

Electronic Supplementary Information

Experimental Section

Materials: Potassium hydroxide (KOH), sodium chloride (NaCl), sodium carbonate (Na_2CO_3), urea ($\text{CO}(\text{NH}_2)_2$), 20 wt.% Pt/C, ruthenium oxide (RuO_2), *N, N*-diethyl-*p*-phenylenediamine (DPD), and Nafion (5 wt.%) were bought from Aladdin Ltd. (Shanghai, China). Nickel nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), sodium hydroxide (NaOH), iron nitrate nonahydrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$), sodium hexafluoroantimonate (NaSbF_6), and ammonium fluoride (NH_4F) were obtained from Shanghai Macklin Biochemical Technology Co., Ltd. Ni foam (NF) was obtained from Shenzhen Green and Creative Environmental Science and Technology Co., Ltd. Ethanol, sulfuric acid (H_2SO_4), and hydrochloric acid (HCl) were procured from Beijing Chemical Reagent Co., Ltd. (Beijing, China). Natural seawater was collected from Qingdao, Shandong, China, most of the magnesium and calcium salts were removed by adding Na_2CO_3 to natural seawater before use. All reagents were used as received, without further purification.

Preparation of NiFe LDH/NF and SbF_6^- -NiFe LDH/NF: Initially, a piece of NF was cut into small pieces measuring $2\text{ cm} \times 3\text{ cm}$ and sonicated sequentially in 3 M HCl, water, and ethanol for 15 min. The pretreated NF was put into a solution containing 0.404 g $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 0.6 g urea, 0.582 g $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, 0.148 g NH_4F , and 40 mL water in a Teflon-lined autoclave, treated at $120\text{ }^\circ\text{C}$ for 6 h to produce NiFe LDH/NF. Then, the obtained NiFe LDH/NF was soaked in a 0.1 M NaSbF_6 solution for 30 min to get SbF_6^- -NiFe LDH/NF.

Preparation of RuO_2 or Pt/C on NF: 5 mg of RuO_2 (or 20% Pt/C) was added in a solution containing 30 μL of Nafion, 485 μL of ethanol, and 485 μL of water with the aid of ultrasonication (30 min) to form a homogeneous ink (5 mg mL^{-1}). 100 μL of catalyst ink was dropped onto a piece of pretreated NF ($0.5\text{ cm} \times 0.5\text{ cm}$) with a loading mass of 2 mg cm^{-2} .

Characterizations: XRD data were obtained via X-ray diffraction (XRD, Philip D8)

with Cu K α source ($\lambda = 1.54056 \text{ \AA}$). Raman spectroscopy was recorded on the Lab RAM HR Evolution confocal microscope with a 532 nm laser. The water contact angle measurements were performed using an OCA 50 AF (Dataphysics). Scanning electron microscopy (SEM, ZISS 300) equipped with an energy dispersive X-ray (EDX) facility, transmission electron microscopy (TEM, JEM-F200, JEOL Ltd.), and X-ray photoelectron spectroscopy (XPS, ESCALAB 250 Xi) were utilized to research the morphology and compositions of samples. UV-visible (UV-vis) spectrophotometry (Shimadzu UV-2700) was utilized for absorbance measurements. The elemental composition was determined by Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES, SPECTRO ARCOS II MV, Germany).

Electrochemical measurements: All electrochemical experiments were carried out using a CHI 660 analyzer (CH Instruments, Inc., Shanghai). All measured potentials were standardized to the reversible hydrogen electrode (RHE) using the equation: $E(\text{RHE}) = E(\text{Hg/HgO}) + 0.098 + 0.059 \times \text{pH}$. Tafel slopes were calculated by the Tafel equation: $\eta = b \log j + a$, where η represents overpotential, j is the current density (mA cm^{-2}), and b is the Tafel slope (mV dec^{-1}). The double-layer capacitance (C_{dl}) values were measured via cyclic voltammetry (CV) curves with scan rates of 20 to 120 mV s^{-1} . Electrochemical impedance spectroscopy tests were conducted between 10^5 to 0.01 Hz with an amplitude of 0.005 V. The iR -compensated potential was obtained from the following equation: $E_{\text{corr}} = E - iR$, where E is the original potential, R is the solution resistance, and i is the corresponding current.

Determination of active chlorine: The concentration of active chlorine in the electrolyte was determined using the DPD colorimetric method, measured by UV-vis spectrophotometry.¹ First, the 100 μL of stability-tested electrolyte was successively mixed with 50 μL of H_2SO_4 (1.0 M), 50 μL of NaOH (2.0 M), and 4.8 mL of water. Then, 250 μL of DPD reagent and 250 μL of PBS ($\text{pH} = 6.5$) were added to the above solution. After allowing 2 minutes for color development in a light-shielded environment, the solution turned pink. The absorbance at 550 nm was then measured

using UV-visible absorption spectroscopy, enabling the analysis of different concentrations of active chlorine.

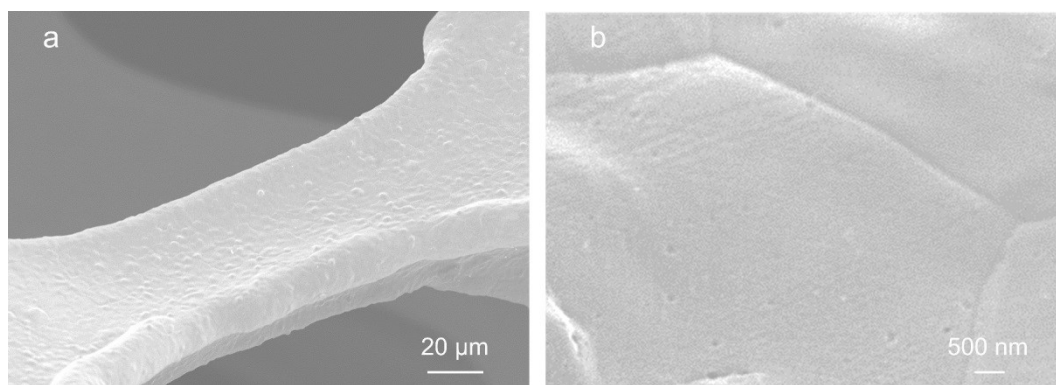


Fig. S1. (a) Low- and (b) high-magnification SEM images of NF.

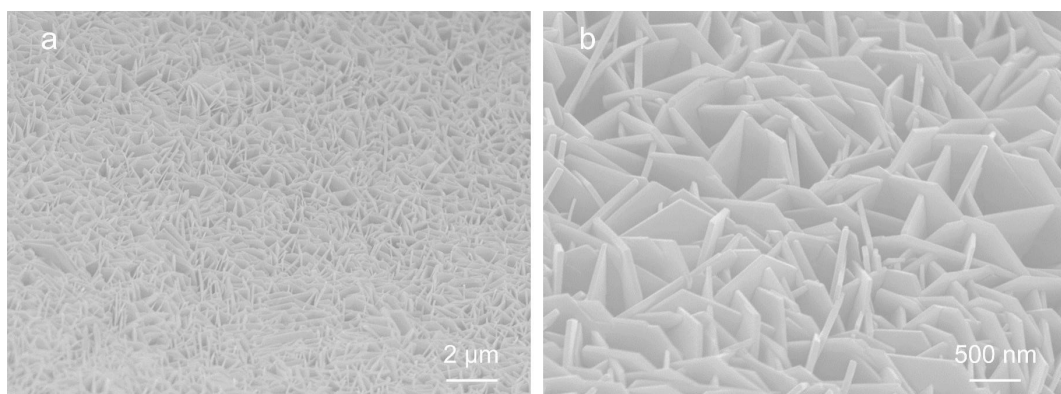


Fig. S2. (a) Low- and (b) high-magnification SEM images of NiFe LDH/NF.

