Supplementary Information

Rapid "self-healing" behavior induced by chloride anions to renew Fe-

Ni(oxy)hydroxide catalytic surface for long-term alkaline seawater

electrolysis

Ruo-Yao Fan, Xin-Yu Zhang, Ning Yu, Feng-Ge Wang, Hui-Ying Zhao, Xin Liu,

Qian-Xi Lv, Da-Peng Liu*, Yong-Ming Chai, Bin $\operatorname{Dong}\nolimits^*$

State Key Laboratory of Heavy Oil Processing, College of Chemistry and Chemical Engineering,

China University of Petroleum (East China), Qingdao 266580, PR China

Experimental section

^{*} Corresponding author. Email: <u>liudp@upc.edu.cn (D.P. Liu); dongbin@upc.edu.cn (B. Dong)</u>

Tel: +86-532-86981156, Fax: +86-532-86981156

All chemicals, including acetone (\geq 99.5% AR), ethyl alcohol (\geq 99.7% AR) and sodium chloride (\geq 99.5% AR) were purchased from Sigma-Aldrich and used as received without any purification. The commercial Fe-Ni foam and Ni foam were purchased from Kunshan Lvchuang Electronic Technology Co., LTD. The vacuum drying chamber used in the experiment is DZG-6050.

Characterization

The crystal phase of the obtained catalysts was studied via X-ray diffraction (XRD) on a Rigaku D/max-2500pc device with Cu K α radiation ($\lambda = 1.54$ Å). The scanning electron microscope (SEM) (Hitachi S-4800) and Transmission Electron Microscope (TEM) (FEI Tecni G20, 200 kV) were applied to collect the information of morphological and structural information of all samples. The element composition and distribution on catalysts were detected by means of the Energy Dispersive System (EDS) and detected on the Hitachi S-4800. The X-ray photoelectron spectroscopy (XPS) is characterized by a Thermo Fisher Scientific II spectrometer with an Al K α source (1486.6 eV).

Electrochemical measurements

The OER performance of all the catalysts was evaluated in a standard three-electrode cell configuration on Gamry Reference 1000 electrochemical equipment at room temperature. This process was performed with the Pt foil, saturated calomel electrode and catalyst grown in situ on Fe-Ni foam as the counter electrode, reference electrode, and working electrode, respectively. The linear sweep voltammetry (LSV) were gained at same condition with scan rate of 10 mV s⁻¹. All the potentials vs. SCE were converted

into a standard reversible hydrogen electrode (RHE) by means of the Nernst equation: $E_{RHE} = E_{SCE} + 0.242 + 0.059 \text{ pH}$ (in this work, 1M KOH pH= 13.65; 1M KOH+Seawater pH= 13.62). The cyclic voltammetry (CV) curves were tested at 20, 40, 60, 80 and 100 mV s⁻¹ and were used to determine the electrical double-layer capacitances (C_{dl}). And electrochemical impedance spectroscopy (EIS) was performed at 0.43 V vs. SCE and the frequency ranges from 10⁵ Hz to 0.01 Hz. The stability of the final sample was performed by chronopotentiometry (CP) at 200 mA cm⁻²



Fig. S1 The photo and XRD of Fe_3O_4 powder (After anodic corrosion).



Fig. S2 The SEM EDX-Mapping of Fe₃O₄ powder (After anodic corrosion).



Fig. S3 The SEM of Fe-Ni foam.



Fig. S4 The (a-c) SEM and (d-g) SEM-energy distribution plane scanning (EDS) Mapping images of the Fe-Ni foam after anodic corrosion.

Elements	Family	Atomic Fraction (%)	Mass Fraction (%)
Ni	к	25.85	55.83
Fe	К	0.35	0.72
0	К	73.81	43.45

 Table S1 Element content statistics of AC-FeNi(O)OH nanosheets.



Fig. S5 The (a) LSV and (b) Tafel slope of AC-FeNi(O)OH-250, AC-FeNi(O)OH-150, AC-FeNi(O)OH-70 and AC-FeNi(O)OH-350. (c) The photographs of Fe₃O₄ powders under different corrosion currents.



Fig. S6 The (a) LSV and (b) Tafel slope of AC-FeNi(O)OH-Cl, FeNi(O)OH, AC-FeNi(O)OH-F, FeNi(O)OH-I and FeNi(O)OH-Br.



Fig. S7 The Bode phase diagram of AC-FeNi(O)OH and FeNi(O)OH.



Fig. S8 The CV of AC-FeNi(O)OH, FeNi(O)OH, AC-Fe(O)OH and Fe(O)OH in 1M KOH.



Fig. S9 The pH value of the solutions used in this work were measured using a standard

pH meter.



Fig. S10 The SEM images of AC-FeNi(O)OH in (a) 1M KOH, (b) 1M KOH+Seawater

and (c) 1M KOH+1M NaCl after 100 hours of stability testing.



Fig. S11 The XPS characterizations of the electrocatalysts before stability test, after 100 hours stability test in 1M KOH and after 100 hours stability test in 1M KOH+Seawater.



Fig. S12 (a)The polarization curves of obtained sample (AC-FeNi(O)OH) toward overall seawater splitting at 1M KOH+Seawater. (b) Corresponding voltages at 10 and 50 mA cm⁻². (c) Long-term stability test of AC-FeNi(O)OH || CoP for water splitting at 50 mA cm⁻² for 25 h.

	Electrocatalysts	Substrate	Overpotential (at 100 mA cm ²)/mV	Tafel slope/ mV dec ⁻¹	Stability/ hours
[31]	FeOOH-NiBDC-NF	Ni foam	280	54.9	100
[33]	Ni-Fe LDH/NF	Ni foam	297	60.8	-
[34]	NiFe(OH) _x -10	Ni foam	277	54	100
[35]	NiFe ₃ Nb ₂ -OH	Ni foam	294	47	90
[36]	S-(Ni,Fe)OOH	Ni foam	300	48.9	100
[37]	FeNiO _x (OH) _y -2	Ni foam	293	39	50
[38]	FeOOH(Se)/IF	Fe foam	364	54	15
[39]	Fe _{0.4} Ni _{0.6} alloy fiber paper	Alloy fiber paper	287	67	80
[40]	(Fe,Co)OOH/MI	Ni foam	290	53	155
[41]	NiFe ₃ Nb ₂ -OH	Ni foam	294	47	90
[42]	Fe-(Ni-MOFs)/FeOOH	Ni foam	278	50	120
This work	AC-FeNi(O)OH	Fe-Ni foam	252	55.25	100

Table S2 Comparison of the OER activity and stability of AC-FeNi(O)OH with otherNi-Fe or Co-Fe based bimetallic electrocatalysts in alkaline electrolyte.