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REVIEW

Recent advancements in direct seawater electrolysis

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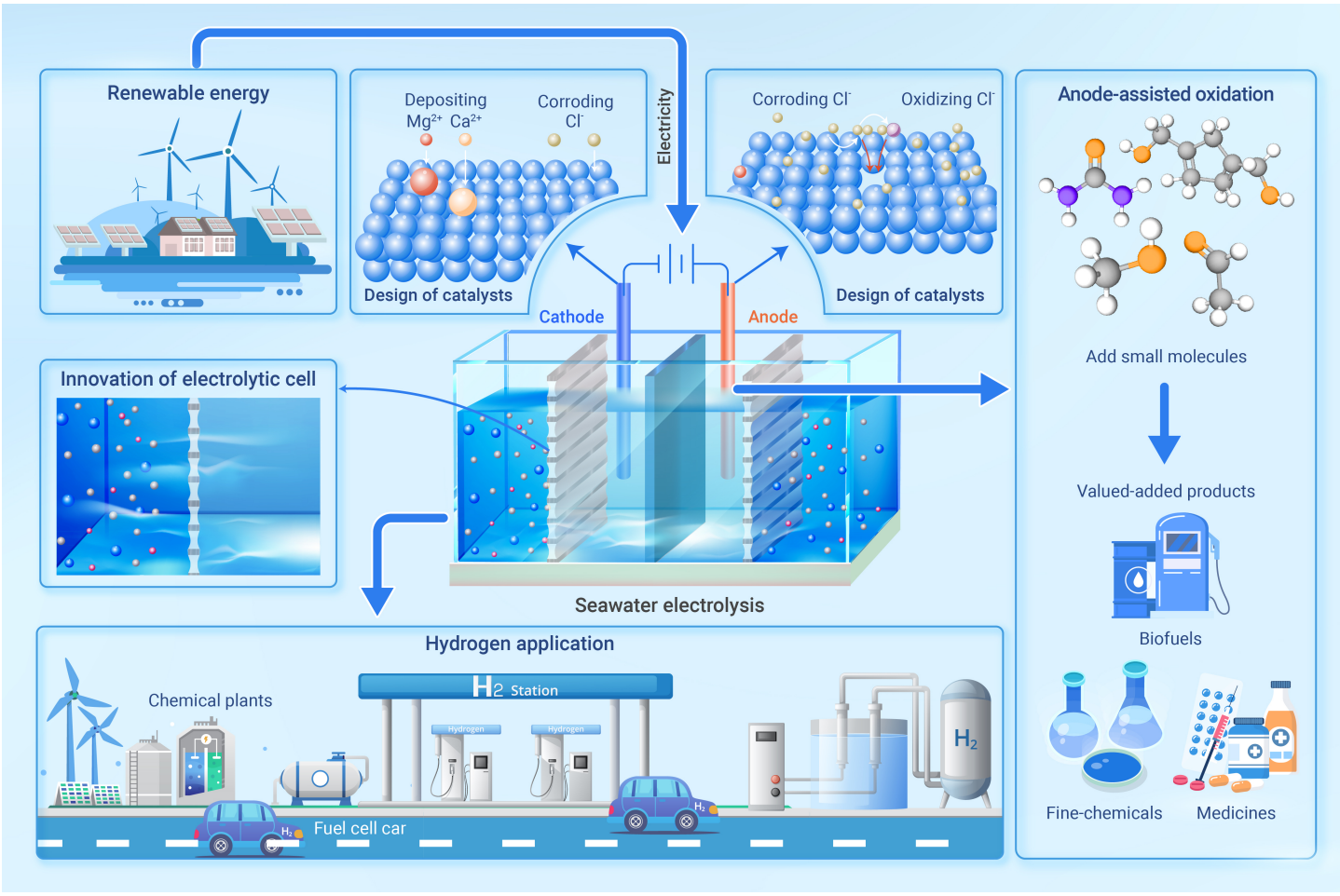
Received: April 3, 2025; Accepted: July 4, 2025; Published Online: October 9, 2025; https://doi.org/10.59717/j.xinn-energy.2025.100110

GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- Challenges of direct seawater electrolysis are summarized.
- Optimization strategies of catalysts for direct seawater electrolysis are summarized.
- Innovative electrolyzer architectures for direct seawater electrolysis are summarized.
- The superiority of direct over indirect seawater electrolysis is provided.
- The application of direct seawater electrolysis for energy storage is summarized.

REVIEW

Recent advancements in direct seawater electrolysis

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Citation: Ran S., Zhao C., Yan X., et al. (2025). Recent advancements in direct seawater electrolysis. *The Innovation Energy* **2**:100110.

Hydrogen production through water electrolysis represents one of the most promising low-carbon energy alternatives. However, current water electrolysis technologies depend on high-purity water feedstocks and their large-scale commercialization could exacerbate global freshwater scarcity. Direct seawater electrolysis (DSE) not only addresses this water resource challenge but also offers a potential solution for integrating deep-sea renewable energy sources. In this review, we first examine the challenges of DSE. Subsequently, we emphasize DSE technology and associated modification strategies, followed assessed the feasibility and economic viability of DSE. Finally, we summarize the potential applications of DSE in energy storage systems. These insights are intended to inform and guide future advancements in hydrogen production. We posit that on-site hydrogen production via DSE represents an optimal model for future renewable energy utilization and continuous research in this field should be actively promoted.

INTRODUCTION

Hydrogen has emerged as a highly promising candidate for low-carbon energy alternatives due to its high energy density and clean combustion characteristics. Additionally, hydrogen production through the electrolysis of water is a sustainable method of hydrogen production. However, current water electrolysis technology relies on high-purity water feedstock; large-scale commercialization would exacerbate global freshwater scarcity. Furthermore, the uneven distribution and limited availability of freshwater resources impede its large-scale implementation.[1] In contrast, direct seawater electrolysis (DSE) presents an ideal solution to this issue, as the oceans account for 96.5% of the Earth's water reserves.[2,3] Moreover, the rapid global development of marine renewable energy, particularly the large-scale expansion of offshore wind power, has introduced challenges in transmitting and consuming offshore electricity, especially from deep-sea renewable sources. Techno-economic models indicate that when offshore wind farms are located beyond 40~50 km from the coast, direct hydrogen production through seawater electrolysis at the wind farm site demonstrates greater competitiveness compared to transmitting electricity to shore.[4] Utilizing offshore wind power for DSE to produce hydrogen emerges as an effective solution to the challenges associated with large-scale grid integration of offshore wind energy and the high costs of deep-sea power transmission.

Despite its considerable potential, DSE encounters significant challenges stemming from the complex composition of seawater, which impedes stable and efficient hydrogen production.[3] This review begins by examining the critical challenges currently confronting DSE, followed by an in-depth analysis of modification strategies to enhance the efficiency of DSE, encompassing advancements in catalyst design, anode-assisted oxidation reaction and electrolysis cell innovation. We then assessed the feasibility and economic viability of DSE. Finally, we summarize the applications of DSE in energy storage. These insights are intended to establish a comprehensive framework to guide the advancement of DSE in hydrogen production.

CHALLENGES OF SEAWATER ELECTROLYSIS

The composition of seawater is highly complex, encompassing a multitude of impurity ions, including Cl^-, Mg^{2+} and Ca^{2+}, as well as bacteria and microorganisms, which present substantial challenges to seawater electrolysis (Figure 1). These ions and impurities can induce system corrosion and initiate detrimental side reactions, thereby complicating the electrolysis process. This section examines these challenges in detail, elucidating the difficulties arising from the intricate composition of seawater in achieving efficient and stable hydrogen production through seawater electrolysis.

Anodic competitive reactions

During seawater electrolysis, alongside the hydrogen evolution reaction (HER) and oxygen evolution reaction (OER), the presence of diverse impurity ions frequently induces numerous side reactions. Specifically, the abundant Cl^- ions in seawater lead to the chlorine evolution reaction (CER) at the anode, which exhibits a thermodynamic overpotential comparable to that of OER, resulting in competitive interactions between CER and OER at the anode.[6] The reaction pathway and products of CER are influenced by the pH of the medium, reaction temperature and applied potential.[6,7] (Equation 1 and 2)

$$\text{Acidic: } 2Cl^- \rightarrow Cl_2\,(g) + 2e^- \quad (1)$$

$$\text{Alkaline: } Cl^- + OH^- \rightarrow ClO^- + H_2O\,(l) + 2e^- \quad (2)$$

In 2016, Dionigi et al. introduced the Pourbaix diagram[8] (Figure 2), they analysis the relationship between the thermodynamic reaction potentials of the OER and the CER as a function of pH in aqueous solutions with a fixed chloride concentration of 0.5 mol L^{-1} at room temperature. The diagram reveals that at low pH (<3), chlorine gas (Cl_2) is predominantly released at the anode; at high pH (>7.5), hypochlorite (ClO^-) is more likely to form; and within the pH range of 3 to 7.5, the primary product is hypochlorous acid (HOCl) (Figure 3). Furthermore, across the entire pH range, OER exhibits a slight thermodynamic advantage over CER in terms of potential. As pH increases, the potential difference between the two reactions also progressively widens. At pH > 7.5, the potential difference between CER and OER reaches its maximum of 480 mV, theoretically enabling OER to occur at a potential of 1.71 V relative to the reversible hydrogen electrode (RHE), thereby favoring OER while suppressing CER. This phenomenon is termed the "alkaline design principle" of DSE, which explains why most seawater electrolysis experiments are conducted in alkaline environments. Consequently, to enhance OER selectivity in seawater electrolysis, additives or buffers are frequently employed to maintain the pH within the neutral or alkaline range, thereby preventing the formation of ClO^-.

Corrosion and deposition

In addition to the competing side reactions at the anode, the complex composition of seawater induces corrosion and degradation of electrolyzer components. In seawater electrolysis, catalysts are frequently exposed to seawater, which induces corrosion. Seawater contains corrosive agents, such as impurity ions, oxygen and microorganisms. These agents erode the catalyst surface, leading to structural degradation and performance deterioration. Corrosion mechanisms are diverse.[10] Electrochemical corrosion is accelerated by high anodic potentials and the presence of chloride ions. Intergranular corrosion occurs when there is preferential dissolution of grain boundaries at elevated potentials, which compromises the integrity of the material. Stress corrosion involves the initiation and propagation of cracks, resulting from a combination of mechanical stress and an electrochemical environment. Surface oxidation refers to the reaction between reactive oxygen produced during electrolysis and the catalyst, leading to the formation of unstable oxides that deactivate the catalyst. Ion etching is caused by various impurity ions in seawater, such as Cl^-, Mg^{2+} and Ca^{2+}, which poison the active sites and corrode the catalyst. This phenomenon is a primary factor contributing to the decline in the performance of the electrolyzer. Ion etching will be discussed in further detail later.

