Solar-driven, highly sustained splitting of seawater into hydrogen and oxygen fuels

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Electrolysis of water to generate hydrogen fuel is an attractive renewable energy storage technology. However, grid-scale freshwater electrolysis would put a heavy strain on vital water resources. Developing cheap electrocatalysts and electrodes that can sustain seawater splitting without chloride corrosion could address the water scarcity issue. Here we present a multilayer anode consisting of a nickel–iron hydroxide (NiFe) electrocatalyst layer uniformly coated on a nickel sulfide (NiSx) layer formed on porous Ni foam (NiFe/NiSx-Ni), affording superior catalytic activity and corrosion resistance in solar-driven alkaline seawater electrolysis operating at industrially required current densities (0.4 to 1 A/cm²) over 1,000 h. A continuous, highly oxygen evolution reaction-active NiFe electrocatalyst layer drawing anodic currents toward water oxidation and an in situ-generated polyatomic sulfate and carbonate-rich passivating layers formed in the anode are responsible for chloride repelling and superior corrosion resistance of the salty-water-splitting anode.

Significance

Electrolysis of water to generate hydrogen fuel could be vital to the future renewable energy landscape. Electrodes that can sustain seawater splitting without chloride corrosion could address the issue of freshwater scarcity on Earth. Herein, a hierarchical anode consisting of a nickel–iron hydroxide electrocatalyst layer uniformly coated on a sulfide layer formed on Ni substrate was developed, affording superior catalytic activity and corrosion resistance in seawater electrolysis. In situ-generated polyanion-rich passivating layers formed in the anode are responsible for chloride repelling and high corrosion resistance, leading to new directions for designing and fabricating highly sustained seawater-splitting electrodes and providing an opportunity to use the vast seawater on Earth as an energy carrier.


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Adsorption of $\text{Cl}^-$ by surface polarization:
\[
\text{M} + \text{Cl}^- \rightarrow \text{MCl}_{\text{ads}} + \text{e}^- \tag{3}
\]

Dissolution by further coordination:
\[
\text{MCl}_{\text{ads}} + \text{Cl}^- \rightarrow \text{MCl}^-_x \tag{4}
\]

Conversion from chloride to hydroxide:
\[
\text{MCl}^-_x + \text{OH}^- \rightarrow \text{M(OH)}_x + \text{Cl}^- \tag{5}
\]

To avoid relying on costly desalination processes, development of electrodes that are corrosion-resistant for splitting seawater into $\text{H}_2$ and $\text{O}_2$ is crucial to the advancement of seawater electrolysis, an undertaking with limited success thus far. Mn-based oxides deposited on $\text{IrO}_2$-Ti showed stability for OER in NaCl-containing electrolytes but were limited by its high cost (14–16). NiFe-layered double hydroxide (NiFe-LDH) (17), Co-borate, and Co-phosphate materials have all shown high OER activity in NaCl-containing electrolytes, but long-term stability at industrial current densities $\sim 1 \text{ A/cm}^2$ has not been achieved for seawater electrolysis (11, 18, 19).

This work developed a multilayer electrode that upon activation evolved into a polyanion sulfate/carbonate-passivated NiFe/ NiS$_x$-Ni foam anode with high activity and corrosion resistance for OER in chloride-containing alkaline electrolytes. The negatively charged polyanions incorporated into the anode were derived from anodization of the underlying nickel sulfide layer and carbonate ions in the alkaline solution, repelling $\text{Cl}^-$ anions in seawater and hence imparting corrosion resistance. When the anode is paired with an advanced Ni-NiO-Cr$_2$O$_3$ electrolyte, the electrolyzer can operate at low voltages and high currents and last for more than 1,000 h.

