# **Supporting Information**

# Stable complete seawater electrolysis by using interfacial chloride ion blocking layer on catalyst surface

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#### Methods

**Preparation of Ni-Co@NF.** NF was sonicated in conc. HCL for 3 min to remove the nickel oxide layer on the surface, rinsed with DI water, isopropanol, and acetone, then left to dry in air. Then the 3 NF piece (size 1 X 2.5 cm) was submerged in 100 mL DI water in the beaker; to the above solution, 24 mmol DETA was added, then the above solution kept for 15 min. Next, to the above solution, 4 mmol Co(NO<sub>3</sub>)<sub>2.6H2</sub>O, 2 mmol Ni(NO<sub>3</sub>)<sub>2.6H2</sub>O was added, and the solution was sonicated for 15 min. The above solution kept for 3h, and then a 50 mL 1M KOH solution was added. Then, the mixture was transferred into a 120 mL pressure-tight glass bottle and heated at 120°C for 2h, 4h, 6h, and 8h (for Optimization of Ni-Co-Hydroxide deposition). After that, the bottle was air-cooled to room temperature, and the resulting product was filtered through distilled water washing. Then dried under vacuum at 60°C for 5 h, and the obtained precursor is Ni-Co layered double hydroxide with Co: Ni ratio is approximately 1: 0.12. The obtained product was annealed at 300°C in argon atmosphere (heating rate of 3°C/min) for 2 h, and another one at 500 °C under air for 3 h, and the resulting product named Ni-Co@NF, Ni-Co<sub>3</sub>O<sub>4</sub>@NF respectively. (Loading: 1.8 mg cm<sup>-2</sup>).

**Electrodeposition of graphene-oxide FeOOH onto Ni-Co@NF.** The obtained Ni-Co@NF electrode was submerged in the solution containing graphene oxide (GO) and Fe(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O (GO 1mg/ml, 0.003 M Fe(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O solution in water) for a different time interval (1, 3 h). The electrodeposition was carried in a standard three-electrode electrochemical cell, employing asobtained Ni-Co@NF as a working electrode, Ag/AgCl (1 M KCl) reference electrode and Pt wire as a counter electrode. The electrochemical bath contained Fe(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O (3 X 10<sup>-3</sup>M) and graphene oxide (1 mg/mL) in DI water (50 mL). The deposition potential was controlled

continuously at -1.0 V versus Ag/AgCl for 300, 500, and 900 sec. After the deposition, the obtained composite electrode was rinsed with DI water and ethanol and then left dry in air. Resulting electrode (500 sec) named as GO@Fe@Ni-Co@NF.

**Electrodeposition of FeOOH on Ni-Co@NF.** The electrodeposition was carried in a standard three-electrode electrochemical cell, employing as-obtained Ni-Co@NF as a working electrode, Ag/AgCl (1 M KCl) reference electrode and Pt wire as a counter electrode in the electrochemical bath contained Fe(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O (0.003 M). The deposition potential was controlled continuously at -1.0 V versus Ag/AgCl for 500 sec. After the deposition, the obtained composite electrode was rinsed with DI water and ethanol and then left dry in air. Resulting electrode named as Fe@Ni-Co@NF

**Synthesis of NiFe-LDH on Ni foam (NiFe@NF).** The NiFe-LDH was electrodeposited on Ni foam in a typical three-electrode configuration using Ni foam as the working electrode, Pt mesh as the counter electrode, and Ag/AgCl as the reference electrode. The electrodeposition was carried out at -1.1 V vs. Ag/AgCl for 500 sec in an electrolyte containing 0.04 M Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and 0.04 M Fe(NO3)<sub>3</sub> • 9H2O in 50 mL water, denoted as NiFe-LDH/Nif. The samples were then washed with deionized water several times and dried in air (loading: 1.9 mg cm<sup>-2</sup>).

**Electrodeposition of graphene-oxide NiFe@NF.** The obtained NiFe@NF electrode was submerged in the solution containing graphene oxide (GO) (GO 1mg/ml solution in water) for a 3h. The electrodeposition was carried in a standard three-electrode electrochemical cell, employing as-obtained NiFe@NF as a working electrode, Ag/AgCl (1 M KCl) reference electrode and Pt wire as a counter electrode. The electrochemical bath contained graphene oxide 0.5 mg/mL in 7 pH phosphate buffer. The deposition potential was controlled continuously at -1.1 V versus Ag/AgCl for 500 sec.