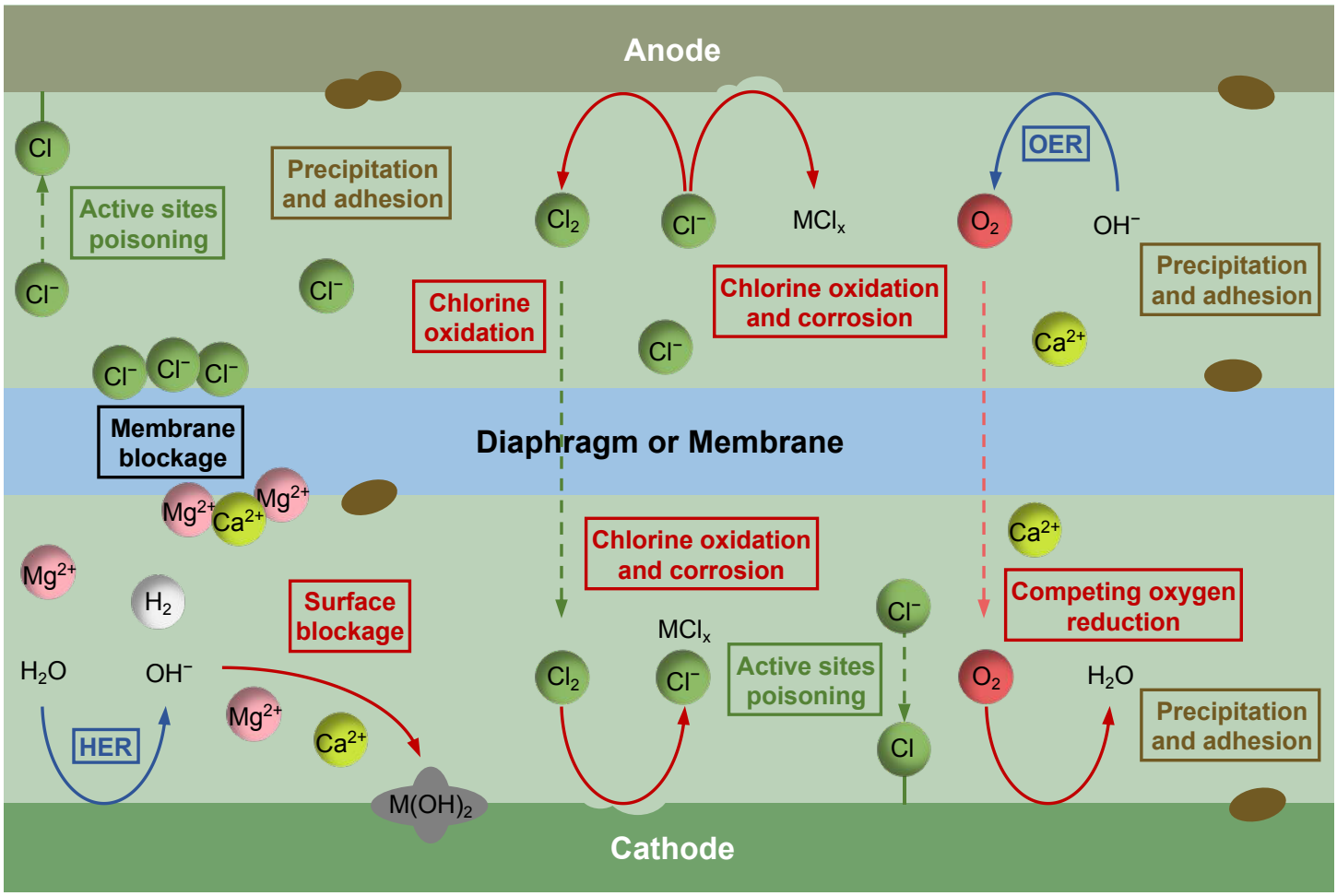

Figure 1. Challenges of DSE.[5]

Cl^- exhibits high corrosivity due to the aggressive oxidative byproducts generated during its oxidation process, which erode and dislodge lattice atoms of catalyst, ultimately leading to catalyst degradation.[11] Meanwhile, during the electrolysis process, Cl^- is adsorbed onto the metal active sites of the catalyst, forming intermediate species. These intermediates coordinate with Cl^- to generate MCl_x complexes, which dissolve in seawater. Subsequently, the dissolved MCl_x complexes precipitate as hydroxides in the alkaline environment of seawater, resulting in the degradation of catalyst performance.[12] The specific reaction pathway is as follows (Equation 3):

$$\begin{aligned} M + Cl^- &\rightarrow MCl_{ads} + e^- \\ MCl_{ads} + Cl^- &\rightarrow MCl_x^- \\ MCl_x^- + OH^- &\rightarrow M(OH)_x + Cl^- \end{aligned} \quad (3)$$

Furthermore, the electrode substrate, typically composed of nickel or its alloys, is prone to exposure to chloride ion, forming soluble chloro-complexes that lead to electrode corrosion. During electrolysis, the applied voltage induces strong polarization, significantly accelerating this corrosion. Notably, even when using acid/alkali-resistant titanium (Ti) electrodes or carbon/graphite electrodes with higher stability than metallic current collectors, the electrochemical corrosion process ultimately causes electrode failure.[13]

Lu et al.[14] investigated the influence of ions other than Cl^- on seawater electrolysis. Their findings revealed that, in addition to Cl^-, Br^- poses a more significant threat to nickel-based anodes due to its lower corrosion resistance and faster corrosion kinetics compared to chlorides. Moreover, for nickel-based electrodes with surface-loaded catalysts, Br^- induces extensive detachment of the catalyst layer, leading to rapid degradation of electrolysis performance. Consequently, in addition to addressing Cl^--induced corrosion, the development of anode materials resistant to Br^- corrosion is imperative for the advancement of seawater electrolysis technologies.

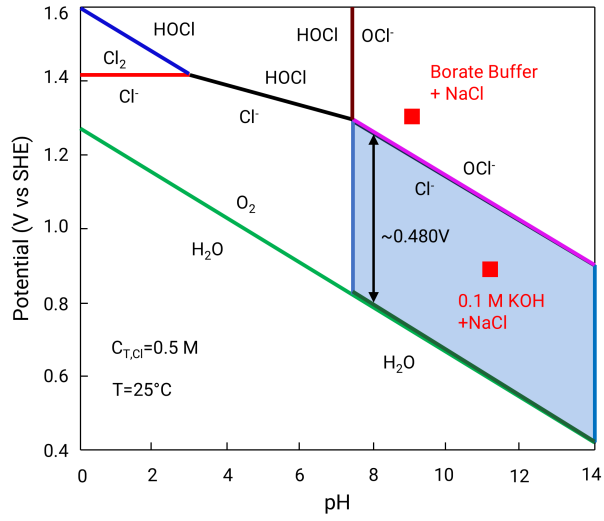

Figure 2. Competing OER and CER Pourbaix graphs.[8]

Notably, although the cathode operates under reducing potentials and

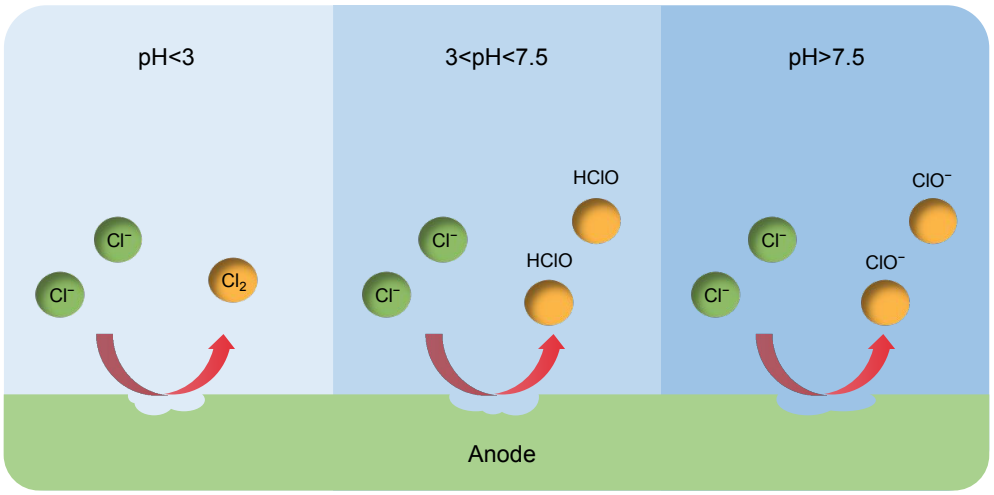

Figure 3. CER reaction mechanism under different pH.[9]

remains immune to electrode dissolution or chloride corrosion reactions during seawater electrolysis.[15,16] As seawater electrolysis typically utilizes renewable energy, which is intermittent, variable and random. This variability leads to frequent start-shutdown operations if renewable electricity is employed to power seawater electrolysis. During idle periods, the cathode undergoes self-discharge processes while simultaneously being susceptible to oxidative damage from reverse currents, resulting in irreversible oxidative degradation of cathode catalyst. Concurrently, chloride ions migrate toward the cathode under potential-driven forces, accelerating corrosion of the cathode material.[17]

The presence of significant concentrations of soluble cations, such as Na^+, Mg^{2+} and Ca^{2+}, poses a major challenge for seawater electrolysis. These cations are corrosive and may poison catalysts, electrodes and membranes, thereby shortening the stable service life of electrolyzers.[18] During seawater electrolysis, water molecules decompose at the cathode surface to produce hydrogen and OH^-, which results in a localized pH increase around the cathode. At the same time, Mg^{2+} and Ca^{2+} in the solution, due to their two positive charges, experience stronger electric field forces, migrate more rapidly toward the cathode, and thereby form precipitates of $Ca(OH)_2$ and $Mg(OH)_2$ that adhere to the surface. As the duration of electrolysis time increases and raw seawater is continuously replenished, metal hydroxide solids gradually accumulate on the surface. These deposits adhere to the surface of catalysts, obstructing water and mass transport and destabilizing the hydrogen production reaction process, ultimately leading to catalysts poisoning and deactivation.[19,20] Furthermore, research has demonstrated that ions such as Mg^{2+} and Ca^{2+} can interact with the lattice oxygen of the catalyst and, under specific conditions, even displace certain cations within the catalyst structure, thereby compromising its stability.[21]

Beyond corrosion, catalysts undergo electrochemical degradation through various surface reactions, including oxidation, reduction and other electrochemical processes. The presence of reactive species, variations in potential differences across the catalyst, and the composition of electrolyte collectively contribute to this degradation.[10] Meanwhile, the oxidized products of chloride and bromide ions in seawater, which possess strong oxidizing properties, accelerate membrane degradation.[22] Furthermore, the adhesion of bacteria, microorganisms and particulate matter to electrodes and membranes can result in clogging, presenting significant impediments to the advancement of seawater electrolysis utilizing conventional membrane electrode assemblies. These issues constitute critical challenges that must be addressed to enhance the feasibility and efficiency of the technology.

