Supporting Information

Experimental Section

2.1 Preparation of Ni-P/NF

Firstly, a piece of Nickle foam (NF, 2×3 cm^2) was washed thoroughly through ultrasonic cleaning in 3 M HCl, ethanol and deionized water for 10 min, respectively. Similar to previously reported [1], the clean NF was transferred into 20 mL Teflon-lined stainless autoclave with 15 mL deionized water and kept at 120 ℃ for 12 h. After cooling down to room temperature, the as-fabricated NiOOH/NF was washed several times using water and dried at 40 ℃ for 2 h. Finally, the resultant Ni-P/NF- was obtained via a typical low-temperature phosphating process with a piece of NiOOH/NF placed in a ceramic boat in downstream of the tube furnace and 1 g $NaH_2PO_2{\cdot}H_2O$ placed in other one ceramic boat in upstream of the tube furnace. The tube furnace was maintained at 350℃ for 2 h under N_2 flow with a heating speed of 5℃/min, and then naturally cooled down to room temperature.

2.2 Construction of Cr-FeOOH@Ni-P/NF

Firstly, 0.5 mmol $Cr(NO_3)_3\cdot 9H_2O$, 5 mmol $FeSO_4\cdot 7H_2O$, 0.5 g Sodium citrate ($C_6H_5Na_3O_7$) as surfactant were dissolved into 40 mL deionized water under vigorous stirring for 30 min to form dark green homogeneous solution. Then thin film of Cr-FeOOH was electrodeposited in the homogeneous solution with above-prepared Ni-P/NF electrode, a carbon rod, and a Hg/HgO electrode served as the counter electrode, and reference electrode, respectively. All electrodeposition processes were all maintained at the potential of -0.1 V for 3 min. Before this, the effect about the different electroplating time on electrocatalytic activity was discussed. Similarly, Cr-FeOOH layer was electrodeposited on clean NF at the potential of -0.1 V for 60 s, 90 s, 120 s, 3 min, 4 min and 5 min, respectively.

2.3 Fabrication of Cr-doped Ni-P/NF

A typical two-step process prepared Cr-doped Ni-P/NF. Similarly, 0.5 mmol $Cr(NO_3)_3{\cdot}9H_2O$ was dissolved into 15 mL deionized water to form green homogeneous solution with vigorous stirring. Then, transferring this solution into 20 mL Teflon-lined stainless autoclave with a clean

NF immersed in it and maintained 120 ℃ for 12 h. After cooling down at room temperature, above-prepared product was phosphated at 350 ℃ for 2 h with 1 g $NaH_2PO_2 \cdot H_2O$ as P source under N_2 atmosphere.

2.4 Synthesis of RuO_2/NF

RuO_2/NF electrode was synthesized via the previously reported method[2]. A mixture of 5 mg prepared RuO_2, 1 mL 1% Nafion and isopropanol is continuous ultrasound for 30 min to obtain a uniformly dispersed mixed solution, and then 1~2 mg XC-72R was added into the above solution to enhance the intrinsic conductivity. After the powder is well dispersed by ultrasound, it is added in drops onto a dry NF substrate (2×3 cm^2), subsequent drying in air overnight.

2.5 Characterizations

The morphologies and the compositions of the catalysts were characterized by Scanning electron microscope (SEM, GeminiSEM 300 scanning electron microscope) and X-raydiffraction (XRD, LabX XRD-6100 X-ray diffractometer with Cu Kα radiation (40 kV, 30 mA) of wavelength 0.154 nm (SHIMADZU, Japan). Meanwhile, the structures of the products were determined by Transmission electron microscopy (TEM, Talos F200x (Thermo Fischer) electronmicroscopy, 200 kV). X-ray photoelectron spectroscopy (XPS) was carried on by an ESCALABMK II x-ray photoelectron spectrometer using Mg as the exciting source.

