



Bioinspired hierarchical porous metallic glass for ultra-stable large-current hydrogen evolution in alkaline seawater

Wenqing Ruan^a, Jihan Jiang^a, Xiaoyu Wu^a, Zhe Chen^{a,b}, Jinbiao Huang^a, Pengjie Ren^a, Shihao Wen^a, Xingran Zhao^a, Jianan Fu^a, Xiaodi Liu^a, Jiang Ma^{a,*}

^a Shenzhen Key Laboratory of High Performance Nontraditional Manufacturing, College of Mechatronics and Control Engineering, Shenzhen University, Shenzhen, 518060, China

^b School of Materials Science and Engineering, Central South University, 932 South Lushan Road, Changsha, 410083, China

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ABSTRACT

Efficient, robust, large-current-capable electrocatalysts are essential for industrial green hydrogen production via seawater electrolysis, where chloride corrosion and hydrogen bubble accumulation challenge catalyst performance and longevity. Inspired by the mass transport-optimized hierarchical pores of natural diatom walls, we develop a facile one-step dissolution strategy to fabricate self-supported hierarchical porous Pt-based metallic glass (HPMG) electrocatalysts. The bioinspired structure maximizes active site exposure, accelerates electrolyte infiltration and hydrogen bubble desorption, and the amorphous matrix enhances corrosion resistance and optimizes HER kinetics. Benefiting from this synergy, HPMG exhibits ultra-long durability, retaining 97.9% activity after 1100 h at 10 mA cm^{-2} in 1 M KOH, with nearly full HER recovery via 1 M HCl treatment. Notably, in alkaline simulated seawater (1 M KOH + seawater), it delivers excellent large-current HER ($\eta_{10} = 44.8 \text{ mV}$, $\eta_{1000} = 345.7 \text{ mV}$, $\eta_{2000} = 613.7 \text{ mV}$) and stabilizes for 100 h at 1000 mA cm^{-2} . Additionally, HPMG shows superior full-pH HER activity ($\eta_{10} = 20.6 \text{ mV}$ in $0.5 \text{ M H}_2\text{SO}_4$, 26.2 mV in 1 M KOH, 31.8 mV in 1 M PBS), outperforming commercial Pt/C. This work provides a feasible route for developing bioinspired robust catalysts, which hold great promise for efficient hydrogen production from seawater.

1. Introduction

Hydrogen is a promising fossil fuel alternative owing to its high calorific value, zero carbon emissions, and renewability [1,2]. Water electrolysis is a scalable route for green hydrogen production by coupling with renewable energy (e.g., wind, solar) [3–5], and alkaline water electrolysis (AWE) dominates industrial applications due to its mature process, low cost, and good device compatibility [6,7]. However, AWE still faces critical challenges: noble metals (e.g., Pt) exhibit inferior HER activity in alkaline media due to unfavorable water dissociation and hydrogen adsorption kinetics [8], while conventional catalysts suffer from high overpotential at large current densities and poor long-term operational stability [9]. These issues are exacerbated in seawater by chloride corrosion and hydrogen bubble accumulation, making the development of robust, high-performance HER catalysts the key to advancing industrial alkaline/seawater electrolysis.

Constructing hierarchical porous structures is an effective strategy to boost HER performance by maximizing active site exposure and mass

transport efficiency [10,11], but traditional fabrication methods (template, dealloying, redox etching) suffer from tedious processing, harsh conditions, poor pore controllability, and low mechanical strength [12–16], hindering industrial scalability. Although metallic glasses (MGs) are promising HER candidates with abundant defect sites and excellent corrosion resistance from their short-range ordered/long-range disordered structure [10,17–21], reports on well-defined self-supported hierarchical porous MG catalysts remain scarce. Most existing porous catalysts also rely on polymer binders, which increase interfacial resistance and cause agglomeration/delamination during long-term operation [22]—a fatal flaw for industrial large-current-density applications. Thus, a scalable fabrication strategy for self-supported hierarchical porous MG catalysts is urgently needed to address the above issues.

Although Zhao et al. employed NaCl templates to fabricate porous structures via thermoplastic forming, the resulting architectures were predominantly microporous with only a limited fraction of nanopores, which is insufficient to maximize the specific surface area [21]. Inspired

* Corresponding author.

E-mail address: majiang@szu.edu.cn (J. Ma).

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by the hierarchical pore architecture of diatom walls—nature's optimized design for efficient mass transport [23,24] (Fig. 1a)—we develop a facile one-step dissolution strategy by leveraging the unique thermoplastic forming ability of MGs [25]. This strategy softens the MG in its supercooled liquid region ($\approx 275^\circ\text{C}$) under mild pressure, shapes it into a monolithic composite, and yields a micro-nano hierarchical porous structure via selective template dissolution. Compared with traditional methods, this approach features mild processing conditions (ambient pressure, no strong acids/alkalis), short preparation time, and controllable large-area porous construction [26], while the self-supported architecture eliminates the need for polymer binders, fundamentally solving the binder-induced performance degradation problem. Notably, this bioinspired fabrication strategy yields the target self-supported micro-nano hierarchical porous Pt-based metallic glass (HPMG) catalyst, whose structural advantages directly translate to exceptional catalytic performance.

This bioinspired self-supported micro-nano HPMG catalyst synergistically optimizes active site accessibility and mass transport: its nanopores maximize active site exposure, while interconnected micrometer-scale through-pores facilitate rapid mass transport. The hierarchical structure not only enables superior HER activity across full pH media but also endows the catalyst with ultra-long durability, achieving 97.9% activity retention after 1100 h of continuous operation, along with structural regenerability and excellent large-current-density performance in alkaline seawater—exemplified by stable operation for 100 h at 1000 mA cm^{-2} . This work integrates MG thermoplasticity with bioinspired hierarchical porous and self-supported design for HER catalysis, which holds great promise to address the key challenges of catalytic activity and long-term stability. It establishes a new paradigm for self-supported hierarchical porous catalyst design and provides a promising pathway for high-efficiency green hydrogen production from industrial alkaline seawater electrolysis.

2. Materials and methods

2.1. Fabrication of the HPMG catalyst

The MG ribbons were first cut and ball-milled into fine powder with a particle size of approximately 0.12 mm. The resulting MG powder was then mixed with 30 nm BaTiO_3 nanoparticles at a mass ratio of 4:1 using

ultrasonic blending. To ensure a homogeneous distribution, the mixed powders were further processed by ball milling followed by an additional 5-min ultrasonic treatment. The synthesis was carried out in a hot-pressing mold where the uniformly mixed powder was spread onto the lower mold plane (Fig. S1) to a controlled thickness of approximately 0.37 mm using a powder thickness control tool (Fig. S2). The powder was lightly compacted with the upper mold, fixed with a positioning mold, and then transferred to a desktop hot press. Under a preload of 100 N, the system was heated to 265°C while gradually increasing the pressure to 20 kN. The pressure was maintained until the temperature reached 275°C , after which the pressure was rapidly released and heating was stopped for cooling. The resulting MG- BaTiO_3 composite was immersed in 1 M HCl for 12 h to completely dissolve the BaTiO_3 nanoparticles, followed by thorough rinsing with deionized water and air-drying, yielding the final HPMG catalyst.

The remaining experimental sections, including fabrication of metallic glasses, preparation of Pt/C electrodes, physical characterizations, electrochemical measurements, and DFT calculations, are detailed in the Supporting Information.

3. Results and discussion

3.1. HPMG electrocatalyst

Pt-based MG ribbons served as the precursor for fabricating the self-supported micro-nano HPMG electrocatalyst. The catalysts are systematically named based on their elemental composition, which was accurately determined by inductively coupled plasma optical emission spectrometry (ICP) to ensure precise correlation between chemical composition and material performance. The precursor material was MG powder with a nominal composition of $\text{Pt}_{57.2}\text{Ni}_{5.7}\text{Cu}_{16.2}\text{P}_{20.9}$ (at%, Fig. S3a). This specific composition was designed to balance electrocatalytic activity with cost-effectiveness while maintaining the glass-forming ability of the amorphous matrix, consistent with previous studies on Pt-based HER catalysts [10,27]. After adopting the dissolution-based manufacturing strategy, the resulting HPMG electrocatalyst exhibited a nominal composition of $\text{Pt}_{57.2}\text{Ni}_{5.8}\text{Cu}_{16.5}\text{P}_{20.5}$ (at%, Fig. S3b). The negligible change in elemental composition pre- and post-processing, together with the subsequent XRD, HRTEM and XPS analyses confirms the exceptional compositional controllability of this

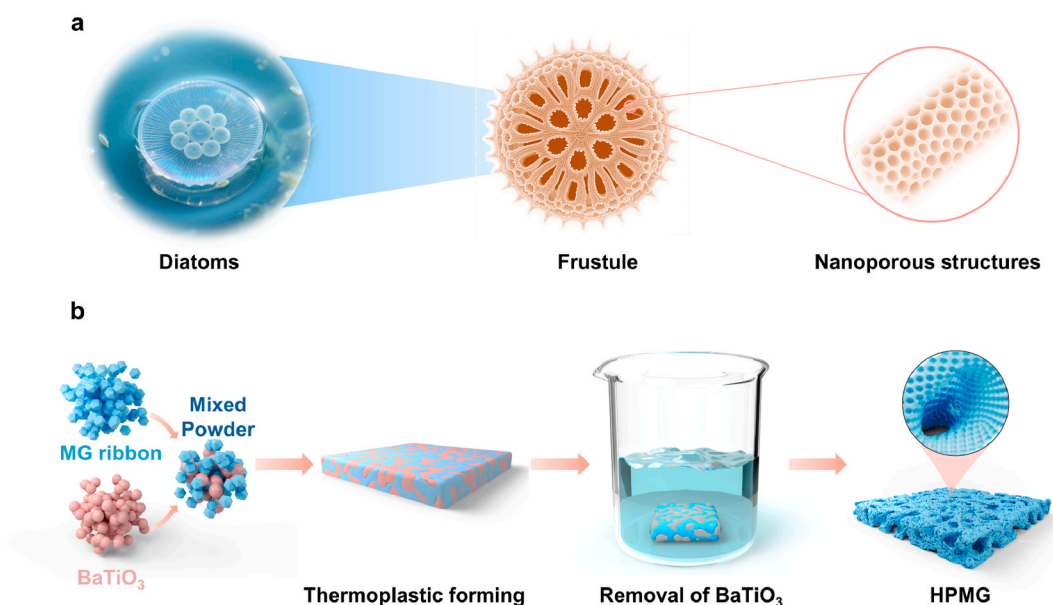


Fig. 1. Bio-inspired design and preparation of hierarchically porous structures. (a) The hierarchical pore architecture of diatom walls in nature. (b) The fabrication strategy for the HPMG electrocatalyst.

fabrication approach, ensuring the preservation of the electronic structure throughout the catalyst preparation process.

The fabrication strategy for the HPMG electrocatalyst is illustrated in Fig. 1b, which primarily leverages the thermoplastic forming ability of the MG. Differential scanning calorimetry (DSC) confirmed the amorphous structure of the MG precursor, revealing a glass transition temperature (T_g) of 222.17°C and a crystallization temperature (T_x) of 282.52°C (Fig. S4). The resulting supercooled liquid region (SLR: $\Delta T = T_x - T_g = 60.35^\circ\text{C}$) enables drastic plastic softening upon heating. When compressed within this temperature window, the softened MG powder particles adhere together and conform around co-dispersed BaTiO₃ nanoparticles. Subsequent dissolution of the nanoparticle template yields an integrated hierarchical porous structure in a single step. Phase evolution throughout this process was tracked by X-ray diffraction (XRD). The MG ribbon displays a characteristic broad diffraction halo between $2\theta = 30^\circ$ and 50° (Fig. S5), indicative of short-range order without long-range periodicity, confirming its fully amorphous nature [28]. Importantly, the HPMG retains this amorphous structure after fabrication, as evidenced by its nearly identical XRD pattern. This preserved amorphous nature and hierarchical porous structure underpin

the material's beneficial catalytic properties, including excellent corrosion resistance, abundant active sites, a high-energy state and a tailored local microenvironment that facilitates bubble release, while the associated electronic structure further contributes to its intrinsic catalytic activity [29–32].

