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Article

Hierarchical coral-like $\text{FeNi}(\text{OH})_x/\text{Ni}$ via mild corrosion of nickel as an integrated electrode for efficient overall water splitting

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ABSTRACT

Efficient, stable, and noble-metal-free electrocatalysts for both the oxygen evolution reaction and the hydrogen evolution reaction are highly imperative for the realization of low-cost commercial water-splitting electrolyzers. Herein, a cost-effective and ecofriendly strategy is reported to fabricate coral-like $\text{FeNi}(\text{OH})_x/\text{Ni}$ as a bifunctional electrocatalyst for overall water splitting in alkaline media. With the assistance of mild corrosion of Ni by $\text{Fe}(\text{NO}_3)_3$, in situ generated $\text{FeNi}(\text{OH})_x$ nanosheets are intimately attached on metallic coral-like Ni. Integration of these nanosheets with the electrodeposited coral-like Ni skeleton and the supermacroporous Ni foam substrate forms a binder-free hierarchical electrode, which is beneficial for exposing catalytic active sites, accelerating mass transport, and facilitating the release of gaseous species. In 1.0 mol L⁻¹ KOH solution, a symmetric electrolyzer constructed with $\text{FeNi}(\text{OH})_x/\text{Ni}$ as both the anode and the cathode exhibits an excellent activity with an applied potential difference of 1.52 V at 10 mA cm⁻², which is superior to that of an asymmetric electrolyzer constructed with the state-of-the-art $\text{RuO}_2\text{-PtC}$ couple (applied potential difference of 1.55 V at 10 mA cm⁻²). This work contributes a facile and reliable strategy for manufacturing affordable, practical, and promising water-splitting devices.

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1. Introduction

The threat of energy shortages is increasingly becoming the Sword of Damocles hanging over the head of human communities [1]. To fill the gap between limited fossil fuel reserves and increasing energy consumption, much attention has been paid to the development of techniques for the conversion and storage of renewable energy (i.e., solar, wind, and tidal energy). Among the various systems, electrolyzers for water splitting, which convert electricity into bond energy stored in chemicals such as H_2 and O_2 , are believed to be a promising alternative

[2–4]. Nevertheless, methods for improving the progress of the oxygen evolution reaction (OER) and the hydrogen evolution reaction (HER) remain a huge challenge. Ru/Ir- and Pt-based compounds have been found to be the most effective catalysts for the OER and the HER, respectively, but the scarcity and high cost of these materials has considerably hampered their large-scale utilization in commercial electrolyzers [5–7]. Therefore, it is highly desirable to develop efficient catalysts based on earth-abundant elements. Until now, numerous non-precious metal-based materials have been screened. Besides the most commonly explored first-row transition metal ele-

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ments, some compounds based on transition metals in group VI and carbon have also been found to be functional as individual OER or HER catalysts [8–15].

Using a bifunctional catalyst active for both the OER and the HER in one electrolyzer can undoubtedly reduce equipment costs and benefit commercialization [16–18]. However, owing to the mismatch of pH ranges in which the catalysts are stable and active, only a few compounds are capable of catalyzing overall water splitting in alkaline electrolytes, including metal phosphides (CoP [19]), metal selenides (NiSe [20]), metal sulfides (Ni₃S₂ [21]), and metal hydroxides (VOOH [22]). Unfortunately, the efficiencies of most of the aforementioned catalysts are still far from satisfactory, as the electrolyzer voltage generally needs to be higher than 1.6 V to afford a current density of 10 mA cm⁻² [10]. In addition, the synthetic procedures for most reported catalysts are tedious and sometimes require the use of hazardous raw materials. Therefore, highly efficient bifunctional catalysts for alkaline electrolyzers with more economic and environmentally friendly synthesis pathways are eagerly desired from the perspective of large-scale commercialization.

In recent years, Fe/Ni mixed compounds, especially hydroxides, have drawn much attention as bifunctional catalysts for the OER or the HER. The merits of this kind of material, such as high intrinsic activity, high solubility resistance to alkaline electrolytes, earth abundance, and nontoxicity, have been revealed by many fundamental research works [23,24]. By contrast, a gap still exists between fundamental explorations based on thin catalyst films and practical systems with thick catalyst films. For example, the electrical resistance of the catalyst film for a practical electrode is a considerable issue, but it is often ignored in fundamental research for simplicity. Thus, various elegant strategies have been developed to enhance the conductivity by constructing hybrid materials with carbon and metals as dopants [25–28]. However, the vast majority of products obtained by such hybridizations are in a powder form, which introduces new problems such as limited surface area, low mass transfer rate, and poor mechanical stability. One possible solution is to assemble metal hydroxides on self-standing metal/carbon nanowire/sheet arrays to construct integrated electrodes [29]. To achieve this goal, hydrothermal and electrodeposition techniques have been commonly adopted, but the relatively harsh reaction environments restrict their wide utilization.