2.6 Electrochemical Measurement

All related measurements about electrocatalytic oxygen evolution was carried out according to the details reported in our previous work. The OER performances of the catalysts were measured on the CHI 760E electrochemical workstation (Chenhua, Shanghai) equipped with a three-electrode system. In a H-type electrolysis cell, the Hg/HgO electrode was equipped as the reference electrode, and a carbon rod was equipped as the counter electrode, and as-fabricated catalysts as work electrode, and 1 mol/L KOH solution as electrolyte. All tests were carried on at room temperature. All measured potentials were calibrated by reversible hydrogen electrode (RHE): E(RHE) = E(Hg/HgO) + 0.059 pH + 0.098. Polarization curves of electrocatalysts were recorded using cyclic voltammetry (CV) with a scan rate of 2 $mV \cdot s^{-1}$.

2.7 O_2 amount Measurement

The amount of O_2 produced from Cr-FeOOH@Ni-P/NF electrode at a continuous current density of 50 mA cm^{-2} in 1 M KOH solution measured with a three-electrode system. The electrochemical water splitting was carried out in a sealed electrolyzer with delivery tube to transfer generated oxygen. The volume of oxygen was measured by an inverse graduated cylinder filled with water. The Faradic efficiency is calculated through measured volume and theoretical volume of oxygen. Meanwhile, the geometric areas of Cr-FeOOH@Ni-P/NF electrode were 2×2 cm^2.

[1] Q.Li, J.Ma, H.Wang, X.Yang, R. Yuan, Y.Chai, *Electrochim. Acta* **2016**, 213, 201-206.

[2] S.Xu, Y. Du, X. Liu,X. Yu, C.Teng, X. Cheng, Y. Chen, Q. Wu, *J. Alloys and Compds.* **2020**, 826, 154210.

Figures:

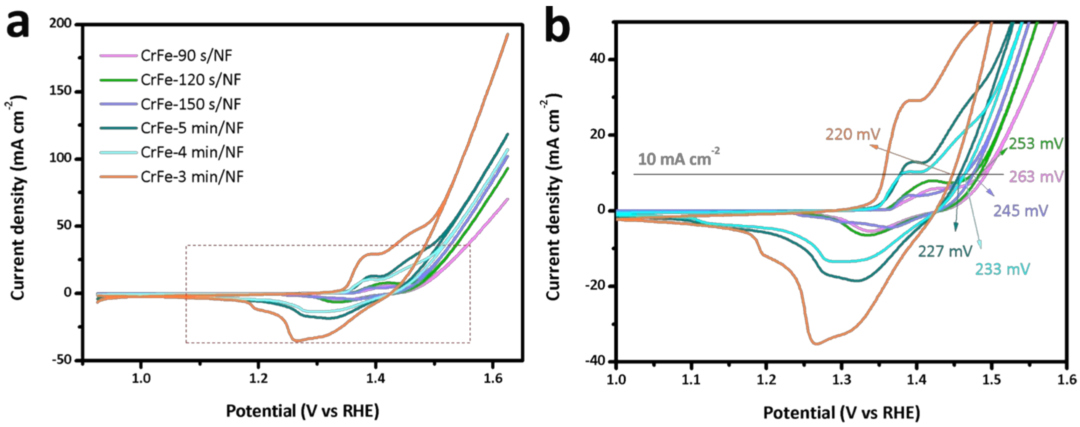
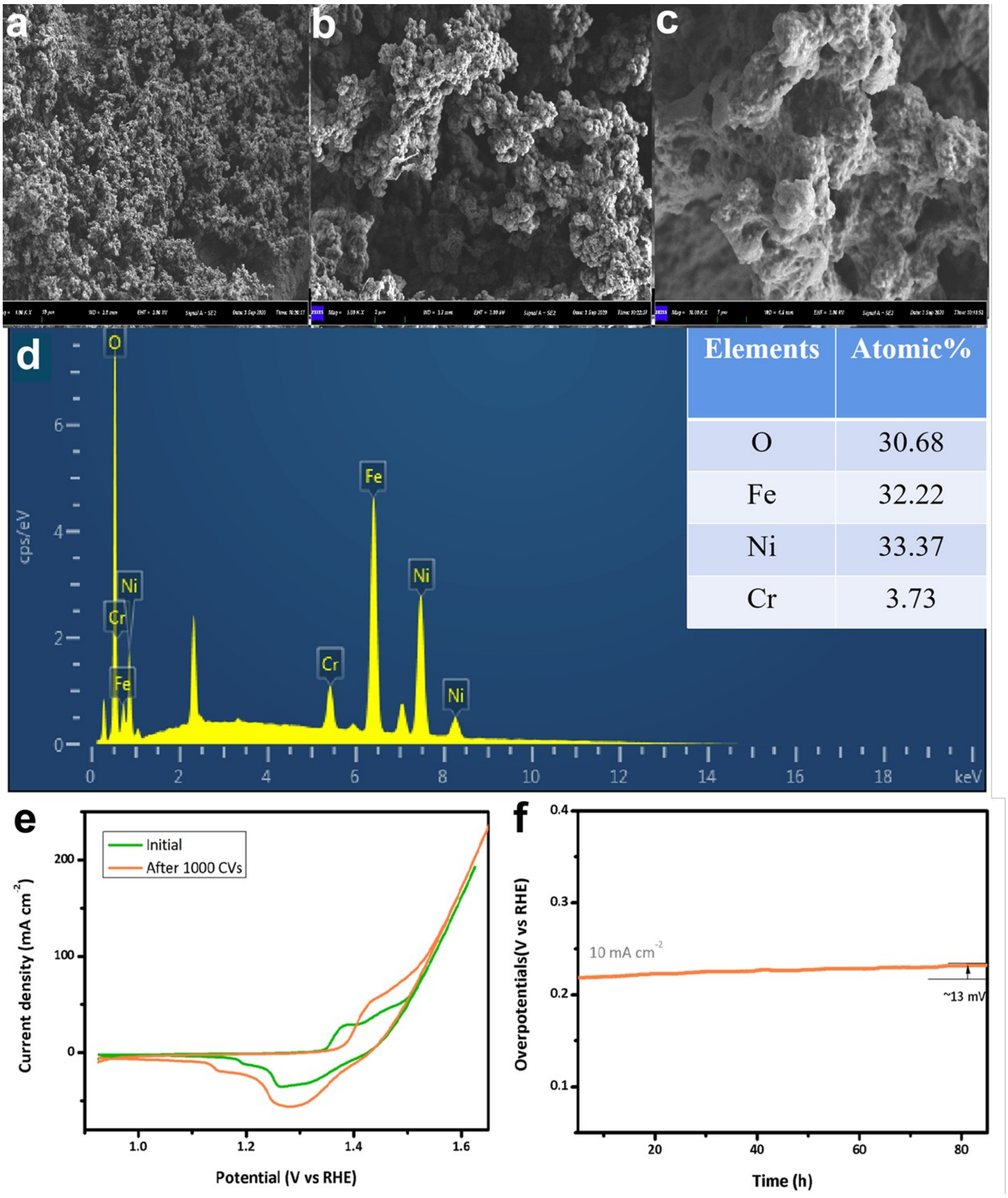


Figure S1. (a) CV curves of different plating times and (b) its partial enlarged detail of CrFe/NF.



Elements	Atomic%
O	30.68
Fe	32.22
Ni	33.37
Cr	3.73

Figure S2. (a-c) SEM images, (d) the element analysis spectrum (inserted chart: element content) and (e, f) stability test of of CrFe/NF-3 min.

