Vanadium-regulated nickel phosphide nanosheets for electrocatalytic sulfion upgrading and hydrogen production

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Experimental section

Chemicals

$Ni(NO)_3·6H_2O$, VCl_3, $CO(NH_2)_2$, NH_4F and $NaH_2PO_2·H_2O$ were bought from Aladdin and $Na_2S·9H_2O$ was supplied Sinopharm group. NaOH and nickel foam (NF) were supplied by Adamas-beta® and Taiyuan source of power company.

Synthesis of V-Ni_2P

In a typical synthesis, 1.6 mmol $Ni(NO_3)_2$, 0.4 mmol VCl_3, 6 mmol NH_4F, 10 mmol $CO(NH_2)_2$ were dissolved in 35 mL water with stirring, which was transferred to autoclave with the NF put into in the solution. The autoclave was and kept at 120 °C for 6 h in an electric oven to prepare Ni-based layered double hydroxide (NiV LDH) precursor. Then, the NiV LDH precursor and $NaH_2PO_2·H_2O$ were placed in furnace and heated up to 300 ℃ for 2h to obtain V-Ni_2P. In contrast, the Ni_2P, V-Ni_2P-1 and V-Ni_2P-2 were synthesized in a similar process without adding VCl_3 and with adding various molar ratios of Ni and V (9:1, 7:3), respectively.

Materials characterization

The crystalline structures, morphology, element distribution and chemical states of the samples were examined on the X-ray diffraction (XRD, Bruker D8), scanning electron microscopy (SEM, FIB-SEM GX4) equipped with energy dispersive spectroscopy, transmission electron microscopy (TEM, FEI Tecnai G20) and X-ray photoelectron spectrometer (XPS, Thermo ESCALAB 250Xi).

Electrochemical measurements

The electrochemical measurements of the samples were carried out on a CHI 760E electrochemical workstation using synthesized samples as working electrodes in a typical three-electrode system to test the electrochemical performances of HER, OER and SOR. The polarization curves were measured at a scan of 2 mV s^{-1} and the electrochemical impedance spectroscopy was performed in the frequency range from 0.01 Hz to 100 kHz.

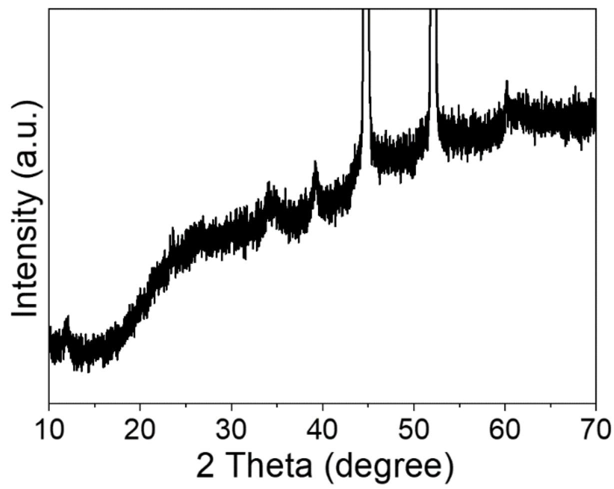

Fig. S1 XRD pattern of NiV LDH precursor.

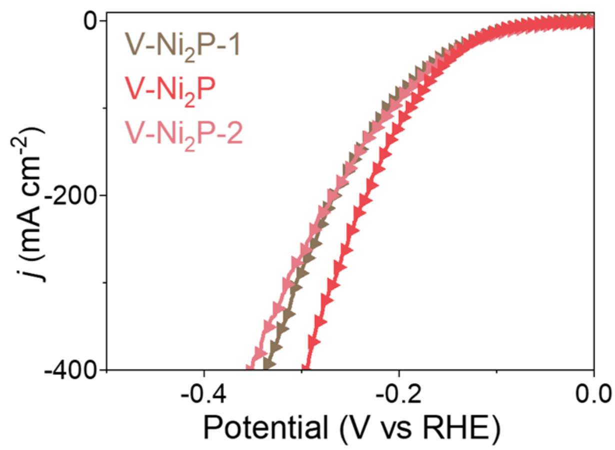

Fig. S2 LSV curves of V-Ni_2P-1, V-Ni_2P and V-Ni_2P-2 in 1.0 M KOH.