Representative scanning electron microscopy (SEM) images of the HPMG electrocatalyst surface are presented in Fig. 2a–c, with additional micrographs at various magnifications provided in Fig. S6. The surface exhibits abundant macropores with diameters ranging from 5 to 100 μm . These macropores not only significantly increase the specific surface area but also provide ideal channels for electrolyte penetration, thereby enhancing mass transport efficiency during the HER process [33]. Notably, the walls of these large pores are uniformly decorated with nanopores measuring approximately 40–80 nm in diameter (Fig. S7). These nanostructures further increase the accessible surface area, while the inherent structural defects associated with them expose a greater number of catalytically active sites [20,34]. This hierarchical porosity synergistically provides abundant active sites and improves electrolyte accessibility, structurally optimizing the catalyst for enhanced HER performance. Cross-sectional SEM images (Fig. 2d–f and Fig. S8) confirm

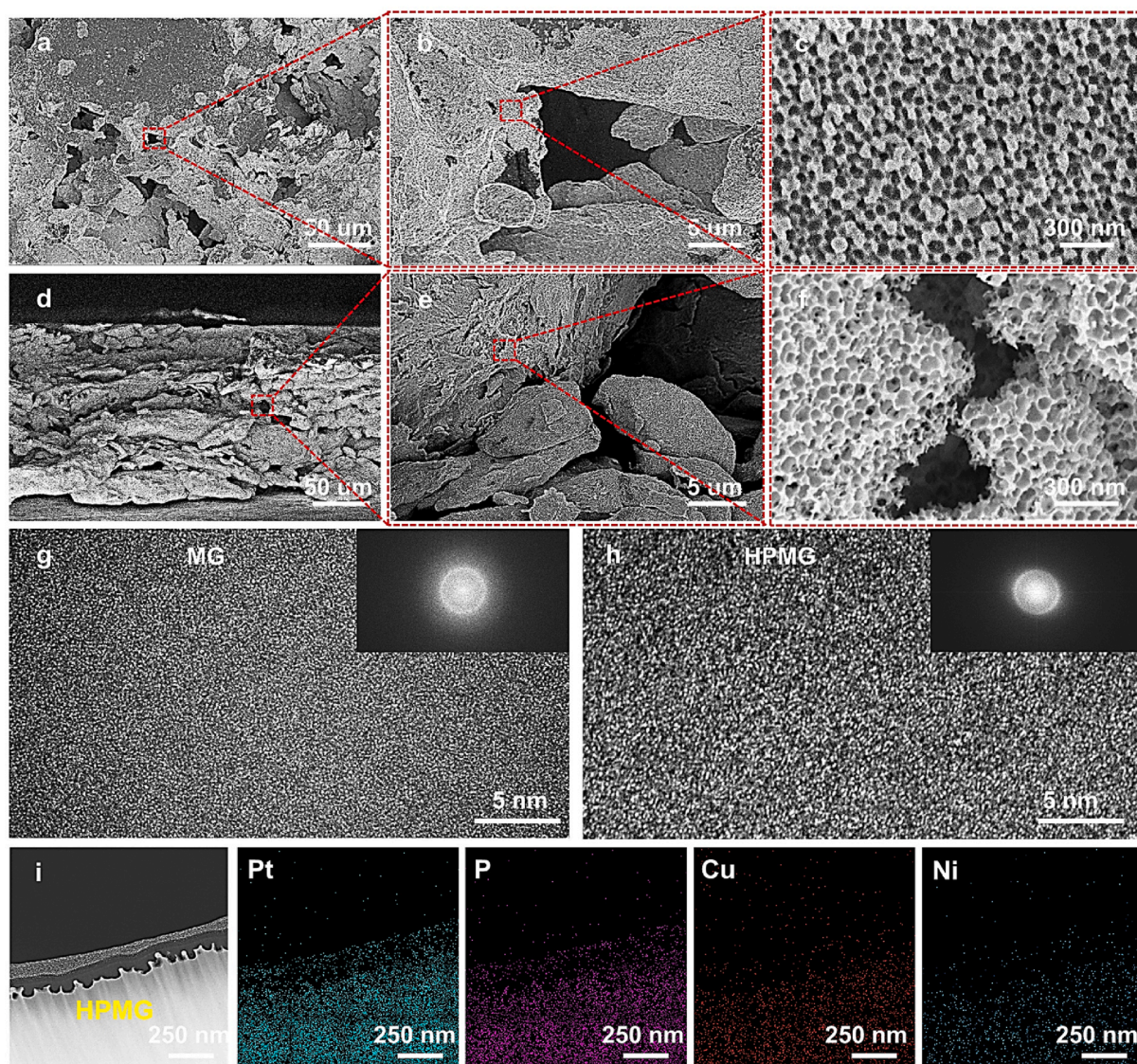


Fig. 2. Micro-nano hierarchical pore structure of HPMG. (a–c) SEM images of different magnifications on the surface of HPMG. (d–f), SEM images of different magnifications on the cross-section of HPMG. HRTEM and FFT image of the (g) MG, (h) HPMG. (i) STEM-HAADF images of the cross-section structure of the HPMG sample and the corresponding EDS mapping of elements Pt, Ni, Cu, and P.

the presence of interconnected through-pores throughout the catalyst bulk. The surfaces of these microchannels are also uniformly covered with nanoscale porosity. Consequently, the HPMG electrocatalyst integrates both micrometer-scale through-pores and nanoscale pores, a composite architecture that synergistically optimizes active site distribution, facilitates electrolyte infiltration, and promotes hydrogen bubble release [35]. Furthermore, this nanoporous structure is monolithic and self-supported, laying the groundwork for low interfacial resistance in subsequent electrochemical measurements [36]. The specific surface area of the HPMG, as evaluated by Brunauer-Emmett-Teller (BET) measurements, was determined to be $3.45 \text{ m}^2 \text{ g}^{-1}$ (Fig. S9). Although lower than that of commercial Pt/C, the hierarchical interconnected pore structure significantly improves the accessibility of active sites, as evidenced by its ECSA being 2.1 times that of Pt/C, ensuring sufficient reaction interfaces for HER.

The atomic-scale structure of the HPMG electrocatalyst was further investigated using high-resolution transmission electron microscopy (HRTEM). Fig. 2g and h presents the HRTEM images and corresponding Fast Fourier Transform (FFT) patterns of the precursor MG (g) and the final HPMG (h), respectively. The comparison of their FFT patterns, which show diffuse halos without distinct diffraction spots, confirms that the HPMG retains its amorphous state after the dissolution-based manufacturing process. The atomic arrangement remains short-range ordered but long-range disordered [37], indicating that the HPMG preserves the characteristic properties of MGs, including excellent corrosion resistance and a high density of active sites. Furthermore, the transmission electron microscopy (TEM) image and corresponding elemental mapping in Fig. 2i clearly reveal the monolithic nature of the nanoporous architecture. The elements Pt, P, Cu, and Ni are uniformly distributed throughout both the porous framework and the solid matrix, confirming the compositional homogeneity of the self-supported catalyst.

The above structural characterizations confirm that HPMG possesses a self-supported micro-nano hierarchical porous structure and uniform elemental distribution. Its unique amorphous phase and porous architecture lay the structural foundation for enhanced electrochemical performance. Below, we systematically evaluate its HER activity, kinetics, and long-term stability across full-pH electrolytes, with comparisons to commercial Pt/C and precursor MG.

3.2. Electrochemical performance of the HPMG

The electrocatalytic performance of the HPMG catalyst was systematically evaluated using a standard three-electrode electrochemical system. The HPMG served directly as the working electrode and was tested at room temperature in three different electrolytes: 1.0 M KOH, 1.0 M PBS, and 0.5 M H_2SO_4 . Comparative linear sweep voltammetry (LSV) tests were conducted on the HPMG, a commercial Pt/C (20 wt% Pt) catalyst, and the precursor MG ribbon. All current densities were normalized to the geometric surface area of the electrodes.

In 1.0 M KOH (Fig. 3a and b), the HPMG electrode demonstrated superior activity, requiring overpotentials of only 26.2 mV and 87.1 mV to achieve current densities of 10 and 100 mA cm^{-2} respectively. These values are significantly lower than those of the Pt/C catalyst (50.1 mV and 214.1 mV) and the MG ribbon (106.5 mV and 549.4 mV). This indicates that the exceptional HER activity of the HPMG electrode in alkaline media—characterized by its remarkably low onset overpotential, which surpasses most noble-metal-based benchmarks—is primarily attributed to its unique hierarchical micro-nano porous architecture. This structure effectively addresses the persistent challenge of high HER overpotentials typically associated with amorphous alloys under alkaline conditions. Notably, the HPMG maintained excellent performance at a high current density of 500 mA cm^{-2} , with an overpotential of 297.1 mV, vastly outperforming the MG (1332.4 mV). It is worth noting that the Pt/C catalyst suffered from severe performance degradation beyond 100 mA cm^{-2} , primarily due to poor hydrogen bubble release, which caused significant noise and instability in the LSV curves [38].

In a neutral 1.0 M PBS solution (Fig. S10a and b), HPMG exhibits the best η_{10} (31.8 mV) among the three catalysts, but its advantage diminishes at high current densities ($\eta_{100} = 293.8 \text{ mV}$, $\eta_{500} = 1213.5 \text{ mV}$) due to inherent high water dissociation and hydrogen desorption barriers in neutral media [39].

In the acidic 0.5 M H_2SO_4 electrolyte (Fig. S11a and b), HPMG shows exceptional η_{10} (20.6 mV) slightly outperforming Pt/C (21.3 mV), with $\eta_{100} = 90.6 \text{ mV}$ and $\eta_{500} = 297.6 \text{ mV}$ —significantly lower than MG (321.8 mV, 963.7 mV), confirming its robust acidic HER performance.