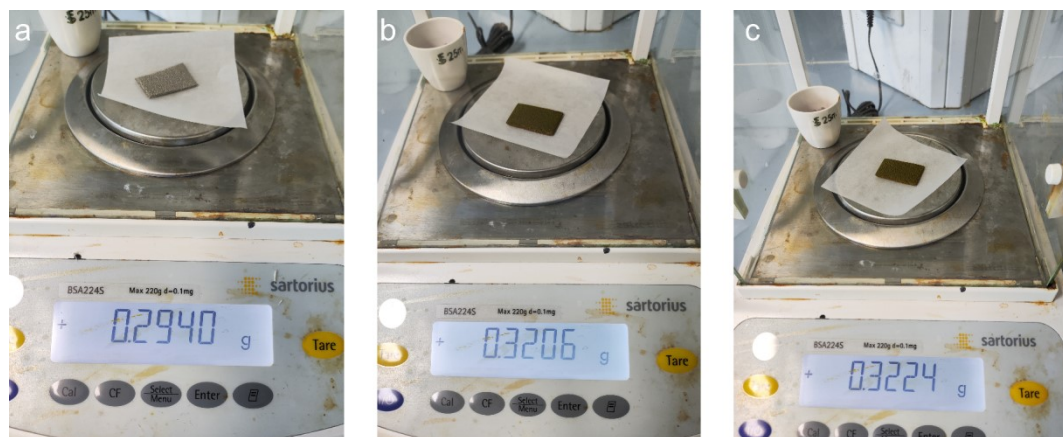


Fig. S3. Digital photographs of (a) NF, (b) NiFe LDH/NF, and (c) SbF_6^- -NiFe LDH/NF.

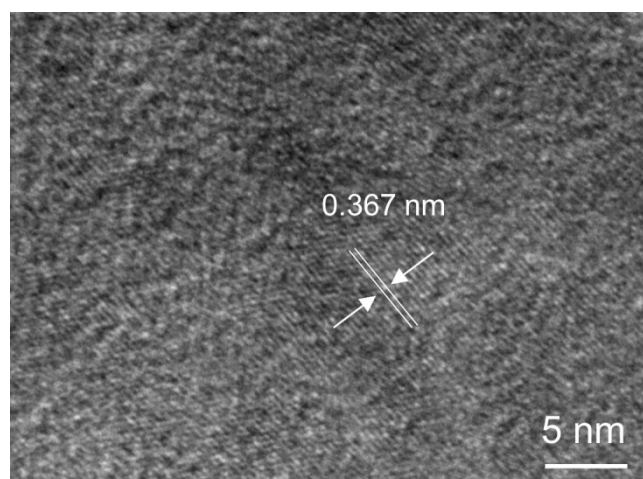


Fig. S4. HRTEM image of NiFe LDH.

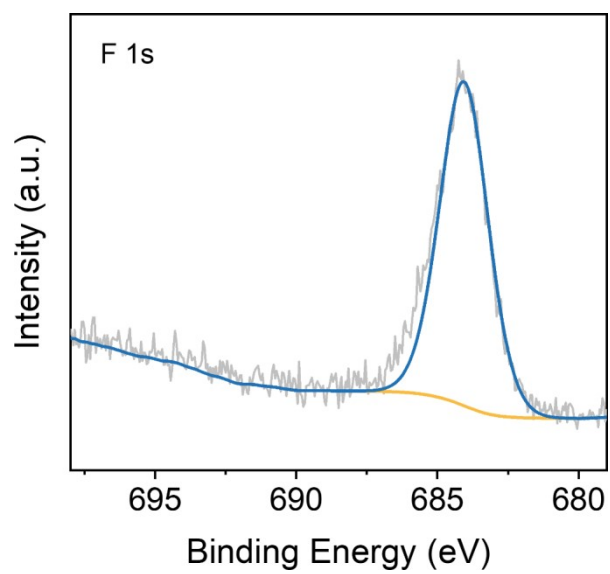


Fig. S5. High-resolution XPS spectrum of SbF_6^- -NiFe LDH in F 1s region.

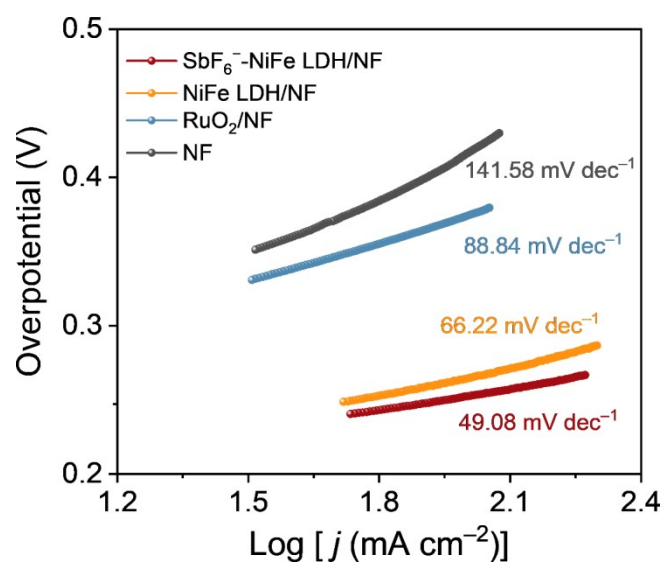


Fig. S6. Tafel plots of different electrocatalysts in 1 M KOH.

