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Research on the Application of High-Entropy Alloys in Water Electrolysis for Hydrogen Production

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Abstract. High-entropy alloys (HEAs) exhibit remarkable catalytic potential for hydrogen evolution (HER) and oxygen evolution (OER) reactions in water electrolysis, owes to their tunable electronic structures and synergistic multi-element effects. This paper systematically reviews the design strategies, performance advantages, and applications of HEAs: stabilizing solid solution phases through the high-entropy (HE) effect, optimizing adsorption energies of intermediate species via lattice distortion, and enhancing active sites synergistically by leveraging the cocktail effect. Studies show that optimally designed HEAs (such as NiFeCoCrMn, AlCoCrFeNi, etc.) exhibit catalytic activities and excellent stabilities close to those of noble metals in alkaline and acidic media. However, their practical application still faces challenges including difficulty in identifying active sites, obstacles in scalable synthesis, and insufficient long-term operational stability. Future research should focus on atomic-level active center design, the innovation of sophisticated characterization methods, and the optimization of industrial-scale fabrication processes to promote the practical application of HEA catalysts in the clean energy field. This work provides new insights for the design of high-performance water electrolysis catalysts and holds important significance for advancing renewable energy conversion technologies.

Keywords: High-entropy Alloys, Preparation Methods, Water electrolysis, Hydrogen Generation

1. Introduction

Hydrogen energy has become a highly popular form of green energy. Based on the carbon emissions produced during its preparation, hydrogen can be classified into gray hydrogen, blue hydrogen, and green hydrogen. Currently, gray hydrogen accounts for approximately 96% of the global hydrogen production, whereas environmentally friendly and clean green hydrogen constitutes only a small proportion. According to different hydrogen production methods, hydrogen can be categorized as fossil fuel-based hydrogen [1], industrial by-product hydrogen [2], water electrolysis hydrogen [3], and renewable energy hydrogen [4]. In China, hydrogen is mainly produced from fossil fuels or as an industrial by-product. Fossil fuel-based hydrogen generation produces a large amount of carbon dioxide while consuming substantial energy, and industrial by-product hydrogen suffers from insufficient purity, which prevents its application in battery manufacturing. Hydrogen produced by

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water electrolysis is characterized by its green and clean nature, making it a clean and efficient approach. Therefore, the effective utilization of water resources for hydrogen production via electrolysis represents the future development direction. However, the use of platinum electrodes during water electrolysis is costly, significantly increasing the cost of hydrogen production. How to effectively reduce the electrolysis cost remains a long-standing challenge.

The preparation of high-entropy alloys (HEAs) has gained popularity over the past two decades. HEAs are generally composed of five or more metallic elements arranged in a specific manner, forming alloys with high configurational entropy [5]. Through systematic studies of HEA systems, their characteristics can be summarized into four main effects: the high-entropy (HE) effect, lattice distortion effect, sluggish diffusion effect, and cocktail effect. (1) HE effect: HEAs consist of multiple metallic components, significantly increasing the configurational entropy of the alloy. By reducing the system's Gibbs free energy, this promotes stable solid solution phase formation and significantly improves the material's thermal stability. (2) Lattice distortion effect: The multiple elements are randomly distributed within the crystal lattice, resulting in uneven strain fields, known as the lattice distortion effect. This effect hinders dislocation movement, stabilizing the structure and significantly improving the strength and hardness of the HEAs. (3) Sluggish diffusion effect: Due to the presence of multiple components with large differences in bonding energies, atomic migration rates are markedly reduced. Consequently, HEAs can maintain their fine-grained microstructure even at high temperatures, effectively preventing recrystallization and improving high-temperature stability. (4) Cocktail effect: The synergistic interaction among multiple elements endows HEAs with performance surpassing that of any individual constituent, enabling adaptation to more complex environments and exhibiting superior overall properties—similar to a cocktail. For example, in the Al-CoCrFeNi system, Al promotes the formation of the BCC structure, while Cr enhances corrosion resistance. Compared with the Al-CoCrFeNi system, the CoCrNiFeAlCu system [6] contains a certain amount of Cu. Since Cu can promote heterogeneous nucleation of L12 precipitates, it can enhance the synergistic effect between strength and ductility, and improve the comprehensive mechanical properties of the material. Thus, by adjusting the types and proportions of constituent elements, HEAs can be directionally designed to optimize material performance. The combined action of these four effects endows HEAs with excellent properties such as high strength and toughness [7], high-temperature resistance [8], corrosion resistance [9], and radiation resistance [10], breaking through the performance limits of traditional alloys and making them promising candidates as a new generation of high-performance materials.