The HER kinetics and underlying mechanism were further probed by analyzing Tafel plots (overpotential η vs. $\log|j|$). The Tafel slope, an intrinsic characteristic of a catalyst, provides critical insight into the rate-determining step [40]. As shown in Fig. 3c, the HPMG

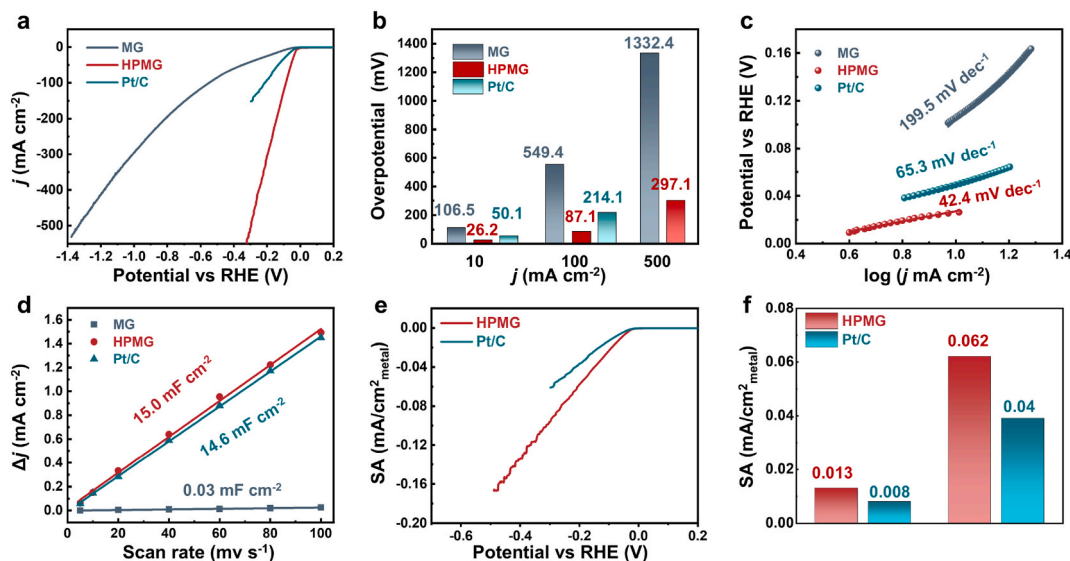


Fig. 3. HER performance in 1 M KOH. (a) LSV curves (i_R -corrected) of MG, HPMG and Pt/C in 1 M KOH at a scan rate of 2 mV s^{-1} . (b) Overpotentials at 10, 100, and 500 mA cm^{-2} of MG, HPMG, and Pt/C in 1 M KOH. (c) Corresponding Tafel plots of MG, HPMG and Pt/C in 1 M KOH. (d) Capacitive current densities against scan rates of MG, HPMG and Pt/C in 1 M KOH. (e-f) SA of HPMG and Pt/C in 1 M KOH.

electrocatalyst exhibited remarkably low Tafel slopes of 42.4 mV dec^{-1} in 1 M KOH, 82.8 mV dec^{-1} in 1 M PBS (Fig. S10c), and 21.2 mV dec^{-1} in $0.5 \text{ M H}_2\text{SO}_4$ (Fig. S11c). These values are substantially lower than those of the MG electrocatalyst (199.5, 185.2, and $144.1 \text{ mV dec}^{-1}$, respectively) and also significantly surpass the commercial Pt/C benchmark (65.3, 114.3, and 35.6 mV dec^{-1} , respectively), demonstrating superior HER kinetics across a wide pH range. This exceptional performance originates from the synergistic effects of the self-supported micro-nano hierarchical porous architecture, which accelerates electrolyte penetration and hydrogen bubble desorption [41], combined with the amorphous structure that promotes water dissociation and optimizes the hydrogen adsorption/desorption energy barrier [42]. The synergistic optimization of both efficiency and activity collectively enhances its adaptability for the hydrogen evolution reaction under universal pH conditions.

The electrochemical ECSA of the HPMG electrocatalyst, Pt/C (20 wt % Pt), and the precursor MG was estimated from cyclic voltammetry measurements (Fig. S12). The linear relationship between $\Delta j/2$ and scan rate yields the double-layer capacitance (C_{dl}), which is directly proportional to the ECSA [43], enabling quantitative comparison of active site availability among different catalysts. In 1.0 M KOH (Fig. 3d and Fig. S13a), the HPMG electrode exhibits a C_{dl} of 15.0 mF cm^{-2} , substantially higher than those of MG (0.03 mF cm^{-2}) and Pt/C (14.5 mF cm^{-2}). This corresponds to approximately 500 times more accessible active sites in HPMG compared to the precursor MG. In 1.0 M PBS (Fig. S13b), the C_{dl} of HPMG (8.3 mF cm^{-2}), while lower than that of Pt/C (21.1 mF cm^{-2}), still significantly exceeds the value for MG (0.1 mF cm^{-2}), representing an 80-fold increase in active site density. Similarly, in $0.5 \text{ M H}_2\text{SO}_4$ (Fig. S13c), HPMG demonstrates a remarkably high C_{dl} of 49.8 mF cm^{-2} , greatly surpassing both Pt/C (13.9 mF cm^{-2}) and MG (0.04 mF cm^{-2}), with an impressive 1000-fold enhancement over the precursor material. These results demonstrate that the hierarchical porous architecture, fabricated via the thermoplastic forming and dissolution-based strategy, dramatically expands the electrochemically active area, exposing a substantially greater number of active sites [44]. This approach provides a new paradigm for designing high-performance electrocatalysts through structural engineering.

The specific activity (SA), a key metric for evaluating the intrinsic activity of electrocatalysts, is presented in Fig. 3e. Higher SA values indicate superior catalytic performance [45]. In 1 M KOH at an overpotential of -70 mV vs. RHE (Fig. 3f), the HPMG electrode demonstrates a remarkable SA of $0.013 \text{ mA cm}_{ECSA}^{-2}$, representing a 1.6-fold enhancement compared to commercial Pt/C ($0.008 \text{ mA cm}_{ECSA}^{-2}$). When the overpotential increases to -210 mV , the SA of HPMG reaches $0.062 \text{ mA cm}_{ECSA}^{-2}$, again 1.5 times the value achieved by Pt/C ($0.04 \text{ mA cm}_{ECSA}^{-2}$). Additionally, the mass activity (MA) and turnover frequency (TOF) are critical metrics for evaluating the utilization efficiency of noble metals [46–48]. The MA of HPMG is $0.753 \text{ mA mgPt}^{-1}$. The TOF values for HPMG and Pt/C are 0.46 s^{-1} and 0.17 s^{-1} , respectively (Fig. S14). These results indicate that the HPMG exhibits a higher intrinsic catalytic activity than the commercial benchmark. Notably, HPMG maintains its exceptional catalytic performance even at elevated overpotentials, highlighting its robust activity across a wide potential window. At an overpotential of -70 mV vs. RHE in 1 M PBS electrolyte (Fig. S10d), the HPMG electrode delivers a remarkable specific activity of $0.01 \text{ mA cm}_{ECSA}^{-2}$, which is 2.5 times that of the commercial Pt/C catalyst ($0.004 \text{ mA cm}_{ECSA}^{-2}$). This result underscores the superior HER activity of the HPMG catalyst in a neutral environment. In $1 \text{ M H}_2\text{SO}_4$ at an overpotential of -70 mV vs. RHE (Fig. S11d), the HPMG electrode does not demonstrate a significant advantage in specific activity ($0.004 \text{ mA cm}_{ECSA}^{-2}$) over the commercial Pt/C catalyst ($0.022 \text{ mA cm}_{ECSA}^{-2}$). Nevertheless, the HPMG catalyst still delivers excellent overall HER performance, attributable to its considerably large ECSA that compensates for the lower per-site efficiency [49].

Electrochemical impedance spectroscopy (EIS) was employed to gain further insight into the catalytic performance, with all test potentials

summarized in Table S1. The Nyquist plots of HPMG, MG, and commercial Pt/C were fitted using a simple equivalent circuit model comprising a constant phase element (CPE), series resistance (R_s), and charge-transfer resistance (R_{ct}). The HPMG electrode exhibits a lower series resistance (R_s) than commercial Pt/C (Fig. S15), indicating superior electrical conductivity and more efficient charge transfer. This advantage is attributed to the self-supported nanoporous architecture, which is thermally pressed into a monolithic structure, thereby minimizing interfacial resistance commonly associated with binder-assisted Pt/C coatings prepared by drop-casting. More importantly, the synergistic effect between the nanoporous framework and the Pt active sites in HPMG significantly enhances charge transfer kinetics [50], further contributing to the enhanced catalytic activity.

Beyond catalytic activity, operational durability is a critical parameter for evaluating electrocatalysts in practical applications. Alkaline conditions are generally recognized as the most challenging environment for hydrogen evolution reactions due to their unfavorable reaction kinetics and highly corrosive nature [51]. Therefore, we primarily focused on evaluating the long-term stability of our catalyst under alkaline conditions, achieving exceptional durability exceeding 1100-h-a benchmark duration commonly reported in the field [52]. Commercial Pt/C catalysts often suffer from significant performance degradation, exhibiting a 50% decline in HER activity after only 2 h of operation (Fig. 4a). This rapid decay primarily stems from poor hydrogen bubble detachment, where accumulated bubbles block active sites during prolonged HER operation. In stark contrast, the HPMG catalyst developed in this work demonstrates exceptional stability during a rigorous 1100-h HER of CP test (Chronopotentiometric testing) in 1 M KOH, maintaining a constant current density of 10 mA cm^{-2} throughout the entire period (Fig. 4a). Prolonged HER consumes substantial amounts of water, necessitating the active addition of ultrapure water to maintain a consistent electrolyte concentration. This operational necessity, which restores the concentration and improves the electrolyte environment, causes minor recoveries in HER performance and consequently the observed fluctuations. Remarkably, the catalyst shows negligible activity decay, with the operating overpotential (η_{10}) increasing by only approximately 12 mV, from an initial 26.2 mV to 38.2 mV after testing (Fig. S16). This minimal performance loss is attributed to a surface self-reconfiguration process. As confirmed by comprehensive post-test characterization, the catalyst surface undergoes significant chemical evolution during prolonged operation. Energy-dispersive spectroscopy (EDS) mapping and line-scan EDS analyses (Figs. S17–19) revealed the formation of oxide nanoparticles and a change in the local elemental composition. Quantitative EDS analysis further showed that the Pt content increased from 15% in the as-prepared HPMG to 17% after removal of the surface oxide species by HCl treatment (Table S2), indicating Pt enrichment during the prolonged HER operation. This surface evolution may be associated with local dealloying and subsequent Pt enrichment, while the formation of oxide nanoparticles may be related to the subsequent relaxation to the open-circuit potential rather than the cathodic HER conditions [53]. This surface oxidation was further verified through X-ray photoelectron spectroscopy (XPS) analysis (Fig. S20). While this surface evolution causes minor HER activity loss, it simultaneously induces local dealloying and secondary nanostructure formation, enabling dynamic reconstruction that continuously generates fresh active sites [54]. This self-optimizing process further creates high-density defects that enhance catalytic performance through increased active site density, improved conductivity, and a favorable shift of the Pt d-band center toward the Fermi level ($\epsilon_d \rightarrow E_F$) [55,56]. Such self-renewal mechanism significantly mitigates performance degradation, enabling HPMG to maintain exceptionally stable HER operation.