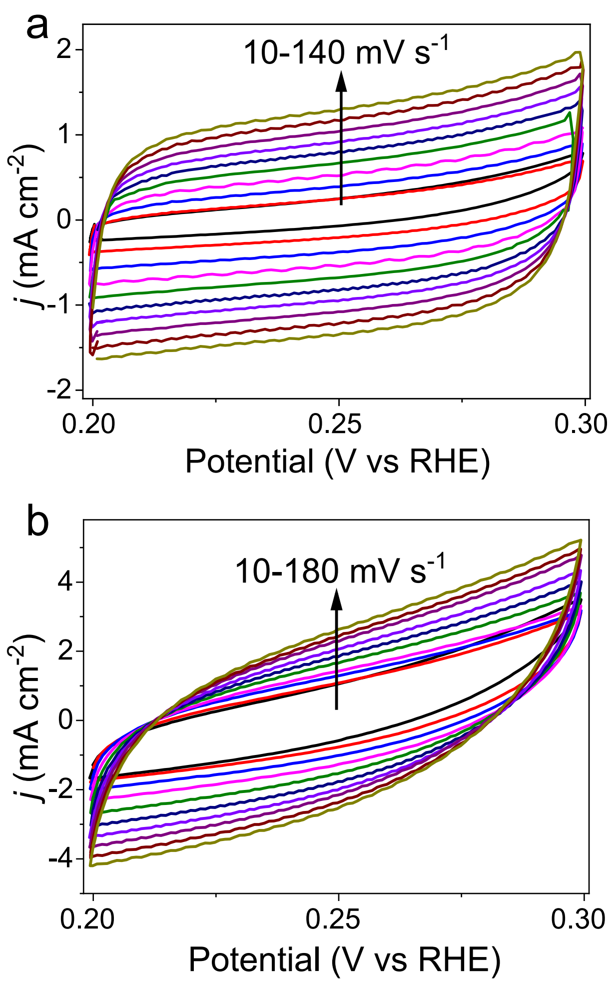

Fig. S3 Cyclic voltammetry curves of (a) Ni_2P and (b) V-Ni_2P.

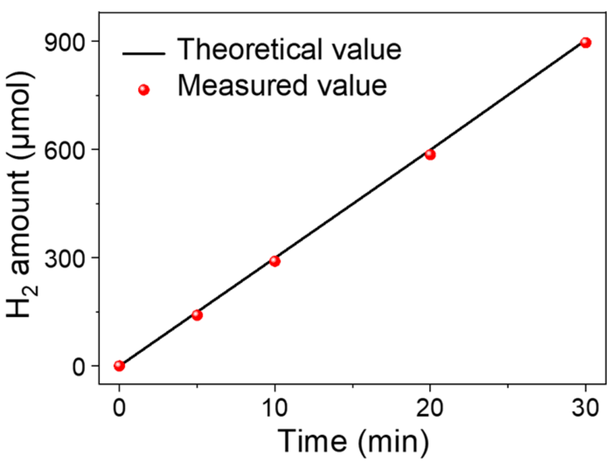

Fig. S4 Experimental and theoretical calculated amount of H_2 over the V-Ni2P.