DESIGN OF CATALYSTS

Rational electrocatalytic design is critical for advancing seawater electrolysis technologies. However, current electrocatalysts often demonstrate inadequate performance, particularly under high current densities in practical applications. This section systematically reviews strategic approaches to enhance catalyst activity, selectivity and stability through optimized design methodologies (Figure 4). In addition, in situ analysis techniques are essential for screening and evaluating novel seawater electrolysis catalysts.

Catalyst activity optimization

Optimizing the catalytic activity not only enhances the efficiency of seawater electrolysis by accelerating the reaction kinetics and reducing energy consumption but also facilitates a substantial OER current density at an overpotential below 480 mV. This effectively mitigates the occurrence of competitive side reactions, such as the CER, in DSE, thereby improving both selectivity and stability. To achieve this, efforts should be directed not only toward enhancing the intrinsic activity of the catalyst but also toward increasing the density of active sites and improving electrical conductivity. Key strategies include heteroatom doping, the construction of heterostructures, morphology engineering and defect engineering.

Optimising the adsorption properties of reaction intermediates improves the intrinsic activity of catalysts. In accordance with the Sabatier principle, the catalytic activity is intrinsically linked to the adsorption energy of reaction intermediates (e.g., *H). Optimal adsorption energy should exhibit a balanced strength—neither excessively strong nor excessively weak—to ensure efficient reactant activation and product desorption. Wang et al.[23] engineered a self-supported Fe_3O_4 electrode incorporating P doping and oxygen vacancies (P-Fe_3O_{4-x}), which demonstrated exceptional efficiency and stability as a HER catalyst under alkaline seawater conditions. The introduction of P doping and oxygen vacancies effectively modulates the local atomic environment of Fe_3O_4, thereby optimizing the hydrogen adsorption/desorption energy at Fe active sites on P-Fe_3O_{4-x} and significantly enhancing the intrinsic HER activity in seawater electrolysis. The alkaline OER activity is influenced by the diffusion rate of OH^- through the catalyst layer and the adsorption rate of OH^- at active sites. Consequently, the development of OER catalysts capable of promoting rapid OH^- diffusion and adsorption is of paramount importance. Lu et al.[24] synthesized a Ni_xCr_yO electrocatalyst, which exhibits enhanced activity through dynamic self-reconstruction. The leaching of chromium increases the catalyst's porosity, facilitates electrolyte penetration, and strengthens the electronic interaction between OH^- and active sites. In 1 M KOH, the Ni_xCr_yO catalyst achieves current densities of 100 and 500 mA cm^{-2} at overpotentials of 270 and 320 mV, respectively, and maintains stability for up to 475 h at 100 mA cm^{-2}.

Morphological engineering serves as a pivotal strategy to augment the surface area and active site density of catalysts, facilitate electrolyte diffusion and enhance catalytic performance. Zheng et al.[25] engineered an electrocatalyst featuring amorphous-crystalline interface bifunctional sites through morphological engineering, specifically designed for the HER in alkaline seawater electrolysis. By constructing an amorphous-crystalline heterointerface on a nanoparticle cage, they successfully formed isolated PdCo clusters integrated with Co_3S_4 bifunctional sites. The hierarchical architecture of hollow porous nanocages and nanoparticle clusters significantly increased the catalyst's surface area and active site availability, thereby promoting efficient electrolyte diffusion. This catalyst demonstrated exceptional HER activity under alkaline seawater conditions.

Defect engineering serves as a critical approach to not only increase the exposure of active sites on catalysts but also optimize their electronic structure, thereby enhancing conductivity. Wang et al.[26] introduced a strategy involving P vacancy (Pv) engineering in NiCoP (NiCoPv@NF) to manipulate electronic redistribution, which demonstrated exceptional catalytic activity and stability for both the HER and OER in alkaline seawater. The incorporation of Pv reduces the energy barrier for surface electrochemical oxidation reconstruction, facilitating the localized transformation of the crystal into active oxygen (hydroxide) species during OER. Under alkaline seawater conditions, NiCoPv@NF exhibited superior HER and OER catalytic performance, achieving a current density of 500 mA cm^{-2} at a low cell voltage of

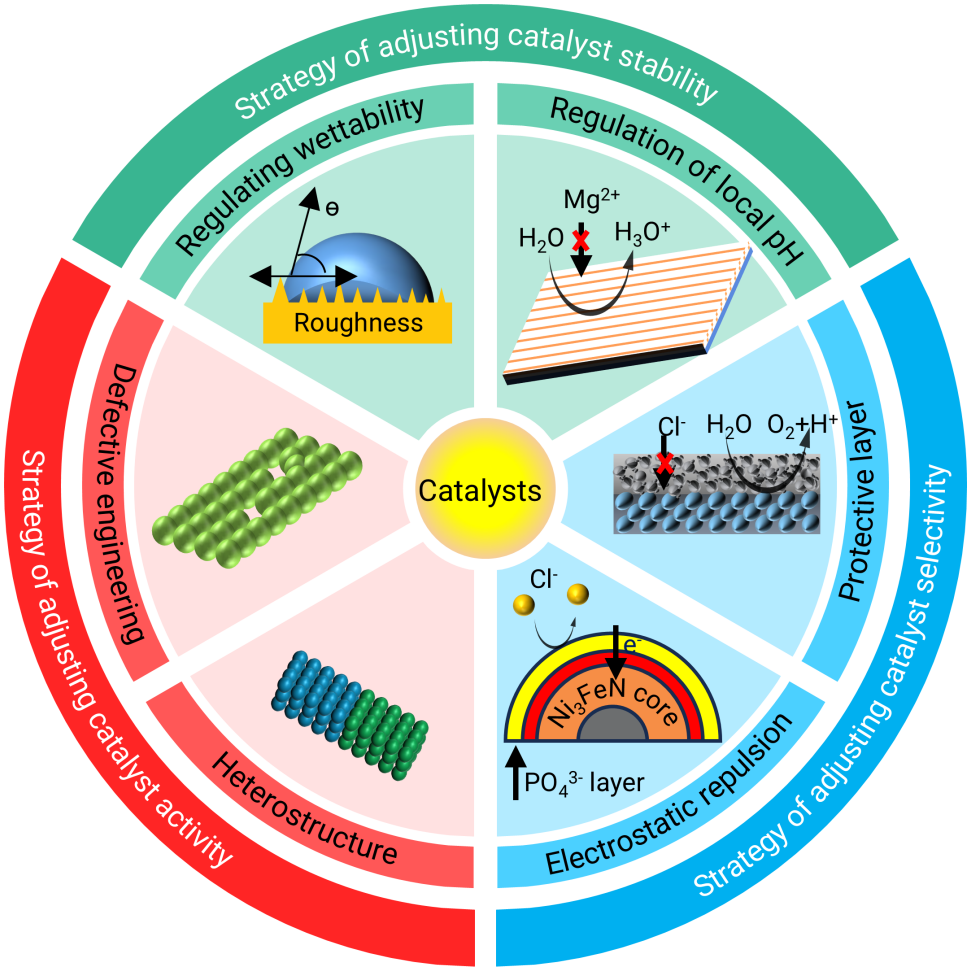

Figure 4. Strategies to improve catalyst performance.

2.43 V while maintaining stability for 110 h.

Enhancing conductivity facilitates accelerated electron transfer rates, thereby improving catalytic kinetics. Common strategies to augment the intrinsic conductivity of catalysts include optimizing their electronic structure and incorporating heteroatom doping. Such optimization of the surface electronic structure can effectively reduce reaction energy barriers, leading to enhanced catalytic performance. Wang et al.[27] developed a bifunctional electrocatalyst for seawater electrolysis, featuring a microcolumn array of Os-Ni_4Mo/MoO_2 that exhibits robust metal-support interaction. The microcolumn architecture promotes efficient electron and mass transfer, catalytic reaction steps and seawater electrolysis efficiency. Both theoretical and experimental analyses demonstrated that the strong MSI between Os and Ni_4Mo/MoO_2 optimizes the catalyst's surface electronic structure, reduces reaction energy barriers and ultimately improves catalytic activity. Wang et al.[28] devised a strategy to modulate the electronic structure of FeP by incorporating manganese heteroatoms and P vacancies into transition metal phosphides. The synthesized Mn-FePV catalyst exhibited remarkably low overpotentials. In situ Raman spectroscopy revealed that P vacancies and manganese doping synergistically altered the electronic structure of FeP, inducing rapid phase reconstruction and thereby enhancing the OER performance of Mn-FePV. When introducing heteroatoms, careful consideration must be given to the binding strength between the heteroatoms and the catalyst, as well as between the heteroatoms and impurity ions. The former influences the catalyst's activity, while the latter impacts its selectivity, both of which can be effectively predicted through density functional theory calculations.

Furthermore, the construction of heterogeneous structures and the deposition of catalysts onto highly conductive substrates can significantly enhance electron transfer rates during catalytic processes, improve interfacial charge transfer efficiency and optimize the adsorption of OER intermediates. These modifications accelerate the reconstruction and formation of active sites during OER. Liang et al.[29] synthesized a $FeOOH/Ni(OH)_2$ heterogeneous structure catalyst on Ni foam coated with Ni_3S_2. The formation of the $FeOOH/Ni(OH)_2$ heterogeneous structure effectively modulated the electronic properties of both FeOOH and $Ni(OH)_2$, thereby enhancing OER activity and selectivity. Additionally, the Ni_3S_2 layer facilitated efficient electron transfer between the $FeOOH/Ni(OH)_2$ heterogeneous structure and the nickel foam substrate, substantially improving catalytic performance. When employed directly as an OER electrode, this catalyst achieved current densities of 100 mA cm^{-2} and 800 mA cm^{-2} at overpotentials of 266 mV and 368 mV, respectively, in a 1 M KOH seawater electrolyte.