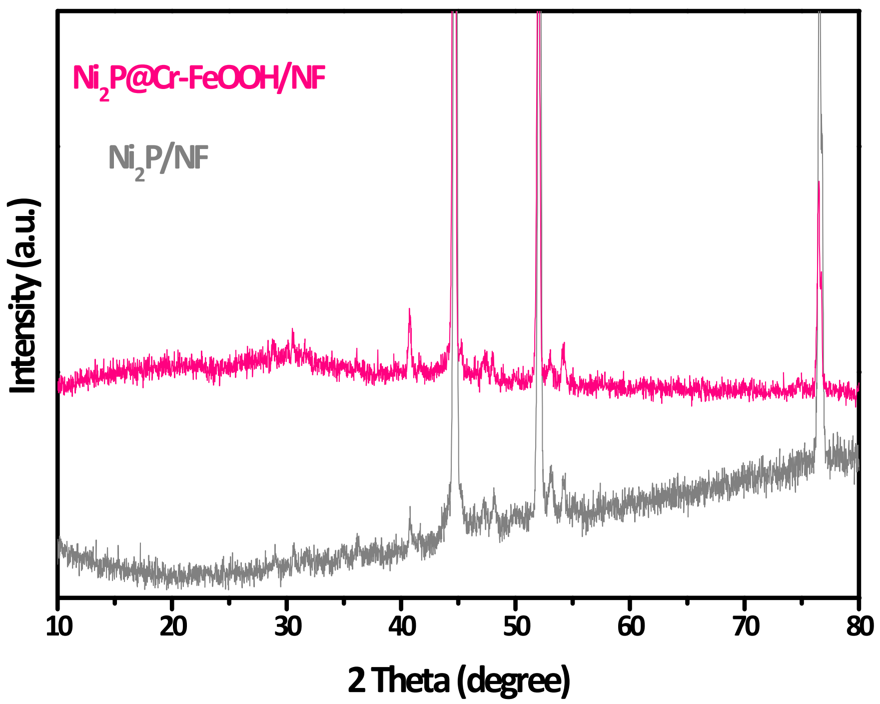
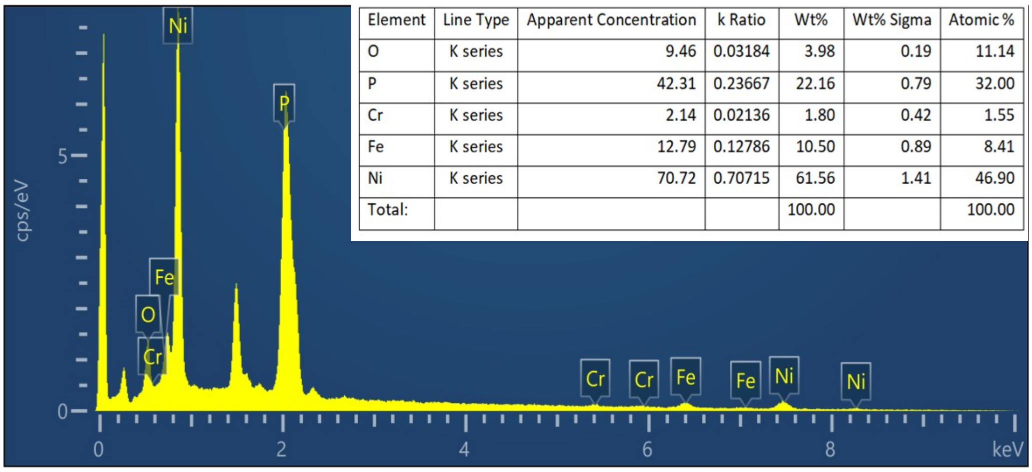


Figure S3. XRD patterns of Ni-P/NF and Cr-FeOOH@Ni-P/NF.



Element	Line Type	Apparent Concentration	k Ratio	Wt%	Wt% Sigma	Atomic %
O	K series	9.46	0.03184	3.98	0.19	11.14
P	K series	42.31	0.23667	22.16	0.79	32.00
Cr	K series	2.14	0.02136	1.80	0.42	1.55
Fe	K series	12.79	0.12786	10.50	0.89	8.41
Ni	K series	70.72	0.70715	61.56	1.41	46.90
Total:				100.00		100.00

Figure S4. EDS spectrum of Cr-FeOOH@Ni-P/NF and corresponding element content.