The primary preparation methods of HEAs—carbon-thermal shock, electrochemical synthesis, and flow synthesis—are introduced in this paper. HEAs are classified into noble metal-based HEAs and non-noble metal-based HEAs according to the presence or absence of noble metals. Several examples of each type are presented and their performances are compared with those of Pt-based alloys. It is found that HEAs, owing to their higher stability and relatively lower cost, hold promising prospects in electrocatalytic water splitting applications. However, the comparison also reveals that there remains a significant gap between current HEA catalysts and practical industrial-scale production. Further improvements are necessary in catalytic active site identification, scalable manufacturing, and long-term stability. This work analyzes the industry prospects and proposes corresponding solutions to existing challenges, emphasizing that exploring advanced characterization techniques and leveraging artificial intelligence for catalyst design will be indispensable paths for future development.

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2. Preparation methods of HEAs

The overall approach to preparing HEAs involves uniformly integrating multiple target metals through specific methods. The fundamental principle is based on conventional alloy fabrication techniques. According to the latest research reports, this paper summarizes several commonly used preparation methods for HEAs and analyzes the advantages and disadvantages of each synthesis technique.

2.1. Carbon-thermal shock method

The carbon-thermal shock (CTS) method involves rapidly heating and cooling metal precursors under conditions providing oxygen and carbon. Under the combined catalytic effect of high temperature and liquid metal, fast-moving particles undergo continuous "fission" and "fusion," achieving uniform mixing of multiple elements. This method shows broad application prospects in the preparation of HEAs. Yao et al [11]. pioneered this method through the mixing of metal salt precursors into a solution, followed by loading onto carbon nanofibers (CNFs). In Figure 1, after heating to 2000 K, a thermal shock was applied for 55 ms, with heating and cooling rates approaching 105 K/s, resulting in the formation of HEA nanoparticles containing up to eight different metal elements. This method has the advantages of low raw material costs, and by adjusting CTS parameters, nanoparticles with different structures and application values can be synthesized. However, the preparation of the CNF precursor and the high-temperature pulsed equipment required incur significant costs.

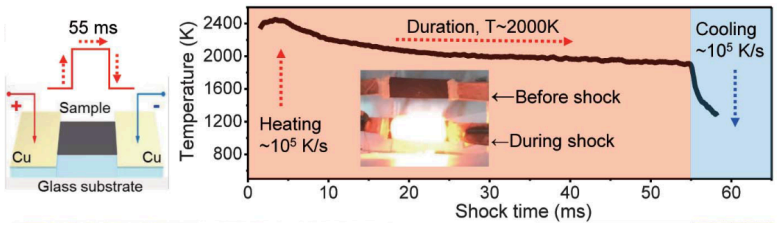

Figure 1. Schematic diagram of the carbon-thermal shock method workflow [11]

Building on prior work, Li et al [12].employed the Carbide-Induced Thermal Shock (CITS) process to synthesize ultra-small high-entropy alloy nanoparticles (HEA-NPs) containing Rh, Ru, Fe, Co, Ni, and Mo on tungsten trioxide (WO_3) nanofibers. This method involves a sequential approach, spun composite nanofibers (NFs) are prepared via electrospinning using a Tungsten (W) precursor incorporated with Polyacrylonitrile (PAN). These composite NFs are then carbonized to form WC NFs. Leveraging the ability of WC NFs to maintain a one-dimensional (1D) fibrous nanostructure under high-temperature conditions, they are impregnated with a solution containing precursors of various metal elements. This results in functionalized WO_3 NFs with uniformly distributed HEA-NPs on their surface. This innovation effectively addresses the limitations of the traditional carbon-thermal shock method, which relies on carbon-based supports, by overcoming constraints related to oxidation and high-temperature environments. The functionalization of WO_3 NFs results in elevated catalytic activity and stability, rendering these nanomaterials highly applicable in advanced catalysis.