The NiFe/NiS$_x$-Ni foam anode (referred to as Ni$^3$ for brevity) was made by first converting the surface of Ni foam to NiS$_x$ by devising a solvothermal reaction of Ni foam with elemental sulfur in toluene (SI Appendix, Materials and Methods). After formation of the NiS$_x$ layer, an OER-active NiFe hydroxide (20–24) was electrodeposited via the reduction of nitrate from a solution of Ni(NO$_3$)$_2$, and Fe(NO$_3$)$_2$ (Ni:Fe = 3:1) (Fig. 14) (20, 25). We propose that the matched crystalline phase and $\delta$ spacing could allow epitaxial growth of NiFe hydroxide laminates on NiS$_x$ surface, resulting in a uniform coating of vertically grown LDH on top of the NiS$_x$ layer (Fig. 1C and SI Appendix, Fig. S1). Electron diffraction patterns revealed that the NiS$_x$ and NiFe layers were amorphous in nature (SI Appendix, Fig. S1). SEM (Fig. 1B and C) and cross-sectional elemental mapping revealed a $\sim 1$- to 2-$\mu$m-thick NiS$_x$ layer formed on Ni foam (SI Appendix, Fig. S2), with a $\sim 200$-nm-thick NiFe layer on top of the NiS$_x$ layer (Fig. 1D and E).

After anodic activation of the Ni$^3$ anode in an alkaline simulated seawater electrolyte (1 M KOH plus 0.5 M NaCl, a mimic of seawater adjusted to alkaline), OER performance was measured in a three-electrode configuration in a freshly prepared alkaline simulated seawater electrolyte (Fig. 24). Cyclic voltammetry (CV) showed a 30 mV lower onset overpotential (taken to be the potential vs. RHE where the reverse scan reaches 0 mA/cm$^2$), an improvement over the original electrodeposited NiFe catalyst and NiFe-LDH (17, 20, 21). The large increase in the Ni$^{2+}$ $\rightarrow$ Ni$^{3+}$ oxidation peak ($\sim 1.44$ V; Fig. 24) suggested increased electrochemically active nickel sites for OER through the activation process. A high current density of 400 mA/cm$^2$ at an overpotential of $\eta = 510$ mV was reached (no iR compensation, $R = 0.7 \pm 0.05$ ohm; Fig. 2B). After iR compensation, the actual overpotential applied on the Ni$^3$ anode to achieve an OER current density of 400 mA/cm$^2$ was $\sim 0.3$ V, well below the 0.49 V overpotential required to trigger chloride oxidation to hypochlorite.

Fig. 1. Fabrication and structure of the dual-layer NiFe/NiS$_x$-Ni foam (Ni$^3$) anode for seawater splitting. (A) Schematic drawing of the fabrication process, including a surface sulfuration step and an in situ electrodeposition of NiFe. (B–D) SEM images of untreated nickel foam, NiS$_x$ formed on nickel foam, and electrodeposited NiFe on the NiS$_x$ surface. (E) Elemental mapping of a cross-section of NiFe/NiS$_x$ on an Ni wire in the Ni foam, revealing Ni wire, NiS$_x$, and NiFe layers.

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We then paired the activated/passivated Ni$_3$ anode with a highly active Ni-NiO-Cr$_2$O$_3$ hydrogen evolution reaction (HER) cathode (26) for two-electrode high-current electrolysis of alkaline seawater. Three-electrode linear scan voltammetry of the Ni-NiO-Cr$_2$O$_3$ showed that −0.37 V overpotential was required to drive an HER current density of 500 mA/cm$^2$ (SI Appendix, Fig. S3A, without iR compensation). After iR compensation, the overpotential applied on the catalyst to drive a current density of 500 mA/cm$^2$ was as low as ~160 mV, which was among the best nonprecious-metal-based HER catalysts reported so far. In addition, chloride did not degrade the activity and stability during the HER process (SI Appendix, Fig. S3B). The two-electrode electrolysis experiment was first carried out in 1 M KOH added to seawater from the San Francisco Bay at room temperature (23°C). The electrolyzer operated at a current density of 400 mA/cm$^2$ under a voltage of 2.12 V (Fig. 2C, without iR compensation; $R = 0.95 \pm 0.05$ ohm) continuously for more than 1,000 h without obvious decay (Fig. 2D), corroborated by three-electrode measurements before and after the 1,000-h stability test (Fig. 2B).