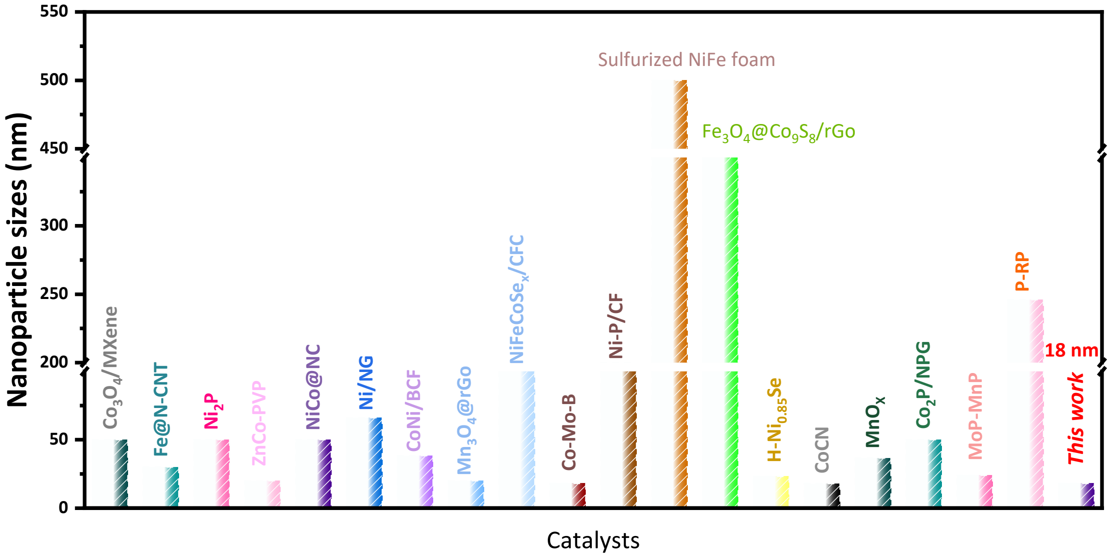


Figure S5. Contrast histogram of different size for nanoparticle form various electrocatalysts.

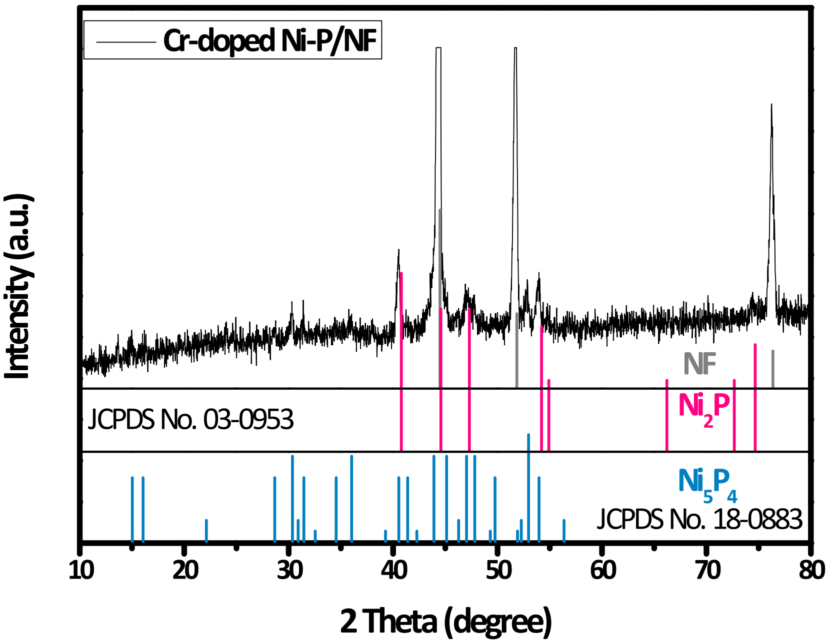


Figure S6. XRD pattern of Cr-Ni-P/NF catalyst.