Materials characterizations. The FE-SEM images were obtained using JEOL 7500F FESEM. The EDS spectra were recorded in an Oxford Instruments X-Max with the INCA software coupled to the FESEM. TEM images were recorded with a JEOL JEM-2100F. X-ray photoelectron spectroscopy (XPS) measurements were carried out with a Versa Probe II system (Ulvac-PHI) by using a monochromatic Al K $\alpha$  source. The base pressure was below 3×10-10 Torr. The instrument work function was calibrated based on ISO 15472:2001 using Au, Ag, and Cu pure films. The X-ray beam diameter was set to 50 µm, and a charge neutralizaer was used to minimize an undesirable charging effect. The binding energy for all spectra was calibrated by referring the C 1s peak from ubiquitous hydrocarboncontamination to 284.8 eV. XRD pattern was acquired using a D8-Advance instrument (Germany) with Cu-Ka radiation. Raman spectroscopy measurements were recorded using a Renishaw RM 1000-Invia micro-Raman system with an excitation wavelength of 532 nm. Inductively Coupled Plasma-Optical Emission Spectroscopy (Agilent ICP-OES) is used to check dissolved metal ion amount during long term durability test of seawater splitting, using element label Co (238.892 nm), Fe (238.204 nm), Ni (231.604 nm).

**Electrochemical characterization.** VMP3 workstation (Biologic Science Instruments, France) was used to assess electrochemical performance. The two half-reactions of OER and HER were each carried out at room temperature (~25 °C) in a standard three-electrode system with our prepared sample as the working electrode, a Pt mesh as the counter electrode, and a standard Ag/AgCl electrode as the reference electrode. The electrode size is around 0.25 cm<sup>-2</sup>. OER polarization curve measurements were performed by linear sweep voltammetry (LSV) at a scan rate of 2 mV s<sup>-1</sup>, and stability tests were carried out under constant current. HER polarization curve measurements were performed by LSV at a scan rate of 2 mV s<sup>-1</sup>. Electrochemical impedance spectra (EIS) were measured at an overpotential of 240 mV from 0.1 Hz to 100 kHz with an

amplitude of 10 mV. For the two-electrode seawater electrolysis, the as-prepared GO@Fe@Ni-Co@NF electrode was used as anode and cathode. The polarization curves were measured at room temperatures, and stability tests were carried out under constant current densities of 1000 mA cm<sup>-2</sup> at room temperature. For water photolysis using direct sunlight, a commercial Si solar cell with an efficiency of 26.53% was employed and connected with the electrolyzer in series. The current of the water photolysis was recorded by chronoamperometry under AM 1.5G illumination without applying any external bias.

Gas chromatography measurement. To check OER selectivity over COR, gas chromatography (YL6500GC, YL Instruments) tests were performed in a gas-tight electrochemical cell with 1 M KOH + 1 M NaCl as the electrolyte at room temperature (26 °C). A thermal conductivity detector (TCD) was used to detect and quantify the oxygen generated. Chronopotentiometry was applied with a constant current density of 200 mA cm<sup>-2</sup> to maintain oxygen and hydrogen generation. For each measurement over an interval of 5 min, 0.25  $\mu$ L gas sample was carefully extracted from the sealed cell and injected into the GC instrument using a glass syringe.

**Chlorine/hypochlorite determination by o-Tolidine.** The chlorine solubility in water is very high (3.26 g  $L^{-1}$  at 25 °C), which forms hypochlorous acid (HClO) and hydrochloric acid (HCl) (eq. 1)

 $Cl_2 + H_2O \leftrightarrow HCl + HClO$  (1)