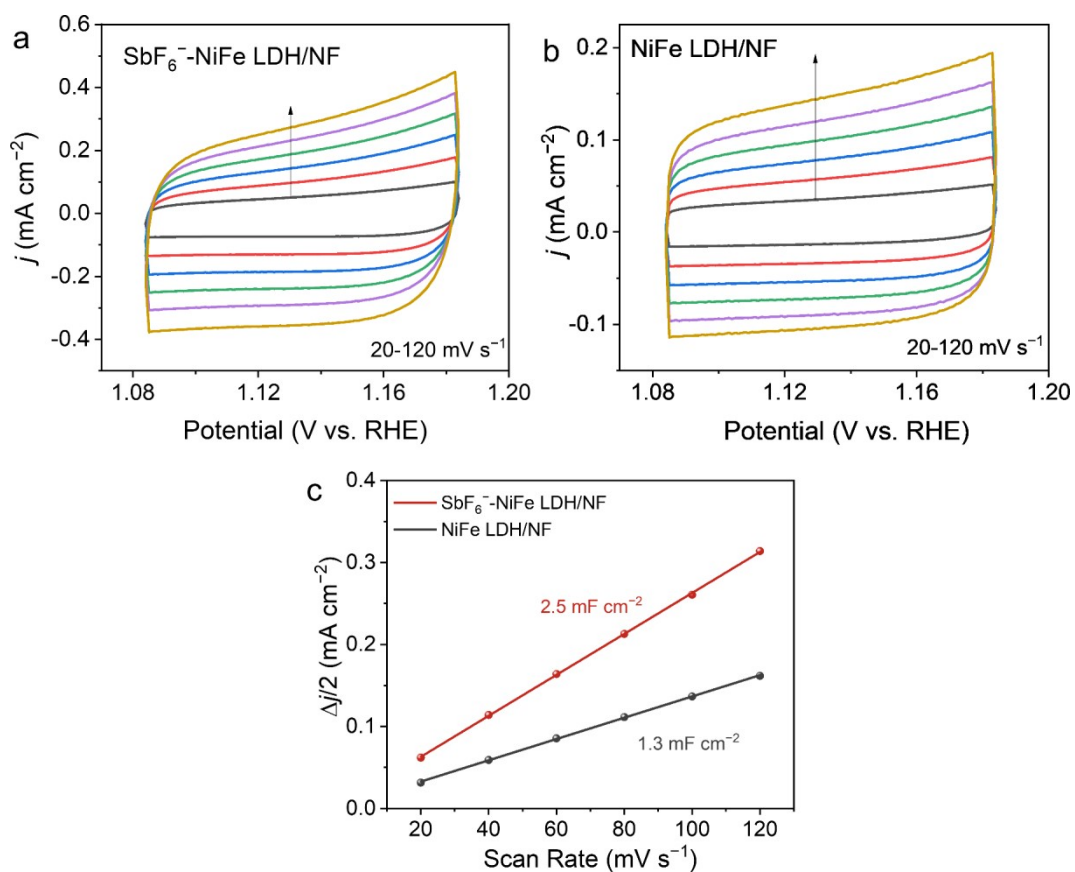


Fig. S7. CV curves of (a) SbF_6^- -NiFe LDH/NF and (b) NiFe LDH/NF in the double layer region at different scan rates of 20, 40, 60, 80, 100, and 120 mV s^{-1} in 1 M KOH. (c) C_{dl} curves of SbF_6^- -NiFe LDH/NF and NiFe LDH/NF.

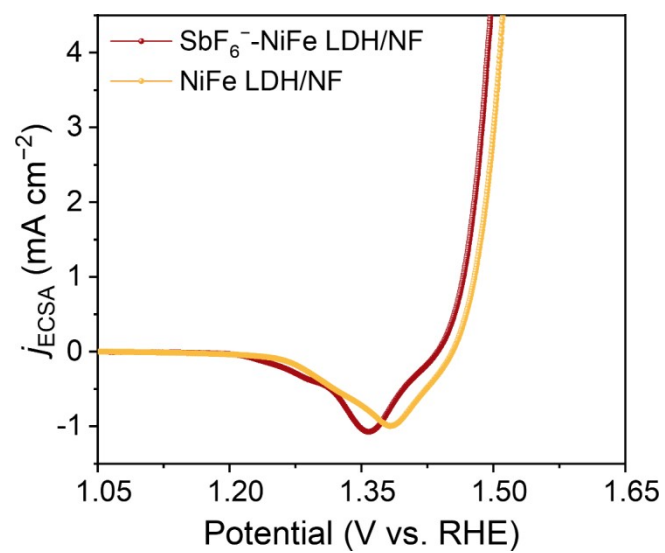


Fig. S8. ECDSA-normalized LSV curves for SbF_6^- -NiFe LDH/NF and NiFe LDH/NF in 1 M KOH.

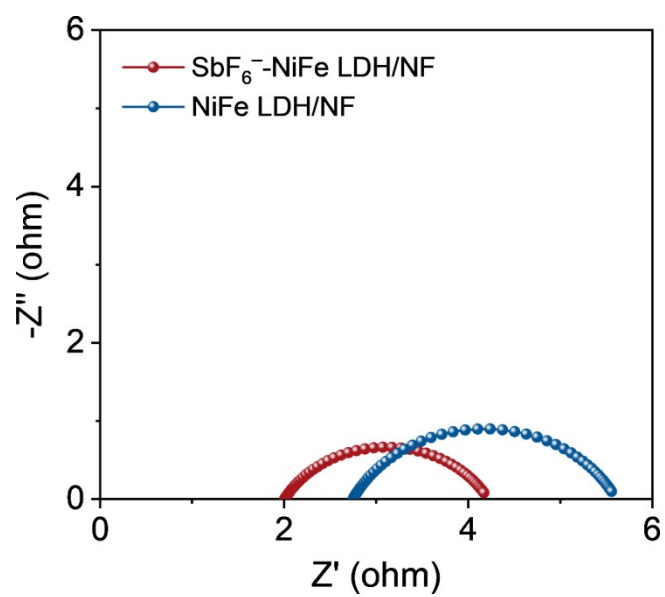


Fig. S9. Nyquist plots of SbF_6^- -NiFe LDH/NF and NiFe LDH/NF in 1 M KOH.

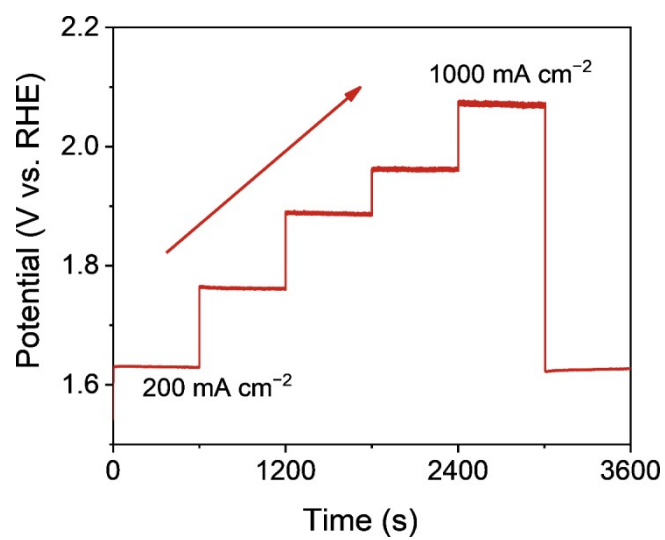


Fig. S10. Multistep chronopotentiometric curve of SbF_6^- -NiFe LDH/NF without iR correction in 1 M KOH.