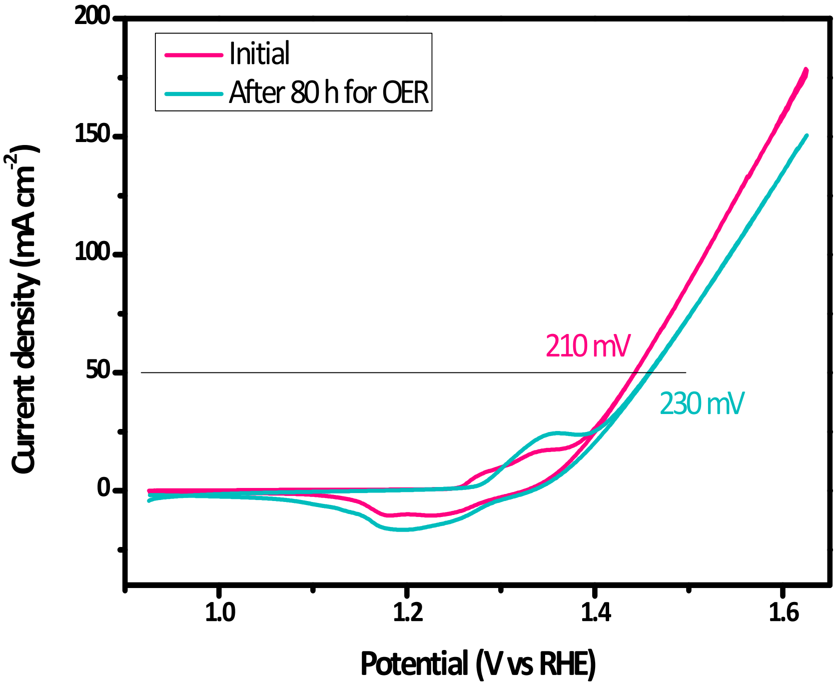


Figure S7. The CV curves of Cr-FeOOH@Ni-P/NF before and after stability measurement over 80 h.

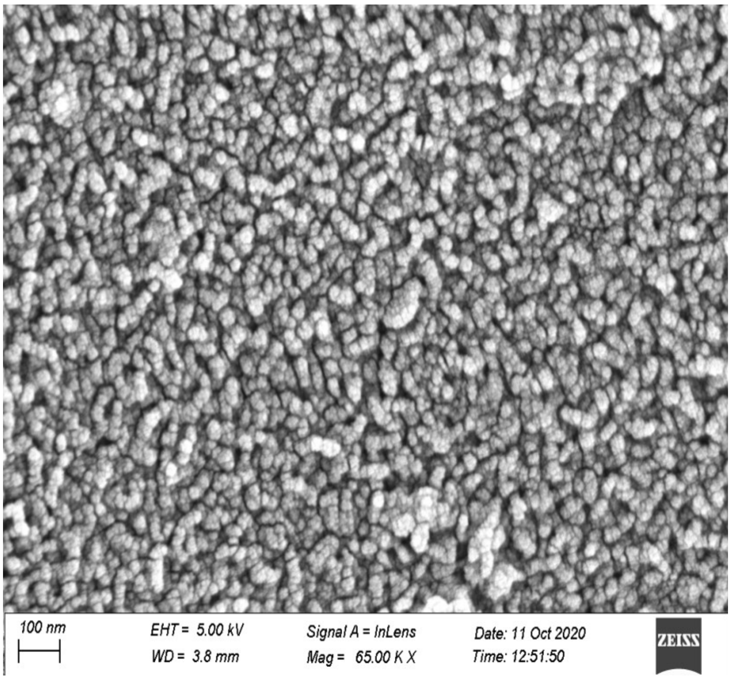


Figure S8. SEM image of Cr-FeOOH@Ni-P/NF after OER test over 80 h.