Hypochlorous acid is difficult to isolate because of rapid equilibrium with water and Cl<sub>2</sub>, but it can be detected by *o*-tolodine test.<sup>1</sup> (reaction a, Fig. S13) The testing solution contained 10 mL of Deionized (DI) water and 0.5 mL of *o*-tolodine solution (0.05 M in ethanol). The calibration curve for Cl<sub>2</sub> obtained by testing different amounts of freshly generated CL<sub>2</sub> (10, 20, 30, 40, and 50  $\mu$ L) in a testing solution (Fig. S13 b). [Production of Cl<sub>2</sub> was done by mixing 1 g of KMnO<sub>4</sub> and 10 mL of concentrated HCl in a sealed vial for 10 min, and gas was collected using a plunger-inneedle syringe and injected into the testing solution]. Seawater OER/complete splitting experiment was carried out in 50 mL of 1M KOH + 0.5 M NaCl electrolyte using different electrode (OER:-Fe@Ni-Co@NF, GO@Fe@Ni-Co@NF while full seawater splitting GO@Fe@Ni-Co@NF// GO@Fe@Ni-Co@NF). 0.2 mL of the electrolyte was collected after 1h of continuous electrolysis with a constant current condition (1000 mA cm<sup>-2</sup>) and then added into the testing solution. For the determination of hypochlorite, the detection was done by using standard NaOCl. For calibration of ClO<sup>-</sup>, the different molar amount of NaOCl (5.96, 11.92, 17.89, 23.85, 29.82 µmol) was injected to the testing solution.<sup>2</sup> (Fig. S13 c)

**Calibration of reference electrodes and conversion to RHE.** The calibration of Ag/AgCl electrodes was performed in a standard three-electrode system with polished Pt disc and Pt wires as the working and counter electrodes, respectively, and the Ag/AgCl electrode as the reference electrode. Electrolytes are pre-purged and saturated with high purity H<sub>2</sub>. Linear scanning voltammetry (LSV) is then run at a scan rate of 0.5 mV s<sup>-1</sup>, and the potential at which the current crossed zero is taken to be the thermodynamic potential for the hydrogen electrode reactions (Fig. S1). For example, in 1 M KOH, the zero current point is at -0.995 V, so E (RHE) = E (Ag/AgCl) + 0.995 V.

#### **Turnover Frequency (TOF) Calculations for OER**

The turnover frequency is defined as the rate of evolved molecular  $O_2$  per surface active site per second, which can be calculated by the following equation:<sup>3</sup>

$$TOF(s^{-1}) = \frac{\text{number of oxygen turnovers } / \text{ cm}^2 \text{ of geometric area}}{\text{number of active sites} / \text{cm}^2 \text{ of geometric area}}$$

### **TOF for OER:**

The number of oxygen turnover was calculated from the current density according to:

number of oxygen turnover = 
$$\left(j\frac{mA}{cm^2}\right)\left(\frac{1C/s}{1000\ mA}\right)\left(\frac{1\ mol\ e^-}{96485.3\ C}\right)\left(\frac{1\ mol\ O_2}{4\ mol\ e^-}\right) \times 6.023 \times 10^{23}$$

At an overpotential of 260, 303, and 345 mV the GO@Fe@Ni-Co@NF reached a current density of 90.65, 500, and 1000 mA cm<sup>-2</sup>, respectively.

number of oxygen turnover for Ni-Co@NF at 260 mV overpotential

$$\left(90.65\frac{mA}{cm^2}\right)\left(\frac{1C/s}{1000\ mA}\right)\left(\frac{1\ mol\ e^-}{96485.3\ C}\right)\left(\frac{1\ mol\ O_2}{4\ mol\ e^-}\right)\times 6.023\times 10^{23}\ =\ 1.414\ \times\ 10^{17}\ \left(\frac{1/s}{cm^2}\right)$$

number of oxygen turnover for GO@Ni-Co@NF at 303 mV overpotential

$$\left(500\frac{mA}{cm^2}\right)\left(\frac{1C/s}{1000\ mA}\right)\left(\frac{1\ mol\ e^-}{96485.3\ C}\right)\left(\frac{1\ mol\ O_2}{4\ mol\ e^-}\right) \times 6.023 \times 10^{23} = 7.803\ \times 10^{17}\ \left(\frac{1/s}{cm^2}\right)$$

number of oxygen turnover for GO@Fe@Ni-Co@NF 345 mV overpotential

$$\left(1000\frac{mA}{cm^2}\right)\left(\frac{1C/s}{1000\ mA}\right)\left(\frac{1\ mol\ e^-}{96485.3\ C}\right)\left(\frac{1\ mol\ 0_2}{4\ mol\ e^-}\right)\times 6.023\times 10^{23} = 15.606\ \times 10^{17}\ \left(\frac{1/s}{cm^2}\right)$$