Herein, we propose a relatively facile but effective approach to synthesize a FeNi(OH)_x/Ni integrated electrode for overall water splitting. A coral-like Ni skeleton was first electrodeposited on Ni foam, and then mild corrosion and hydrolysis processes were conducted in the presence of Fe(NO₃)₃. Though metal corrosion and hydrolysis have been explored for centuries, the fabrication of structure-diversified nanomaterials in a controlled corrosion environment is a relatively new concept [30]. By coupling the corrosion of the substrate and the hydrolysis of the metal ions, we converted the waste (i.e., metal ions produced by corrosion) into a functional material. The merits of this approach can be summarized as follows: (1) corrosion of Ni by Fe³⁺ is conducted in a mild manner in solution, which is economical, easy to process, and environmentally

friendly; (2) the controlled corrosion and hydrolysis leads to the autologous growth of FeNi(OH)_x species, endowing the final hybrid electrode with high conductivity and mechanical stability; and (3) the rationally designed coral-like morphology favors efficient mass transfer. Therefore, the resultant electrode exhibited impressive activity in terms of low overpotentials for the OER ($\eta = 254$ mV at 50 mA cm⁻²) and the HER ($\eta = 68$ mV at 10 mA cm⁻²) as well as a low cell voltage for overall water splitting ($E_{\text{cell}} = 1.52$ V at 10 mA cm⁻²) in alkaline media.

2. Experimental

2.1. Catalyst preparation

Ni foam (1.5 mm) was purchased from Shanxi Lizhiyuan Battery Material Company (China). Nickel chloride (NiCl₂·6H₂O), potassium hydroxide (KOH), ammonium chloride (NH₄Cl), and ferric nitrate (Fe(NO₃)₃·9H₂O) were purchased from Chengdu Kelong Chemical Reagent Factory (China). RuO₂, PtC (20 wt% Pt on Vulcan XC-72R), and Nafion (5 wt%) were purchased from Sigma-Aldrich. All chemical reagents were analytically pure. The water used in all experiments was purified through a Millipore system.

Synthesis of coral-like Ni. The electrodeposition of coral-like Ni was conducted following a published procedure with modifications [31]. First, commercial Ni foam was sequentially washed with absolute ethanol and 3 mol L⁻¹ HCl for 10 min under ultrasonication to eliminate NiO_x species and other contaminants on the surface. After acid washing, the Ni foam was sequentially washed with pure water and ethanol, and then dried in a N₂ atmosphere. Subsequently, the Ni foam (1 × 1 cm²) and a Pt plate (1 × 1 cm²) were used as a cathode and an anode, respectively, and cathodic electrodeposition was conducted in a NH₄Cl (2 mol L⁻¹) and NiCl₂ (0.1 mol L⁻¹) aqueous solution for 3 min at a current of 2.5 A. The obtained coral-like Ni was immersed in a HCl solution (0.1 mol L⁻¹) for several minutes, washed with deionized water and ethanol, and then dried in a N₂ atmosphere.

Synthesis of coral-like FeNi(OH)_x/Ni. Coral-like Ni was immersed in a Fe(NO₃)₃ aqueous solution in a glass bottle, which was then placed in an electric oven at various temperatures for different times. The resultant FeNi(OH)_x/Ni electrode was washed thoroughly with deionized water and air dried overnight.

Synthesis of FeNi(OH)_x/Ni. The resultant coral-like FeNi(OH)_x/Ni was calcined in a muffle furnace in air at 350 °C for 2 h with a heating rate of 5 °C min⁻¹.

Synthesis of FeNi(OH)_x@Ni foam. Commercial Ni foam was cleaned using the aforementioned process. Then, the Ni foam was dipped into a Fe(NO₃)₃ aqueous solution (0.01 mol L⁻¹) at room temperature. The resultant FeNi(OH)_x@Ni foam electrode was washed thoroughly with deionized water and air-dried overnight.

Preparation of RuO₂/Ni and PtC/Ni. Commercial RuO₂ (4 mg) was dispersed in a mixture of 800 μL of ultrapure water, 190 μL of isopropanol, and 10 μL of 5 wt% Nafion. After ultrasonication for 30 min, 500 μL of the resultant catalyst ink was

loaded onto coral-like Ni. The catalyst loading was calculated to be 2 mg cm⁻¹. The same procedure was used to prepare 20 wt% PtC/Ni.

2.2. Physical characterization

The surface morphologies and the microstructures of the catalysts were analyzed by X-ray diffraction (XRD-6000, Shimadzu), X-ray photoelectron spectroscopy (XPS, PHI 550 ESCA/SAM), field-emission scanning electron microscopy (FE-SEM, JSM-7800, Japan), and energy-dispersive X-ray spectroscopy (EDS, Oxford Link-ISIS-300). Transmission electron microscopy (TEM) measurements were conducted on a Hitachi H-8100 electron microscope (Hitachi, Japan) with an accelerating voltage of 200 kV. The Raman spectra were recorded on the Raman scattering spectroscopy system (Renishaw 2000) with excitation by a diode laser (514 nm, laser power of 25 mW) in the backscattering geometry.