Notably, after the 1100-h HER test, the HPMG catalyst could be largely restored to its original micro-nano hierarchical porous structure (Fig. 4b and Fig. S21) through a simple post-treatment process involving immersion in 1 M HCl followed by rinsing with deionized water. As

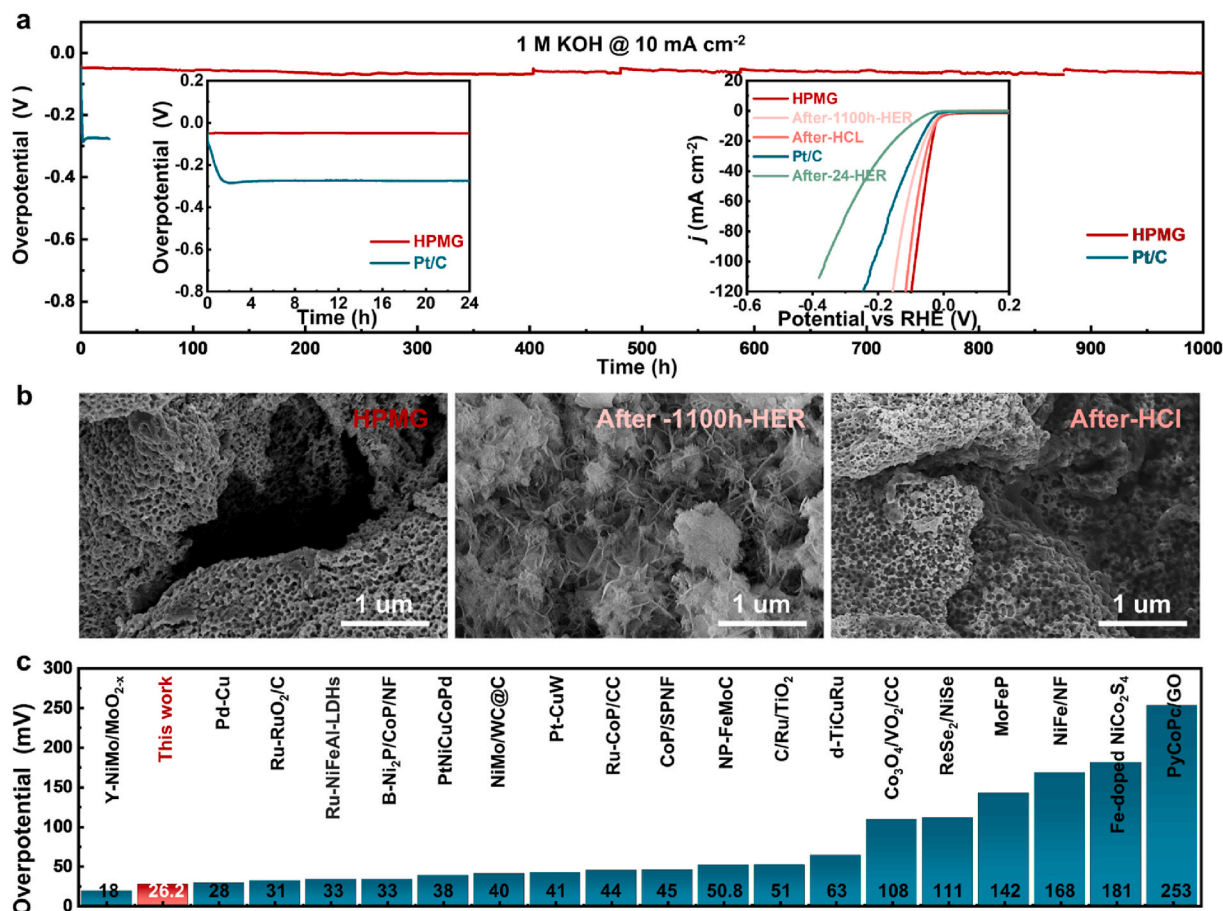


Fig. 4. Stability and structural regeneration of HPMG. (a) Stability test of the HPMG catalysts under a static overpotential of 10 mA cm⁻² in 1 M KOH. (b) Structural regeneration of the HPMG catalyst. (c) The comparison of the overpotential η_{10} of HER of the previous catalysts in 1 M KOH.

evidenced by EDS mapping (Fig. S22) and line-scan EDS profiles (Fig. S23), the surface oxide particles were almost completely removed after this regeneration step. Correspondingly, the HER performance was nearly fully recovered, with the overpotential ($\eta_{10} = 28.8$ mV) returning close to its initial value (26.2 mV). This demonstrates the excellent recoverability of both the structure and catalytic function of HPMG, highlighting its reusability. The remarkable stability originates from the unique self-supported configuration of the HPMG catalyst [57], combined with the protective effect of the hierarchical pores against degradation by reactants and products [58]. This architecture effectively mitigates the common issues of nanoparticle agglomeration and active site loss observed in conventional catalysts. By maintaining ultrastable HER performance (less than 5% activity loss after 1100 h) while preserving structural integrity, HPMG emerges as an ideal candidate material for industrial water electrolysis systems. These findings significantly surpass the durability benchmarks set by traditional Pt/C catalysts and effectively address one of the major bottlenecks in current hydrogen production technology. The slow performance degradation, recoverable nanoporous structure, and restorable HER activity of HPMG establish a new paradigm for designing cost-effective, high-performance, and renewable HER catalysts.

The HER performance of the HPMG catalyst was benchmarked against a wide range of reported electrocatalysts, including noble metals, transition metal oxides, dichalcogenides, nitrides, phosphides, carbides, borides, and MGs (Fig. 4c and Table S3). In alkaline media, the HPMG electrode exhibits a lower overpotential at 10 mA cm⁻² than most documented catalysts, demonstrating superior HER activity. Furthermore, it displays exceptional long-term stability, outperforming numerous counterparts (Fig. S24 and Table S4), demonstrating its

potential as an efficient HER catalyst. The HPMG also maintains competitive performance in both neutral and acidic electrolytes, surpassing the majority of reported catalysts in these media (Tables S5 and S6). This comprehensive comparison underscores the outstanding activity and durability of the HPMG catalyst across a wide pH range.

The excellent performance of HPMG at high current densities (e.g., 500/1000 mA cm⁻²) and rapid reaction kinetics (Fig. 3c) is primarily attributed to the mass transfer advantage of its hierarchical porous structure. This section will further clarify how this structure synergistically enhances HER catalytic efficiency by regulating hydrophilic/aerophobic properties, accelerating electrolyte penetration, and promoting hydrogen bubble release.

3.3. Enhanced HER performance through the hierarchical porous structure

During the HER process, both Pt/C (Fig. S25b and Video 2) and MG (Fig. S25c and Video 3 surfaces exhibit large bubble stagnation (Pt/C: $d \approx 210$ μm; MG: $d \approx 480$ μm), which blocks the active sites. In contrast, HPMG demonstrates exceptionally high hydrogen bubble desorption efficiency, generating only minute bubbles ($d \approx 60$ μm) without pinning (Fig. S25a and Video 1). This property ensures sustained contact between active sites and the electrolyte, thereby significantly enhancing the HER efficiency. The exceptional HER performance of the HPMG electrocatalyst is fundamentally attributed to its rationally designed micro-nano hierarchical porous architecture, which effectively modulates the number of surface coordination sites, facilitates electrolyte penetration, and promotes hydrogen gas desorption, thereby significantly enhancing the catalytic reaction kinetics [59].

To elucidate the structural advantages of HPMG, contact angle (CA) measurements were performed and compared with the precursor MG. The water contact angle (WCA), which critically influences the initial wetting behavior and thus the HER dynamics [60], was measured as $20 \pm 1^\circ$ for HPMG versus $83 \pm 1^\circ$ for MG in air (Fig. S26a and b). This pronounced hydrophilicity of the HPMG surface ensures rapid electrolyte infiltration into the porous network, enhancing mass transport efficiency. Simultaneously, the hierarchical pores significantly improve the gas-repelling ability. The bubble contact angle (BCA) measured under water was $146 \pm 1^\circ$ for HPMG, substantially higher than the $122 \pm 1^\circ$ observed for MG (Fig. S26c and d). This superior aerophobicity facilitates the efficient detachment and release of hydrogen bubbles from the catalyst surface—reducing bubble detachment time and bubble size (Fig. S25 and Video 1-3)—minimizing active site blocking and concentration polarization, which directly supports HPMG's excellent performance at $500/1000 \text{ mA cm}^{-2}$ (Fig. 3b and Fig. S11b).

Dynamic wetting characterization further revealed the rapid electrolyte penetration capability of the HPMG architecture. As shown in Fig. S26e and a water droplet completely infiltrated the hierarchical porous surface within 0.5 s, demonstrating its superhydrophilic nature. Concurrently, the BCA remained stable at $146 \pm 1^\circ$ (Fig. S26f), confirming the persistent aerophobicity of the HPMG surface. This optimal combination of superhydrophilicity and aerophobicity plays a crucial role in enhancing HER performance [61].

The structural advantages of HPMG are schematically illustrated in Fig. S26g and h, highlighting the fundamental differences between the hierarchical porous structure and the flat MG surface during HER operation. The HPMG architecture enables efficient electrolyte penetration throughout its porous network, providing abundant accessible active sites and facilitating water molecule interaction with catalytic centers [62], thereby accelerating reaction kinetics and supporting its low overpotential at high current densities. In contrast, the poor gas-repelling property of the flat MG surface leads to excessive bubble accumulation (Fig. S26g), which physically blocks active sites and isolates the catalyst from the electrolyte, significantly impairing reaction efficiency. Conversely, the excellent aerophobicity of HPMG promotes immediate bubble detachment (Fig. S26h), preventing active site coverage and maintaining uninterrupted catalytic interfaces. Therefore, the micro-nano hierarchical porous structure synergistically enhances both electrolyte accessibility and gas product removal, substantially improving the overall HER catalytic performance.

The mass transfer optimization of the macrostructure has clearly supported the performance advantages of HPMG, while the intrinsic mechanism of its 1100 h long-term stability (Fig. 4a) and structural regenerability (Fig. 4b) still needs to be analyzed from the perspective of elemental evolution. Below, XPS characterization is used to investigate the changes in elemental valence states and composition on the catalyst surface after stability testing, revealing its "oxidation-self-reconstruction" stability mechanism.

3.4. Elemental evolution after stability testing

XPS was employed to investigate the evolution of elemental composition and valence states in catalysts under different conditions, comparing the pristine MG, as-prepared HPMG, and the HPMG sample after 1100 h of CP testing (CP1100 h) (Fig. S20). In the Pt 4f spectra (Fig. S20a), the intensity of the Pt^0 peaks in HPMG showed no significant change compared to MG, while the Pt^{2+} peaks slightly intensified. After the prolonged HER test (CP1100 h), the Pt^0 peaks experienced a minor attenuation with a corresponding increase in Pt^{2+} signal; however, the valence state of Pt remained largely stable, indicating its robust nature during the extended HER operation [40]. For Ni 2p (Fig. S20b), the Ni^0 peak intensity in HPMG was initially similar to that in MG. After testing (CP1100 h), the Ni^0 peak nearly vanished, and the Ni^{2+} peak intensity markedly increased, indicating surface oxidation of Ni^0 to Ni^{2+} —consistent with the CuO/NiO oxide particles observed on the

post-test surface. The Cu 2p spectra (Fig. S20c) revealed minimal changes in the Cu^0 and Cu^{2+} peaks for the as-prepared HPMG. After the stability test, the Cu^0 peak decreased substantially, accompanied by a slight enhancement of the Cu^{2+} signature, suggesting oxidation and partial removal of Cu from the surface. The P 2p spectra (Fig. S20d) showed no obvious change in HPMG relative to MG, but a pronounced weakening of the P signal occurred after long-term testing. This suggests that the coordination environment of P atoms at the nanoporous surface may be less stable than in the bulk, making it more susceptible to removal during prolonged operation.

These results indicate that Ni, Cu, and P atoms undergo oxidation and dealloying during extended HER, while Pt remains relatively stable. This surface evolution may lead to the loss of some active sites, while the simultaneous dynamic self-reconfiguration of the micro-nano structure generates new active regions [54], collectively resulting in the observed slow performance decay and excellent long-term HER stability. Subsequent removal of the surface oxides with HCl solution may induce Pt enrichment on the HPMG surface, creating a more abundant and under-coordinated environment favorable for HER [63]. This explains the significant recovery of HER performance, aligning with the previously discussed regenerative capability of the HPMG catalyst.

Elemental evolution analysis indicates that the stability of Pt and the selective oxidation of Ni/Cu collectively ensure the long-term service capability of HPMG, but the electronic structure advantages of amorphous active sites remain unclear. This section will use DFT calculations to reveal the intrinsic electronic mechanism of HPMG's catalytic activity from the aspects of water dissociation energy barrier, hydrogen adsorption free energy, and charge transfer characteristics.