Based on the electrochemical active surface area along with the unit cell (volume of 142.65 Å<sup>3</sup>) of FeHO<sub>2</sub> crystal structure (Fe is considered as the active site) in the case of in the GO@Fe@Ni-Co@NF catalyst active sites per real surface area:

Active sites = 
$$\left(\frac{2 \text{ atom/unit cell}}{142.65/\text{unit cell}}\right)^{2/3}$$

 $= 0.058144 \text{ x } 10^{16} \text{atoms cm}^{-2}_{\text{real}}$ 

$$TOF(n260) = \frac{1.414 \times 10^{17} \left(\frac{1/s}{cm^2}\right)}{Surface \ sites \ \times ECSA}$$
$$TOF(n260) = \frac{14.14 \times 10^{16} \left(\frac{1/s}{cm^2}\right)}{0.058144 \times 10^{16} \ atoms \ cm^{-2} real \times 3266.25 cm^2} = 0.02 \ s^{-1}$$
$$TOF(n303) = \frac{78.03 \times 10^{16} \left(\frac{1/s}{cm^2}\right)}{0.058144 \times 10^{16} \ atoms \ cm^{-2} real \times 3266.25 cm^2} = 0.10 \ s^{-1}$$
$$TOF(n345) = \frac{156.06 \times 10^{16} \left(\frac{1/s}{cm^2}\right)}{0.058144 \times 10^{16} \ atoms \ cm^{-2} real \times 3266.25 cm^2} = 0.21 \ s^{-1}$$



Fig. S1. LSV plot for calibration of Ag/AgCl electrodes.



Fig. S2. A thermo-kinetically controlled synthesis of Ni-Co hydroxide and via thermal relaxation of the DETA-M complex. Here, the spontaneous formation of metaldiethylenetriamine (DETA) complex takes place, and the cage-like complex of  $M^{2+}$  (Co/Ni) ion can withstand in a strongly alkaline environment from hydroxide precipitation at room temperature. When the complex alkaline solution is heated, the DETA ligand relaxation takes place, which provides free access to OH<sup>-</sup> ion towards  $M^{2+}$  ion leading to the slow nucleation and growth of metal hydroxide.



**Fig. S3. Optimization of metal hydroxide deposition by hydrothermal deposition time** [the corresponding SEM images of Ni-Co hydroxide on Ni foam synthesized with 2 h (2a-2d), 4 h (4a-4d), 6 h (6a-6d), and 8 h (8a-ad), hydrothermal time interval. As depicted in Fig. S3, after 2h, the formation of the micro-rod array on the surface of Ni foam can be seen. As hydrothermal reaction time increases, the secondary growth of petal-like Ni-Co hydroxide takes place (4h), such a rod-like structure with the surface formed by petal can effectively improve the active surface area of the catalyst which is very crucial for generating high current density for OER/HER. However, when the hydrothermal time increased more than 4h, the additional precipitation of NI-Co hydroxide occurs, which blocks the 3D network of Ni foam. So, 4 h hydrothermal samples was used for the next experiments.]



Fig. S4. *In-situ* XRD measurement. XRD patterns of the as-prepared Ni-Co hydroxide (4h hydrothermal time) at different temperatures. [The peak highlighted in grey color background correspond to nickel foam, In the XRD pattern of Ni-Co hydroxide shows two prominent central peaks at 18.88 Å (d spacing 0.476 nm, calculated from Bragg's law) and 37.72 Å. After 250 °C metal oxide phase starts to form. As the temperature increases, the metal oxide phase becomes dominant.[# peaks correspond to  $\beta$  Co<sub>1</sub>-Ni<sub>0.12</sub> (OH)<sub>2</sub>,<sup>4,5 §</sup> peaks correspond to Ni-doped Co<sub>3</sub>O<sub>4</sub>,<sup>6</sup>, the peak highlighted in the grey color background corresponds to nickel foam.]