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2.2. Electrochemical synthesis method

The electrochemical synthesis method utilizes the influence of potential differences to drive ions to move in specific directions and produce alloys through redox reactions. The FFC Cambridge Process typically involves a dual system composed of graphite and metal oxides, converting metal ions into elemental metals via electrical energy to synthesize metals and alloys. Yao et al [13]. first synthesized high aspect ratio, densely surfaced nanorods by potentiostatic electrodeposition in a DMF (N,N-dimethylformamide)–CH_3CN organic system. The resulting Bi-Fe-Co-Ni-Mn HEA thin films exhibited hard magnetic properties and anisotropy after deposition and annealing, while the as-deposited films displayed soft magnetic characteristics. However, due to the differing deposition potentials of various metal elements, controlling the elemental distribution within the alloy is challenging. Additionally, the synthesized alloys often contain other electrolytic impurities, making purification difficult. The relationship between Bi content and deposition potential is shown in Figure 2.

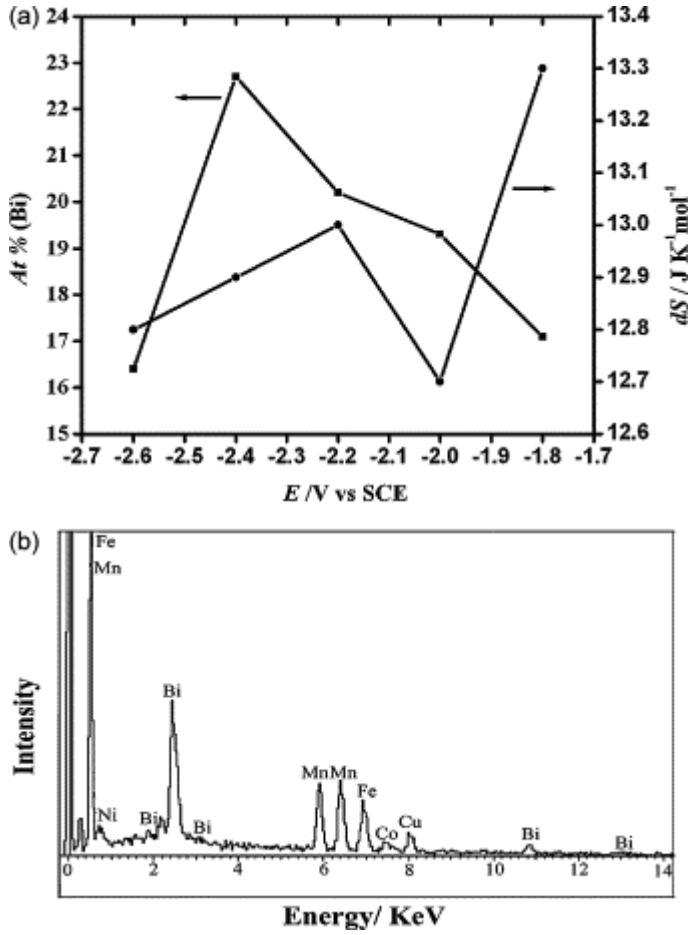

Figure 2. Effect of deposition potential on Bi content and molar entropy of mixed HEAs

Building on this, Jiao et al [14]. synthesized the AlCrNbTaTi HEA using the FFC method. This approach can directly convert metal oxides into alloys, and by employing non-graphite anode materials, it can avoid the generation of greenhouse gas CO_2, making the production process more environmentally friendly and sustainable. The raw materials for electrochemical synthesis are metal oxides, which significantly reduce synthesis costs compared to elemental metals. Electrochemical synthesis consumes electrical energy and does not require maintaining high reaction temperatures,

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further enhancing its green and eco-friendly nature. However, the electrochemical synthesis method is not yet well-suited for large-scale production.

2.3. Flow synthesis method

The flow synthesis method combines gas foaming agents with high-temperature channel reactors to generate HEAs containing internal voids through transient high-temperature heating. Wang et al [15]. first prepared an aerosol by mixing metal chlorides dissolved in ethanol with citric acid as a foaming agent, then blowing the aerosol stream into a tubular furnace. At 100°C, ethanol volatilization induces precipitation of solid particles with homogenized metal chlorides-citric acid mixtures. Subsequent temperature elevation to 300°C decomposes citric acid, releasing H_2O and COx to form porous architectures. Upon reaching 1080°C, metal salt particles decompose into metals, forming hollow nanostructured HEA particles. Li et al [16]. used dispersed carbon nanofiber (CNF) precursors as large-area supports, dissolving metal precursor salts in ethanol and uniformly coating them on the CNF surface. Laser heating melts the metal salt particles to form small metal droplets, which crystallize into AuPdFeCuNi nanoparticles. By adjusting the laser irradiation time, the structure and elemental composition of HEA nanoparticles can be controlled, enabling the preparation of HEA materials with different functionalities. This method offers fast synthesis speed, produces stable hollow nanostructured HEA particles with higher activity per unit mass, and is cost-effective with promising development potential. However, it is currently limited to mixing only a few metals to form hollow nanostructured HEA particles, and faces challenges such as high difficulty in control under high-temperature conditions and expensive instrumentation.