2.3. Electrochemical measurements

Electrochemical measurements were conducted using a Princeton Applied Research Parstat 4000 potentiostat at room temperature, unless otherwise stated. A standard three-electrode system was used. Ag/AgCl (3 mol L⁻¹ KCl) was used as the reference electrode, which was calibrated in a high-purity H₂-saturated electrolyte with a Pt wire as the working electrode (Fig. S1(a)). The polarization curves were collected without iR correction.

OER test. In a three-electrode cell, the prepared catalysts were used as the working electrode and a Pt plate was used as an auxiliary electrode. Prior to data collection, 20 cyclic voltammetry (CV) cycles were conducted between 0.2 and 0.6 V (vs Ag/AgCl). Linear sweep voltammetry (LSV) measurements were performed at a scan rate of 1 mV s⁻¹ between 0.2 and 0.8 V (vs Ag/AgCl). AC impedance measurements were conducted at an overpotential of 200 mV with frequencies from 10 kHz to 0.01 Hz, unless otherwise stated. The data were analyzed and fitted using the ZSimpWin software. To determine the double-layer capacitance (*C_{dl}*), CV scans between 0.25 and 0.3 V (vs Ag/AgCl) were conducted at 4, 6, 8, 10, and 12 mV s⁻¹. To test the stability, galvanostatic measurements were conducted at a fixed current for 20 h.

HER test. A carbon rod was used as an auxiliary electrode

instead of the Pt plate. Prior to data collection, 20 CV cycles were conducted between -0.8 and -1.2 V (vs Ag/AgCl). LSV measurements were performed at a scan rate of 0.5 mV s⁻¹ between -0.8 and -1.5 V (vs Ag/AgCl). AC impedance measurements were conducted at an overpotential of 50 mV with frequencies from 10 kHz to 0.01 Hz, unless otherwise stated. The data were analyzed and fitted using the ZSimpWin software. To determine the *C_{dl}*, CV scans between -0.85 and -0.9 V (vs Ag/AgCl) were conducted at 4, 6, 8, 10, and 12 mV s⁻¹. To test the stability, galvanostatic measurements were conducted at a fixed current for 10 h.

Overall water splitting. A standard two-electrode system was constructed with coral-like FeNi(OH)_x/Ni as both the anode and the cathode in a KOH aqueous solution (1.0 mol L⁻¹). For comparison, RuO₂/Ni and PtC/Ni were used as the anode and the cathode, respectively, in the same electrolyzing system. Ten CV cycles were conducted between 1.2 and 1.6 V to reach a steady state. The polarization curves were recorded at a scan rate of 1 mV s⁻¹ between 1.2 and 2.0 V. To test the stability, galvanostatic measurements were conducted at a fixed current for 20 h.

3. Results and discussion

3.1. Synthesis and characterization of coral-like FeNi(OH)_x/Ni

A two-step procedure was proposed to fabricate the FeNi(OH)_x/Ni electrode. First, coral-like Ni was electrodeposited on macroporous commercial Ni foam with massive hydrogen bubbles generated in situ as dynamic templates (step I in Fig. 1(a) and Fig. S1(b)) [31]. Then, the resultant coral-like Ni was used as both substrate and reactant in the chemical corrosion process (step II in Fig. 1(a)). Fig. 1(b) illustrates the detailed reaction pathways for the chemical corrosion and hydrolysis processes. Specifically, after immersing metallic Ni in a Fe(NO₃)₃ solution, oxidation of Ni by Fe³⁺ and reduction of O₂ to generate OH⁻ species occurs according to Eqs. (1) and (2), respectively. Then, simultaneous hydrolysis of Fe³⁺ and Ni²⁺ by OH⁻ results in the nucleation and growth of FeNi(OH)_x nanosheets on the surface of the Ni substrate (Eq. (3)). Fe³⁺ is usually used as an oxidative etching reagent for dealloying owing to its high standard electrode potential (Fe³⁺ + e⁻ → Fe²⁺, *φ*^θ = 0.77 V) [32]. Further, the hydrolysis of Fe³⁺ has also been widely explored for the preparation of pigments owing to its

Fig. 1. Schematic illustration of the two-step procedure for the fabrication of coral-like FeNi(OH)_x/Ni (a) and the corrosion and hydrolysis processes for the in situ decoration of FeNi(OH)_x nanosheets on the Ni substrate (b).

quite low solubility product ($\text{Fe}^{3+} + 3\text{OH}^- \rightarrow \text{Fe}(\text{OH})_3$, $K_{\text{sp}} = 2.8 \times 10^{-39}$) [33]. In our particular case, we coupled the oxidative etching and hydrolysis processes of Fe^{3+} by selecting mild reaction conditions (i.e., a low concentration of $\text{Fe}(\text{NO}_3)_3$ and moderate heating) to balance the speed of oxidative etching and hydrolysis of metal ions, which plays a key role in the successful construction of the coral-like $\text{FeNi}(\text{OH})_x/\text{Ni}$ electrode.