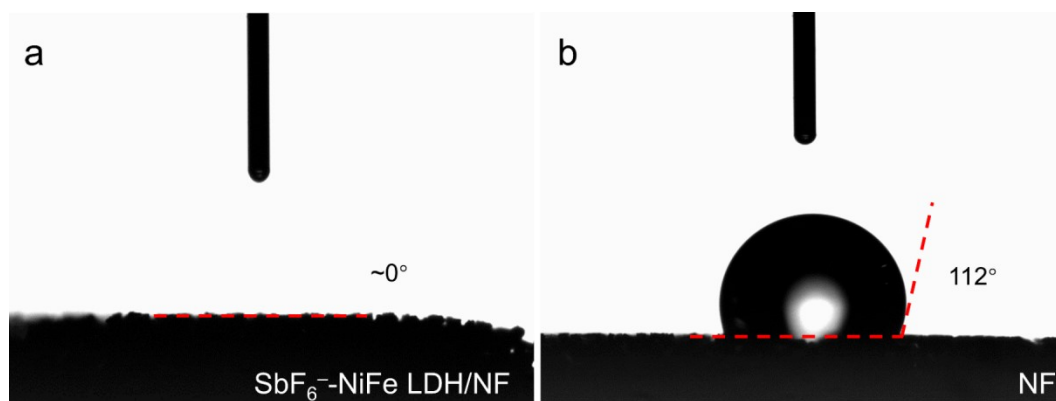


Fig. S11. The water contact angle tests of (a) SbF₆⁻-NiFe LDH/NF and (b) NF.

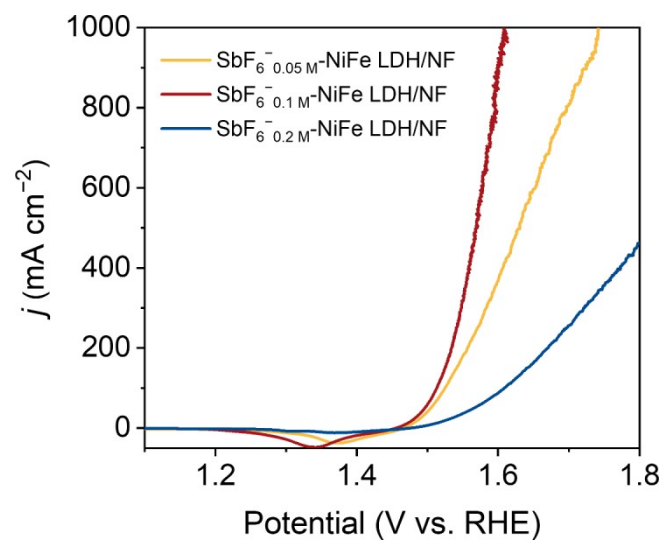


Fig. S12. LSV curves of different concentrations of NaSbF₆ immersed electrocatalysts in 1 M KOH + seawater.

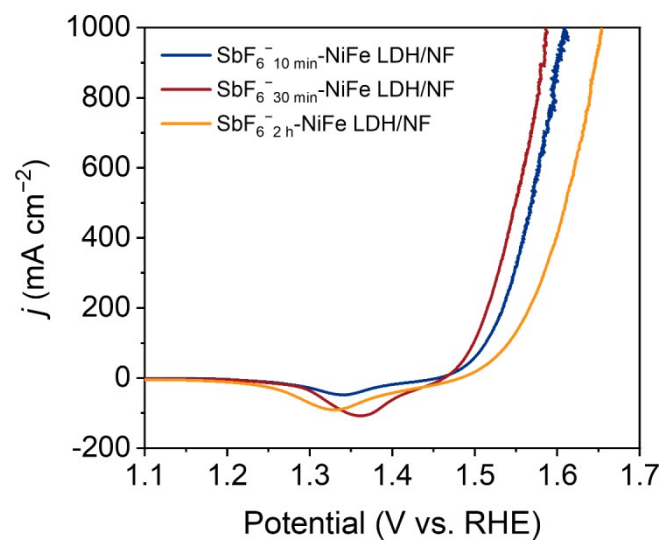


Fig. S13. LSV curves of electrocatalysts immersed in 0.1 M NaSbF₆ for different times in 1 M KOH + seawater.

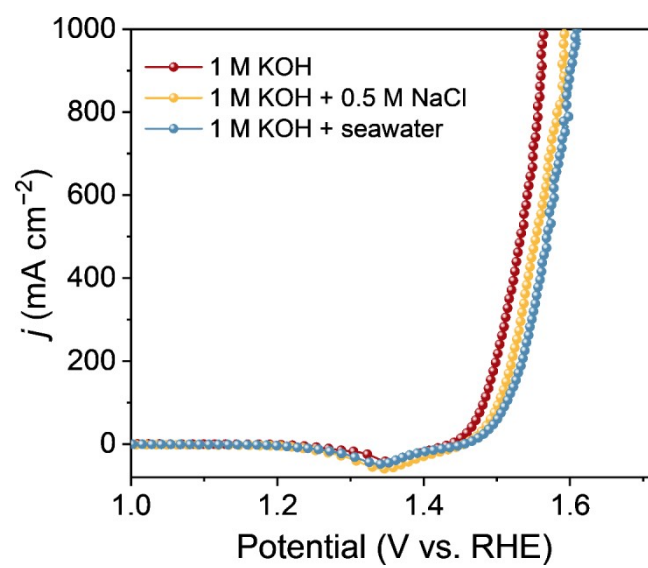


Fig. S14. LSV curves of SbF₆⁻-NiFe LDH/NF in different electrolytes.

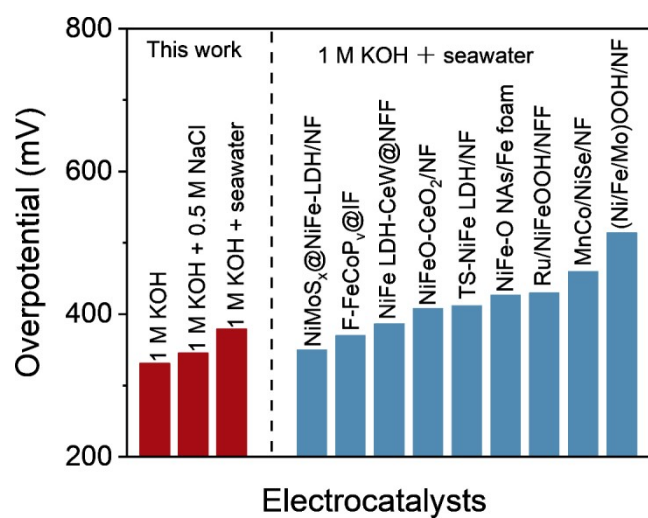


Fig. S15. Comparison of overpotentials required to achieve a j of 1000 mA cm^{-2} between SbF_6^- -NiFe LDH/NF and other electrocatalysts in 1 M KOH + seawater.