3. Applications of HEAs in water electrolysis for hydrogen production

In recent years, water electrolysis has become an important method for hydrogen production. The electrolysis of water consists of two main processes: one of them is hydrogen evolution reaction and the other one is oxygen evolution reaction. The HER process can be accelerated using Pt catalysts. Nevertheless, the scarcity and prohibitive expense of platinum restrict the further development of water electrolysis technology. The emergence of HEAs has greatly improved the utilization efficiency of precious metals. By introducing different metals, two types of HEAs—precious metal-based and non-precious metal-based—have been developed. Both exhibit excellent catalytic performance and promising prospects in water electrolysis hydrogen production. The following sections will introduce these two categories in detail.

3.1. Precious metal-based HEAs

Precious metal HEAs are alloys composed of one precious metal such as Pt, Pd, or Ru, alloyed with multiple non-precious metals. By leveraging the synergistic effects between the precious metals and other elements [17] cost and fulfillment of catalytic performance requirements.

Pt-based HEAs are a popular research focus. Compared to pure Pt and binary or ternary Pt alloys, Pt-based multicomponent HEA catalysts contain lower Pt content while exhibiting outstanding electrocatalytic performance. Li et al [18]. synthesized $Pt_{18}Ni_{26}Fe_{15}Co_{14}Cu_{27}$ nanoparticles (NPs) with an average diameter of approximately 3.4 ± 0.6 nm using a one-pot oil-phase synthesis method. Testing showed that the area-specific activity of these HEAs reached 83.78 mA cm^{-2} at −0.07 V vs. RHE, and the mass activity reached 10.96 A $mg^{-1}Pt$, far surpassing that of Pt/C catalysts. Li et al [17]. also prepared Al-Cu-Ni-Pt-Mn HEAs using a top-down equiatomic synthesis approach. These

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HEAs maintain high oxygen reduction reaction (ORR) catalytic performance under various conditions with low Pt content and exhibit excellent electrochemical cycling durability. Their dark-field STEM-EDS characterization can be seen in Figure 3.

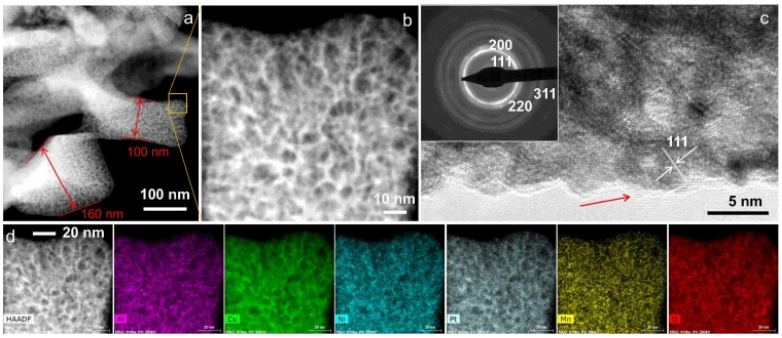

Figure 3. Dark-field STEM-EDS mapping of Al-Cu-Ni-Pt-Mn

Pd-based HEAs also demonstrate excellent catalytic performance. PdFeCoNiCu/C [19] exhibits remarkable stability and outstanding alkaline HER catalytic activity: at a current density of 10 mA cm^{-2}, its overpotential is only 18 mV, and its mass activity at −0.07 V vs. RHE reaches 6.51 A mg_Pd^{-1}.

Additionally, Jia et al [20]. designed a $Pd_{20}Pt_{20}Cu_{20}Ni_{20}P_{20}$ high-entropy metallic glass (HEMG), which shows superior synergistic catalytic functions compared to the previously developed quaternary PdCuNiP metallic glass (MG). As the Figure 4 shows, the overpotentials at 10 mA cm^{-2} current density in 1.0 M and 0.1 M KOH solutions are 32 mV and 93 mV, respectively—significantly lower than those of pure Pt and the quaternary PdCuNiP MG. Furthermore, this HEMG exhibits higher HER stability based on testing results.