Catalyst selectivity optimization

The high concentration of Cl^- in seawater significantly impacts the efficiency of seawater electrolysis. Cl^- not only adsorbs onto the catalyst surface, obstructing the accessibility of active sites, but also engages in the competing CER alongside the anodic OER. This competition results in the generation of pollutants and increased energy consumption.[30] Consequently, enhancing the selectivity of the catalyst is imperative to mitigate the detrimental effects induced by CER.

Enhancing the selectivity of the catalyst is primarily accomplished by increasing the activity of the OER relative to the CER. As previously discussed, within the general pH range, OER exhibits a slight thermodynamic advantage over CER. This potential difference between the two reactions progressively widens as the pH increases. When the pH exceeds 7.5, the potential difference between CER and OER reaches its maximum value of 480 mV, theoretically enabling OER to occur at a voltage of up to 1.71 V relative to the RHE. This condition favors OER while entirely suppressing CER, making seawater electrolysis more feasible under alkaline conditions with selective oxidation. Consequently, regulating the pH of the electrolyte and the applied voltage has become a widely adopted strategy for achieving selective oxidation.[31] The addition of buffer solutions to seawater to neutralize protons generated during OER not only simplifies the operational process but also maintains the reaction within the high potential gap between OER and CER,[32] thereby minimizing the occurrence of undesirable side reactions.

An alternative approach to improving OER selectivity involves reducing the local concentration of Cl^- near the electrode, typically achieved by constructing a Cl^- blocking layer on the catalyst surface to impede the migration and diffusion of Cl^- to the active sites. Lu et al.[33] demonstrated this strategy by depositing silver nanoparticles on the catalyst surface, which in situ generated AgCl nanoparticles. This modification facilitated the specific repulsion of Cl^- within the electric double layer at the electrode interface. Both experimental and theoretical simulation results indicate that the formation of AgCl on the surface significantly diminishes the concentration of free Cl^- ions in proximity to the anode, thereby effectively inhibiting chloride-induced catalyst corrosion.

Electrostatic repulsion mechanisms can also effectively inhibit the adsorption of Cl^- and mitigate the occurrence of competitive side reactions. Yang et al.[34] developed a highly stable transition metal nitride catalyst through a rapid phosphorization strategy, demonstrating exceptional performance in seawater electrolysis. PO_4^{3-} effectively repel Cl^- through electrostatic interactions, significantly enhancing the catalyst's resistance to chloride-induced degra-

dation. This modification facilitated improved electron transfer between PO_4^{3-} and Ni_3FeN, thereby stabilizing the active sites and optimizing the overall catalytic performance.

Incorporating Cl^- into catalyst design, rather than eliminating them, represents a promising strategy to mitigate the competitive CER. Sun et al.[35] demonstrated this approach by decorating highly dispersed Pt on a CoP nanoarray surface (Pt/CoP) for the HER in Cl^--containing electrolytes. Their findings reveal that Cl^- induces electron redistribution on the Pt sites through the formation of Pt-Cl coordination, effectively reducing the HER overpotential by half in simulated seawater electrolyte. At a current density of 1 A cm^{-2}, the seawater electrolyzer equipped with a commercial anion exchange membrane (AEM) exhibited stable operation for over 100 h with a remarkably low voltage of 1.75 V.

Catalyst stability optimization

The stability and durability of catalysts in seawater electrolysis are critical for ensuring sustained efficiency and cost-effectiveness in hydrogen production. Catalyst degradation not only reduces the overall efficiency of the electrolysis process but also necessitates frequent replacements, escalating operational expenditures and compromising the system's overall sustainability.[10]

Enhancing the selectivity of the catalyst can effectively inhibit the competitive side reactions triggered by chloride ions and thus improve the stability, and the related strategy has been mentioned in the previous section. For example, adding sulfates or phosphates to the electrolyte enables their adsorption onto the cathode surface, forming a negatively charged layer that delays Cl^- corrosion and significantly enhances electrode stability.[36] However, oxygen-containing anions, such as PO_4^{3-} and SO_4^{2-}, persist only when their adsorption energies are comparable to or lower than those of competing species, such as OER intermediate. Excessive oxygen-containing anions may block OER active sites, leading to reduced activity.[37] Consequently, the in situ design and synthesis of oxygen-containing anions that can maintain maintaining prolonged stability during catalytic processes, hold critical significance for addressing long-term catalyst durability.[38] Deng et al. successfully fabricated molybdate (MoO_4^{2-})-modulated nickel-iron oxide electrodes for seawater electrolysis via a rapid thermal shock method.[38] The in situ generated MoO_4^{2-} on the electrode surface regulates, stabilized the catalytically active phase, enhancing OER activity and crucially protecting the electrode against chloride corrosion to extend operational longevity. The catalyst demonstrated an exceptionally slow degradation rate of 20 $\mu V h^{-1}$ during prolonged operation (>1500 h) at 100 mA cm^{-2}. This work provides novel insights into oxyanion-modified catalyst design, offering broad applicability for addressing challenges in seawater electrolysis.

Dissolved ions, such as Ca^{2+} and Mg^{2+} in natural seawater, can form precipitates due to localized pH increases during the HER, leading to catalyst deactivation and electrolyzer clogging. Consequently, regulating the local electrode environment to suppress precipitate formation represents a viable strategy for improving stability. Qiao et al.[39] developed a Pt/WO_2 catalyst that dynamically generates H_xWO_y as a proton reservoir, creating a localized acidic environment. This modification significantly enhances HER performance while inhibiting precipitate formation. Compared to conventional catalysts, Pt/WO_2 exhibits exceptional catalytic activity and long-term stability, operating continuously in natural seawater for over 500 h at a current density of 100 mA cm^{-2}. Similarly, Ling et al.[20] introduced a Lewis acid (LA) layer of CrO_x on transition metal (TM) oxides, achieving remarkable HER activity and anti-precipitation performance. The LA layer dynamically splits water molecules and captures hydroxide anions (OH^-), rapidly increasing the local pH at the cathode surface and thereby accelerating HER kinetics. Furthermore, utilizing the intermediate *H species generated during the HER can effectively inhibit precipitate formation in seawater, thereby enhancing catalyst stability. Gao et al. reported a Mo_2N catalyst.[40] On this catalyst's surface, N atoms on the surface dissociate and react with the *H intermediate from HER to form ammonium ions (NH_4^+), which subsequently adsorb onto the catalyst surface. The presence of these NH^{4+} not only optimizes the hydrogen-bond network connectivity but also effectively mitigates localized pH increases, thereby preventing precipitate formation and significantly improving the performance and stability of the catalyst.

Intermittent seawater electrolysis, powered by renewable electricity, induces cathodic oxidation and halide ion corrosion during shutdown periods, whose challenges that were previously overlooked. Sun et al. discovered that the introduction of non-metallic species with broad oxidation states and high oxygen coordination numbers, in combination with metal oxides, enables in situ generation of passivation layers.[17] This effectively suppresses cathode deactivation during shutdown phases of intermittent electrolysis. When seawater is used as feedstock, the spontaneously formed phosphate species on the cathode surface resists halide ion adsorption/corrosion toward both catalyst and substrate during idle periods. Based on this principle, they engineered a NiCoP-Cr_2O_3 HER electrocatalyst that achieves 10000 h intermittent seawater electrolysis at 0.5 A cm^{-2} with a voltage decay rate below 0.5% khr^{-1}. Their research establishes a pathway for addressing the degradation of HER electrocatalysts during intermittent electrolysis. This approach may directly mitigate the challenges faced by AEM electrolysers and alkaline electrolysers when coupled with fluctuating renewable energy sources.

The seawater electrolysis process often involves two-phase or three-phase interfacial challenges. Consequently, the generation and release of hydrogen bubbles play a crucial role in hydrogen production.[41] However, the sluggish growth and retention of gas bubbles can create "dead zones" at the active sites of electrocatalysts, which persistently block contact with the electrolyte solution over extended durations, thereby degrading the HER performance. Furthermore, hydrogen bubbles generate substantial interfacial adhesion forces that can readily damage the catalyst's surface structure during detachment. These effects become exacerbated at high current densities, as excessive bubble generation and release severely compromise both catalytic activity and stability.[42] Consequently, enhancing catalyst stability can be achieved by optimizing surface and interface wettability to facilitate gas desorption. Ren et al.[43] developed a highly porous sulfur-doped Ni/Fe (oxy)hydroxide catalyst grown on nickel foam, characterized by multilevel porosity and excellent hydrophilic properties. The synthesized catalyst demonstrated advantages such as low charge transfer resistance and efficient bubble release, exhibiting superior stability in alkaline natural seawater electrolyte. Furthermore, well-designed hierarchical structures with exceptional superhydrophilicity and underwater superaerophobicity, expose more active sites, accelerate bubble nucleation, and enable smaller bubble detachment sizes, representing a promising strategy for enhanced stability.[44] Zhang et al. systematically engineered core/shell nanoarrays to architect bubble transport channels, achieving precise modulation of interfacial H_2O activation dynamics while enabling ultrafast hydrogen bubble nucleation and detachment.[45] The self-supported catalyst holds uniform ultra-low Ru active sites of 0.38 wt% and promotes the rapid formation of plentiful small hydrogen bubbles, which are rapidly released by the upright channels, mitigating the blockage of active sites and avoiding surface damage from bubble movements. The integrated electrolyzer achieves industrial-grade performance with 800-h stability at 2 A cm^{-2} in alkaline seawater. Notably, variations in pH, temperature, and pressure can influence catalyst stability by altering reaction kinetics, surface morphology and adsorption properties. Selecting appropriate pH values and meticulously controlling temperature and pressure are critical for optimizing both the performance and longevity of catalysts in seawater electrolysis.[10]

At present, the development of catalysts for seawater electrolysis remains an ongoing challenge, primarily due to the intricate interplay between activity, selectivity and stability. Most catalysts fail to simultaneously achieve optimal performance across these three critical parameters. For instance, defect engineering, while enhancing catalytic activity, often compromises stability. Similarly, the construction of barrier layers, while improving selectivity and stability, may introduce mass transfer limitations, thereby reducing activity. Consequently, future catalyst designs should adopt a targeted approach, focusing on specific performance attributes tailored to the unique requirements of different electrolysis systems, rather than attempting to simultaneously enhance all properties.