3.5. DFT calculations

The water dissociation free energy (ΔG_B) and the hydrogen adsorption free energy (ΔG_{H^*}) are key descriptors for evaluating the HER activity of electrocatalysts. Values approaching zero for these parameters indicate optimal water dissociation kinetics and hydrogen adsorption/desorption behavior, respectively [64,65]. To elucidate the influence of the amorphous structure on the HER performance, first-principles density functional theory (DFT) were conducted for both the amorphous HPMG and its crystalline Pt/C counterpart.

The chemical adsorption models of key intermediates (e.g., H_2O , *OH , and H^*) on the HPMG surface were optimized, and the corresponding free energy barriers were evaluated (Fig. 5). As shown in Fig. 5a, the free energy barrier for water dissociation on HPMG is -0.029 eV , significantly lower than that on the Pt/C (0.528 eV). This indicates that HPMG active sites more effectively facilitate the complete dissociation of water into H^* and *OH , thereby accelerating the sluggish Volmer step in alkaline media [66]. Furthermore, the hydrogen adsorption free energy (ΔG_{H^*}), another critical descriptor for HER electrocatalytic activity, reflects the efficiency of converting H^* intermediate into H_2 . The ΔG_{H^*} value of HPMG is -0.108 eV (Fig. 5b), which is closer to the thermoneutral ideal than that of Pt/C (-0.127 eV), suggesting more favorable hydrogen desorption kinetics [67].

The optimized H^* adsorption configuration on the HPMG surface (Fig. 5c) exhibits a distinctive charge density distribution, where the H atom gains 0.116 e^- , notably less than that on the Pt/C surface (0.121 e^- , Fig. 5d). This reduced electron accumulation at the H^* intermediate suggests weakened H-H bonding and promoted H_2 molecule desorption, thereby optimizing the hydrogen adsorption free energy [68]. Differential charge density analysis further confirms that the Pt-Ni and Pt-Cu alloy active sites within the amorphous domains facilitate significant electron transfer from the adsorbed H^* to adjacent Ni and Cu atoms. This electronic rearrangement weakens the H-H bonding and reduces the hydrogen desorption energy barrier, effectively overcoming the kinetic limitations in the HER process [69].

The exceptional performance of the HPMG electrocatalyst originates from its unique amorphous structure, which provides abundant

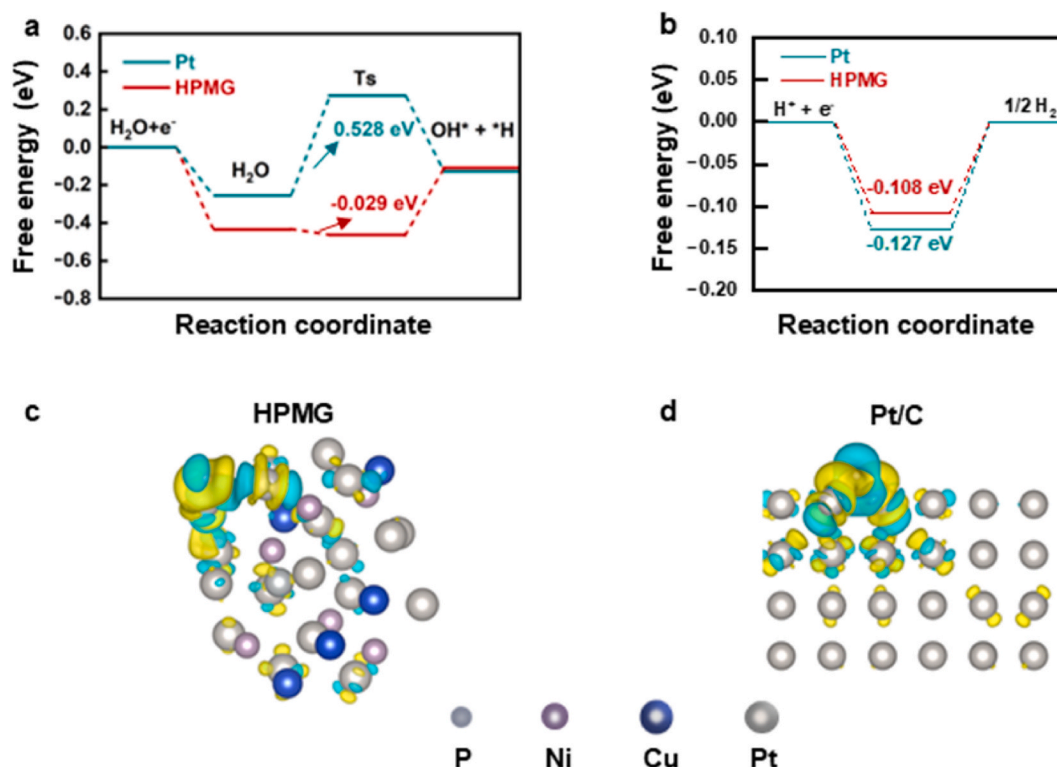


Fig. 5. Gibbs free energy barriers of water electrolysis on amorphous structures. (a) The free energy diagram of G_{BS} of HPMG and Pt. (b) The free energy diagram of H^* adsorption of HPMG and Pt. Iso-surface of the charge difference before and after adsorption of (c) HPMG and (d) Pt. The accumulation and reduction of the charge were denoted by yellow and cyan colors, respectively.

unsaturated coordination environments and enhances active site density. DFT calculations confirm that the amorphous phase reduces the water dissociation free energy and optimizes the hydrogen adsorption energy profile—these results, combined with the macrostructural mass transfer advantage (Section 3) and elemental stability mechanism (Section 4), collectively reveal the multi-scale synergy of HPMG: hierarchical pores optimize mass transport, selective oxidation of Ni/Cu stabilizes the structure, and amorphous active sites reduce reaction energy barriers—all contributing to its exceptional HER performance.

The synergistic effect of macrostructural mass transfer advantages, elemental stability mechanism, and microscopic electronic structure optimization provides theoretical and experimental support for the industrial application of HPMG. Below, we focus on alkaline seawater—the core industrial scenario—to verify its large-current-density HER performance and durability in environments containing chloride ions and complex ionic compositions, highlighting its potential for large-scale hydrogen production.

3.6. Hydrogen production from alkaline seawater

Building on the remarkable HER performance of the HPMG electrode in alkaline, neutral, and acidic electrolytes, its potential for seawater electrolysis was further evaluated to assess its viability for industrial-scale hydrogen production from seawater. Seawater electrolysis represents an attractive alternative method, leveraging the abundance of seawater as well as its high conductivity and salinity (~3.5 wt%) [70, 71]. However, natural seawater contains high concentrations of hard cations (e.g., Ca^{2+} , Mg^{2+}), which can precipitate on the catalyst surface during HER, blocking active sites and leading to rapid performance degradation [72]. In alkaline seawater, these cations are converted into insoluble $Mg(OH)_2$ and $Ca(OH)_2$ solids and effectively removed [73]. Furthermore, a high pH environment suppresses the competing chlorine evolution reaction, thereby minimizing the generation of chlorine gas

and hypochlorite at high current densities [74]. Consequently, alkaline seawater was selected as the electrolyte for this investigation due to its distinct advantages over natural seawater for water electrolysis applications.

Herein, simulated alkaline seawater (1 M KOH + Seawater) was employed as the electrolyte to evaluate the HER performance of the HPMG electrode in the presence of corrosive chloride ions and complex ionic matrix. As shown in Fig. 6a and b, the HPMG electrode demonstrates superior HER performance with remarkably low overpotentials ($\eta_{10} = 44.8$ mV, $\eta_{100} = 96.8$ mV, $\eta_{500} = 209.6$ mV, $\eta_{1000} = 345.7$ mV, $\eta_{2000} = 613.7$ mV). These values significantly outperform those of the Pt/C electrode ($\eta_{10} = 62.1$ mV, $\eta_{100} = 311.6$ mV) and the pristine MG ribbon ($\eta_{10} = 203.8$ mV, $\eta_{100} = 516.7$ mV, $\eta_{500} = 806.8$ mV, $\eta_{1000} = 1036.8$ mV)—benefiting from the hierarchical porous structure that resists chloride corrosion and promotes mass transport. The HPMG electrode exhibited a significantly lower Tafel slope (49.2 mV dec^{-1}) than the Pt/C benchmark (96.1 mV dec^{-1}) in alkaline seawater (Fig. 6c), indicating more favorable HER kinetics. Notably, the HPMG electrode maintains exceptional HER activity even at ultrahigh current densities (e.g., 2000 mA cm^{-2}), demonstrating its potential to significantly enhance the efficiency of seawater-to-hydrogen production.

The C_{dl} of HPMG was determined from CV measurements (Fig. S27), enabling calculation of its SA for comparison with Pt/C. As shown in Fig. 6d and Fig. S28, the SA values provide additional evidence of HPMG's superior HER performance. Compared to Pt/C, which exhibits SA values of 0.016 mA cm_{ECSA}^{-2} at -70 mV, 0.065 mA cm_{ECSA}^{-2} at -210 mV, and 0.115 mA cm_{ECSA}^{-2} at -350 mV (all vs. RHE), HPMG demonstrates significantly enhanced intrinsic activity with SA values of 0.016 mA cm^{-2} , 0.125 mA cm_{ECSA}^{-2} , and 0.245 mA cm_{ECSA}^{-2} at the corresponding overpotentials. Notably, the exceptional specific activity of HPMG becomes particularly pronounced at higher overpotentials, consistent with the experimentally observed outstanding HER performance at large current densities.

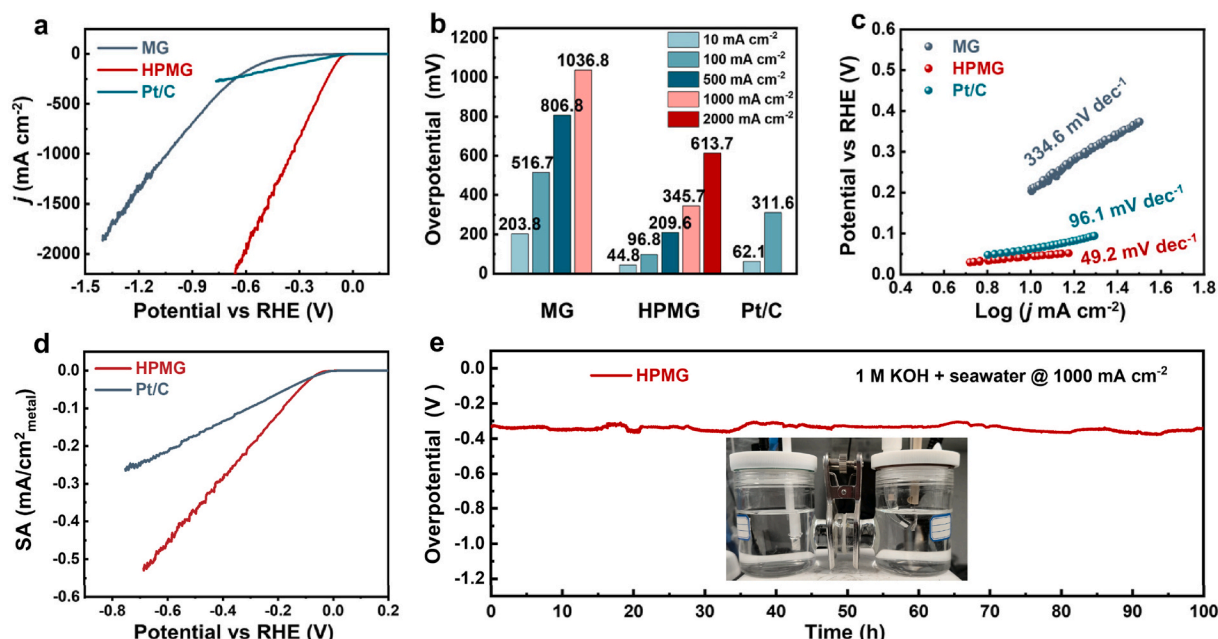


Fig. 6. HER performance in alkaline Seawater. (a) LSV curves (i_R -corrected) of MG, HPMG and Pt/C in 1 M KOH + seawater. (b) Overpotentials at 10, 100, 500, 1000, and 2000 mA cm⁻² of MG, HPMG, and Pt/C in 1 M KOH + seawater. (c) Corresponding Tafel plots of MG, HPMG and Pt/C in 1 M KOH + seawater. (d) SA of MG, HPMG and Pt/C in 1 M KOH + seawater. (e) Stability test of the HPMG catalysts under a constant current density of 1000 mA cm⁻² in 1 M KOH + seawater.