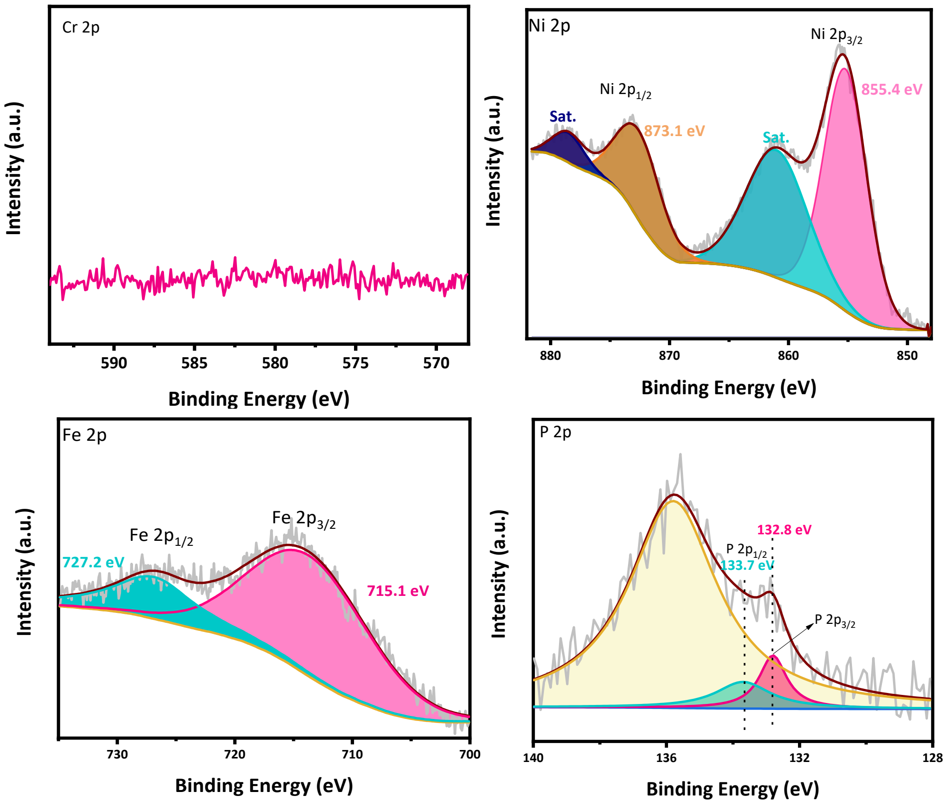


Figure S9. XPS spectra of Cr-FeOOH@Ni-P/NF after OER test over 80 h.