The importance of in situ analysis technology in catalyst design

Electrochemical processes are fundamentally dynamic and occur in non-equilibrium conditions. They are characterized by complex interfacial phenomena, a high dependence on the reaction state, the presence of

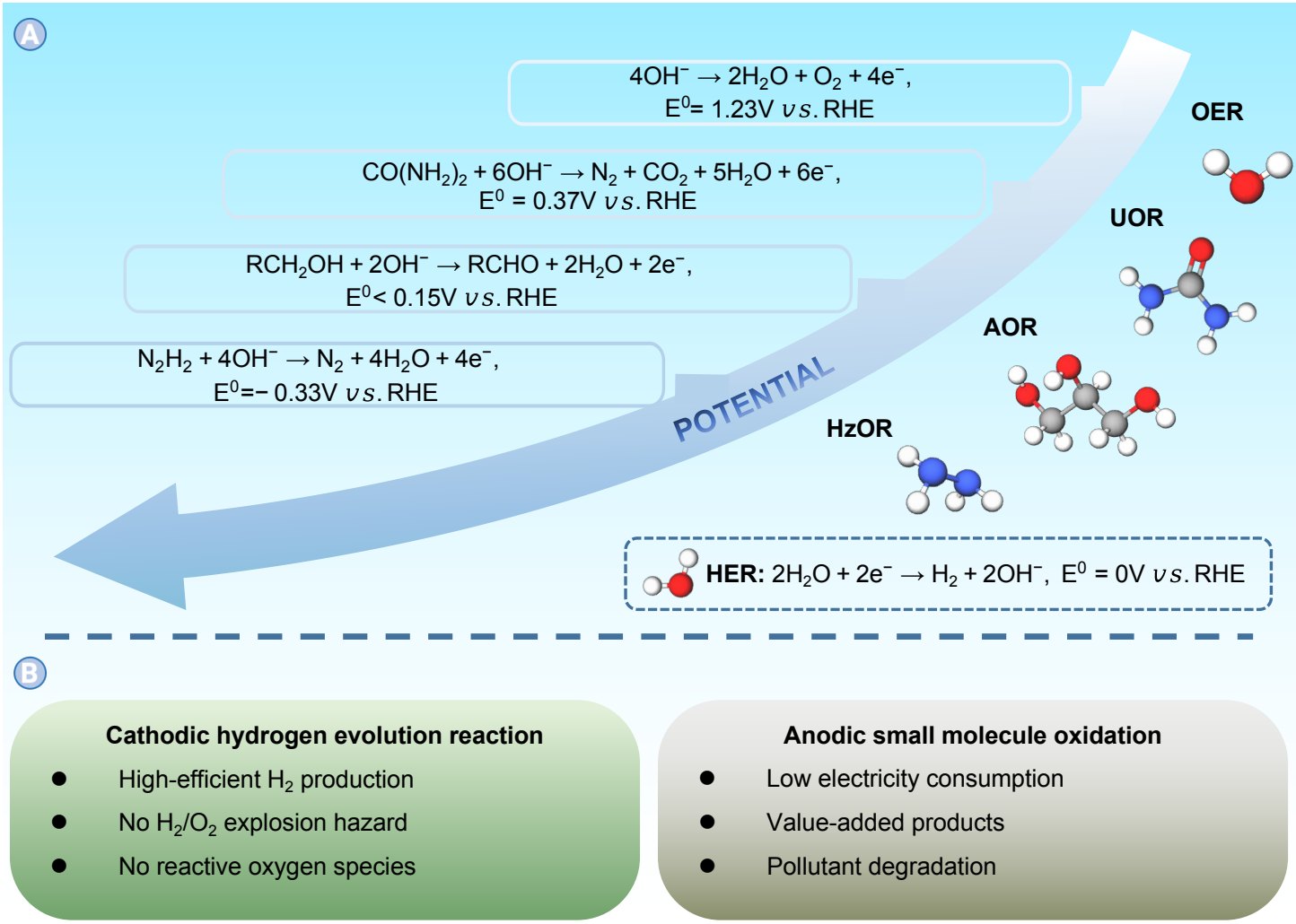

Figure 5. (A) Comparison of HER and other small molecule oxidation reactions with low onset potentials, (B) Advantages of small molecule assisted oxidation for hydrogen production.

metastable and transient species, and localized reactions. Compared to in situ characterization, ex situ analysis inevitably loses critical transient information. During seawater electrolysis, complex chemical reactions occur at the active sites of electrocatalysts. In situ analysis techniques, such as electrochemical impedance spectroscopy (EIS), X-ray photoelectron spectroscopy (XPS) and Raman spectroscopy can operate within the actual reaction environment, allowing for real-time monitoring microstructural changes on catalyst surfaces, the formation and transformation of intermediate products, and variations in performance.

For example, in situ transmission electron microscopy (TEM) enables the direct observation of dynamic processes with high spatial and temporal resolution.[46] In situ XPS can analyze the changes of valence states of elements in catalysts under varying potentials.[47] In situ EIS allows for real-time monitoring of impedance variations during reactions, facilitating the investigation of catalyst surface reconstruction and electrochemical reaction kinetics, as well as comparisons of performance differences among catalysts.[47] In situ Raman spectroscopy detects short-lived reactive intermediates generated during seawater electrolysis, identifying rate-limiting steps and critical active sites.[48] Fu et al. designed a high-efficiency OER electrocatalyst ((Ni,Fe)S_2@Ti_3C_2) by integrating MXene (Ti_3C_2) with NiFe sulfide (Ni,Fe)S_2, which significantly enhanced both catalytic activity and stability.[47] They employed in situ EIS to analyze the catalyst's surface reconstruction and electrochemical reaction kinetics, comparing performance differences among various catalysts. Shao et al. constructed a non-precious metal heterostructured catalyst rich in oxygen vacancies, (Ni,Fe)O(OH)@NiCoS NAs, on a nickel foam self-supporting electrode via a "hydrothermal + annealing" synthesis strategy.[48] They employed in situ Raman spectroscopy to probe the OER reaction mechanism.

In situ analysis techniques allow for more accurate screening and evaluation of novel electrocatalysts by providing real-time monitoring of catalysts' true operational states and performance under actual seawater electrolysis conditions. This approach significantly accelerates the development of high-performance catalysts and delivers critical material support for the practical application and large-scale deployment of seawater electrolysis technologies.

WAYS TO IMPROVE DSE EFFICIENCY

Anode-assisted oxidation

Despite the development of numerous effective electrocatalysts to enhance the OER, seawater electrolysis still necessitates a high overpotential to achieve the desired high current density. Moreover, the oxygen generated at the anode holds limited economic value due to its abundance in the atmosphere, and its mixing with hydrogen produced at the cathode poses a potential explosion hazard.[49,50] In light of these challenges, researchers are investigating the use of small molecule electrooxidation reactions as an alternative to OER at the anode. This approach is grounded in the principle that the theoretical oxidation potential of most small molecules is substantially lower than the thermodynamic onset potential of OER. Consequently, readily oxidizable soluble small molecules, such as urea, hydrazine (N_2H_4) and alcohols, have been employed to replace traditional OER in seawater electrolysis for hydrogen production. This strategy significantly reduces the operating voltage of the electrolyzer and mitigates the explosion risk associated with hydrogen and oxygen mixing. Additionally, the oxidation of these small molecules at

lower potentials effectively prevents interference from the CER at the anode, substantially lowering energy consumption and improving hydrogen production efficiency.[51] Undoubtedly, the integration of small molecule-involved anodic oxidation reactions in seawater electrolysis represents a highly promising advancement (Figure 5).

Urea-assisted oxidation. Urea, owing to its favorable energy density and high hydrogen content, has been established as an effective hydrogen carrier for long-term sustainable energy supply.[52] Industrial, agricultural and domestic wastewater contain substantial quantities of urea, and untreated wastewater can pose significant environmental risks.[53] Conventional urea treatment methods typically involve thermodynamic hydrolysis and chemical oxidation.[54] While thermodynamic hydrolysis can fully recover N from urea wastewater, it incurs high technical and operational costs. Chemical oxidation demands considerable manpower and resources, and the reaction products are often difficult to control. Consequently, the treatment of urea remains a persistent challenge. In recent years, urea oxidation reaction (UOR) has garnered considerable attention due to its ability to be coupled with the HER in seawater electrolysis, enabling the production of high-energy-density hydrogen while treating domestic and industrial wastewater.[55] Furthermore, given urea's low raw material cost and a theoretical potential of 0.37V, electrochemical UOR represents a promising approach for advancing various environmental and clean energy technologies.[56,57]

The six-electron transfer process of UOR results in inherently sluggish reaction kinetics.[58] Among various metal catalysts, Ni-based catalysts demonstrate superior activity in UOR,[59] with Ni^{3+} in NiOOH identified as the primary catalytic active site. Extensive research has focused on enhancing catalytic activity by modulating the electronic structure of Ni^{3+} to optimize the adsorption behavior of key intermediates. In 2009, Botte et al.[60] pioneered the use of nickel-based materials to facilitate hydrolysis in UOR, revealing that TM-based materials exhibit notable catalytic activity in UOR. This discovery spurred further investigation into the electrocatalytic performance of various TM-based materials in UOR, with Ni-, Co-, Mn-, and Mo-based materials emerging as the most promising candidates due to their unique electronic structures and favorable earth abundance for electrocatalytic applications.[61] Guo et al.[62] reported the synthesis of Ru, P co-doped $NiMoO_4$ multi-channel nanorods grown in situ on nickel foam, which enable chlorine-free hydrogen production by coupling seawater decomposition with thermodynamically favorable UOR. The synthesized catalyst achieved a UOR working potential of only 1.46 V and delivered a current density of 1000 mA cm^{-2} in an electrolyte containing 1 mol L^{-1} KOH and 0.5 mol L^{-1} NaCl.

N_2H_4-assisted oxidation. N_2H_4 is a widely utilized industrial material, commonly employed as a fuel in rockets and fuel cells. Research indicates that the N_2H_4 oxidation reaction (HzOR), with a low theoretical potential of -0.33V, serves as an effective alternative to the OER and can significantly reduce energy consumption for energy-efficient hydrogen production.[63] Furthermore, HzOR typically demonstrates faster kinetics, involving a four-electron transfer process, compared to the six-electron transfer process of UOR. Notably, the sole product of the HzOR reaction is N_2 gas, ensuring that this process is not only carbon-neutral but also eliminates safety concerns associated with the mixture of N_2 gas at the anode and hydrogen gas at the cathode.[64] Consequently, replacing OER-based hydrogen production with N_2H_4 oxidation, which exhibits favorable thermodynamic and kinetic properties, is regarded as a highly promising energy-saving alternative.