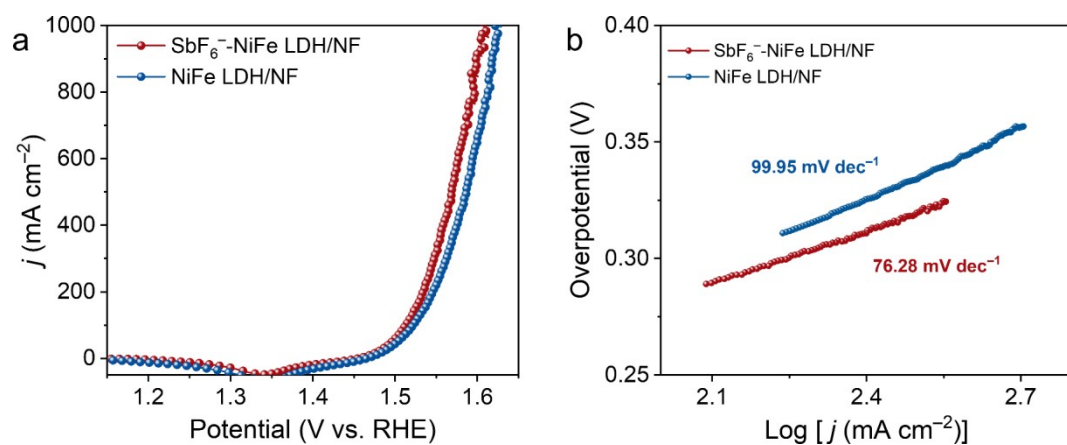


Fig. S16. (a) LSV curves and (b) corresponding Tafel plots of SbF_6^- -NiFe LDH/NF and NiFe LDH/NF in 1 M KOH + seawater.

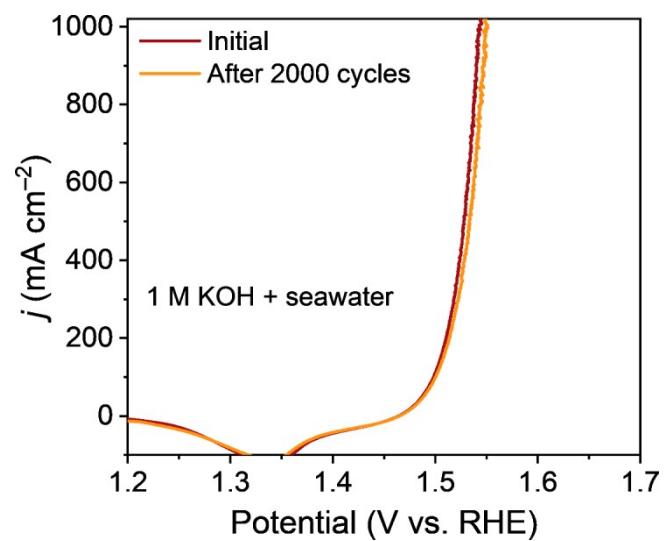


Fig. S17. LSV curves of SbF_6^- -NiFe LDH/NF before and after 2000 CV cycles in 1 M KOH + seawater.

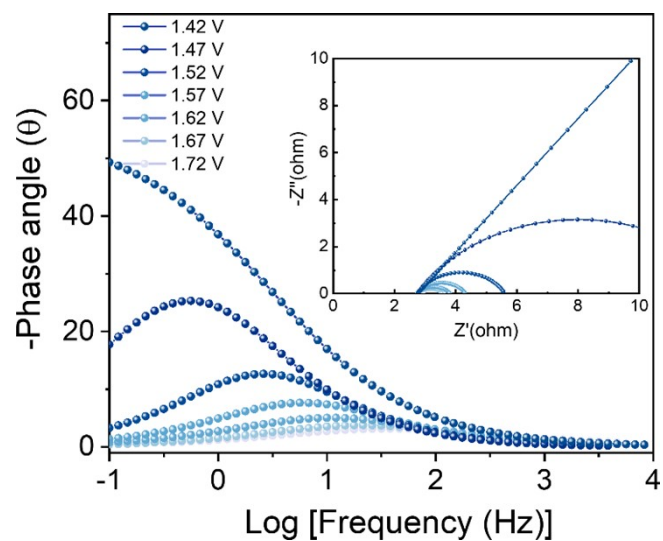


Fig. S18. Bode plots and the corresponding operando Nyquist plots of NiFe LDH/NF at different potentials versus reversible hydrogen electrode.

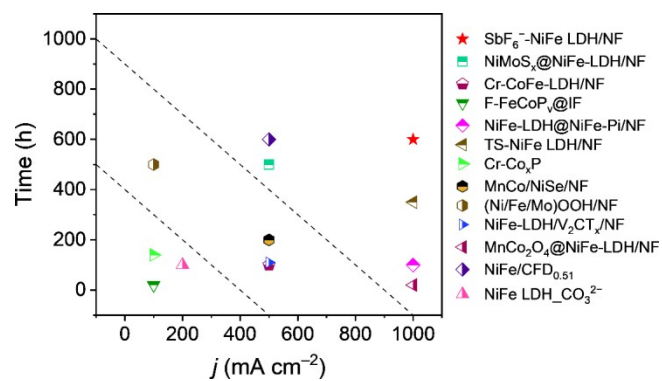


Fig. S19. Comparison of the stability of SbF₆⁻-NiFe LDH/NF with other catalysts in 1 M KOH + seawater.

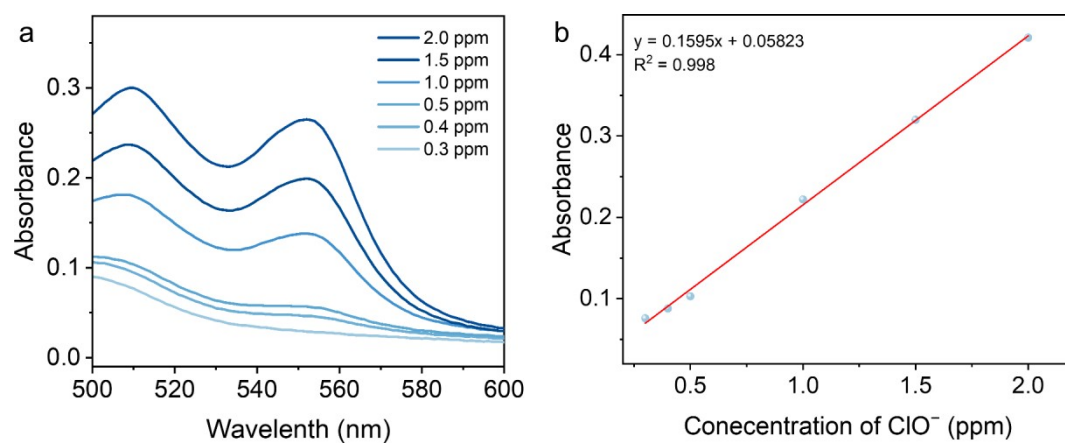


Fig. S20. (a) UV-vis absorption spectra of various concentrations of active chlorine.
(b) Calibration curve.

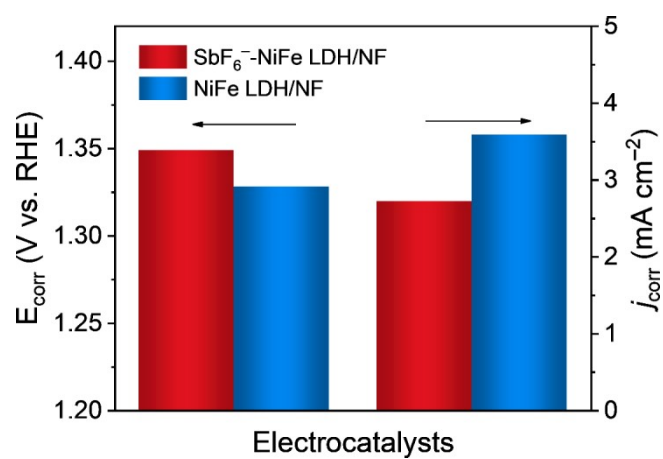


Fig. S21. Corrosion potentials and corrosion j of SbF_6^- -NiFe LDH/NF and NiFe LDH/NF in 1 M KOH + seawater.