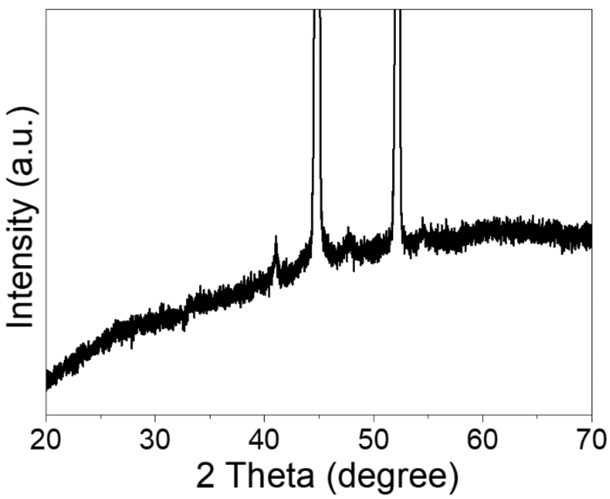

Fig. S5 XRD pattern of V-Ni_2P after HER test.

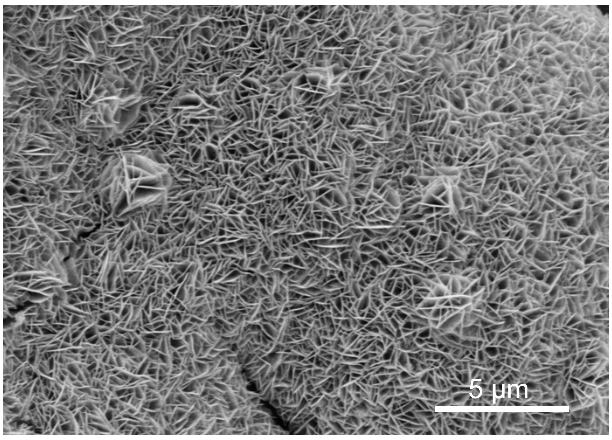

Fig. S6 SEM image of V-Ni_2P after HER test.

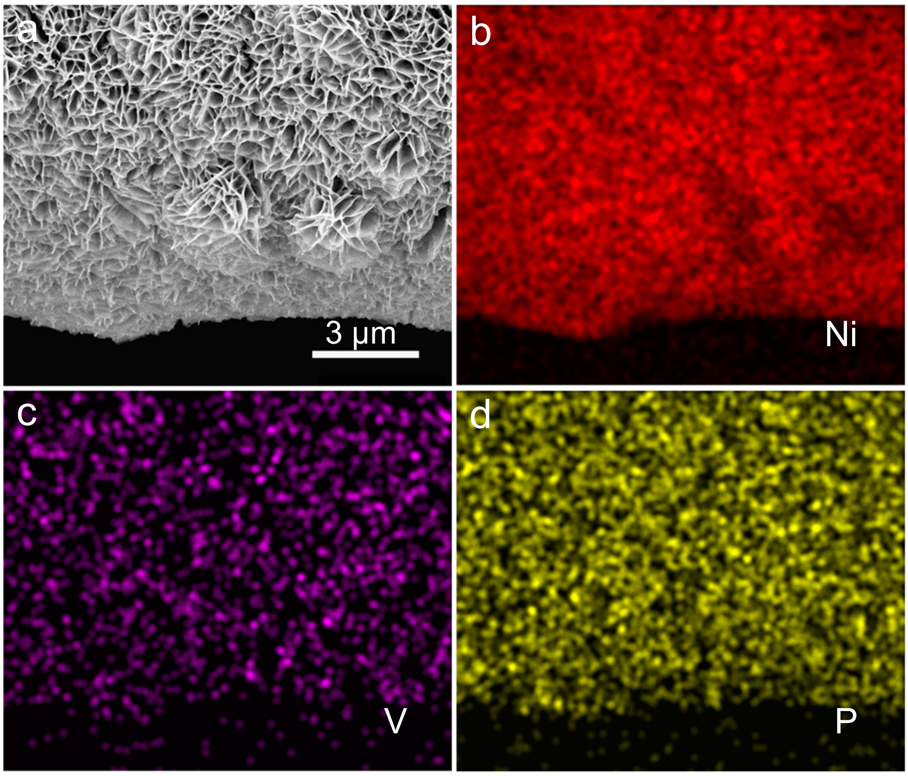

Fig. S7 SEM and corresponding element mapping images of V-Ni_2P after HER test.