To ensure efficient operation of N_2H_4-assisted seawater electrolysis systems, bifunctional electrocatalysts with both HER and HzOR activity are required. Precious metals like ruthenium and palladium show excellent performance in both HER and HzOR, enabling efficient hydrogen production via N_2H_4-assisted water splitting. However, their high cost and scarcity greatly limit their practical applications.[65] Consequently, there is a pressing need to design and develop cost-effective, high-efficiency bifunctional electrocatalysts based on non-precious metals to enhance the kinetics of both HER and HzOR.[66] Yuan et al.[67] synthesized a sea urchin-shaped iron-nickel co-doped CoP HER/HzOR bifunctional electrocatalyst supported on nickel foam and applied it to N_2H_4-assisted seawater electrolysis systems. This catalyst achieved an industrial-level current density of 1.5 A cm^{-2} at 70°C, and the system demonstrated stable operation for hundreds of hours in seawater environments. Furthermore, the construction of crystalline/amorphous interfaces in electrocatalysts has been demonstrated as an effective strategy,[68] it could enhance the kinetics of hydrogen evolution and N_2H_4 oxidation.[69–71] Jiang et al.[72] developed a straightforward in-situ synthesis method utilizing multi-metal oxyacids to construct a crystalline/amorphous bifunctional interface in Pt/MoO_{3-x} nanosheets, achieving exceptional HER and HzOR performance. In a seawater electrolyte, the HER and HzOR working potentials at a current density of 10 mA cm^{-2} were as low as -23 mV and -41 mV, respectively.

Alcohol-assisted oxidation. The urea and N_2H_4 employed in the aforementioned anode-assisted oxidation reactions primarily function as sacrificial agents. Their oxidation facilitates the reduction of the energy-intensive OER and mitigates the generation of pollutants from the CER, thereby lowering energy consumption and minimizing environmental impact.[73] This approach presents a viable strategy for seawater electrolysis. However, the products generated at the anode through these reactions generally possess limited economic value. In recent years, anode reactions assisted by alcohol oxidation (AIOR) have garnered considerable attention from researchers. These reactions not only share the benefits of UOR and HzOR, such as reduced energy consumption and environmental compatibility, but also yield high-value chemicals or fuels at the anode.[74,75]

Methanol, a simple organic molecule, is both cost-effective and characterized by an exceptionally low thermodynamic oxidation potential (0.016 V vs. RHE).[76,77] Over the past few decades, MOR has been the subject of extensive research, as a substitute for the OER in mixed seawater electrolysis.[78] Complete oxidation of methanol yields carbon dioxide and water, while partial oxidation produces formic acid or formaldehyde. The corresponding reaction equations are as follows (Equation 4-6):

$$\text{Complete oxidation to } CO_2 \text{ and water: } CH_3OH + O_2 \rightarrow CO_2 + 2H_2O \quad (4)$$

$$\text{Partial oxidation to formic acid: } CH_3OH + H_2O \rightarrow HCOOH + 2H^+ + 2e^- \quad (5)$$

$$\text{Partial oxidation to form formaldehyde: } CH_3OH + \frac{1}{2}O_2 \rightarrow HCHO + H_2O \quad (6)$$

The key to harnessing the benefits of methanol-assisted oxidation reactions lies in the development of efficient MOR catalysts. Liu et al.[79] achieved significant advancements in the study of multimetallic compound methanol electrooxidation catalysts for facilitating efficient mixed seawater electrolysis for hydrogen production. They synthesized a novel quaternary $Pt_{1.8}Pd_{0.2}CuGa/C$ intermetallic nanoparticle catalyst, which demonstrated exceptional catalytic activity and stability. When applied to MOR-assisted seawater electrolysis, the catalyst operated continuously for 240 h in simulated seawater and 120 h in natural seawater at 1.23 V without notable degradation, showcasing remarkable stability. Moreover, understanding the catalytic behavior of methanol at specific bifunctional sites is critical for advancing electrocatalysis. Wang et al.[80] introduced the concept of dual-directional electronegativity-driven d-band center modulation at Pt-Rh bifunctional sites to precisely tailor the catalytic behavior of the MOR. This work represents a significant breakthrough in the design of advanced bifunctional electrocatalysts for complex catalytic reactions.

In addition to methanol, the anode oxidation reaction involving ethylene glycol (EGOR) also exhibits considerable potential for seawater electrolysis. Wang et al.[81] synthesized porous, ultrathin, defect-rich, and heterogeneous Rh metal-ene materials via a one-step hydrothermal method for EGOR-assisted seawater electrolysis in energy-efficient hydrogen production. The unique two-dimensional metal-ene structure, combined with the synergistic effect of the Rh/RhOOH heterogeneous interface, facilitates the provision of abundant catalytic active sites, optimizes the adsorption and desorption of reaction intermediates, and enhances the mass transfer process. This study provides a novel strategy for the design and construction of efficient catalysts for electrochemical small molecule oxidation-assisted seawater splitting.

Innovation of electrolytic cell

Advancing DSE technology necessitates not only innovations in catalysts and progress in small molecule-assisted oxidation reactions but also the

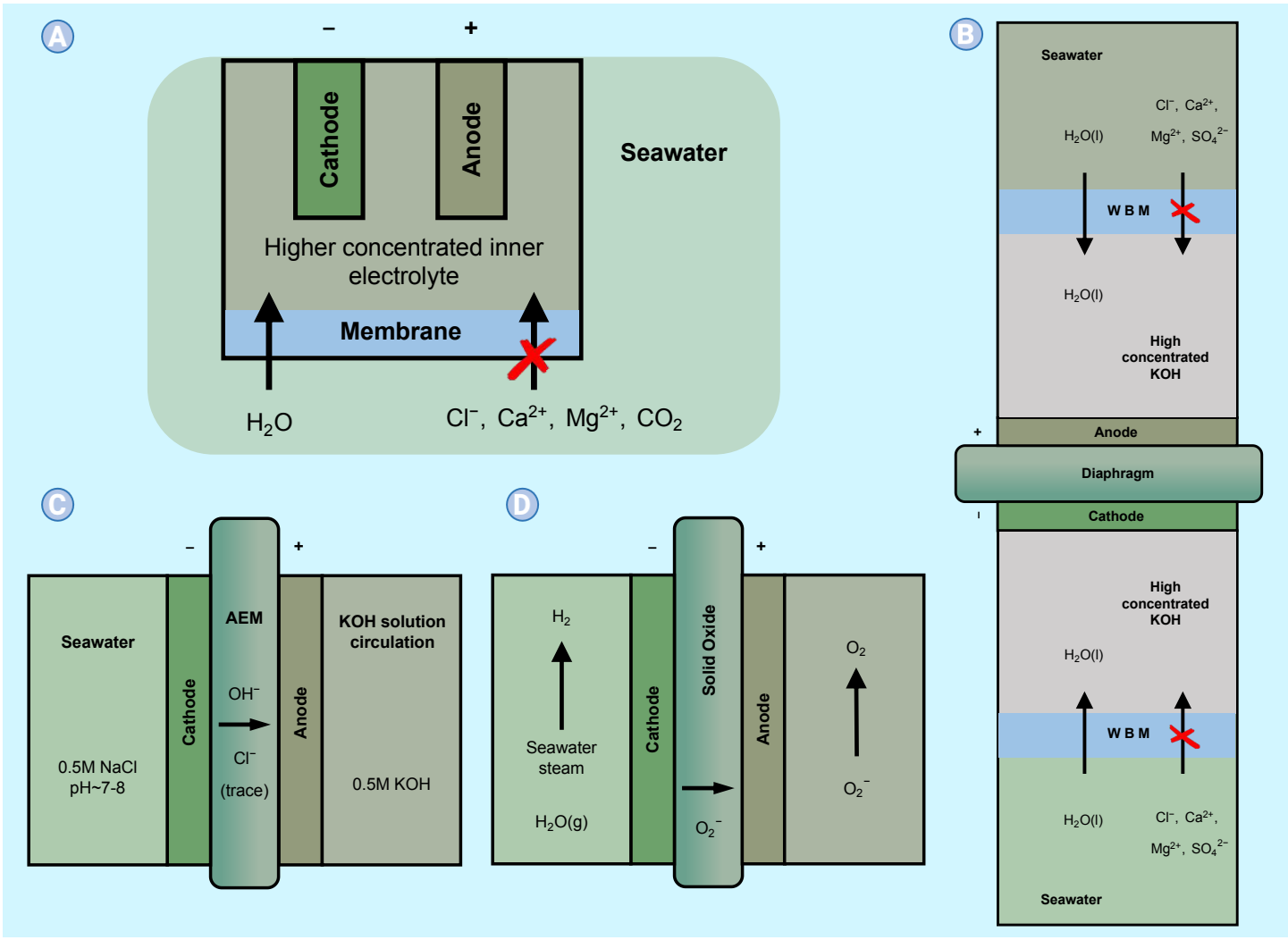

Figure 6. Innovations in electrolysis tanks (A) Forward osmosis-seawater electrolyzer,[83] (B) Direct electrolysis of seawater driven by phase transition migration,[84] (C) Asymmetric electrolytic cell,[85] (D) High temperature solid oxide electrolytic cell.

development of practical and efficient electrolysis cells, which are critical components of an optimized DSE system. Currently, the primary types of water electrolysis hydrogen production systems include alkaline water electrolysis (AWE), AEM electrolysis, proton exchange membrane (PEM) electrolysis and high-temperature solid oxide electrolysis cells (SOECs). However, these systems impose stringent requirements on electrolytes, and the presence of numerous complex ions and impurities in seawater results in suboptimal hydrogen production efficiency. Consequently, there is an urgent need to design more efficient and practical systems tailored specifically for seawater electrolysis. In this section, we introduce several innovative cells (Figure 6,7).