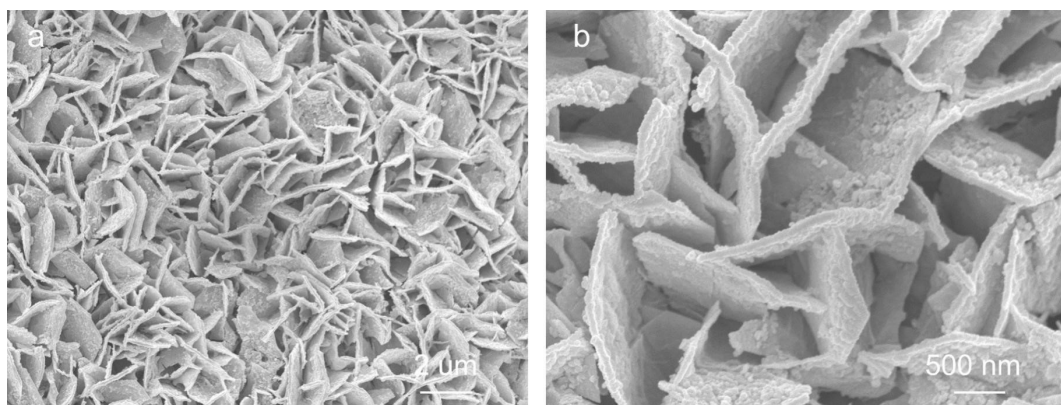


Fig. S22. (a) Low- and (b) high-magnification SEM images of SbF_6^- -NiFe LDH/NF after stability test.

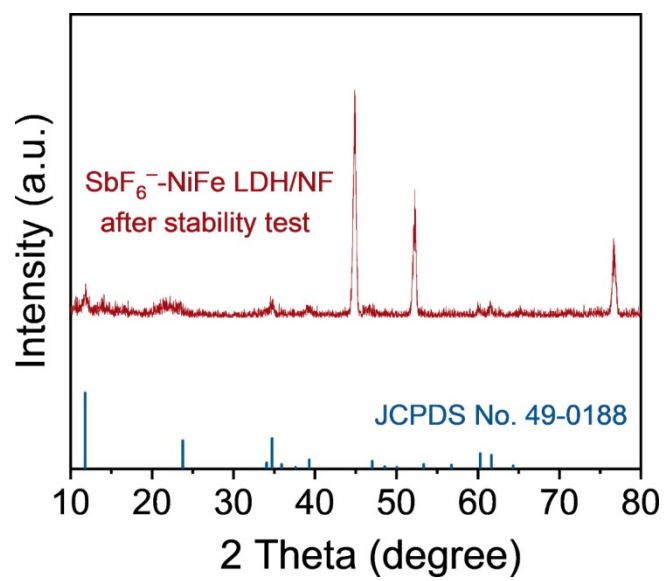


Fig. S23. XRD pattern of SbF₆⁻-NiFe LDH/NF after stability test in 1 M KOH + seawater.

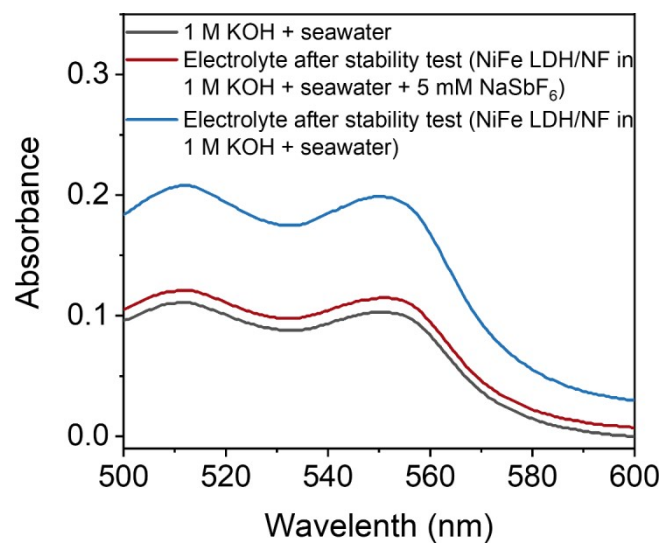


Fig. S24. UV-vis absorption spectra of collected electrolytes from NiFe LDH/NF stability test in alkaline seawater electrolytes with and without 5 mM NaSbF₆.

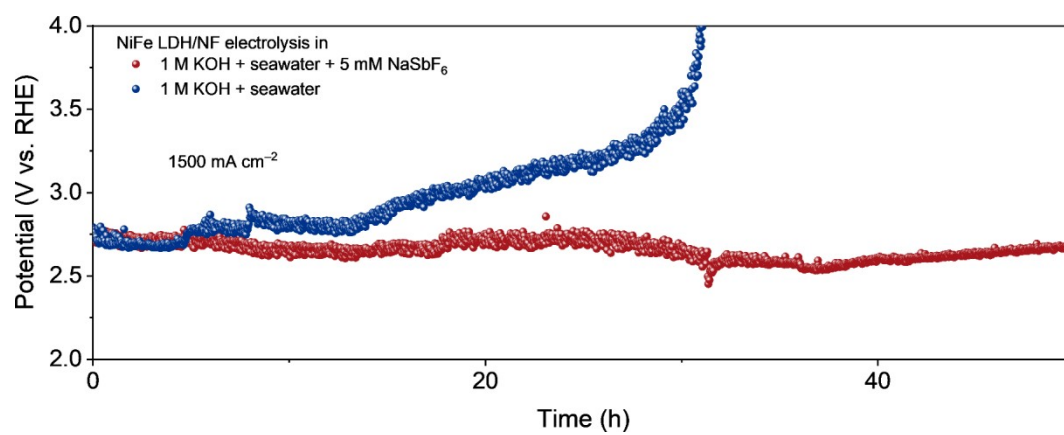


Fig. S25. The chronopotentiometry curves of NiFe LDH/NF tested in 1 M KOH + seawater with and without 5 mM NaSbF₆.