In a real electrolysis application, salt may accumulate in the electrolyte if seawater is continuously fed to the system and water is converted to H$_2$ and O$_2$. To this end, we investigated electrolytes with higher NaCl concentrations than in seawater, using deionized water with 1 M KOH + 1 M NaCl or even 1.5 M NaCl (Fig. 2D and SI Appendix, Fig. S4). Meanwhile we increased NaCl concentration during Ni$_3$ anodic activation/passivation to match the conditions of the stability test (i.e., the second phase of anodization would use 1 M KOH + 1 M or 1.5 M NaCl if the stability test was going to be carried out in this electrolyte). Due to the higher ionic strengths with NaCl concentrations of 1 M and 1.5 M, the cell resistance decreased and the voltages of the electrolyzers using the activated Ni$_3$ anode afforded a current density of 400 mA/cm$^2$ at lower cell voltages of 2.09 V and 2.02 V, respectively. The electrolysis was still stable for more than 1,000 h, with no obvious corrosion or voltage increase observed, suggesting impressively active and stable anode for electrolysis in high-salinity water. Further, the electrolyzer operated stably under conditions typically used in industry (high temperature and concentrated base) (27), needing only 1.72 V to reach a current density of 400 mA/cm$^2$ in 6 M KOH + 1.5 M NaCl at 80°C for >1,000 h (Fig. 2 C and D). Mass spectrometry showed no anodic Cl$_2$ evolution and gas chromatography showed a relative Faradaic efficiency (R FE, defined as the ratio of O$_2$ produced in KOH + NaCl electrolyte over O$_2$ produced in KOH electrolyte) of O$_2$ production of ~100% (SI Appendix, Figs. S5 and S6), suggesting selective OER in alkaline-adjusted salty water using the passivated Ni$_3$ anode.

We performed control experiments and found that in a “harsh” electrolyte of 1 M KOH with a high concentration of 2 M NaCl, an activated Ni$_3$ anode paired with an Ni-NiO-Cr$_2$O$_3$ cathode lasted for ~600 h (SI Appendix, Fig. S7A) before breakdown under a constant current of 400 mA/cm$^2$ (during which R FE was ~99.9% for O$_2$; SI Appendix, Fig. S7C). Bare Ni foam and sulfur-treated Ni foam (to form NiS) without NiFe lasted for less than 20 min (SI Appendix, Fig. S7A, Inset) with R FE <35% for O$_2$ (SI Appendix, Fig. S7C). NiFe hydroxide-coated Ni foam without the NiS interlayer was also inferior to Ni$_3$ and lasted 12 h (SI Appendix, Fig. S7A) with an R FE for O$_2$ of 99% at 400 mA/cm$^2$ (SI Appendix, Fig. S7C). These results together with three-electrode constant voltage testing (SI Appendix, Fig. S7D) suggested that the

**Fig. 2.** Sustained, energy-efficient seawater splitting continuously over 1,000 h. (A) CV scans of an Ni$_3$ anode before and after activation in 1 M KOH at 400 mA/cm$^2$ for 12 h and 1 M KOH + 0.5 M NaCl for 12 h at 400 mA/cm$^2$; the CV curves were taken in 1 M KOH, resistance 0.75 \pm 0.05 ohm. (B) CV scans of an Ni$_3$ anode (activated in 1 M KOH at 400 mA/cm$^2$ for 12 h followed by 1 M KOH + 0.5 M NaCl at 100 mA/cm$^2$ for 12 h) before and after 1,000-h seawater splitting in an alkaline simulated seawater electrolyte (1 M KOH with 0.5 M NaCl in deionized water), $R = 0.7 \pm 0.05$ ohm. (C) Linear sweep voltammetry (LSV) scans of a seawater-splitting electrolyzer (Ni$_3$ paired with an Ni-NiO-Cr$_2$O$_3$ cathode) taken in alkaline seawater electrolyte (1 M KOH + real seawater) at room temperature (23°C, resistance 0.95 \pm 0.05 ohm) and in near-saturated salt concentration (1.5 M NaCl) under industrial electrolysis conditions (6 M KOH electrolyte at 80°C, resistance 0.55 \pm 0.05 ohm). The Ni$_3$ anode was first activated under 400 mA/cm$^2$ in 1 M KOH for 12 h followed by 1 M KOH + 0.5 M NaCl at 100 mA/cm$^2$ for 12 h. (D) Durability tests (1,000 h) recorded at a constant current of 400 mA/cm$^2$ of the seawater-splitting electrolyzer under 1 M KOH + real seawater at room temperature ($R = 0.95 \pm 0.05$ ohms), 1 M KOH + 1.5 M NaCl at room temperature ($R = 0.8 \pm 0.05$ ohms), and 6 M KOH electrolyte at 80°C ($R = 0.55 \pm 0.05$ ohms), respectively. Data were recorded after activation of Ni$_3$ anode under 400 mA/cm$^2$ in both 1 M KOH and 1 M KOH + 0.5 M NaCl (or 1.5 M NaCl for the test in 1 M/6 M KOH + 1.5 M NaCl) electrolytes for 12 h.
combination of NiFe hydroxide on top of NiS$_x$/Ni to form a Ni$^3^+$ structure was key to superior chloride corrosion resistance.