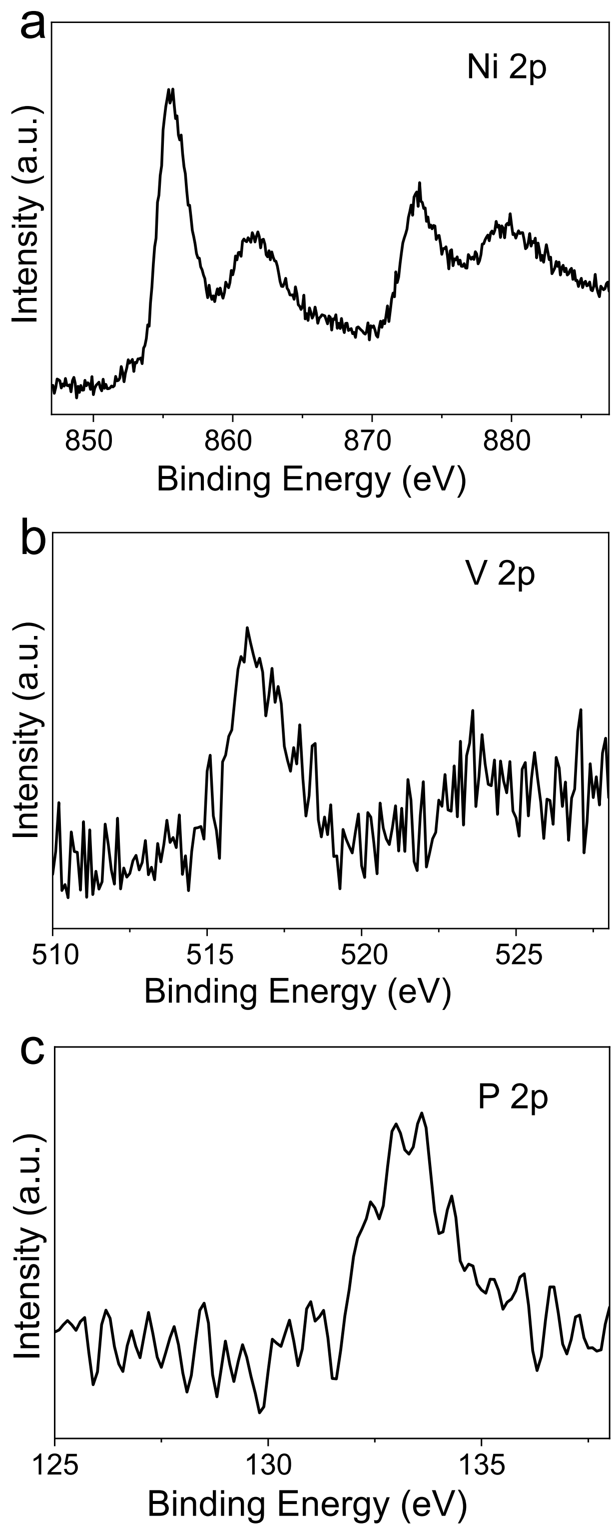

Fig. S8 XPS spectra of (a) Ni 2p, (b) V 2p and (c) P 2p of V-Ni_2P after HER test.

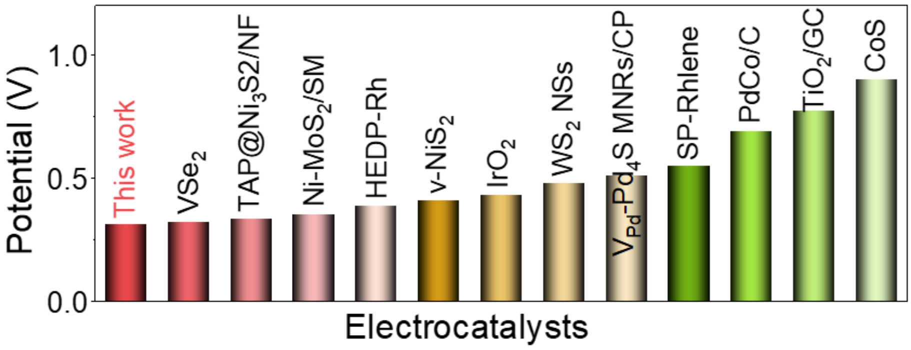

Fig. S9 Potentials comparison of V-Ni_2P with reported catalysts.