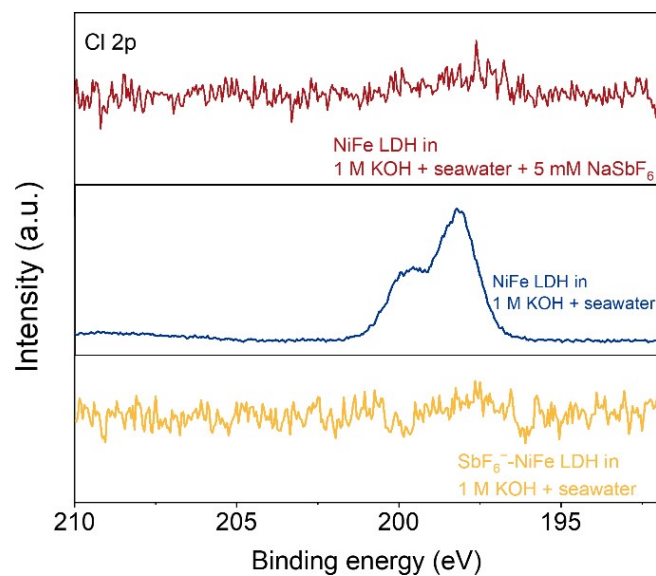


Fig. S26. Comparison of XPS spectra in the Cl 2p region between NiFe LDH in 1 M KOH + seawater + 5 mM NaSbF₆, NiFe LDH and SbF₆⁻-NiFe LDH in 1 M KOH + seawater after stability test.

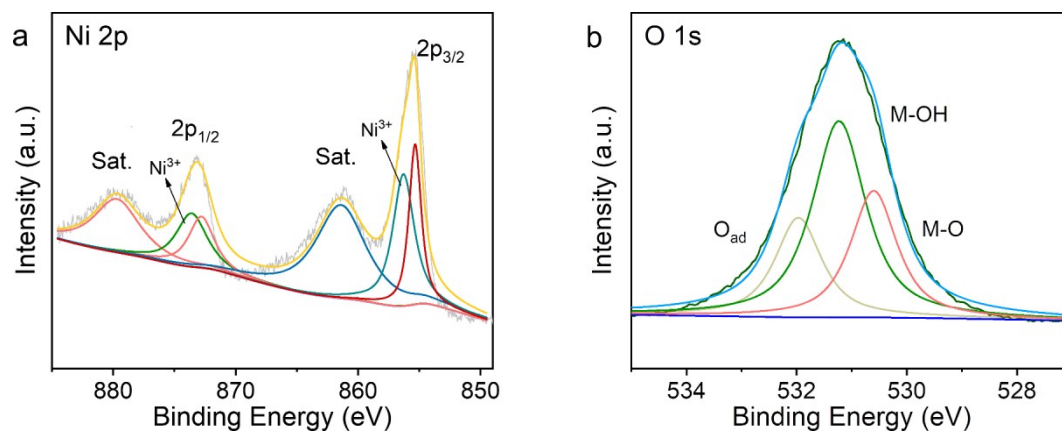


Fig. S27. High-resolution XPS spectra in the (a) Ni 2p and (b) O 1s regions of SbF₆⁻-NiFe LDH after stability test in 1 M KOH + seawater.

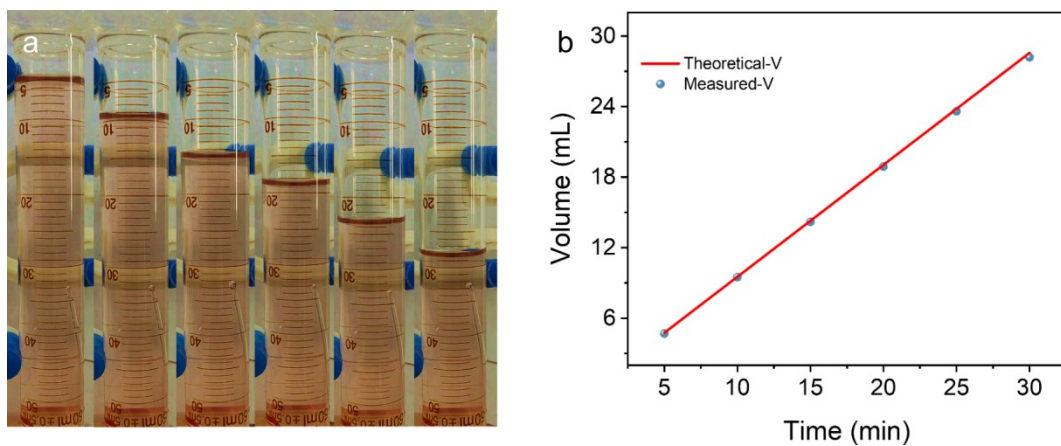


Fig. S28. (a) Digital photographs of the collected O_2 . (b) Comparison between the amount of collected O_2 and theoretical O_2 for SbF_6^- -NiFe LDH/NF at the j of 1000 mA cm^{-2} in $1 \text{ M KOH} + \text{seawater}$.

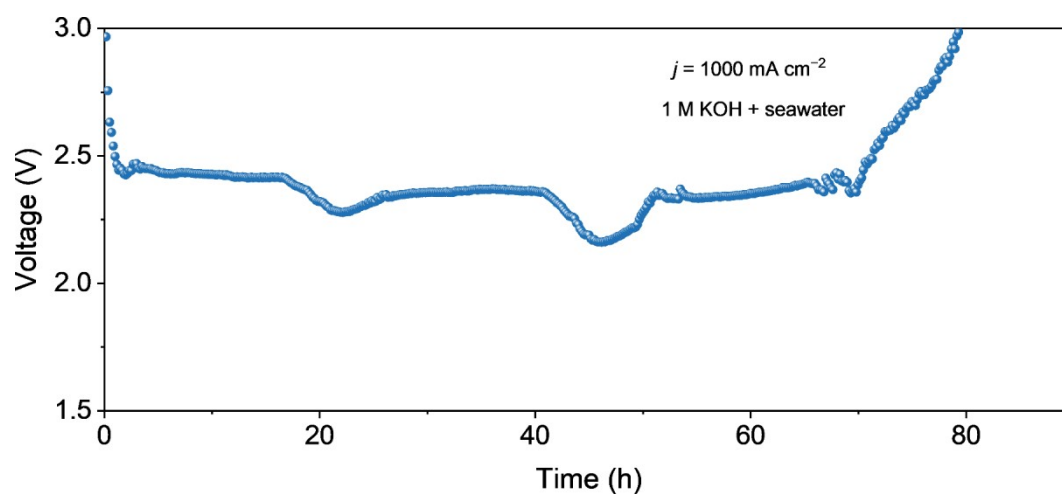


Fig. S29. The chronopotentiometry curve of NiFe LDH/NF||Pt/C/NF tested in 1 M KOH + seawater electrolyte.