Forward osmosis-seawater electrolytic cell. Reverse osmosis (RO), one of the most widely employed methods for seawater desalination,[82] traditionally separates RO seawater desalination from freshwater electrolysis in conventional DSE systems. Nocera et al.[83] proposed a forward osmosis seawater electrolysis system (Figure 6A), offering a streamlined approach to DSE compared to indirect systems. This system integrates forward osmosis-based seawater desalination directly into the electrolysis cell, enabling simultaneous seawater desalination and water electrolysis. The system operates by placing the forward osmosis water decomposition electrolysis cell in a pH-neutral, high-concentration electrolyte, utilizing a cellulose acetate semi-permeable membrane to separate untreated seawater. The resulting concentration gradient drives water flow from seawater to the more concentrated electrolyte, while the cellulose acetate membrane effectively repels Cl^-, Ca^{2+}, Mg^{2+} and CO_2. The osmotic inflow of water balances the outflow through electrolysis, facilitating continuous seawater purification and electrolysis. This

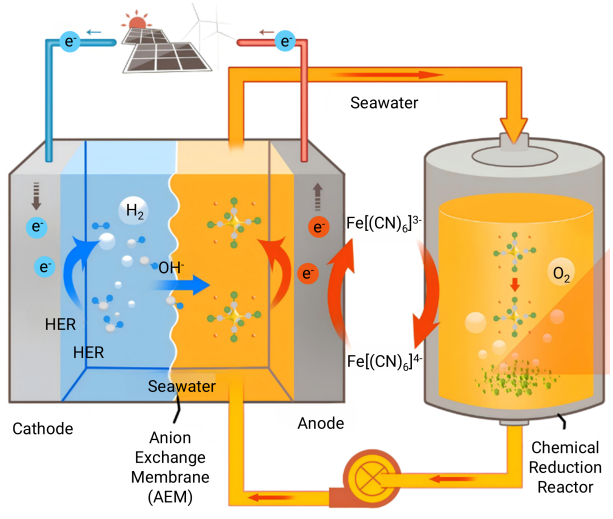

Figure 7. Redox-mediated seawater direct decoupling electrolyzers.[95]

approach allows the use of freshwater decomposition electrocatalysts, significantly reducing desalination costs and simplifying the process. Moreover, since the forward osmosis seawater electrolysis cell selectively extracts

water molecules from seawater via the concentration gradient through the forward osmosis membrane, it circumvents the high energy consumption associated with the elevated pressure required for RO membranes, presenting a cost-effective alternative.

Phase-transition-migration electrolytic cell. Although the direct penetration seawater electrolysis system exhibits significant potential for seawater electrolysis, ion mutual diffusion between seawater and the electrolyte persists during prolonged operation. To ensure the complete separation of pure water and seawater, Xie et al.[84] integrated molecular diffusion, interfacial phase equilibrium, and other physicochemical processes with electrochemical reactions, achieving the spontaneous phase transition and migration of water in seawater (Figure 6B). This represents a breakthrough in DSE. The technology enables stable DSE without additional energy input, effectively isolates seawater ions, and facilitates efficient in-situ DSE for hydrogen production, eliminating the need for desalination, mitigating side reactions, and avoiding extra energy consumption. The system operates by utilizing a waterproof and breathable layer to construct micron-scale "gas phase" isolation domains in seawater. It leverages the natural vapor pressure difference between the self-humidifying electrolyte and seawater as the mass transfer driving force, enabling the spontaneous phase transition and migration of water from seawater to vapor, diffusion through the membrane, and absorption and liquefaction on the electrolyte side. Simultaneously, the intrinsic hydrophobicity of the waterproof breathable layer ensures the complete separation of liquid seawater and its impurities, permitting seawater to diffuse solely in the form of water molecules. The high saturated vapor pressure of seawater and the low saturated vapor pressure of the concentrated electrolyte generate a driving force, promoting the transport of water molecules through the membrane to liquefy on the electrolyte side. As hydrogen production consumes water, it further sustains the interfacial pressure difference across the membrane, thereby inducing the continuous replenishment of water from seawater to the electrolyte. Ultimately, when equilibrium is established between the water migration rate and the electrolysis rate, the system stably generates hydrogen from seawater.

In 2024, Xie et al.[86] integrated offshore wind power renewable energy with DSE for hydrogen production, utilizing offshore wind power to drive seawater hydrogen production in marine environments. They established a theoretical model for seawater hydrogen production through phase transition migration under real sea wave fluctuations, achieving high stability exceeding 500 h in a laboratory-simulated marine environment.

Asymmetric electrolytic cell. Strasser et al.[85] introduced an innovative asymmetric seawater electrolyzer for DSE, which enhances electrolysis efficiency and stability through an asymmetric electrolyte supply strategy (Figure 6C). In this configuration, neutral seawater is directly utilized at the cathode, while a separately circulated pure KOH electrolyte is employed at the anode. This design effectively prevents the formation of a highly alkaline environment at the cathode and minimizes Cl^- transfer to the anode. Shi et al.[87] developed a pH-asymmetric electrolyzer incorporating a Na^+ exchange membrane for DSE. This Na^+ exchange membrane effectively blocks Cl^- migration from the cathode electrolyte (natural seawater) to the anode electrolyte (NaOH), thereby circumventing the CER. Additionally, it mitigates the issue of Mg^{2+} and Ca^{2+} precipitation in near-neutral seawater. However, this pH-asymmetric electrolysis technology necessitates the use of ion exchange membranes and high-performance electrocatalysts to ensure both high efficiency and stability during the electrolysis process.

High-temperature solid oxide electrolytic cell. High-temperature SOECs, operating at temperatures between 800-1000°C and driven by thermal energy, exhibit considerable potential in the field of seawater electrolysis (Figure 6D). In contrast to conventional seawater electrolyzers operating at lower temperatures, the elevated temperatures significantly enhance reaction kinetics and improve overall electrolysis efficiency.[88] Research indicates that SOECs, compared to traditional alkaline and PEM electrolyzers, possess higher theoretical hydrogen production efficiency and unit area production rates, positioning them as a promising technology for seawater hydrogen production.[89,90]

Redox-mediated decoupling electrolytic cell. Decoupled water electrolysis represents an innovative approach that facilitates stepwise reactions by employing an auxiliary redox mediator as an electron/ion buffer, thereby enabling the spatial and temporal separation of the HER and OER.[91,92] This decoupling strategy is frequently utilized to mitigate limitations associated with conventional electrolyzers, including low load capacity, high operational voltage, and compromised gas purity.[93,94]

Xie et al.[95] integrated the decoupling strategy into DSE, developing a decoupled seawater electrolysis system for hydrogen production directly from seawater (Figure 7). This system comprises a distinct oxygen production reactor and a DSE cell, effectively preventing Cl_2 generation without the need for additional chemical agents. The system employs ferrocyanide and ferrous ferrocyanide ($[Fe(CN)_6]^{3-/4-}$) as electron mediators, with the mediator functioning as a charge carrier that circulates between the electrolyzer and the reactor. Experimental results demonstrate that the $[Fe(CN)_6]^{3-/4-}$ redox couple maintains reversible redox kinetics and stability in seawater. The system operates at a voltage of approximately 1.37 V at 10 mA cm^{-2} and around 1.57 V at 100 mA cm^{-2}, achieving stable operation for over 250 h at an industrial current density of 200 mA cm^{-2}. The reaction process is as follows (Equation 7-9):

$$\text{Cathodicreaction (HER): } 2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad (7)$$

$$\text{Anodereaction (FeCN oxidation): } 2[Fe(CN)_6]^{4-} - 2e^- \rightarrow 2[Fe(CN)_6]^{3-} \quad (8)$$

$$\text{Reactor: } 4[Fe(CN)_6]^{3-} + 4OH^- \rightarrow 4[Fe(CN)_6]^{4-} + 2H_2O + O_2 \quad (9)$$

This decoupled seawater direct electrolysis system offers a novel approach for achieving efficient and sustainable hydrogen production from seawater. Future advancements in the optimization of high-performance redox mediators and catalysts hold the potential to further enhance the cost-effectiveness and sustainability of the system.

FEASIBILITY AND ECONOMIC VIABILITY OF DIRECT SEAWATER ELECTROLYSIS

Current seawater electrolysis predominantly employs an indirect approach, specifically involving desalination prior to hydrogen production via electrolysis. Hausmann et al. argue that DSE technology is overhyped, as the purified water currently used in electrolyzers can be obtained from seawater and other water sources through established technologies like RO. Seawater purification accounts for only 0.03% of the total energy consumption in water electrolysis for hydrogen production, resulting in a negligible impact on overall costs. Therefore, DSE was considered to offer no significant energy or cost advantages.[96] However, they did not discuss scale effects.[97] The RO systems employed in seawater desalination are large-scale facilities, typically producing between 10 thousand to nearly 1 million cubic meters of water per day.[98] The maximum scale of commercial electrolyzers is 10 MW, requiring 125 cubic meters of water per day. There exists a significant scale mismatch between these systems. DSE is typically utilized in small and decentralized hydrogen production systems, such as those found in coastal communities or offshore wind farms. These scenarios typically do not necessitate large-scale RO facilities; rather, they require smaller and more compact solutions to accommodate varying geographical conditions and production needs. On the other hand, compared with DSE, conventional water electrolysis demonstrates only a marginal advantage in terms of energy consumption (Table S2).[99] However, in many cases, the purity of the obtained reverse osmosis water still fails to meet the requirements for membrane-based freshwater electrolysis. This necessitates additional purification steps. Notably, PEM membranes particularly require ultra-purified water with partial ion exchange for stable operation. Consequently, seawater electrolysis using conventional membrane electrolyzers demands extra energy input, additional space allocation and increased capital investment. Furthermore, when factoring in associated costs along with safety considerations such as establishing restricted zones around hydrogen facilities, the compact footprint of small-scale integrated one-step systems becomes particularly crucial.[100]

The indirect seawater electrolysis process exhibits significant drawbacks in offshore areas. Its reliance on RO generates highly concentrated brine discharge, which adversely impacts marine ecosystems. During seawater desalination, over 3.5 million cubic meters of brine are produced daily and discharged back into the ocean.[101] In contrast, DSE can circumvent these