The electrode structures (before and after seawater splitting in various electrolytes) were investigated by 3D X-ray microtomography (SI Appendix, Fig. S8). After 1,000 h of seawater electrolysis, the Ni$^3^+$ anode showed a structural integrity (SI Appendix, Fig. S8B) similar to that before electrolysis (SI Appendix, Fig. S8A). Even under a harsh condition with four times the salt concentration of real seawater for 300 h, the anode still maintained the Ni foam skeleton structure (SI Appendix, Fig. S8C). However, Ni foam without NiS$_x$ but with electrodeposited NiFe (the best control sample) showed severe corrosion after only an 8-h test in 1 M KOH + 2 M NaCl (SI Appendix, Fig. S8D). Clearly, both the NiFe OER catalyst coating and the nickel sulfide layer underneath the catalyst are critical to long-term stability of the anode against chloride corrosion.

Given that the activated Ni$^3^+$ anodes were quite stable in various electrolytes under the current density of 400 mA/cm$^2$, we then tested the system at even higher current densities. Three-electrode testing of an activated Ni$^3^+$ electrode in simulated alkaline seawater electrolyte showed a 750 mV overpotential (without iR compensation) for reaching a high OER current density of 1,500 mA/cm$^2$ (Fig. 3A). After iR compensation, the actual overpotential applied on the Ni$^3^+$ anode to achieve the OER current density of 1500 mA/cm$^2$ in simulated alkaline seawater was ~0.38 V (Fig. 3B), still

![Fig. 3. Seawater electrolysis running at current density up to 1 A/cm$^2$.](image-url)

(A) CV scans of a 0.25-cm$^2$ Ni$^3^+$ anode before and after activation in 1 M KOH and 1 M KOH + 0.5 M NaCl (both at 400 mA/cm$^2$); the CV curves were taken in simulated alkaline seawater (1 M KOH + 0.5 M NaCl), resistance 1.2 ± 0.05 ohm. (B) CV scans with and without iR compensation of the 0.25-cm$^2$ Ni$^3^+$ anode shown in A. (C) Two electrode RFEs of oxygen generation in seawater electrolyzer (Ni$^3^+$ paired with Ni-NiO-Cr$_2$O$_3$) running at 400 mA/cm$^2$, 800 mA/cm$^2$, and 1,000 mA/cm$^2$ in 1 M KOH + 0.5 M NaCl electrolyte. (D) Durability tests of the seawater-splitting electrolyzer (0.5 cm$^2$ Ni$^3^+$ paired with Ni-NiO-Cr$_2$O$_3$) recorded in 1 M KOH + 0.5 M NaCl electrolyte at room temperature under constant currents of 400 mA/cm$^2$ ($R = 1.5 ± 0.05$ ohms), 800 mA/cm$^2$ ($R = 1.6 ± 0.05$ ohms), and 1,000 mA/cm$^2$ ($R = 1.6 ± 0.05$ ohms), respectively. Data were recorded after activation of Ni$^3^+$ anode under 400 mA/cm$^2$ in both 1 M KOH and 1 M KOH + 0.5 M NaCl electrolytes for 12 h. (E) Current density–potential curve (J–V) of the seawater electrolyzer and two perovskite tandem cells under dark and simulated AM 1.5-G 100 mW/cm$^2$ illumination. The illuminated surface area of each perovskite cell was 0.12 cm$^2$ (0.24 cm$^2$ total), and the catalyst electrode areas (geometric) were 1 cm$^2$ each. The Ni$^3^+$ were first activated in 1 M KOH under 400 mA/cm$^2$ for 12 h then in 1 M KOH + 0.5 M NaCl under 100 mA/cm$^2$ for 12 h. After that the electrolyzer was held at 20 mA/cm$^2$ for 5 h before pairing with the solar cell. (F) Twenty-hour stability test of perovskite solar cell-driven seawater electrolysis and corresponding solar-to-hydrogen (STH) efficiency. (G) A photo showing a commercial silicon solar cell-driven electrolysis (1-cm$^2$ electrodes) of seawater running at 876 mA under a voltage of 2.75 V ($R = 1.0 ± 0.05$ ohms). No iR compensation was applied to any experiment.
110 mV lower than the 0.49 V threshold for chloride oxidation. The R_FE tests running at 400, 800, and 1,000 mA/cm² all showed nearly 100% for oxygen generation (Fig. 3C). Two-electrode electrolysis in 1 M KOH + 0.5 M NaCl showed a cell voltage of 2.06 V, 2.27 V, and 2.44 V for a current density of 400, 800, and 1,000 mA/cm² (Fig. 3D), respectively, all without obvious performance decay for more than 500 h at such high currents.