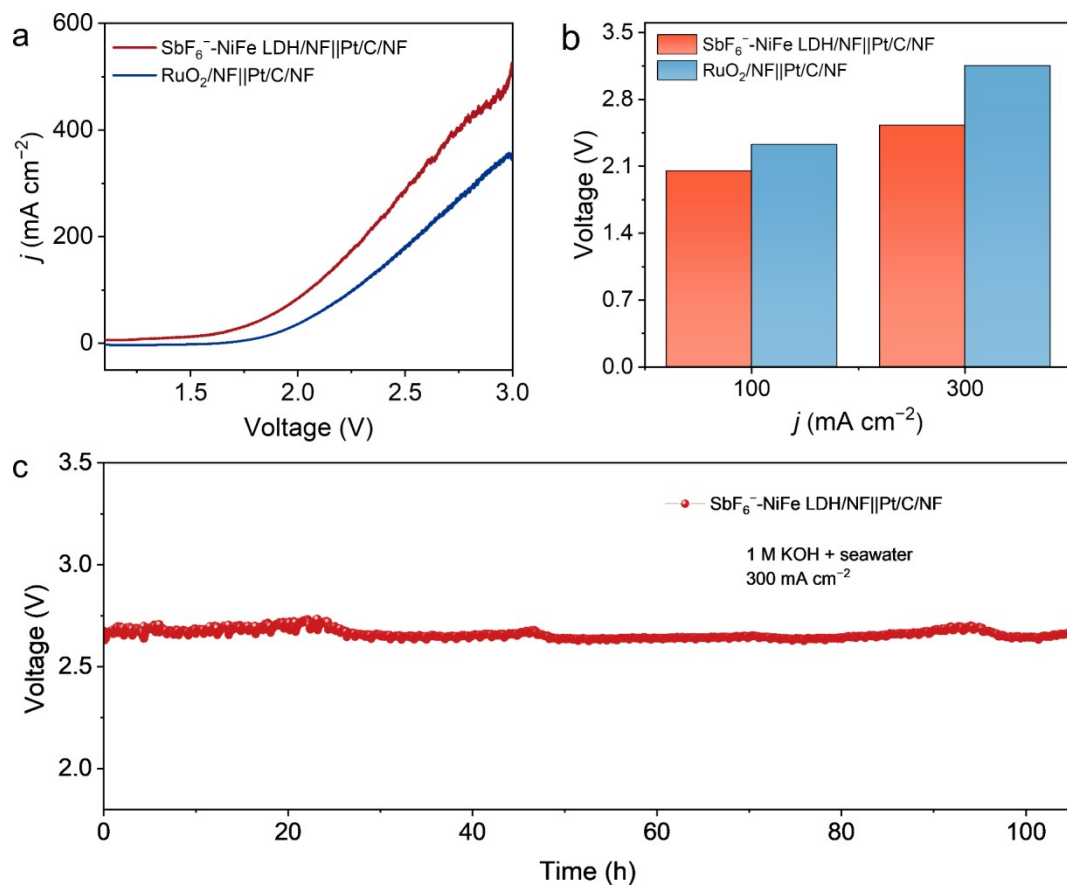


Fig. S30. (a) The AEM-based electrolysis performance of SbF₆⁻-NiFe LDH/NF||Pt/C/NF and the RuO₂/NF||Pt/C/NF pairs in 1 M KOH + seawater. (b) Cell voltages required for the two pairs of electrodes to achieve different j . (c) The chronopotentiometry curve of SbF₆⁻-NiFe LDH/NF||Pt/C/NF tested in 1 M KOH + seawater electrolyte.

Table S1a. Element analysis of SbF_6^- -NiFe LDH by ICP-OES.

Element	Wt. (%)
Ni	22.42
Fe	6.21
Sb	0.67

Table S1b. Element analysis of NiFe LDH by ICP-OES.

Element	Wt. (%)
Ni	21.76
Fe	6.08
Sb	0

Table S1c. Element analysis of SbF_6^- -NiFe LDH after stability test by ICP-OES.

Element	Wt. (%)
Ni	17.99
Fe	5.66
Sb	0.55

Table S2. Comparison of the OER performance of SbF_6^- -NiFe LDH/NF with other reported electrocatalysts.

Catalysts	Electrolyte	η_{1000} (mV)	Stability@ <i>j</i>	Ref.
SbF_6^- -NiFe LDH/NF	1 M KOH + seawater	379	1000 h@1000 mA cm ⁻²	This work
Cr-Co _x P	1 M KOH + seawater	423	140 h@100 mA cm ⁻²	1
FeMo-NiP _x /NF	1 M KOH + 0.5 M NaCl	309	150 h@1000 mA cm ⁻²	2
NiMoS _x @NiFe-LDH/NF	1 M KOH + seawater	350	500 h@500 mA cm ⁻²	3
Mn-doped Ni ₂ P/Fe ₂ P	1 M KOH + seawater	358	200 h@500 mA cm ⁻²	4
Cr-CoFe-LDH/NF	1 M KOH + seawater	369	100 h@500 mA cm ⁻²	5
F-FeCoP _v @IF	1 M KOH + seawater	370	20 h@100 mA cm ⁻²	6
NiFe-LDH@NiFe-Pi/NF	1 M KOH + seawater	370	100 h@1000 mA cm ⁻²	7
NiFe LDH-CeW@NFF	1 M KOH + seawater	386.7	100 h@1000 mA cm ⁻²	8
NiFe-MOF@Ni ₂ P/Ni(OH) ₂ /NF	1 M KOH + seawater	394	/	9
NiFeO-CeO ₂ /NF	1 M KOH + seawater	408	/	10
TS-NiFe LDH/NF	1 M KOH + seawater	412	350 h@1000 mA cm ⁻²	11
NiFe-O NAs/Fe foam	1 M KOH + seawater	427	/	12
Ru/NiFeOOH/NFF	1 M KOH + seawater	430	100 h@100 mA cm ⁻²	13
MnCo/NiSe/NF	1 M KOH + seawater	460	200 h@500 mA cm ⁻²	14
(Ni/Fe/Mo)OOH/NF	1 M KOH + seawater	514	500 h@100 mA cm ⁻²	15
NiFe-LDH/V ₂ CT _x /NF	1 M KOH + seawater	/	110 h@500 mA cm ⁻²	16
MnCo ₂ O ₄ @NiFe-LDH/NF	1 M KOH + 0.5 M NaCl	/	20 h@1000 mA cm ⁻²	17

NiFe/CFD _{0.51}	1 M KOH + seawater	/	600 h@500 mA cm ⁻²	18
NiFe LDH_CO ₃ ²⁻	1 M KOH + seawater	/	100 h@200 mA cm ⁻²	19

η_{1000} represents the overpotentials required to attain a j of 1000 mA cm⁻².

Table S3. Comparison of overall seawater splitting performance for SbF_6^- -NiFe LDH/NF with other reported electrocatalysts.

Catalysts	Electrolyte	j (mA cm^{-2})	Voltage (V)	Ref.
SbF_6^- -NiFe LDH/NF Pt/C/NF	1 M KOH + seawater	500 1000	1.82 2.05	This work
NiMoS_x @NiFe-LDH/NF NiMoS_x /NF	1 M KOH + seawater	500	1.84	3
NiFe-P NAs NiFe-O NAs	1 M KOH + seawater	500 1000	1.86 1.96	12
NiMoN NiFe-PBA	1 M KOH + seawater	500	1.78	20
Ni-MoN SSM	1 M KOH + seawater	500 1000	1.78 1.88	21
CoFe-Ni ₂ P CoFe-Ni ₂ P	1 M KOH + seawater	500	1.84	22
CoP_x CoP_x @FeOOH	1 M KOH + seawater	500	1.86	23
Go@Fe@NiCo@NF Go@Fe@NiCo@NF	1 M KOH + seawater	500 1000	1.94 2.02	24
NiCoHPi@Ni ₃ N/NF NiCoHPi@Ni ₃ N/NF	1 M KOH + seawater	500	1.99	25
HE-LH Pt/C	6 M KOH + seawater	500	2.20	26

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