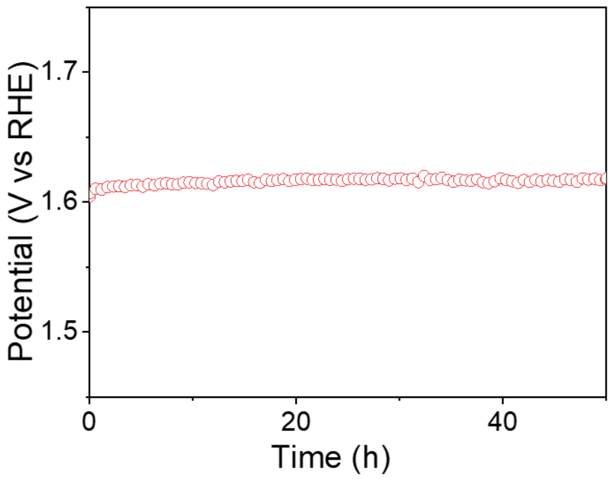

Fig. S10 Durability test of V-Ni_2P for OER.

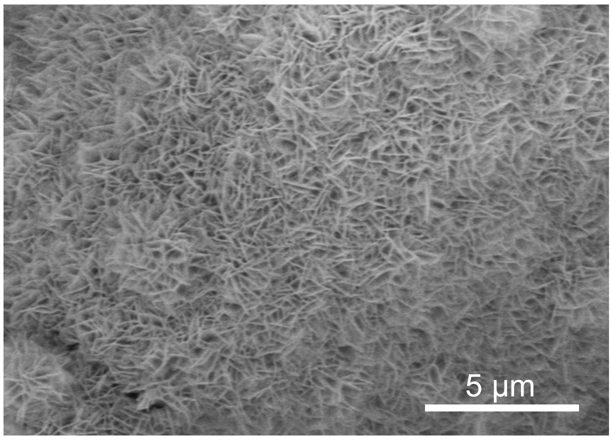

Fig. S11 SEM image of V-Ni_2P after OER test.

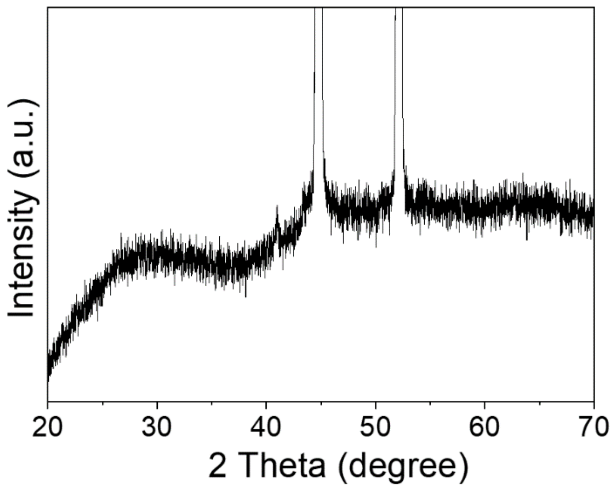

Fig. S12 XRD pattern of V-Ni_2P after OER test.