The durability of the HPMG electrode in alkaline seawater was further investigated using an H-type electrolytic cell equipped with a Pt sheet counter electrode, an Ag/AgCl (saturated KCl) reference electrode, and an anion exchange membrane. As shown in Fig. S29, the HPMG electrode exhibited exceptional stability during a 250-h CP test at 10 mA cm⁻², with negligible loss. More importantly, even under the demanding condition of 1000 mA cm⁻², the electrode maintained remarkable performance over a 100-h test, with an overpotential decay of only ~20 mV (from initial 345.7 mV to 365.5 mV, Fig. 6e)—a decay rate of 0.2 mV h⁻¹, confirming its industrial applicability. The significant fluctuations are primarily due to the rapid water consumption under high-current-density and prolonged HER. This necessitates the frequent replenishment of ultrapure water to maintain a consistent electrolyte concentration, which in turn leads to minor recoveries in HER performance and thereby causes the observed fluctuations. Moreover, no discernible Cl signal was detected in the HPMG electrode after the stability test, indicating negligible Cl accumulation within the detection limit of EDS (Fig. S30). These results demonstrate both the outstanding chloride corrosion resistance of the HPMG electrode in alkaline seawater and the feasibility of high-current-density seawater electrolysis for hydrogen production—consistent with its hierarchical porous structure and amorphous matrix that synergistically resist harsh environments. The hierarchical porous architecture maintains excellent HER performance even under harsh simulated seawater conditions, offering a promising solution for seawater hydrogen production [75]. This work is expected to provide an effective catalyst for hydrogen production from seawater.

4. Conclusion

Inspired by the hierarchical pore architecture of diatom walls—nature's optimized design for efficient mass transport—we developed a bioinspired one-step dissolution strategy to fabricate self-supported micro-nano HPMG electrocatalysts. This bioinspired design addresses the key challenges of balancing catalytic activity and long-term durability for HER catalysis. Benefiting from the synergistic effect of bionic hierarchical pores—maximizing active site exposure and accelerating electrolyte infiltration and hydrogen release—the HPMG delivers

superior full-pH HER activity with η_{10} values ranging from 20.6 to 31.8 mV and exceptional large-current-density performance in alkaline seawater, with η_{1000} of 345.7 mV and η_{2000} of 613.7 mV. The catalyst retains 97.9% of its initial activity after 1100 h of operation in 1 M KOH and can be regenerated via 1 M HCl treatment, achieving nearly full recovery of catalytic performance—attributed to the protective effect of the bionic structure and the electronic modulation of the amorphous matrix. This study demonstrates the potential of nature-inspired design for developing high-performance electrocatalysts, provides a promising route for hydrogen production from seawater, and offers new insights into the design of advanced electrocatalysts for energy conversion.

CRediT authorship contribution statement

Wenqing Ruan: Writing – review & editing, Resources, Formal analysis. **Jihan Jiang:** Writing – original draft, Methodology, Conceptualization. **Xiaoyu Wu:** Supervision, Data curation. **Zhe Chen:** Software, Investigation, Data curation. **Jinbiao Huang:** Methodology. **Pengjie Ren:** Formal analysis. **Shihao Wen:** Software. **Xingran Zhao:** Software. **Jianan Fu:** Investigation. **Xiaodi Liu:** Validation, Project administration. **Jiang Ma:** Writing – review & editing, Resources, Funding acquisition.

Data availability statement

All data needed to evaluate the conclusions in the paper are present in the paper and/or the supplementary information. Data are also available from the corresponding author upon request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.tramat.2026.100440>.

References

- J.J. Li, Z.Y. Jing, H.T. Bai, Z.H. Chen, A.I. Osman, M. Farghali, D.W. Rooney, P. S. Yap, Optimizing hydrogen production by alkaline water decomposition with transition metal-based electrocatalysts, *Environ. Chem. Lett.* 21 (2023) 2583–2617, <https://doi.org/10.1007/s10311-023-01616-z>.
- D.Y. Jin, F. Qiao, H.Q. Chu, Y. Xie, Progress in electrocatalytic hydrogen evolution of transition metal alloys: synthesis, structure, and mechanism analysis, *Nanoscale* 15 (2023) 7202–7226, <https://doi.org/10.1039/D3NR00514C>.
- Z.Y. Yu, Y. Duan, X.Y. Feng, X.X. Yu, M.R. Gao, S.H. Yu, Clean and affordable hydrogen fuel from alkaline water splitting: past, recent progress, and future prospects, *Adv. Mater.* 33 (2021) 2007100, <https://doi.org/10.1002/adma.202007100>.
- D. Wu, H. Du, Z. Liu, G. Bagliu, J. Lai, L. Wang, Construction of high-loading WO₃-x sub-nanometer clusters via orderly-anchored top-down strategy boost acidic hydrogen evolution, *EcoEnergy* 2 (2024) 724–735, <https://doi.org/10.1002/eece2.63>.
- R. Deng, M. Guo, C. Wang, Q. Zhang, Recent advances in cobalt phosphide-based materials for electrocatalytic water splitting: from catalytic mechanism and synthesis method to optimization design, *Nano Mater. Sci.* 6 (2024) 139–173, <https://doi.org/10.1016/j.nanoms.2022.04.003>.
- S. Anantharaj, S. Noda, V.R. Jothi, S. Yi, M. Driess, P.W. Menezes, Strategies and perspectives to catch the missing pieces in energy-efficient hydrogen evolution reaction in alkaline media, *Angew. Chem. Int. Ed.* 60 (2021) 18981–19006, <https://doi.org/10.1002/anie.202015738>.
- S.J. Liu, Z.G. Zhang, K. Dastafkan, Y. Shen, C. Zhao, M.K. Wang, Yttrium-doped NiMo-MoO₂ heterostructure electrocatalysts for hydrogen production from alkaline seawater, *Nat. Commun.* 16 (2025) 773, <https://doi.org/10.1038/s41467-025-55856-4>.
- J. Zhu, L.S. Hu, P.X. Zhao, L.Y.S. Lee, K.Y. Wong, Recent advances in electrocatalytic hydrogen evolution using nanoparticles, *Chem. Rev.* 120 (2020) 851–918, <https://doi.org/10.1021/acs.chemrev.9b00248>.
- T.W. Zhao, S.H. Wang, C. Jia, C.L. Rong, Z. Su, K. Dastafkan, Q. Zhang, C. Zhao, Cooperative boron and vanadium doping of nickel phosphides for hydrogen evolution in alkaline and anion exchange membrane water/seawater electrolyzers, *Small* 19 (2023) 2208076, <https://doi.org/10.1002/smll.202208076>.
- S.H. Zeng, W.Q. Ruan, Z. Chen, S. Ren, J.H. Jiang, J.Q. Lin, H.T. Zhang, Z.X. Zhang, J.N. Fu, Q. Chen, X. Liang, J. Ma, Dissolution manufacturing strategy for the facile synthesis of nanoporous metallic glass multifunctional catalyst, *Small Methods* 9 (2025) 2401109, <https://doi.org/10.1002/smt.202401109>.
- B. Zhong, P.Y. Kuang, L.X. Wang, J.G. Yu, Hierarchical porous nickel supported NiFeOxHy nanosheets for efficient and robust oxygen evolution electrocatalyst under industrial condition, *Appl. Catal. B Environ.* 299 (2021) 120668, <https://doi.org/10.1016/j.apcatb.2021.120668>.
- S.Q. Xiong, J.C. Fan, Y. Wang, J. Zhu, J.R. Yu, Z.M. Hu, A facile template approach to nitrogen-doped hierarchical porous carbon nanospheres from polydopamine for high-performance supercapacitors, *J. Mater. Chem. A* 5 (2017) 18242–18252, <https://doi.org/10.1039/c7ta05880b>.