As shown in Fig. S2, XPS was conducted to determine the amount of surface Fe. Generally, approximately 40% Fe was found for $\text{FeNi}(\text{OH})_x/\text{Ni}$ electrodes obtained in the early stage of chemical corrosion treatment (< 2.0 h), which is consistent with the content reported for highly active $\text{FeNi}(\text{OH})_x$ species in the literature [34]. This result also indicates that the oxidation of Ni^0 to Ni^{2+} and the hydrolysis of $\text{Fe}^{3+}/\text{Ni}^{2+}$ proceeded in a relatively steady-state manner during the early stage. As the $\text{FeNi}(\text{OH})_x$ nanosheets matured, the migration of Fe^{3+} from the bulk solution to the surface of coral-like Ni became difficult. The balance between oxidative etching and hydrolysis no longer existed, and hydrolysis of Fe^{3+} became the major reaction in the system. This phenomenon might be the reason for more Fe (ca. 56%) being detected with a longer soaking time (2.5 h).

The detailed morphology characteristics of the as-prepared electrodes were revealed by SEM and TEM. After the electro-deposition process, highly porous coral-like attachments were uniformly distributed around the Ni foam backbone (Fig. S3). Large micrometer-sized tunnels extended from the surface of the Ni foam to the top (Fig. S3(c)), and numerous canyons were spread over the attachments. After being subjected to the chemical corrosion treatment, numerous irregular nanosheets were generated on the surface of coral-like Ni, while the coral-like morphology was maintained (Fig. 2(a)–(c)). The TEM image in Fig. 2(d) indicates that the $\text{FeNi}(\text{OH})_x$ nanosheets were quite thin and tended to curl, with a thickness measured at the vertical edges to be ca. 2 nm (Fig. 2(e)). A blurry diffraction ring

was observed in the selected area electron diffraction (SAED) pattern of the corresponding nanosheets (Fig. 2(d), inset), indicating their poor crystalline nature. The phase composition of these deposits was examined by powder XRD (PXRD). As shown in Fig. S4, all the diffraction peaks in the PXRD patterns of coral-like Ni and $\text{FeNi}(\text{OH})_x/\text{Ni}$ can be well-indexed to metallic Ni (Card No. 03-1051), further proving the low crystallinity of the nanosheets, as no other diffraction peaks were observed.

The reaction rates of oxidative etching and hydrolysis, which are affected mainly by the Fe^{3+} concentration and the temperature, should be balanced to obtain well-designed $\text{FeNi}(\text{OH})_x$ nanosheets. As shown in Fig. S5, only sporadic nanosheets were decorated on the Ni surface in 0.005 mol L^{-1} $\text{Fe}(\text{NO}_3)_3$, whereas $\text{FeNi}(\text{OH})_x$ grows into large nanoparticles in 0.05 mol L^{-1} $\text{Fe}(\text{NO}_3)_3$. Further, as shown in Fig. S6, the nanosheet species were poorly developed on the surface of coral-like Ni at 30°C , but became too dense at 90°C . These observations indicate that the chemical corrosion and hydrolysis processes should be performed at a moderate rate under mild conditions (i.e., 0.01 mol L^{-1} $\text{Fe}(\text{NO}_3)_3$ and 60°C) for the fabrication of well-structured coral-like $\text{FeNi}(\text{OH})_x/\text{Ni}$.

Raman spectroscopy and XPS were conducted to assess the composition and structure of $\text{FeNi}(\text{OH})_x/\text{Ni}$. As shown in Fig. 3(a), no Raman signals were observed for coral-like Ni owing to its metallic state. On the contrary, three significant peaks were recorded after corrosion. The peak at 474 cm^{-1} can be attributed to the Ni–O vibrations of $\text{Ni}(\text{OH})_2$, whereas the other two peaks at 550 and 680 cm^{-1} are contributed by Fe incorporation and the more disordered structure of $\text{Ni}(\text{OH})_2$ [35–40]. As shown in Fig. S7, the XPS spectra of coral-like Ni reveal the presence of Ni and O elements on the surface, whereas Fe, Ni, and O elements were detected on the surface of $\text{FeNi}(\text{OH})_x/\text{Ni}$ (Fig. S8). The stoichiometric molar ratio of Fe, Ni, and O was determined to be 0.39:1:2.7. The surface Ni $2p$ signals of coral-like Ni and $\text{FeNi}(\text{OH})_x/\text{Ni}$ were resolved based on the protocol established by Biesinger et al. [41]. As illustrated in Fig. 3(b), the two major peaks at binding energies of 855.5 and 873.2 eV are attributed to either a $\text{Ni}(\text{OH})_2$ or NiOOH phase of $\text{FeNi}(\text{OH})_x/\text{Ni}$ [35,42,43]. Moreover, compared with coral-like Ni (Fig. S7), no Ni $2p$ signal was observed at 852.3 eV , indicating that the entire support surface was covered by $\text{FeNi}(\text{OH})_x/\text{Ni}$. As shown in Fig. 3(c), the broad Fe $2p_{3/2}$ envelope with a large peak splitting of 7.5 eV from its satellite confirmed a high-spin Fe^{III} state, verifying the presence of Fe^{3+} [35,44,45]. According to the XPS fitting protocol for Fe $2p$ signals from Grosvenor et al. [46], the broad Fe $2p_{3/2}$ envelope was fitted as a surface peak with multiplets ranging from 714.3 to 710.2 eV . The O $1s$ spectrum of $\text{FeNi}(\text{OH})_x/\text{Ni}$ was also recorded (Fig. 3(d)), and fitting confirmed the presence of three oxygen species, i.e., metal-oxygen bonds (528.9 eV), OH^- groups of hydroxides (530.6 eV), and defect sites with low oxygen coordination (531.8 eV) [35,42]. SEM-EDS was conducted to further probe the elemental distribution of the coral-like electrode. As shown in Fig. 2(f)–(i), O, Fe, and Ni elements were found to be evenly distributed, demonstrating the uniform incorporation of Fe throughout the material surface. All these results clearly confirm the successful preparation of coral-like