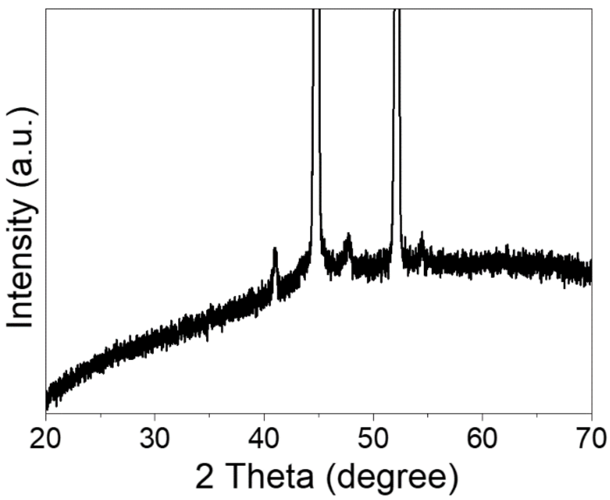

Fig. S13 XRD pattern of V-Ni_2P after SOR test.

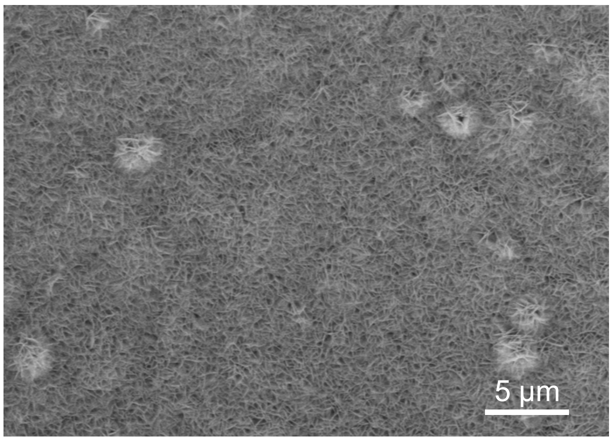

Fig. S14 SEM image of V-Ni_2P after SOR test.

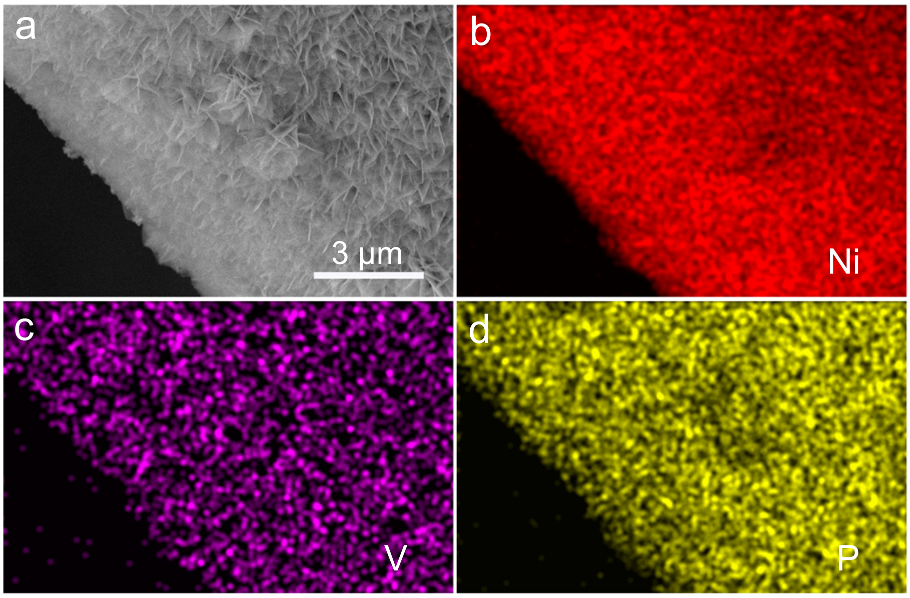

Fig. S15 SEM and corresponding element mapping images of V-Ni_2P after SOR test.