- R.R. Song, J.H. Han, M. Okugawa, R. Belosludov, T. Wada, J. Jiang, D.X. Wei, A. Kudo, Y. Tian, M.W. Chen, H. Kato, Ultrafine nanoporous intermetallic catalysts by high-temperature liquid metal dealloying for electrochemical hydrogen production, *Nat. Commun.* 13 (2022) 5157, <https://doi.org/10.1038/s41467-022-32768-1>.
- M. Tu, D.E. Kravchenko, B.Z. Xia, V. Rubio-Giménez, N. Wauteraerts, R. Verbeke, I. F.J. Vankelecom, T. Stassin, W. Egger, M. Dickmann, H. Amenitsch, R. Ameloot, Template-mediated control over polymorphism in the vapor-assisted formation of zeolitic imidazolate framework powders and films, *Angew. Chem. Int. Ed.* 60 (2021) 7553–7558, <https://doi.org/10.1002/anie.202014791>.
- J. Chen, W.J. Xu, M.Z. Jiang, J. Chen, H.P. Jia, Polyoxometallate functionalizing CeO₂ via redox-etching precipitation to synergistically catalyze oxidation of gaseous chlorinated pollutants: from lab to practice, *Appl. Catal. B Environ.* 278 (2020) 119263, <https://doi.org/10.1016/j.apcatb.2020.119263>.
- J.H. Qin, H. Zhang, W.F. Lu, J.H. Zhu, J.S. Tian, J. Shen, Nanoporous NiPt alloy by dealloying of amorphous NiZrPt metallic glass for alkaline hydrogen evolution reaction, *Chem. Eng. J.* 490 (2024), <https://doi.org/10.1016/j.cej.2024.151700>.
- C.Q. Pei, S.Q. Chen, J.Y. Xie, S.D. Feng, M.Y. Yu, C. Zhan, Y.Y. Qian, G.N. Yang, Y. X. Chen, S. Lan, E.R. Kan, D. Wang, X.K. Mu, H. Hahn, B.A. Sun, G. Wilde, T. Feng, Strain engineering in gradient-structured metallic glasses for excellent overall water splitting, *Mater. Today* 85 (2025) 100–111, <https://doi.org/10.1016/j.mattod.2025.02.024>.
- Y.P. Zhang, F. Gao, D.Q. Wang, Z.L. Li, X.M. Wang, C.Q. Wang, K.W. Zhang, Y. K. Du, Amorphous/crystalline heterostructure transition-metal-based catalysts for high-performance water splitting, *Coord. Chem. Rev.* 475 (2023) 214916, <https://doi.org/10.1016/j.ccr.2022.214916>.
- S. Anantharaj, S. Noda, Amorphous catalysts and electrochemical water splitting: an untold story of harmony, *Small* 16 (2020) 1905779, <https://doi.org/10.1002/smll.201905779>.
- X.Y. Zhang, Y.Y. Yang, Y.J. Liu, Z. Jia, Q.Q. Wang, L.G. Sun, L.C. Zhang, J.J. Kruzic, J. Lu, B.L. Shen, Defect engineering of a high-entropy metallic glass surface for high-performance overall water splitting at ampere-level current densities, *Adv. Mater.* 35 (2023) 2303439, <https://doi.org/10.1002/adma.202303439>.
- X.R. Zhao, J.S. Liu, H.T. Zhang, J.H. Jiang, J.Q. Lin, J.N. Fu, Y. Zhang, X. Li, W. Q. Ruan, J. Ma, Fabrication of self-supporting hierarchical porous metallic glass for pH-universal hydrogen evolution reaction, *Chem. Eng. J.* 515 (2025) 163667, <https://doi.org/10.1016/j.cej.2025.163667>.
- S.L. Jiao, X.W. Fu, S.Y. Wang, Y. Zhao, Perfecting electrocatalysts via imperfections: towards the large-scale deployment of water electrolysis technology, *Energy Environ. Sci.* 14 (2021) 1722–1770, <https://doi.org/10.1039/d0ee03635h>.
- Y.Y. Fang, M.Y. Wu, Q. Zhang, F.Z. Zhou, C. Deng, Y. Yan, H.H. Shen, Y. Tang, Y. J. Wang, Hierarchical covalent organic frameworks-modified diatomite for efficient separation of bisphenol A from water in a convenient column mode, *Sep. Purif. Technol.* 298 (2022) 121611, <https://doi.org/10.1016/j.seppur.2022.121611>.
- C. Su, Y.Z. Mu, H. Zhan, G.S. Chen, K.K. Yang, Q.S. Fu, X.J. Sun, X.G. Chen, K. Shao, C. Feng, Hierarchical micro-nano porous structure of diatom biosilica induces selective absorption and protease inhibition for bleeding control, *ACS Nano* 19 (2025) 26673–26689, <https://doi.org/10.1021/acsnano.5c06075>.
- S. Sohrabi, J.N. Fu, L.Y. Li, Y. Zhang, X. Li, F. Sun, J. Ma, W.H. Wang, Manufacturing of metallic glass components: processes, structures and properties, *Prog. Mater. Sci.* 144 (2024) 101283, <https://doi.org/10.1016/j.pmatsci.2024.101283>.
- J.A. Fu, J. Yang, K. Wu, H.J. Lin, W.N. Wen, W.Q. Ruan, S. Ren, Z.X. Zhang, X. Liang, J. Ma, Metallic glue for designing composite materials with tailorable properties, *Mater. Horiz.* 8 (2021) 1690–1699, <https://doi.org/10.1039/D1MH00521A>.
- Z.W. Hu, K.Y. Chen, Y.H. Zhu, B. Liu, J.H. Shen, Synergistic effects of PtRhNiFeCu high entropy alloy nanocatalyst for hydrogen evolution and oxygen reduction reactions, *Small* 20 (2024) 2309819, <https://doi.org/10.1002/smll.202309819>.
- J. Chen, S. Ren, Z. Chen, J. Dong, Z. Lu, J. Zhu, L. Zhu, Y. Zhan, X. Zhao, W. Wang, Plasticity enhancement in metallic glasses via aging-assisted ultrasonic vibrations, *Mater. Futures* 4 (2025) 035003, <https://doi.org/10.1088/2752-5724/adeeae>.
- S. Su, M.J. Lee, W.H. Ryu, B. Huang, Z.L. Ning, Y.J. Huang, W.X. Huang, Q.X. Yuan, J.F. Sun, D. Sopp, E.S. Park, Tensile plasticity in amorphous microwires: the role of ion irradiation-induced gradient rejuvenation, *Int. J. Plast.* 190 (2025) 104371, <https://doi.org/10.1016/j.ijplas.2025.104371>.
- Y. Yu, Q. Wang, X. Li, Q. Xie, K. Xu, S. Zhang, H. Zhang, M. Gong, W. Lei, Laser-thermal reduction synthesis of high-entropy alloys towards high-performance pH universal hydrogen evolution reaction, *Nano Mater. Sci.* 7 (2025) 400–408, <https://doi.org/10.1016/j.nanoms.2024.05.012>.
- X. Fu, H. Li, Y. Zong, W. Xiao, J. Wang, H. Li, T. Ma, Z. Wu, L. Wang, Ultrafast construct local acidic microenvironment of boron-intercalated osmium with anti-precipitation and corrosion-resistant for seawater-splitting, *Angew. Chem. Int. Ed.* 64 (2025) e202512710, <https://doi.org/10.1002/ange.202512710>.
- W. Meng, J. Jiang, H. Wu, Y. Jiao, X. Zhang, Z. Cheng, X. Wang, Topological electrode Hf₂Se: enhanced hydrogen evolution reaction activity from nontrivial topological Fermi arc, *EcoEnergy* 3 (2025) 432–440, <https://doi.org/10.1002/eece2.82>.
- F. Zhou, Y. Zhou, Z. Zhang, P. Peng, J.M. Liang, H.Y. Gao, M.D. Kang, J. Wang, Hierarchical-heterostructured porous intermetallics in nickel-cobalt-aluminum alloys as efficient electrocatalysts for hydrogen evolution, *Chem. Eng. J.* 477 (2023) 146893, <https://doi.org/10.1016/j.cej.2023.146893>.
- J.K. Tian, Y.H. Song, X.D. Hao, X.D. Wang, Y.Q. Shen, P.Z. Liu, Z.B. Wei, T. Liao, L. Jiang, J.J. Guo, B.S. Xu, Z.Q. Sun, Greatly enhanced oxygen reduction reaction in anion exchange membrane fuel cell and Zn-air battery via hole inner edge reconstruction of 2D Pd nanomesh, *Adv. Mater.* 37 (2025) 2412051, <https://doi.org/10.1002/adma.202412051>.
- X.E. Jiang, V. Kyriakou, B.T. Wang, S.H. Deng, S. Costil, C.Y. Chen, T.K. Liu, C. M. Deng, H.L. Liao, T. Jiang, Hierarchical microporous Ni-based electrodes enable "two birds with one stone" in highly efficient and robust anion exchange membrane water electrolysis (AEMWE), *Chem. Eng. J.* 486 (2024) 150180, <https://doi.org/10.1016/j.cej.2024.150180>.
- M.K. Liu, P. Zhang, Z.H. Qu, Y. Yan, C. Lai, T.X. Liu, S.Q. Zhang, Conductive carbon nanofiber interpenetrated graphene architecture for ultra-stable sodium ion battery, *Nat. Commun.* 10 (2019) 3917, <https://doi.org/10.1038/s41467-019-11925-z>.
- J.Q. Tao, K.X. Zou, J.T. Zhou, H.J. Wu, L.L. Xu, J. Wang, X.W. Tao, H.X. Huang, Z. J. Yao, Phenolic multiple kinetics-dynamics and discrete crystallization