Fig. 2. Local features (a), global features (b), and side (c) view of $\text{FeNi}(\text{OH})_x/\text{Ni}$ in SEM images; (d, e) TEM images of $\text{FeNi}(\text{OH})_x$ nanosheets (the inset of (d) shows the corresponding selected area diffraction (SAED) pattern); (f–i) SEM-EDS images of $\text{FeNi}(\text{OH})_x/\text{Ni}$.

Fig. 3. (a) Raman spectra of FeNi(OH)_x/Ni and coral-like Ni; Ni 2p (b), Fe 2p (c), and O 1s (d) XPS spectra for FeNi(OH)_x/Ni.

FeNi(OH)_x/Ni.

3.2. OER test

The OER catalytic performance of FeNi(OH)_x/Ni, coral-like Ni, RuO₂/Ni, and FeNiO_x/Ni was evaluated by LSV measurements in 1.0 mol L⁻¹ KOH solution using a standard three-electrode configuration. As shown in Fig. 4(a), prior to oxygen evolution, prominent oxidation waves were observed for both FeNi(OH)_x/Ni and coral-like Ni, which can be assigned to the redox couple of Ni²⁺/Ni³⁺ [47]. This observed oxidation peak shifted to a higher potential over FeNi(OH)_x/Ni, which indicates the occurrence of charge transfer from Ni to Fe in Fe/Ni hydroxides, as demonstrated in the literature [48,49]. Apparently, FeNi(OH)_x/Ni showed the highest OER activity, as revealed by the polarization curves. For example, for FeNi(OH)_x/Ni, an overpotential of only 254 mV was required to reach a current density of 50 mA cm⁻², whereas for FeNiO_x/Ni, RuO₂/Ni, and coral-like Ni, the overpotentials were as high as 338, 390, and 443 mV, respectively. Furthermore, a similar activity trend was found after normalizing the activity to the electrochemical surface area (ECSA) (Fig. S9(a)). Correspondingly, the kinetic activities of all the electrodes were assessed using Tafel plots derived from the polarization curves. As

shown in Fig. 4(b) and Table S1, the smallest Tafel slope was acquired over FeNi(OH)_x/Ni (45 mV dec⁻¹). In order to avoid the arbitrary influence of replotting the polarization curve to calculate the Tafel slope [10], an analysis method based on electrochemical impedance spectroscopy (EIS) was also conducted (Fig. S10), and a more comprehensive value of 42.8 mV dec⁻¹ was obtained. Such excellent catalytic activity makes FeNi(OH)_x/Ni among the best reported nonprecious-metal-based OER catalysts (Table S3).

Fe³⁺ plays a key role in forming highly intrinsic active FeNi(OH)_x species. On the one hand, the number of charge carriers is increased by the incorporation of Fe, thus enhancing the conductivity of Ni hydroxides [49]. On the other hand, incorporated Fe³⁺ serves as a valence modulator through a partial-charge-transfer effect between Ni and Fe [39,43,49], which is beneficial for the electrocatalytic OER process.

