe)$_3S_2$ /NF	224	239	277	30	[12]

Table S6. Comparison of the Overall water splitting performance of $P_{8.9\%}$-Ni_3S_2/NF with other well-performed electrocatalysts.

	η@10 mA cm^{-2} mV	η@20 mA cm^{-2} mV	η@100 mA cm^{-2} mV	Ref.
$P_{8.9\%}$-Ni_3S_2/NF	1.63	1.68	1.84	This work
$Fe_{11.1\%}$-Ni_3S_2/NF	1.60	1.66	--	[5]
H-$Co_{0.85}$Se|P	1.64	--	--	[8]
$Ni_{1.5}Fe_{0.5}P$/CF	1.589	1.635	--	[7]
$P_{9.03\%}$-$(Ni, Fe)_3S_2$ /NF	1.54	1.58	1.72	[12]

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