Fig. S5. (a) FE-SEM image and (b) EDS data of Ni-Co hydroxide (4 h sample).



**Fig. S6.** (a) HR-TEM and (b, c) TEM-EDS elemental mapping of Ni-Co@NF [d spacing ~ 0.474 nm measured in HR-TEM is consistence with XRD results.]



**Fig. S7. Graphene oxide deposition.** 1 h (a, b) graphene oxide deep coating, 3 h (c, d) graphene oxide deep coating followed by electrodeposition of FeOOH for 500 sec.



**Fig. S8.** Raman spectra of GO@Fe@Ni-Co@NF catalyst at different electrodeposition time. [The characteristic D and G band of graphene oxide (GO) can be seen at 1339 and 1578 cm<sup>-1,</sup> respectively.<sup>7</sup> There was no shift observed in the D and G band peak with increasing electrodeposition time, but as electrodeposition time increase, the intensity of G band increases. The I<sub>D</sub>/I<sub>G</sub> ratio at 0, 300, 500, and 900 sec were 1.00, 0.998, 0.985, and 0.931, respectively.] As the electrodeposition time increases, the decrease in I<sub>D</sub>/I<sub>G</sub> ratio was found to decrease. This indicates a considerable recovery of the conjugated graphitic framework<sup>8</sup> upon reduction/defunctionalization of hydroxyl, epoxies, or other oxygen-containing groups during the electrodeposition process.



**Fig. S9.** (a) Optimization of annealing temperature metal hydroxide for OER. (b) Optimization of FeOOH/GO electrodeposition time for OER. (LSV curve without iR compensation)



Fig. S10. XRD pattern of Ni-Co@NF, GO@Ni-Co@NF, Fe@Ni-Co@NF, and GO@Fe@Ni-Co@NF electrode. [\* peaks correspond to  $\beta$  Ni<sub>1</sub>-Co<sub>2.9</sub> (OH)<sub>2</sub>,<sup>4,5</sup> # peaks correspond to Ni-doped Co<sub>3</sub>O<sub>4</sub>,<sup>6</sup>, the peak highlighted in the grey color background corresponds to nickel foam.]



**Fig. S11.** XPS full survey (a) and O 1s (b) XPS spectra of Ni-Co@NF, Fe@Ni-Co@NF, and GO@Fe@Ni-Co@NF catalyst.



**Fig. S12.** Nyquist plots obtained by EIS for screened OER catalyst at 0.4 V vs. Ag/AgCl in 1M KOH



**Fig. S13.** CV plots of Ni-Co@NF (a), GO@Ni-Co@NF (b), Fe@Ni-Co@NF (c), and GO@Fe@Ni-Co@NF (d) at different scan rates in 1M KOH



**Fig. S14.** Current (recorded at a fixed potential) as a function of scan rate for Ni-Co@NF (a), GO@Ni-Co@NF (b), Fe@Ni-Co@NF (c), and GO@Fe@Ni-Co@NF (d)



Fig. S15. GC data of gas collected over GO@Fe@Ni-Co@NF anode surface.



**Fig. S16.** *o***-tolidine test for Cl<sub>2</sub>, HCIO, NaOCl.** (a) The chemical reaction between *o*-tolidine and HCIO.<sup>1</sup> The chemical reaction between o-tolidine and NaOCl, leading to the formation of semiquinone or haloquinone.<sup>2</sup> (b) UV-vis spectra of the testing solutions after the addition of different amounts of Cl<sub>2</sub>. (c) UV-vis spectra of the testing solutions after addition of different amounts of NaOCl. (d) Calibration curve obtained by plotting the concentration of free chlorine against the corresponding absorption peak intensity at 410 nm. (e) Calibration curve obtained by plotting the concentration of NaOCl against the corresponding absorption peak intensity at 410 nm. (e) Calibration curve obtained by plotting the concentration of NaOCl against the corresponding absorption peak intensity at 440 nm. (f) Photographs of the testing standard solutions (Cl<sub>2</sub> and NaOCl). (g) UV-vis spectra of the testing solutions after the addition of electrolyte (0.2 mL) sampled from different OER systems (Fe@Ni-Co@NF, GO@Fe@Ni-Co@NF) after 1 hours of continuous seawater electrolysis at 1000 mA cm<sup>-2</sup> current density.