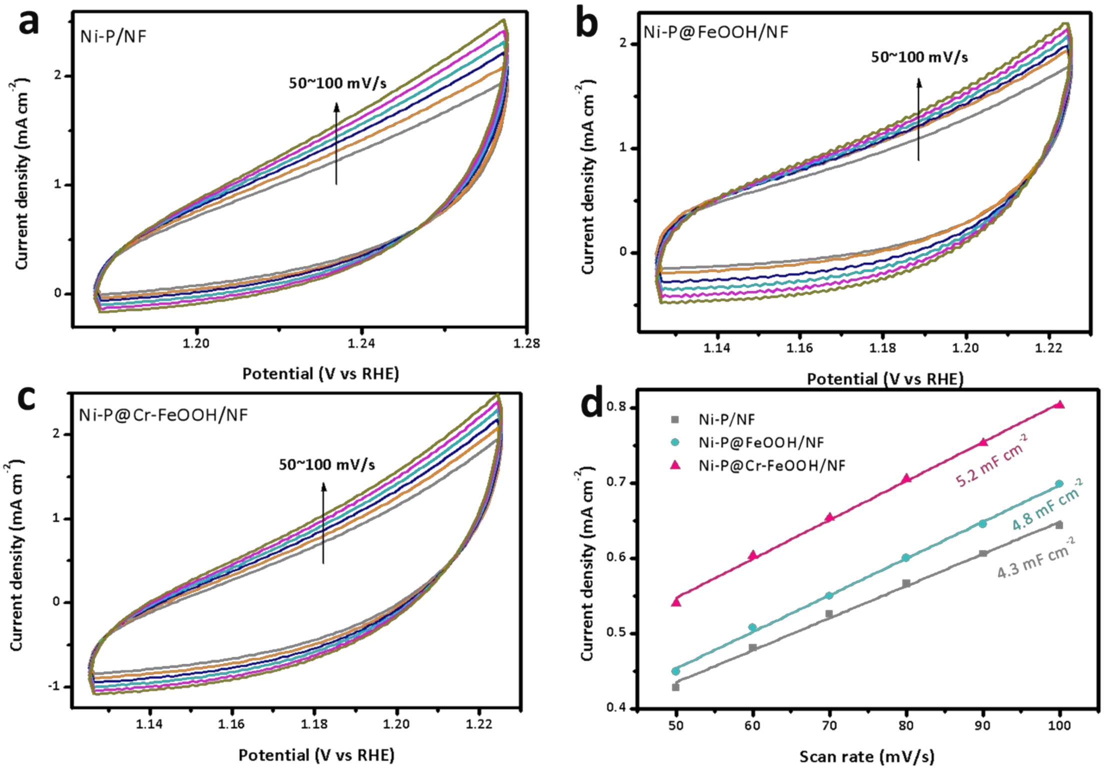


Figure S10. (a-c) CV curves of Ni-P/NF, FeOOH@Ni-P/NF and Cr-FeOOH@Ni-P/NF at potential scan rate form 50 to 100 mV s^{-1}. (d) Double-layer capacitance of Ni-P/NF, FeOOH@Ni-P/NF and Cr-FeOOH@Ni-P/NF.

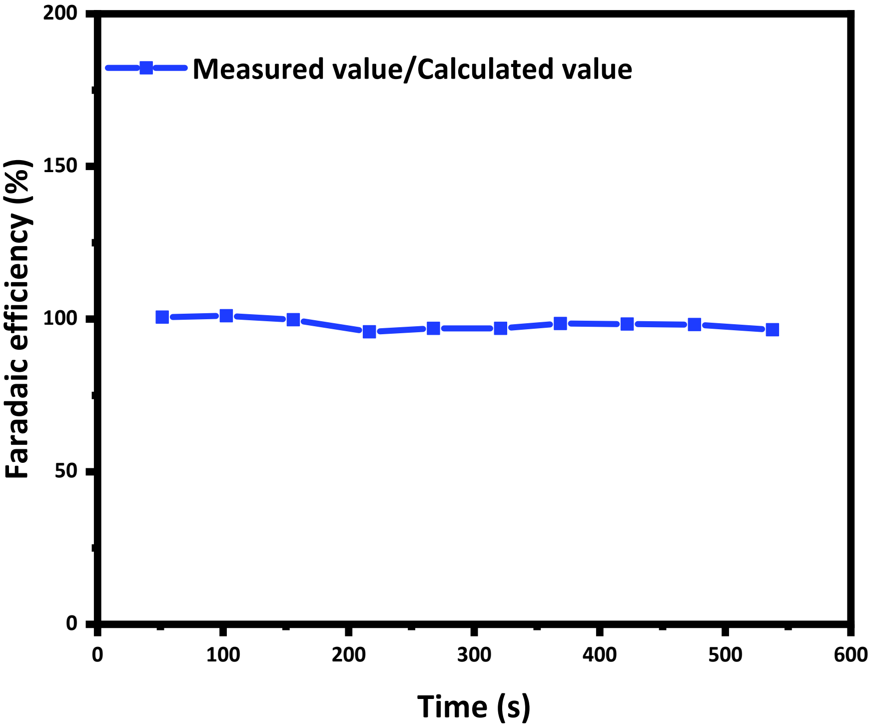


Figure S11. Faradaic efficiency of Cr-FeOOH@Ni-P/NF at 50 mA cm^{-2}.