Besides the high intrinsic activity of FeNi(OH)_x species, the rationally designed microstructure also contributes to the observed activity. In a comparison experiment, FeNi(OH)_x@Ni foam was prepared by synthesizing FeNi(OH)_x species directly on Ni foam. After being soaked in a 0.01 mol L⁻¹ Fe(NO₃)₃ aqueous solution, the relatively smooth surface of the Ni foam was uniformly covered by numerous cross-linked nanosheets (Fig. S11). As revealed in the LSV curves (Fig. S12), the catalytic

Fig. 4. (a) LSV curves of coral-like Ni, RuO₂/Ni, FeNiO_x/Ni, and FeNi(OH)_x/Ni. Reaction conditions: 1.0 mol L⁻¹ KOH, scan rate 1 mV s⁻¹, without iR compensation. (b) Tafel slopes of coral-like Ni, RuO₂/Ni, FeNiO_x/Ni, and FeNi(OH)_x/Ni; (c) Dependence of capacitive current on scan rate for coral-like Ni, FeNiO_x/Ni, RuO₂/Ni, and FeNi(OH)_x/Ni electrodes in 1.0 mol L⁻¹ KOH solution; (d) Current density-time (*j*-*t*) curves for the FeNi(OH)_x/Ni electrode at different potentials in 1.0 mol L⁻¹ KOH solution.

activity with different soaking times demonstrates a trend of first increasing and then decreasing. The best performance was observed at an excellent overpotential of 217 mV to reach a current density of 10 mA cm⁻². However, the activity at higher overpotentials was far from satisfactory. For example, an overpotential of 536 mV was required to reach a current of 200 mA cm⁻² for FeNi(OH)_x/Ni foam, which was much higher than that required for coral-like FeNi(OH)_x/Ni ($\eta_{200} = 379$ mV, Fig. 5(a) and Table S1). This decreased activity might be due to a lower gas diffusion efficiency and a higher bubble resistance [17,29]. Owing to the large pore size of commercial Ni foam, gas generated on the electrode surface tends to accumulate in big bubbles instead of being released into the bulk liquid, leaving less surface area for oxygen evolution. During the electrodeposition process, coral-like Ni generates numerous tunnels with in situ generated H₂ bubbles as dynamic templates (Fig. S1(b)). These small tunnels, in addition to the 3D porous framework of the Ni foam, might be much more beneficial for mass transfer during catalytic gas evolution processes [31].

However, electrodeposition of coral-like Ni and the subsequent chemical corrosion process also make the electrode surface much rougher. As shown in Fig. 5(b), compared with bare Ni foam (12 mF cm⁻²), the *C*_{dl} of coral-like Ni (77 mF cm⁻²) was

almost six times higher (Table S1). After processing with Fe(NO₃)₃, the *C*_{dl} value of FeNi(OH)_x/Ni increased to 126 mF cm⁻², further highlighting the advantage of the chemical corrosion strategy in terms of exposing more active sites. As shown in Figs. S2 and S14, although the Fe/Ni ratios were approximately 40% in the early stage of corrosion (<2 h), the catalytic activity initially increased and then decreased. The observed activities correlate well with the *C*_{dl} values of the electrodes with different corrosion treatment times, indicating that the electrode morphology governs the performance in this particular circumstance. This phenomenon was also confirmed by the electrochemical results for electrodes obtained by corrosion with different concentrations of Fe(NO₃)₃ (Fig. S15).

Hybridization with metal was expected to be an efficient way to enhance the conductivity of metal hydroxides [29]. Such enhancement can also be achieved by the controlled chemical corrosion and hydrolysis growth of FeNi(OH)_x nanosheets on coral-like Ni. As depicted in Fig. S16(a), a small semicircle was observed in the EIS plot for FeNi(OH)_x/Ni, demonstrating a smaller electronic resistance (*R*_{ct} = 6.2 Ω, Table S1).

Long-term durability plays a key role in commercial utilization. The stability of coral-like FeNi(OH)_x/Ni was investigated by bulk electrolysis at room temperature, and the current den-

Fig. 5. (a) LSV curves of bare Ni foam, coral-like Ni, FeNi(OH)_x@Ni foam, and coral-like FeNi(OH)_x/Ni in 1.0 mol L⁻¹ KOH solution at a scan rate of 1 mV s⁻¹; (b) Dependence of the capacitive current at 0.24 V (vs Ag/AgCl) on the scan rate in 1.0 mol L⁻¹ KOH solution.

sities were nearly unchanged at various potentials for 20 h (Fig. 4(d)). A multistep chronoamperometry experiment further demonstrated the strong durability of the resultant FeNi(OH)_x/Ni electrode (Fig. S16(b)).

3.3. HER test

The HER activity of coral-like FeNi(OH)_x/Ni was also examined by LSV in 1.0 mol L⁻¹ KOH solution. As shown in Fig. 6(a), compared with coral-like Ni, the HER was greatly enhanced after decoration with FeNi(OH)_x nanosheets by chemical corrosion. To reach a current density of 10 mA cm⁻², overpotentials of 170, 68, and 27 mV were required for coral-like Ni, FeNi(OH)_x/Ni, and 20 wt% PtC/Ni, respectively. As demonstrated in previous studies, in alkaline solutions, the HER kinetics is determined by a delicate balance between water dissociation and concomitant interactions of the water dissociation products with the surface. When FeNi(OH)_x is decorated on a metallic Ni substrate, the edge of hydroxide clusters promotes the dissociation of water ($\text{H}_2\text{O} + \text{e}^- \leftrightarrow \text{H} + \text{OH}^-$), and the generated H is adsorbed on the nearby Ni surface while OH⁻ is absorbed by the hydroxides, thus enhancing the HER activity in alkaline environments [50,51]. Although FeNi(OH)_x/Ni exhibits lower HER catalytic performance than commercial PtC, it is better than most reported nonprecious-metal-based catalysts in alkaline media (Table S4).