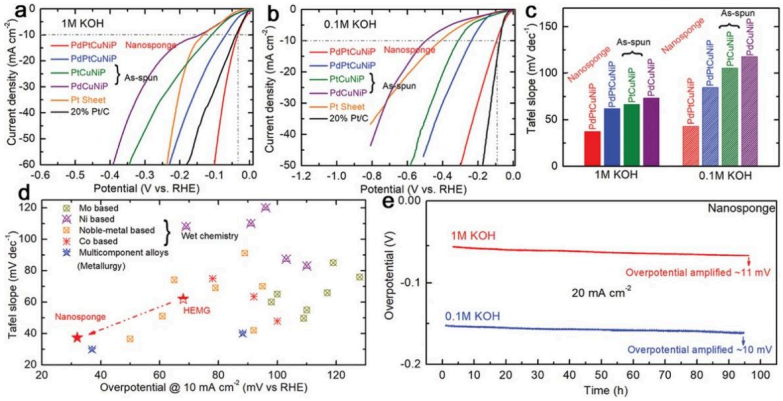

Figure 4. HER performance data of various catalysts in 1.0 M and 0.1 M KOH solutions

Wang et al [21]. deposited raw materials onto carbon fiber cloth via magnetron sputtering to fabricate FeCoNiCuPd thin films with a face-centered cubic (FCC) structure. At a current density of 10 mA cm^{-2}, the FeCoNiCuPd film exhibited a minimum overpotential of 29.7 mV, which is lower than that of Pt/C (35.4 mV). In addition to its excellent electrocatalytic performance, the FeCoNiCuPd film-based alkaline electrolyzer, as shown in Figure 5, demonstrated outstanding stability by continuously operating at a high current density of 100 mA cm^{-2} for 24 hours.

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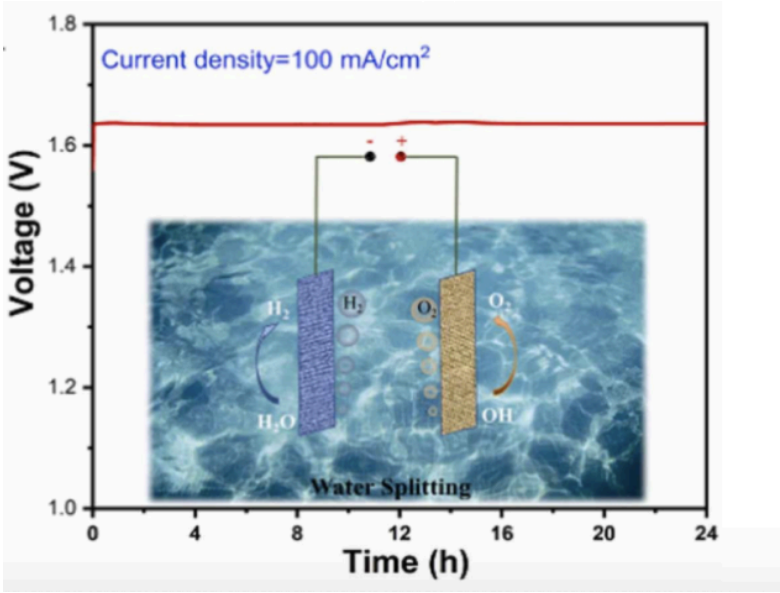

Figure 5. Potential curve of the electrolyzer constructed with FeCoNiCuPd operated at a current density of 100 mA cm^{-2}

Precious metal-based HEAs often exhibit lower overpotentials and higher catalytic activity, along with excellent HER stability. By reducing the content of precious metals, the production cost of catalysts can be significantly lowered. Future research will focus on exploring more optimal compositional ratios and developing HEAs with broader applicability.

3.2. Non-precious metal-based HEAs

The catalytic activity for HER is a function of the M–H bond strength [22]. Compared to HEAs centered on precious metals such as Pt and Pd, HEAs containing partially filled d-orbitals can exhibit comparable M–H bond strengths. Therefore, non-precious metal-based HEAs hold great promise for hydrogen production via water electrolysis, offering cost savings while still meeting catalytic performance requirements.