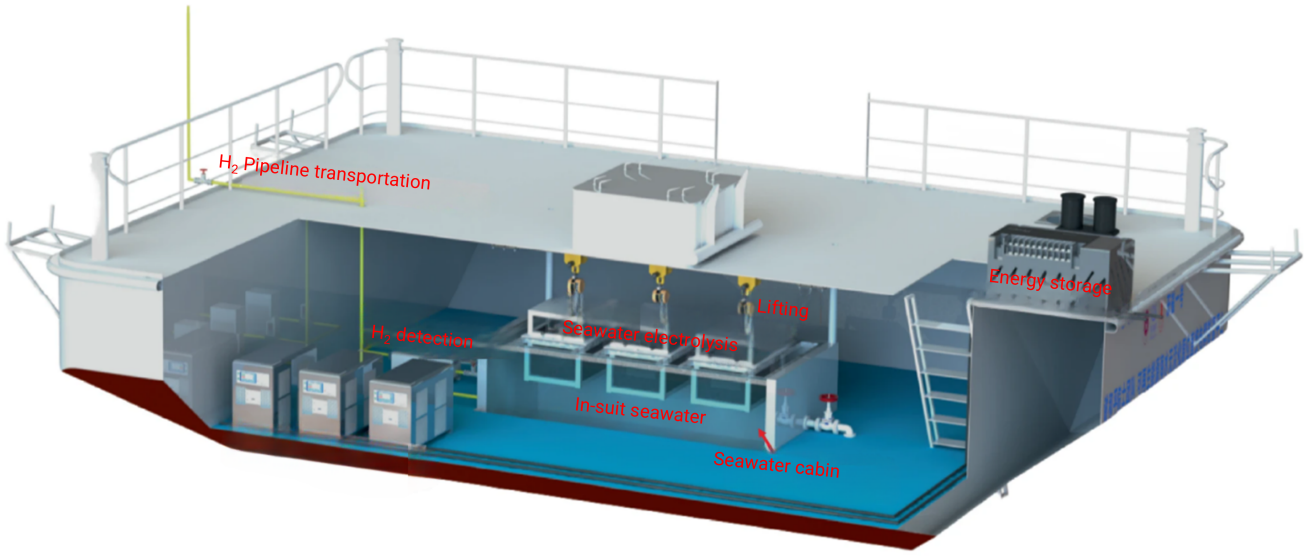

Figure 8. In-situ direct seawater electrolysis using floating platform in ocean with uncontrollable wave motion.[86]

limitations by developing electrolyzers capable of directly utilizing untreated seawater as feedstock. And interest in DSE is particularly high for offshore hydrogen production, where the capital cost is heavily dependent on the installation footprint. Economic viability can be improved by simplifying engineering and eliminating pre-treatment systems.[100]

As of now, all major offshore hydrogen production projects rely on purified water, using desalination units to supply feedstock for electrolyzers. This is mainly because currently available electrolyzer technologies cannot directly process seawater, and impurities with potential fouling issues complicate the process.[100] Xie et al.[86] developed the world's first demonstration of a platform for coupling DSE and offshore wind power for hydrogen production (Figure 8), which is driven by a 10 MW wind turbine and achieves an electrolysis energy consumption of 5.0 kWh/standard cubic metre of hydrogen, which is comparable to freshwater electrolysis. It was experiMentally verified to have operated stably for more than 240 hours in force 8 winds, one-metre waves and torrential rains, thus validating the feasibility and stability of in situ DSE technology under real ocean conditions.

The economic analysis of electrolysis technology primarily focuses on raw material inputs, energy consumption and subsequent equipment maintenance expenses. For DSE, the costs of raw material can be disregarded. As previously mentioned, utilizing the anode for small-molecule oxidation enables the conversion of organic compounds and pollutants in seawater into valuable chemicals.[9] This process not only offsets operational costs but also enhances the return on investment, thereby reducing the payback period of DSE systems. Maintenance costs in the later stages of operation will be influenced by advancements in technology, including innovative materials, new reaction principles and novel device structures.

DSE systems significantly relax water quality requirements, thereby eliminating geographical constraints for terrestrial deployment. This characteristic makes them uniquely suited for large-scale, distributed hydrogen production in offshore areas and coastal regions. Moreover, the advancements in deep-sea development have positioned DSE as an effective solution to address challenges in large-scale grid integration of offshore wind power and to mitigate the exorbitant costs associated with deep-sea power transmission. As DSE technology matures and offshore wind power advances, seawater-based hydrogen production is poised to become a core driver of low-carbon energy systems. However, it is critical to emphasize that all policies and infrastructure related to DSE must undergo rigorous assessments of their impacts on marine ecosystems. The deployment of such systems should accelerate green energy transitions without introducing new potential hazards to oceanic biodiversity.[102]

APPLICATIONS FOR ENERGY STORAGE

DSE employs electrical energy to dissociate seawater into hydrogen and oxygen. Conversely, there is a device capable of reversing this process by converting hydrogen and oxygen back into freshwater while releasing electrical energy. This device, known as a reversible seawater electrolyzer, is constructed by integrating an electrolyzer with a fuel cell.[103] Dionigi et al.[99] calculated that the reversible seawater electrolyzer requires approximately 3370 kW h^{-1} of energy input to produce 1 Nm^3 of freshwater, with the resulting water being of high purity. However, when hydrogen energy is converted into electricity and water via a fuel cell, reversible seawater electrolysis is predominantly regarded as an energy storage technology rather than a freshwater production method. This is because the energy consumption for desalinating and producing 1 Nm^3 of freshwater using reversible seawater electrolysis is significantly higher—by several orders of magnitude—than that of conventional seawater desalination techniques. Furthermore, given the exceptionally high energy density of hydrogen, the reversible electrolyzer device plays a crucial role in storing surplus renewable electricity.

A proposed process scheme for achieving stable and sustainable freshwater production integrated with energy storage, incorporating new energy sources, RO, direct seawater electrolyzers, and fuel cells, is illustrated in the diagram (Figure 9). Using solar energy as an example: during daylight hours, the photovoltaic system supplies electricity to both the RO unit and urban areas with power demands. Following seawater desalination by the RO unit, potable freshwater is generated, while the photovoltaic system concurrently meets the city's electricity requirements. Surplus solar energy is converted into electrical energy by the photovoltaic system and directly powers the direct seawater electrolyzer to produce hydrogen, which is subsequently stored in designated storage devices. During nighttime, when solar power is unavailable, the stored hydrogen is channeled into the hydrogen fuel cell to generate electricity, simultaneously producing freshwater. A portion of the electricity is allocated to the city for power supply, while the remainder drives the RO unit to produce additional freshwater.[99]

Moreover, the time-decoupling mechanism of the decoupled electrolyzer enables energy storage during the charging phase and energy release during the discharging phase. Gan et al.[104] proposed a novel mechanism based on the anode-cathode sequential ACS-OER utilizing nickel-hydroxide. By integrating electrochemical oxidation and reduction processes, this mechanism significantly reduces overvoltage and achieves time-decoupled production of hydrogen and oxygen gases, alongside electrochemical energy storage. Through the redox reaction of nickel-hydroxide, the system stores electrical energy during charging and releases it during discharging, effectively combining water electrolysis and battery functionalities. This hybrid device

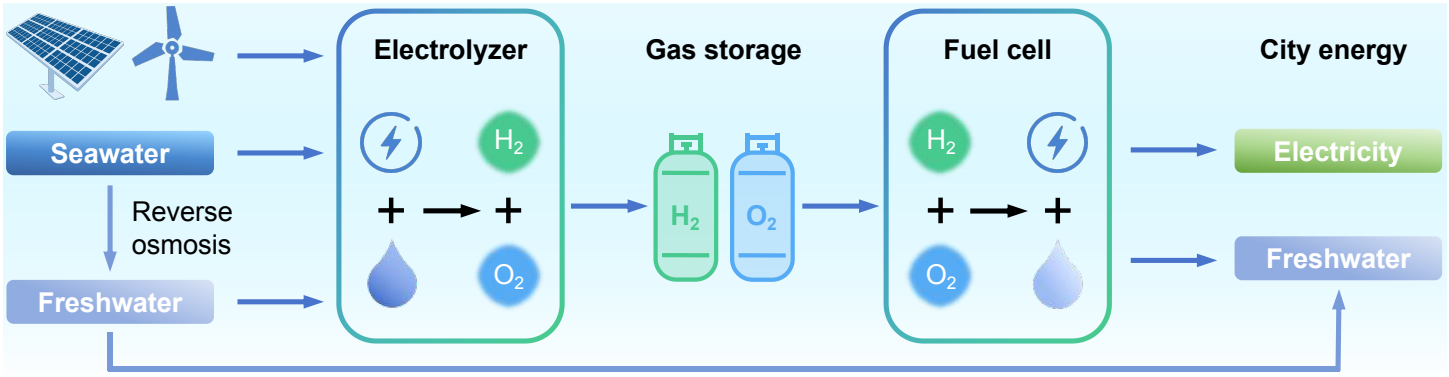

Figure 9. Process scheme of producing fresh water and energy storage based on seawater electrolysis.

not only enhances the efficiency of water electrolysis but also offers a novel approach to the storage and utilization of renewable energy. Recently, Xie et al.[95] extended the decoupling strategy to DSE, demonstrating the feasibility of seawater decoupling electrolyzers. These studies collectively establish the potential of seawater decoupling for energy storage.

CONCLUSION AND PROSPECTS

This review examines advancements in DSE focusing on catalyst design, anodic oxidation, electrolyzer innovations and energy storage integration. Effective catalyst design must address activity, selectivity and stability challenges. Enhancing activity requires optimizing intrinsic properties, active site density and optimizing conductivity through strategies like heteroatom doping, heterostructures, morphology engineering and defect engineering. Selectivity improvements involve promoting OER over CER by increasing OER activity or creating Cl^--repellent interfaces *via* surface coatings or electrostatic effects. Stability is achieved by mitigating impurity-induced side reactions, suppressing precipitate formation through local environment regulation and enhancing corrosion resistance. Adjusting surface wettability to facilitate gas desorption further alleviates mass transfer limitations. However, balancing these three parameters remains challenging, necessitating tailored designs aligned with specific electrolyzer requirements. Furthermore, in situ analysis techniques are essential for screening and evaluating novel catalysts for seawater electrolysis. Anodic-assisted oxidation reactions, replacing OER with small-molecule electrooxidation, reduce operational voltage and eliminate explosive hydrogen/O_2 mixing risks. Concurrently, optimized electrolyzer designs are critical for practical implementation. Indirect hydrogen production *via* desalination offers marginal energy savings but competes with direct methods in spatial-constrained scenarios. Both approaches warrant parallel development to address diverse applications. Integrating seawater electrolysis with renewable energy and fuel cells presents a promising energy storage solution. Continued research into remains essential, despite current technological and economic hurdles, to unlock sustainable hydrogen production in marine environments. As technology matures and offshore wind power progresses, seawater-based hydrogen production is set to become a fundamental driver of low-carbon energy systems.

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FUNDING AND ACKNOWLEDGMENTS

We acknowledge the support from the National Key Research and Development Program of China (No. 2022YFB3803600), National Natural Science Foundation of China (No. 52276212), the Suzhou Science and Technology Program (No. SYG202101), and the China Fundamental Research Funds for the Central Universities. The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

AUTHOR CONTRIBUTIONS

Shijun Ran: Conceptualization, Data analysis, Original Draft. Chenhao Zhao: Conceptualization, Review & Editing, Original Draft. Xueli Yan: Conceptualization, Review & Editing. Jinwen Shi, Dengwei Jing, Maochang Liu: Funding acquisition, Project administration, Writing-review & editing.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

Data are available from the corresponding author upon reasonable request.

SUPPLEMENTAL INFORMATION

It can be found online at https://doi.org/10.59717/j.xinn-energy.2025.100110