The corresponding Tafel plots for these electrodes are shown in Fig. 6(b) and Table S2. A Tafel slope value of 48.6 mV dec⁻¹ was obtained for PtC loaded on coral-like Ni, whereas Tafel slope values of 127.8 and 143.3 mV dec⁻¹ were recorded for FeNi(OH)_x/Ni and coral-like Ni, respectively, indicating that the HER rate-determining step over FeNi(OH)_x/Ni and coral-like Ni is different from that over PtC [52–54]. The electrode kinetics for the HER was also elucidated by EIS measurements at an overpotential of 50 mV. The Nyquist plots of the tested electrodes are shown in Fig. 6(c). The semicircle in the plot of

FeNi(OH)_x/Ni is significantly smaller than that in the plot of coral-like Ni. By fitting the experimental data, a small charge transfer resistance (R_{ct}) of 6.8 Ω was extracted for FeNi(OH)_x/Ni (Table S2), which is possibly responsible for the higher HER activity. The C_{dl} values of these three electrodes for the HER were also determined. As shown in Fig. S18(a)–(c), the C_{dl} of FeNi(OH)_x/Ni is 122 mF cm⁻², which is 1.5 times higher than that of coral-like Ni (80 mF cm⁻²), indicating the presence of more exposed and accessible active sites. Moreover, by comparing the measured C_{dl} values of FeNi(OH)_x/Ni for the OER (126 mF cm⁻²) and the HER (122 mF cm⁻²), we speculate that the active sites for the OER and the HER might be the same [55]. A bulk electrolysis experiment was conducted to assess the long-term stability of FeNi(OH)_x/Ni for the HER (Fig. S18(d)). The current density was nearly unchanged at the end of the test, indicating that FeNi(OH)_x/Ni is a promising cathode for long-term usage.

3.4. Overall water splitting

Based on the above analysis, FeNi(OH)_x/Ni exhibits good bifunctional features and can serve as both an anode for the OER and a cathode for the HER in alkaline solutions. This bifunctionality is most clearly evidenced by the CV experiment shown in Fig. 6(d). Catalytic HER currents were observed when applying a cathodic potential ($E < E^0(\text{H}_2/\text{H}_2\text{O})$), whereas an anodic potential higher than the thermodynamic threshold ($E > E^0(\text{O}_2/\text{H}_2\text{O})$) initiated the oxygen evolution process.

A two-electrode alkaline electrolyzer with FeNi(OH)_x/Ni as both the anode and the cathode was constructed to elucidate its overall water-splitting catalytic activity. At operation potentials higher than 1.5 V, obvious H₂ and O₂ bubbles were released on the surface of both electrodes (Fig. S19(a)). As revealed by the galvanostatic polarization curves (Fig. 6(e)), a cell voltage of only 1.52 V was required to reach a current density of 10 mA cm⁻² for the FeNi(OH)_x/Ni system in 1.0 mol L⁻¹ KOH solution,

Fig. 6. (a) LSV curves of coral-like Ni, FeNi(OH)_x/Ni, and 20 wt% PtC/Ni. Reaction conditions: 1.0 mol L⁻¹ KOH, scan rate 0.5 mV s⁻¹, without iR compensation. (b) Corresponding Tafel plots derived from the LSV curves; (c) Corresponding EIS plots in 1.0 mol L⁻¹ KOH solution at an overpotential of 50 mV; (d) Bifunctional catalytic activity of FeNi(OH)_x/Ni, FeNiO_x/Ni, and 20 wt% PtC/Ni tested by CV in 1.0 mol L⁻¹ KOH solution at a scan rate of 1 mV s⁻¹; (e) LSV curves of two-electrode water-splitting systems constructed with bifunctional FeNi(OH)_x/Ni and the RuO₂-Pt/C couple in 1.0 mol L⁻¹ KOH solution at room temperature; the red line shows the bifunctional FeNi(OH)_x/Ni system in 1.0 mol L⁻¹ KOH solution at an elevated temperature of 60 °C; (f) Bulk electrolysis at a constant applied potential difference of 1.52 V with FeNi(OH)_x/Ni as both the anode and the cathode of a two-electrode system; the inset shows a multicurrent process with a controlled current interval of 20 mA cm⁻² from 10 to 90 mA cm⁻².

which is better than that required for most reported bifunctional catalysts (Table S5) and even superior to that of the noble-metal-based RuO₂-PtC couple (1.55 V). At 60 °C, the kinetics and thermodynamics were further improved for the FeNi(OH)_x/Ni system, with an applied potential difference of 1.41 V affording a current density of 10 mA cm⁻² (Fig. 6(e)), indicating promise for industrial usage [17].

