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Electronic Supplementary Information

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Meterals:Nickel(II) nitrate hexahydrate (Ni(NO₃)₂·6H₂O), ferric(III) nitrate nonahydrate (Fe(NO₃)₃·9H₂O), urea (CH₄N₂O), ammonium fluo-ride (NH₄F), ethanol (C₂H₆O), N, N-Dimethylformamide (DMF), potassium hydroxide (KOH) and sodium hypophosphite (NaH₂PO₂·H₂O) were purchased from Jinan Camolai Trading Company.NF was purchased from Suzhou Taili New Energy Co., Ltd. Pt/C (20 wt% Pt on Vulcan XC-72R) were purchased from Jinan Jiadong Chemical Co., Ltd. The ul- trapure water (UP H₂O) was obtained using the AD3L-05-030OR UP H₂O instrument. All chemicals were analytically pure without further purificatio.

TOF calculation: The TOF is quantified the concentration of active site and calculated by the following equation:

$$TOF = \frac{jA}{4Fm}$$

Where *j* is current density (A cm⁻²) at defined overpotential; A is the geometric area of the testing electrode; 4 indicates the mole of electrons consumed for evolving one mole O₂ from water; F is the Faradic constant (96485 C mol⁻¹); m is the number of active sites (mol), which can be extracted from the linear relationship between the oxidation peak currents and scan rates by the following equation:

$$slope = \frac{n^2 F^2 m}{4RT}$$

Where n is the numbers of electron transferred; R and T are the ideal gas constant and the absolute temperature, respectively.



Fig. S1 SEM images of NF at different magnifications.



Fig. S2 SEM images of Ni(OH)F/NF at different magnifications.



Fig. S3 SEM images of Ni_2P/NF at different magnifications.



Fig. S4 SEM images of Ni(OH)F/NF treated in different concentrations of ink liquid (a) 0.05 g mL⁻¹ (b) 0.20 g mL⁻¹.



Fig. S5 CV curves of (a) NF, (b) Ni_2P/NF and (c) Ni_2P-FeP_x/NF at different scanning rates.



Fig. S6 Volume of oxygen theoretically calculated and actually collected (illustration: gas collection unit).



Fig. S7 (a) CVs for Ni_2P -FeP_x/NF under different scan rates from 10 to 50 mV s⁻¹ in 1.0 M KOH. (b) Linear relationship of the peak currents *vs.* scan rates.



Fig. S8 (a) CV curve recorded by NF in potassium ferriccyanide (5 mM) at a scanning rate of 50 mV s⁻¹; (b) Electrochemical surface area normalization OER polarization curves.



Fig. S9 (a) XPS survey spectrum of Ni_2P -FeP_x/NF after OER test; High-resolution XPS spectra of (b) Ni 2p, (c) Fe 2p and (d) P 2p in Ni_2P -FeP_x/NF after OER test.



Fig. S10 XRD image of Ni_2P -FeP_x after OER test.





Fig. S12 HRTEM image of Ni_2P -FeP_x after OER test.



Fig. S13 Mercury injection curve of Ni_2P -FeP_x/NF.

Catalyst	Electrolyte	<i>j</i> (mA cm ⁻²)	TOF(s ⁻¹)	η (mV)	Ref.
Ni ₂ P-FeP _x	1.0 M KOH	50	0.458	241	This work
V-Ni ₂ P	1.0 M KOH	10	1.85	250	1
FeNi ₂ P	1.0 M KOH	10	/	210	2
Ni ₂ P	1.0 M KOH	10	/	290	3
Ni _{1-x} Fe _x PS	1.0 M KOH	100	0.097	300	4
Ni ₂ P-Fe ₂ P	1.0 M KOH	100	0.925	315	5
NiPS ₃	1.0 M KOH	10	0.0294	343	6

Table S1 Comparison of OER performance of $\rm Ni_2P\mathcal{P}\mat$

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