**Fig. S17.** FE-SEM, EDS, and elemental mapping of Fe@Ni-Co@NF electrode after 12 h OER stability test at 1000 mA cm<sup>-2</sup> current density in simulated alkaline seawater.



**Fig. S18.** FE-SEM, EDS, and elemental mapping of GO@Fe@Ni-Co@NF electrode after 12 h OER stability test at 1000 mA cm<sup>-2</sup> current density in simulated alkaline seawater.



**Fig. S19.** HER LSV plot of GO@Fe@Ni-Co@NF in 1M KOH + 0.5M NaCl (simulated alkaline seawater) at 2 mV sec<sup>-1</sup> scan rate. [LSV curve without iR compensation)]



Fig. S20. LSV spectra (without iR compensation) of GO@Fe@Ni-Co@NF(+) // GO@Fe@Ni-Co@NF(-) electrolyzer in different electrolyte at 25 °C and 60 °C



**Fig. S21.** Identification of insoluble precipitates. EDX spectra of the GO@Fe@Ni-Co@NF catalyst a) before, and b) after seawater electrolysis.



**Fig. S22.** EIS spectra of GO@Fe@Ni-Co@NF// GO@Fe@Ni-Co@NF electrolyzer after and before testing in 1M KOH + 0.5M NaCl (simulated alkaline seawater) for 378 h at 1000 mA cm<sup>-2</sup> current density.



Fig. S23. Schematic presentation of working of HER catalyst with GO outer layer in alkaline seawater.



**Fig. S24.** FE-SEM, EDS elemental mapping of GO@Fe@Ni-Co@NF cathode after 378 h overall simulated alkaline seawater splitting test at 1000 mA cm<sup>-2</sup> current density.



**Fig. S25.** TEM images of GO@Fe@Ni-Co@NF anode catalyst before (a, b) and after (c, d) 378 h CP test at 1000 mA cm<sup>-2</sup> in alkaline seawater.



**Fig. S26.** FE-SEM, EDS, and elemental mapping images of GO@Fe@Ni-Co@NF anode after 378 h overall simulated alkaline seawater splitting test at 1000 mA cm<sup>-2</sup> current density.



**Fig. S27.** XPS C 1s spectra of GO@Fe@Ni-Co@NF anode after and before testing in 1M KOH + 0.5M NaCl (simulated alkaline seawater) for 378 h at 1000 mA cm<sup>-2</sup> current density in 2 electrode configuration.



**Fig. S28.** Faradic efficiency (FE) measurement of GO@Fe@Ni-Co@NF as OER and HER catalyst using three-electrode assembly. Pt mesh as a counter electrode, Ag/AgCl as a reference electrode, and GO@Fe@Ni-Co@NF as a working electrode. (a) Measured H<sub>2</sub> amount and FE of HER over time in 1M NaCl + 1 M KOH at current densities of -200 mA cm<sup>-2</sup>. (b) measured O<sub>2</sub> amount and FE of OER over time in 1M NaCl + 1 M KOH at current densities of -200 mA cm<sup>-2</sup>. (c) Reaction setup for gas measurement.



**Fig. S29.** IV plot of commercial Si solar cell (the J-V characteristic curve of the commercial Si solar cell in the dark and under 100 mW cm<sup>-2</sup> illumination, demonstrating a short-circuit photocurrent density, open-circuit voltage, and film factor of 17.22 mA cm-2, 2.275 V, and 0.677, respectively, yielding a power conversion efficiency (PCE) of 26.53%.)



Fig. S30. FE-SEM images of Ni-FeLDH@NF (a, b, c) and GO@Ni-FeLDH@NF (d, e, f) electrode

## **Faradic efficiency measurement:**

The Faradic efficiency was calculated using the above equation:

$$Faradaiv \ efficiency = \frac{experimental \ \mu mol \ of \ O_2 \ or \ H_2 \ gas}{Theoretical \ \mu mol \ of \ O_2 \ or \ H_2 \ gas} \times 100$$

The theoretical amount of O<sub>2</sub> and H<sub>2</sub> gas was calculated from Faraday's law:

$$n = \frac{I \times t}{z \times F}$$
$$n = \frac{Q}{z \times F}$$

where *n* is the number of mol, *I* is the current in ampere, *t* is the time in seconds, *z* is the transfer of electrons (for O<sub>2</sub> z = 4 and H<sub>2</sub> z = 2), and F is the Faraday constant (96485. 3329 C mol<sup>-1</sup>). Q is charge. And the experimental amount of O<sub>2</sub> and H<sub>2</sub> gas measurement method mentioned in Fig. S 28.