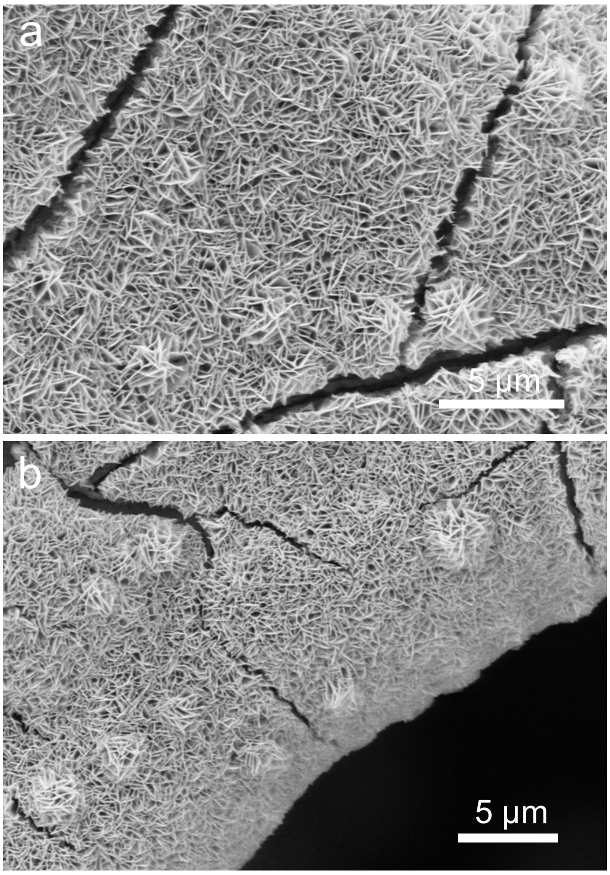

Fig. S16 SEM image of V-Ni_2P at the cathode (a) and at the anode (b) after the HWE test.

Table S1 The comparison of HER performance of V-Ni_2P with reported catalysts.

Catalysts	Overpotential at 10 mA cm^{-2} (mV)	Reference
V-Ni_2P	**93**	**This work**
Ni_2P/CoN–PCP	94	S1
Ni-FeP/C	95	S2
Mo,Fe-$NiCoP_x$/NF	99	S3
Ni-N_3	103	S4
O_3-V_{10}-Ni_2P	108	S5
$Fe(OH)_x$@Cu-MOF	112	S6
NiFeP/SG	115	S7
$Ti_3CN(OH)_x$@MoS_2	120	S8
Al-Ni_2P/TM	129	S9
Cu@Co-CoO/Rh	137.7	S10
MoS_2/CeO_2	147	S11
1T-MoS_2	158	S12
CoP	173	S13
Ni-Mo_2C/NPC	183	S14
MoS_2@$FePS_3$	196	S15
WS_2-NSs	214	S16
Cu-CoFe/Co/NC	217	S17
H-TaS_2	230	S18
MoS_2	248	S19
$Ni_{1.5}Fe_{0.5}P$	282	S20

Table S2 The comparison of SOR performance of V-Ni_2P with reported catalysts.

Catalysts	Potential (V) at 10 mA cm^{-2}	Reference
V-Ni_2P	**0.313**	**This work**
VSe_2	0.32	S21
TAP@Ni_3S_2/NF	0.335	S22
Ni-MoS_2/SM	0.35	S23
HEDP-Rh	0.385	S24
v-NiS_2	0.41	S25
IrO_2	0.43	S26
WS_2 NSs	0.48	S27
V_{Pd}-Pd_4S MNRs/CP	0.511	S28
SP-Rhlene	0.550	S29
PdCo/C	0.69	S30
TiO_2/GC	0.77	S31
CoS	0.9	S32

Table S3 The comparison of HWE performance of V-Ni_2P with reported catalysts.

Catalysts	Voltage at 10 mA cm^{-2} (V)	Reference
V-Ni_2P	**0.389**	**This work**
Cu_2S/NF	0.43	S33
a-RhS_{2-x}	0.442	S34
v-NiS_2	0.49	S25
Ni-MoS_2/SM	0.49	S23
CoS/MoS_2	0.528	S35
HEDP-Rh	0.585	S24
WS_2 NSs	1.17	S27
CoP/NF	1.27	S36

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