To assess the long-term stability, the resultant electrolyzer was operated at a fixed potential for 20 h. As depicted in Fig. 6(f), little attenuation of the current density was observed during the continuous electrolysis process. Moreover, the multi-step electrolysis process shown in the inset of Fig. 6(f) also reveals outstanding stability. The structure and composition integrity of these electrodes after long-term electrolysis was also demonstrated by physical characterization (Fig. S20).

Finally, a single AA battery was used to drive the same two-electrode electrolyzer. As illustrated in Fig. S19(b), the gases evolved at the anode and the cathode were gathered and found to be roughly consistent with the theoretical volume ratio of O₂ and H₂ for overall water splitting (1:2), further suggesting the overall water-splitting ability of FeNi(OH)_x/Ni.

4. Conclusions

In summary, we have successfully developed a cost-effective and environmentally friendly strategy that combines chemical corrosion of a Ni substrate with hydrolysis of Fe³⁺ and Ni²⁺ into one step to prepare a coral-like FeNi(OH)_x/Ni electrode. Under mild corrosion conditions, FeNi(OH)_x/Ni nanosheets grew stably and uniformly on an electrodeposited coral-like Ni skeleton without the usage of any binder, which not only promoted the water-splitting efficiency but also created numerous tunnels for more active sites and easier bubble diffusion.

Electrochemical tests revealed that the prepared FeNi(OH)_x/Ni electrode shows higher activity than RuO₂ in the OER and comparable activity to PtC in the HER in alkaline solutions. Therefore, the cell voltage required to reach 10 mA cm⁻² was only 1.52 V when FeNi(OH)_x/Ni was used as a bifunctional catalyst for overall water splitting, which is even lower than the cell voltage required by the precious metal couple of RuO₂ and PtC. This study is meaningful for the fabrication of noble-metal-free, highly efficient, and stable overall water-splitting catalysts in terms of both fundamental research and practical commercialization.

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Graphical Abstract

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Hierarchical coral-like FeNi(OH)_x/Ni via mild corrosion of nickel as an integrated electrode for efficient overall water splitting

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A coral-like FeNi(OH)_x/Ni integrated electrode for efficient overall water splitting ($V_{\text{cell}} = 1.52 \text{ V}$ at 10 mA cm^{-2}) is prepared with the assistance of an environmental friendly, low-cost strategy, namely, controlled corrosion, hydrolysis.

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通过温和的镍腐蚀制备珊瑚状 $\text{FeNi}(\text{OH})_x/\text{Ni}$ 作为一种一体化高效水分解电极

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摘要: 高效稳定并可同时催化析氧反应(OER)和析氢反应(HER)的非贵金属催化剂对于实现廉价水分解电解槽的商业化十分重要。虽然众多研究表明 $\text{FeNi}(\text{OH})_x$ 是一种极具潜力的催化剂,但是在基础研究与更有实用前景的电极之间仍有许多空白亟待填补。比如,基础研究多基于薄膜电极,其催化剂内部导电性的影响通常可以忽略。而基于实用化的电极则需要负载较厚的催化剂膜以获得更多的活性位,与此同时,其催化剂内部导电性的不利影响将会增大。此外,物质传递方面也会出现类似的情况。因此,一些在基础研究中显示出高本征活性的催化剂,在更加接近实际应用的体系下难以表现出预期的高活性。对于这一问题,目前鲜有相关的研究报道。

基于上述分析,本文报道了一种经济且环保的方法,以制备珊瑚状的 $\text{FeNi}(\text{OH})_x/\text{Ni}$ 催化剂。在碱性条件下,该催化剂具有同时催化OER和HER,从而实现全水分解的能力。在催化剂的制备过程中,具有高本征活性的 $\text{FeNi}(\text{OH})_x$ 纳米片借助 $\text{Fe}(\text{NO}_3)_3$ 对Ni温和的腐蚀过程,被原位负载到珊瑚状镍骨架上。这些纳米片与电沉积制备的珊瑚镍骨架以及3D泡沫镍基底一起构成了一体化的析气电极。这样的电极结构有助于暴露活性位、电解质快速传递和气体产物的迅速释放。此外,与珊瑚状金属镍骨架的复合也有利于减轻较厚的催化剂薄膜所带来的导电性降低的负面影响。在 $1.0 \text{ mol L}^{-1} \text{ KOH}$ 溶液中,以 $\text{FeNi}(\text{OH})_x/\text{Ni}$ 同时作为阳极和阴极而构建的对称电解槽表现出了优异的催化活性,只需要施加1.52 V的槽压即获得 10 mA cm^{-2} 的催化电流密度。其活性甚至优于当前最佳的由贵金属催化剂 RuO_2 和 Pt/C 构建的非对称电解槽所表现出来的活性(10 mA cm^{-2} 的槽压为1.55 V)。本文提供了一种简便易行且十分可靠的制备更加实用、具有潜力且可负担的水分解装置的策略。

关键词: 全水分解; 电催化; Fe/Ni 氢氧化物; 碱性电解槽; 一体化电极

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