Driving electrolysis with inexpensive solar cells is an attractive way to generate hydrogen using a carbon-free energy source. To this end, we paired an activated Ni³⁺ anode with an Ni-NiO-Cr₃O₇ cathode in 1 M KOH + 0.5 M NaCl and the cell was allowed to operate for 5 h at 20 mA/cm², after which the electrolyzer was connected in series with two 0.2-cm² perovskite solar cells (28) (0.12-cm² masked area, single-cell maximum power point tracking data in SI Appendix, Fig. S9) connected in series as a side-by-side tandem cell. Under AM 1.5-G 100 mW/cm² simulated sunlight, the tandem cell (without electrolyzer) achieved an open circuit voltage of 2 V, a short circuit current density of 9.7 ± 0.1 mA/cm², and a maximum power conversion efficiency of 16%. The predicted operating current density of the solar cell–electrolyzer combination was defined by the intersection of the solar cell power curve with the electrolyzer load curve (Fig. 3E), giving a value of 9.7 ± 0.1 mA/cm². The light-driven electrolysis rate responded to a solar-to-hydrogen (STH) efficiency of 11.9 ± 0.1%, comparable to similar systems that utilize purified water (29). The integrated solar-driven seawater-splitting system operated stably for 20 h without obvious STH decay (Fig. 3F). We then paired the seawater electrolyzer with a commercial Si solar cell (Renogy E.FLEX 5W, 5 V, 1 A) for a high-current test using real sunlight. The system operated at an impressively high current of ~880 mA under a photovoltage of 2.75 V (Fig. 3G and Movie S1).

Activation of Ni³⁺ electrode in simulated alkaline seawater played a key role in formation of a chloride-repelling passivation layer. After 3 to 4 h of anodization in 1 M KOH plus 0.5 M NaCl at 400 mA/cm², an obvious voltage dip (drop and partial recovery) was observed (Fig. 4A), during which the R_FE of O₂ production was low (~96%); Fig. 4B), suggesting anodic etching/corrosion during OER in salty water. After the voltage dip and over 12-h continued activation, the voltage stabilized and the oxygen R_FE increased to a stable ~100%, suggesting passivation of the anode with no further corrosion. In contrast, activation of NiFe/Ni foam (without a NiSₓ interlayer) under the same condition (400 mA/cm²) in 1 M KOH + 0.5 M NaCl electrolyte showed continuous voltage increase with oxygen R_FE < 100% (SI Appendix, Fig. S10), suggesting continuous anodic corrosion by the chloride ions and the lack of passivation without the NiSₓ layer in the anode.