Time	H <sub>2</sub> volume	H <sub>2</sub> µmol	Q	n (H <sub>2</sub> µmol	<b>FE<sub>HER</sub></b>
min	mL	Experimental		Theoretical)	
5	1.9	78.70567	15.96	82.70687	95.16219
10	3.8	157.4113	31.2	161.6826	97.35824
15	5.7	236.117	46.93	243.1976	97.08855
20	7.7	318.9651	62.58	324.298	98.35555
25	9.7	401.8132	78.3	405.7612	99.02701

Table S1. Faradic efficiency for HER

Time	O <sub>2</sub> volume	O <sub>2</sub> µmol	Q	n (O <sub>2</sub> µmol	<b>FE<sub>OER</sub></b>
min	mL	Experimental		Theoretical)	
5	0.95	39.4929345	15.67	40.60203	97.26838
10	1.95	81.06444449	31.99	82.88825	97.79968
15	2.95	122.6359545	48.38	125.3558	97.83026
20	3.95	164.2074645	64.7	167.6421	97.95123
25	4.95	205.7789745	81.53	211.2497	97.41029

**Table S3** ICP-OES analysis results of the electrolyte after 12 h CP test at 1000 mA cm<sup>-2</sup> current density test in simulated alkaline seawater

Electro-catalyst	Element Label (nm)	Conc(mg kg <sup>-1</sup> )
Fe@Ni-Co@NF	Со	0.0391
	Fe	0.0293
	Ni	0.0015
GO@Fe@Ni-Co@NF	Со	0.0092
	Fe	0.0315*
	Ni	0.00002

(\*The Fe dissolution concentration is slightly high, which is possibly because of the FeOOH particle, those are deposited on outer graphene oxide layer during electrodeposition)

Table S4 Reported seawater OER catalyst

No	Anode	Electrolyte	OER overpotential	Reference
			mv	
1	GO@Fe@Ni-Co@NF	1M KOH + 0.5 M NaCl	$n_{50} = 247$	This work
			$n_{500} = 303$	
			$n_{1000} = 345$	
2	NiFe/NiSx-Ni	1M KOH + 0.5 M NaCl	$n_{400} = 300$	9
3	NCFPO/C@CC	0.1M KOH +0.5 M NaCl	$n_{50} = 330$	10
4	NiMoN@NiFeN	1M KOH+ natural	$n_{500} = 347$	11
		seawater		
5	NiFe LDH	1M KOH + 0.5 M NaCl	$n_{10} = 359$	12
6	NiP <sub>2</sub> /CC	Natural seawater	$n_4 = 581$	13
7	NiCo-DEA	Natural seawater	$n_{100} = \sim 670$	14
8	CaFeO <sub>x</sub> /FePO <sub>4</sub>	phosphate 0.5 M, NaCl	$n_{10} = 710$	15
		0.36 M and MgCl <sub>2</sub> 0.07 M		
9	S-doped Ni/Fe	1M KOH+ natural	$n_{500} = 398$	16
	(oxy)hydroxide	seawater		

**Table S5** ICP-OES analysis results of GO@Fe@Ni-Co@NF(+) // GO@Fe@Ni-Co@NF(-)electrolyzer electrolyte after 378 h CP at 1000 mA  $cm^{-2}$  current density test in simulated alkalineseawater

Electrolyzer	Element Label (nm)	Conc(mg kg <sup>-1</sup> )
GO@Fe@Ni-Co@NF(+) // GO@Fe@Ni-Co@NF(-)	Со	0.0104
	Fe	0.0332
	Ni	0.0051

**Movie S1** GO@Fe@Ni-Co@NF// GO@Fe@Ni-Co@NF Seawater electrolyzer testing at 1000 mA cm<sup>-2</sup> current density.

#### References

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