- thermodynamics in amorphous carbon nanostructures for electromagnetic wave absorption, *Nat. Commun.* 15 (2024) 10337, <https://doi.org/10.1038/s41467-024-54770-5>.
- [38] M.X. Xu, Y.R. Kang, L.L. Wang, Y.L. Zhang, G. Jiang, Y.F. Cai, Y.C. Tu, Q. Zhao, J. Chi, W. Song, H.M. Yu, J.T. Hu, W. Liu, R. Huang, L. Yu, J.L. Lu, X.H. Bao, D. H. Deng, Enriched asymmetric π electrons confining single-site Pt for acidic hydrogen evolution, *Joule* 9 (2025) 101968, <https://doi.org/10.1016/j.joule.2025.101968>.
- [39] J.Y. Xiao, C. Jiang, H. Zhang, Z. Xing, M. Qiu, Y. Yu, Amorphous core-shell NiMoP@CuNWs rod-like structure with hydrophilicity feature for efficient hydrogen production in neutral media, *Chin. J. Catal.* 63 (2024) 154–163, [https://doi.org/10.1016/S1872-2067\(24\)60086-0](https://doi.org/10.1016/S1872-2067(24)60086-0).
- [40] C.Z. Wan, Z.S. Zhang, J.C. Dong, M.J. Xu, H.T. Pu, D. Baumann, Z.Y. Lin, S. B. Wang, J. Huang, A.H. Shah, X.Q. Pan, T.D. Hu, A.N. Alexandrova, Y. Huang, X. F. Duan, Amorphous nickel hydroxide shell tailors local chemical environment on platinum surface for alkaline hydrogen evolution reaction, *Nat. Mater.* 22 (2023) 1022–1026, <https://doi.org/10.1038/s41563-023-01584-3>.
- [41] C.K. Zhao, Z.Y. Ding, L.M. Lei, Y.B. Sun, M. Hojamberdiev, Y.S. Xue, Y.J. Wang, M. Wang, H. Hu, M.B. Wu, Synergistic kinetics acceleration and mass transport by nanocone-nanoneedle hierarchical electrodes for efficient anion exchange membrane water electrolysis, *Adv. Funct. Mater.* 36 (2026) e13731, <https://doi.org/10.1002/adfm.202513731>.
- [42] Z.S. Hua, J. Wang, X.B. Wu, Z. Zeng, H. Liu, S.W. He, Z.D. Du, S.Y. Feng, M.K. Liu, X.L. Tian, Enhancing hydrogen evolution reaction via heterophase boundaries and stacking faults in molten salt electrodeposited Mg-Ni alloys, *Appl. Energy Mater.* 15 (2025) e03249, <https://doi.org/10.1002/aenm.202503249>.
- [43] D.B. Liu, X.Y. Li, S.M. Chen, H. Yang, C.D. Wang, C.Q. Wu, Y.A. Haleem, S. Duan, J. L. Xu, B.H. Ge, P.M. Ajayan, Y. Luo, J. Jiang, L. Song, Atomically dispersed platinum supported on curved carbon supports for efficient electrocatalytic hydrogen evolution, *Nat. Energy* 4 (2019) 512–518, <https://doi.org/10.1038/s41560-019-0402-6>.
- [44] Y.L. Zhao, X.H. Zhang, In situ activation graphitization to fabricate hierarchical porous graphitic carbon for supercapacitor, *Sci. Rep.* 11 (2021) 6825, <https://doi.org/10.1038/s41598-021-85661-0>.
- [45] G.T. Mao, X.Y. Han, Y. Xiong, L.Q. Luo, Y. Han, F.Q. Zhang, Q.M. Wang, Chemical order engineering in high-entropy alloys for enhanced methanol oxidation reaction via optimized coordination and lattice strain, *Appl. Catal. B Environ. Energy* 382 (2026) 125972, <https://doi.org/10.1016/j.apcatb.2025.125972>.
- [46] J. Li, R. Guo, X. Zhang, Y. Wang, H. Li, W. Xiao, L. Xin, Y. Zong, G. Fu, Z. Wu, Surface-enriched Ru on octahedral CeO₂ with strong electronic interactions as an efficient electrocatalyst for hydrogen generation in alkaline freshwater/seawater, *Inorg. Chem. Front.* 12 (2025) 821–832, <https://doi.org/10.1039/D4QI02552K>.
- [47] X. Wang, H. Zhao, X. Zhang, L. Li, W. Xiao, Q. Wu, L. Wang, Z. Wu, Strain-engineered interfacial water reorganization enables boosted HER/OER via intermediate adsorption modulation, *Nano Res. Energy* 5 (2026) e9120238, <https://doi.org/10.26599/NRE.2026.9120238>.
- [48] Z. Wu, P. Yang, Q. Li, W. Xiao, Z. Li, G. Xu, F. Liu, B. Jia, T. Ma, S. Feng, Microwave synthesis of Pt clusters on black TiO₂ with abundant oxygen vacancies for efficient acidic electrocatalytic hydrogen evolution, *Angew. Chem. Int. Ed.* 62 (2023) e202300406, <https://doi.org/10.1002/anie.202300406>.
- [49] T.X. Yang, Y.W. Zhang, J. Shi, G.P. Yang, J.O. Dang, M.L. Sun, A. Arshad, N. Zafar, S.N. Yun, Theoretical guidance for targeted modulation of metal-nitrogen single atom active sites on 3D porous carbon to optimize electrocatalytic performance in energy conversion applications, *J. Mater. Chem. A* 13 (2025) 5400–5414, <https://doi.org/10.1039/D4TA05738D>.
- [50] L. Ji, F.Z. Guo, K.H. Wang, L. Wang, Surfactant-assisted formation of nanoporous Pt particles as co-catalyst loaded on P25 and enhanced photocatalytic performance, *Chem. J. Chin. Univ.* 40 (2019) 1501–1509.
- [51] N. Mahmood, Y.D. Yao, J.W. Zhang, L. Pan, X.W. Zhang, J.J. Zou, Electrocatalysts for hydrogen evolution in alkaline electrolytes: mechanisms, challenges, and prospective solutions, *Adv. Sci.* 5 (2018) 1700464, <https://doi.org/10.1002/advs.201700464>.
- [52] C. Feng, F.Z. Wang, Z. Liu, M. Nakabayashi, Y.Q. Xiao, Q.G. Zeng, J. Fu, Q.B. Wu, C.H. Cui, Y.F. Han, N. Shibata, K. Domen, I.D. Sharp, Y.B. Li, A self-healing catalyst for electrocatalytic and photoelectrochemical oxygen evolution in highly alkaline conditions, *Nat. Commun.* 12 (2021) 5980, <https://doi.org/10.1038/s41467-021-26281-0>.
- [53] E. Ender, Y. Wang, C. Tseng, H.E. Taylor, L. Dwyer, K. Li, H. Becker, K. Felisari, S. J. Haigh, R. Rao, Surface memory in nickel HER catalysts: hydroxide formation at open-circuit potential, *ACS Catal.* 16 (2026) 14196–14204, <https://doi.org/10.1021/acscatal.6c00325>.
- [54] G. Doubek, R.C. Sekol, J.Y. Li, W.H. Ryu, F.S. Gittleson, S. Nejadi, E. Moy, C. Reid, M. Carmo, M. Linardi, P. Bordeenithikasek, E. Kinser, Y.H. Liu, X. Tong, C. O. Osuji, J. Schroers, S. Mukherjee, A.D. Taylor, Guided evolution of bulk metallic glass nanostructures: a platform for designing 3D electrocatalytic surfaces, *Adv. Mater.* 28 (2016) 1940–1949, <https://doi.org/10.1002/adma.201504504>.
- [55] T.Y. Jeon, S.K. Kim, N. Pinna, A. Sharma, J. Park, S.Y. Lee, H.C. Lee, S.W. Kang, H. K. Lee, H.H. Lee, Selective dissolution of surface nickel close to platinum in PtNi nanocatalyst toward oxygen reduction reaction, *Chem. Mater.* 28 (2016) 1879–1887, <https://doi.org/10.1021/acs.chemmater.6b00103>.
- [56] Y.B. Zhang, J.A. Pan, G. Gong, R.X. Song, Y. Yuan, M.Z. Li, W.F. Hu, P.C. Fan, L. X. Yuan, L.L. Wang, In situ surface reconstruction of catalysts for enhanced hydrogen evolution, *Catalysts* 13 (2023) 120, <https://doi.org/10.3390/catal13010120>.
- [57] J. Li, Z. Wen, Z.X. Hui, Z.W. Chen, C.C. Yang, Q. Jiang, Graphene-MoS₂ vertically anchored on an MXene-derived accordion-like TiO₂/C skeleton: an ultrastable HER catalyst, *J. Mater. Chem. A* 8 (2020) 14223–14233, <https://doi.org/10.1039/c9ta13965f>.
- [58] N. Yang, T.J. Fu, C.T. Cao, X.Q. Wu, H.L. Zheng, Z. Li, Constructing hierarchical ZSM-5 coated with small ZSM-5 crystals via oriented-attachment and in situ assembly for methanol-to-aromatics reaction, *Front. Chem. Sci. Eng.* 18 (2024) 76, <https://doi.org/10.1007/s11705-024-2432-2>.
- [59] Q.L. Kang, M.F. Su, Y.N. Luo, T. Wang, F. Gao, Q.Y. Lu, Chemical fermentation pore creation on multilevel bio-carbon structure with in situ Ni-Fe alloy loading for superior oxygen evolution reaction electrocatalysis, *Nano-Micro Lett.* 17 (2025) 269, <https://doi.org/10.1007/s40820-025-01777-2>.
- [60] L.J. Zhang, H.H. Hu, C. Sun, D.D. Xiao, H.T. Wang, Y. Xiao, S.W. Zhao, K.H. Chen, W.X. Lin, Y.C. Shao, X.Y. Wang, C.W. Pao, L.L. Han, Bimetallic nanolloys planted on super-hydrophilic carbon nanocages featuring tip-intensified hydrogen evolution electrocatalysis, *Nat. Commun.* 15 (2024) 7179, <https://doi.org/10.1038/s41467-024-51370-1>.
- [61] W. An, Z.Y. Liu, T.J. Shen, Y.Z. Du, C. Huang, B.Y. Duan, C.X. Wang, G.Y. Huang, Y. M. Yan, S.M. Xu, Regulating d-p hybridization in Mo-doped CoP nanoflowers with superior wetting surfaces for water splitting at 3000 mA cm⁻², *ACS Catal.* 15 (2025) 10358–10371, <https://doi.org/10.1021/acscatal.4c07485>.
- [62] K. Jiang, Z.X. Liu, Z. Wang, F. Xie, X.Y. Yuan, Y.W. Tan, Manipulating interfacial water by metallic Pt1Co6 sites on self-adaptive metal phosphides to enhance water electrolysis, *Adv. Mater.* 37 (2025) 2419644, <https://doi.org/10.1002/adma.202419644>.
- [63] G. Sun, R. Luo, D. Fu, K. Wu, X. Wang, X. Bian, Z. Lu, X. Chang, Z. Wang, S. Huang, Y. Zhu, J. Zhou, S. Chen, C. Pei, Z.-J. Zhao, J. Gong, Full utilization of noble metals by atom abstraction for propane dehydrogenation, *Science* (2025), <https://doi.org/10.1126/science.adw3053>.
- [64] M.H. Liao, Q.G. Zhu, Y.Y. Li, Q.Z. Yan, S.S. Zhou, J.J. Dai, Y.C. Huang, J. Gu, Polyoxometalates hydrogels derived porous conductive Pt@Mo2C carbon aerogels as self-supporting electrocatalysts for enhancing alkaline hydrogen evolution reaction, *Rare Met.* 44 (2025) 8609–8618, <https://doi.org/10.1007/s12598-025-03515-4>.
- [65] S. Lee, S. Lee, C. Kim, Y.K. Han, Origin of hydrogen adsorption free energy variation in electrochemical hydrogen evolution in transition-metal dichalcogenides, *Mater. Today Energy* 43 (2024) 101581, <https://doi.org/10.1016/j.mtener.2024.101581>.
- [66] Y.Z. Wu, Y.Y. Zhao, P.L. Zhai, C. Wang, J.F. Gao, L.C. Sun, J.G. Hou, Triggering lattice oxygen activation of single-atomic Mo sites anchored on Ni-Fe oxyhydroxides nanoarrays for electrochemical water oxidation, *Adv. Mater.* 34 (2022) 2202523, <https://doi.org/10.1002/adma.202202523>.
- [67] J. Wang, D. Wang, Z.L. Zhao, G.G. Chang, W. Guo, J. Xu, Y.Y. Xiang, J.R. Li, J.S. Li, 2D MOF structure tuning atomic Ru sites for efficient and robust proton exchange membrane water electrolysis, *Small* 21 (2025) 2410657, <https://doi.org/10.1002/smll.202410657>.
- [68] B. Deng, Z.Y. Wu, E.K. Feng, L. Ma, Z. Wang, J.H. Chen, L. Eddy, A. Latham, T. Wang, W.Y. Chen, Y. Cheng, S.C. Xu, Q.M. Liu, B.I. Yakobson, H.T. Wang, Y. F. Zhao, J.M. Tour, Coupling amorphization and compositional optimization of ternary metal phosphides toward high-performance electrocatalytic hydrogen production, *J. Am. Chem. Soc.* 147 (2025) 16129–16140, <https://doi.org/10.1021/jacs.5c00071>.
- [69] M.M. Jiang, J. Xu, Q. Zhou, Y.J. Chen, P. Munroe, L.L. Li, Z.H. Xie, Y.P. Wu, S. J. Peng, Rationally designed asymmetric Pt–O–Cu ligand to stabilize active sites towards superior industrial-standard alkaline hydrogen evolution, *Angew. Chem. Int. Ed.* 64 (2025) e202510259, <https://doi.org/10.1002/anie.202510259>.
- [70] T. Liu, C. Lan, M. Tang, M.X. Li, Y.T. Xu, H.R. Yang, Q.Y. Deng, W.C. Jiang, Z. Y. Zhao, Y.F. Wu, H.P. Xie, Redox-mediated decoupled seawater direct splitting for H₂ production, *Nat. Commun.* 15 (2024) 8874, <https://doi.org/10.1038/s41467-024-53335-w>.
- [71] T. Liu, Z.Y. Zhao, W.B. Tang, Y. Chen, C. Lan, L.Y. Zhu, W.C. Jiang, Y.F. Wu, Y. P. Wang, Z.Z. Yang, D.S. Yang, Q.J. Wang, L.B. Luo, T.S. Liu, H.P. Xie, In situ direct seawater electrolysis using floating platform in ocean with uncontrollable wave motion, *Nat. Commun.* 15 (2024) 5305, <https://doi.org/10.1038/s41467-024-49639-6>.
- [72] H.S. Hu, Z.R. Zhang, L.J. Liu, X.L. Che, J.C. Wang, Y. Zhu, J.P. Attfield, M.H. Yang, Efficient and durable seawater electrolysis with a V2O3-protected catalyst, *Sci. Adv.* 10 (2024) eadn7012, <https://doi.org/10.1126/sciadv.adn7012>.
- [73] C.H.D. Nieto, N.A. Palacios, K. Verbeeck, A. Prévost, K. Rabaey, V. Flexer, Membrane electrolysis for the removal of Mg²⁺ and Ca²⁺ from lithium rich brines, *Water Res.* 154 (2019) 117–124, <https://doi.org/10.1016/j.watres.2019.01.050>.
- [74] W.M. Tong, M. Forster, F. Dionigi, S. Dresch, R.S. Erami, P. Strasser, A.J. Cowan, P. Farràs, Publisher correction: electrolysis of low-grade and saline surface water, *Nat. Energy* 6 (2021) 935, <https://doi.org/10.1038/s41560-021-00851-4>.
- [75] Z.B. Li, H.N. Zhong, X.J. Liu, F.K. Chiang, R. Li, H.W. Chen, X.Z. Wang, C.B. Wan, Y. Wu, H. Wang, S.H. Jiang, X.B. Zhang, Z.P. Lu, Dynamic dissolution-deposition equilibrium enables unprecedented HER stability in acidic PEMWE, *Adv. Mater.* 37 (2025) e10703, <https://doi.org/10.1002/adma.202510703>.