Systematic investigations revealed that the activation/passivation treatment of Ni³⁺ led to transient etching followed by passivation by polyoxometalates through the layers of NiFe/NiSₓ-Ni foam anode. Raman spectroscopy of the Ni³⁺ electrode after anodic activation revealed vibrational modes at 470 cm⁻¹ and 540 cm⁻¹ characteristic of NiFe LDHs (Fig. 4C) (30), with the peaks around 1,230 cm⁻¹, 1,440 cm⁻¹, and 1,590 cm⁻¹ assigned to S-O vibration (31). This result suggested etching of the Ni³⁺ anode during the activation process was mainly oxidation of the sulfides, which was corroborated by the voltage dip in Fig. 4A and reduced R_FE for OER (Fig. 4B). In the activation step, anodic etching of the NiSₓ layer led to the formation of sulfate ions that migrated to the NiFe catalyst layer (free sulfate ions were also observed in the electrolyte; see SI Appendix, Fig. S11). Together with carbonate ions known to exist in KOH solution, the sulfate ions intercalated into the NiFe LDH resulted from anodization of the NiFe coating layer. Note that the NiFe-LDH was near amorphous in nature.
with a low degree of crystallinity (SI Appendix, Fig. S1F; the electron diffraction pattern did reveal ~0.64-nm layer spacing). Indeed, TOF secondary ion mass spectroscopy (TOF-SIMS) mapping on a NiFe electrode after activation unambiguously revealed the presence of both sulfate and carbonate species (Fig. 4D), confirming the formation of a layer of NiFe-LDH intercalated with two types of polyanions (32, 33). For this reason, we performed Raman spectroscopy to investigate the fate of the NiS interlayer underneath the NiFe-based catalyst by activating an NiS-coated Ni foam electrode under the same anodic activation process used for NiFe. We observed strong sulfate vibrational modes (34) at ~930 cm$^{-1}$ after the activation step (Fig. 4C), corroborated by TOF-SIMS mapping. Raman spectroscopy also revealed LDH [i.e., a phase-Ni(OH)$_2$] signatures with TOF-SIMS revealing both sulfate and carbonate ions. These results suggested the formation of a layer of LDH highly enriched with sulfate intercalants formed over NiS, with abundant sulfate anions outweighing carbonate anions compared with the relative abundance of anions in the NiFe catalyst layer after the same anodic activation. For the activated NiFe anode, we concluded that the sulfate and carbonate intercalated NiFe hydroxide catalyst layer together with the underlying sulfate-rich anodized NiS layer were responsible for the high OER activity and corrosion resistance to chlorine ions in seawater. The NiFe hydroxide layer exhibited higher OER activity than the NiS, (SI Appendix, S12) and synergized corrosion resistance with the underlying sulfate-rich Ni-Fe-NiS$_x$ interface. Without the electrodeposited NiFe layer, NiS$_x$/Ni was much less stable for electrolysis in the same salty electrolytes tested (SI Appendix, Fig. S7A). This was based on the fact that multivalent anions on hydros metal oxides surfaces are well known to enhance cation selectivity and afford repulsion and blocking of chloride anions (13, 35, 36). The polyanionic anion (sulfate and carbonate-) passivated Ni layers played a critical role in corrosion inhibition by repelling chloride anions and not allowing them to reach and corrode the underlying structure. In addition to electrode design, we also purposely added sulfated and other polyatomic anions in alkaline seawater electrolytes for electrolysis and also observed stabilization effects for NiFe and other OER anodes. Hence, careful design of anodes and electrolytes can fully solve the chloride corrosion problem and allow direct splitting of seawater into renewable fuels without desalination.

In summary, we have developed a NiFe/NiS/Ni anode for active and stable seawater electrolysis. The uniform electrodeposited NiFe was a highly selective OER catalyst for alkaline seawater splitting, while the NiS$_x$ layer underneath a conductive interlayer and a sulfur source to generate a cation-selective polyatomic anion-rich anode stable against chloride etching/corrosion. The seawater electrolyzer could achieve a current density of 400 mA/cm$^2$ under 2.1 V in real seawater or salt-accumulated seawater at room temperature, while only 1.72 V was needed in industrial electrolysis conditions at 80 °C. Critically, the electrolyzer also showed unmatched durability. No obvious activity loss was observed after up to 1,000 h of a stability test. Such a device provides an opportunity to use the vast seawater on Earth as an energy carrier.

**Materials and Methods**

The materials and methods used in this study are described in detail in *SI Appendix, Materials and Methods*. Information includes synthesis procedures of NiS/Ni foam, NiFe/Ni, Ni$_3$-NiO$_2$-O$_2$, cathode, perovskite solar cell fabrication procedure, electrochemical tests details, gas chromatography measurement details, and materials characterization details.

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