Zhang et al [23]. prepared a series of $FeCoCrNiMo_x$ (x = 0, 0.5, 1, 1.5, 2) HEAs using arc melting. Electrochemical measurements in Figure 6 revealed that in 0.5 M H_2SO_4, the Mo1.5 electrode exhibited an overpotential of 344 mV at a current density of 10 mA cm^{-2}, with a Tafel slope of 150 mV dec^{-1}.

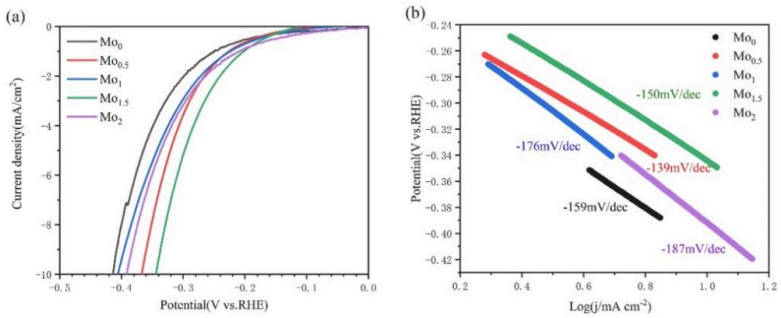

Figure 6. Polarization curves and Tafel plots

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Frank et al [24].overpotential of the HEA was measured to be only −0.32 V. After subjecting the CoCrFeNi alloy to 1000 cyclic voltammetry scans between 0 and −0.45 V at a scan rate of 20 mV s^{-1}, it demonstrated excellent stability. This was primarily attributed to Cr and other stable oxides occupying defect sites on the electrode surface, thereby preventing the dissolution of the HEA. Liu et al [25]. used copper foam as a substrate to support the growth of other metallic materials, forming a CuNiMoWCo HEA. This HEA exhibited a current density of 8.65 mA cm^{-2} at an overpotential of 50 mV, and an overpotential of only 22 mV at a current density of 10 mA cm^{-2}. Non-precious metal-based HEAs can achieve catalytic performance comparable to conventional Pt-based alloys at significantly lower cost, and they exhibit excellent stability, making them highly promising for practical applications. However, they still face challenges such as limited environmental adaptability and relatively weak corrosion resistance, necessitating further in-depth research.

4. Conclusion

The study explores synthetic approaches for HEAs and discusses their utility in hydrogen production through water electrolysis, comparing two key categories of these alloys. The current primary synthesis strategies generally rely on high temperature, electric potential difference, and other driving forces to combine multiple metallic elements. The HE effect facilitates the stabilization of thermodynamically-driven solid solution through multi-element synergy, providing abundant active sites for catalysis; the lattice distortion effect generates local strain fields that optimize the binding strength of catalytic intermediates; the sluggish diffusion effect endows materials with excellent structural stability; and the cocktail effect enables synergistic optimization of properties through compositional tuning. These characteristics allow HEA-based catalysts to exhibit outstanding performance in both activity and stability—approaching that of noble metal-based benchmarks in some systems—while offering significant cost advantages.

HEAs demonstrate great promise in the field of electrocatalytic hydrogen production. However, several challenges still hinder their practical application. First, the complex multi-component systems make it difficult to clarify the mechanisms of active sites, complicating the rational design of catalysts. Second, conventional preparation methods struggle to achieve precise control over nanostructures and face technological bottlenecks in large-scale production. Moreover, under harsh electrolytic conditions, certain constituent elements may dissolve or undergo phase transitions, affecting long-term stability. Addressing these issues requires the advancement of sophisticated analytical methodologies and computational modeling approaches to better understand the structure–property–performance relationships during catalysis.

Looking forward, the synthesis of HEAs should be integrated with advanced technologies such as artificial intelligence (AI). Establishing AI-driven screening systems can leverage big data analytics and computational power to efficiently and precisely design HEAs with specific properties, optimizing element combinations and dramatically reducing the time and cost of empirical trials. Additionally, it is necessary to enhance the stability of HEAs under industrial current densities and to optimize scalable synthesis methods to facilitate industrial-level production. With breakthroughs in these key areas, HEA catalysts are expected to overcome the transition barriers between fundamental research and practical deployment, offering new material solutions for the large-scale production of clean hydrogen energy. This advancement will not only drive progress in catalytic materials science but also have a profound impact on technologies for renewable energy storage and conversion.

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