

Research Progress on Non-Noble Metal Materials for Oxygen Evolution Reaction in Electrolytic Water Splitting

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Abstract. Hydrogen energy, as a zero-carbon clean energy source, has attracted great attention. Due to its hydrogen purity is high, electrolysis of water has become an important way to make green hydrogen. However, most commercial electrolytic water catalysts are rare and costly noble metals. The oxygen evolution reaction (OER) in electrolytic water has four-electron transfer and slow reaction speed, which is the key problem limiting electrolytic water efficiency. So, developing low-cost and high-performance non-noble metal OER catalysts is very urgent. This paper looks at the research progress of transition metal-based and non-metal catalysts, analyzes their performance traits, improvement methods, and ways they speed up reactions. Studies show that through methods like adding different atoms, structural design, and making defects, the activity and stability of some non-noble metal Oxygen evolution catalysts get close to or even surpass those of noble metal catalysts. But they still have problems such as dissolving in acid and not being stable enough at high current densities. In the future, we need to push the wide use of electrolytic water hydrogen production through new material systems, to lower green hydrogen costs and achieve global energy change.

1 Introduction

With the getting worse global energy crisis and environmental pollution problems, the development of clean and sustainable energy sources has become an important aim as a global goal. Hydrogen energy, as a clean energy with no carbon pollution, has the features of high energy concentration and being kind to the environment. So hydrogen energy is seen as a key part of the future energy system. The technology of electrolyzing water to make hydrogen has many special advantages like simple equipment, high hydrogen purity, and green production process, has become a very promising way to make green hydrogen [1]. This technology turns electrical energy into chemical energy stored in hydrogen through the whole water electrolysis reaction powered by electricity. At the same time, it can turn unstable energy from renewable sources into stable chemical energy, achieving effective energy use. Currently, the main technologies for electrolyzing water to make hydrogen

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include alkaline water electrolysis (AWE), proton exchange membrane electrolysis (PEM), and solid oxide electrolysis cells (SOEC) [2]. Right now, the common catalysts used in commercial electrolysis of water to make hydrogen are mainly rare metals and their oxides. For example, Pt and its alloys are used as HER electrocatalysts, and IrO_2 and RuO_2 as OER electrocatalysts. However, these rare metal materials are expensive and hard to find, which greatly limits the wide business use of water electrolysis technology [3]. To solve this problem, researchers are dedicated to developing high-performance and low-cost catalysts made from common metals (like regular metal materials or non-metal materials) in recent years. These materials have advantages such as being plenty in the Earth's surface, low cost, gentle reaction conditions, large surface area, many active parts, changeable structure, fast reaction speed, and good chemical stability. They offer new chances for the development of electrolyzing water to make hydrogen technology.

The electrolysis of water reaction consists of the hydrogen evolution reaction (HER) at the cathode and the oxygen evolution reaction (OER) at the anode. Compared to HER, OER involves a four-electron transfer process, exhibiting slower kinetics and requiring higher overpotentials to achieve practical current densities. Consequently, in the process of green hydrogen production through water electrolysis, OER constitutes the critical bottleneck that limits overall efficiency [4]. Therefore, the development of highly efficient OER catalysts is important for increasing the efficiency of water electrolysis. This paper will review the research progress on transition metal materials and non-metallic materials in the context of overall water splitting for OER, analyzing their performance, mechanisms, and application prospects as non-precious metal catalysts in enhancing the rate of the oxygen evolution reaction.

2 Electrolysis of water

2.1 Mechanism of the oxygen evolution reaction

The oxygen evolution reaction (OER) primarily encompasses four elementary reactions, involving the formation and transformation of multiple intermediates (e.g., OH^* , O^* , OOH^*).

As illustrated in Fig. 1, under alkaline conditions, the anode undergoes the oxygen evolution reaction (OER) to generate oxygen, while the cathode undergoes the hydrogen evolution reaction (HER) to produce hydrogen. These reactions involve the Adsorption Evolution Mechanism (AEM).

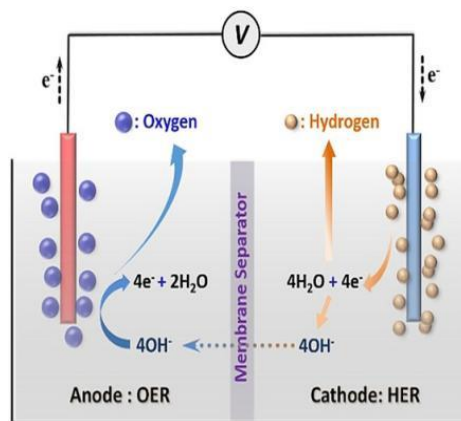


Fig. 1. A typical alkaline water electrolysis cell [4].

Regardless of the reaction environment, the thermodynamic equilibrium potential of the OER at 25 °C is consistently 1.23 V (relative to RHE). However, the OER is a non-spontaneous reaction, characterized by a positive Gibbs free energy change (ΔG), and the electrolysis system inherently possesses internal resistance. So, the real voltage needed to make water electrolysis work is higher than the theoretical voltage (1.23 volts). The gap between the real voltage and the theoretical voltage is called overpotential. The higher the overpotential, the more energy needed for electrolysis—which lowers efficiency—and the opposite is true. That's why the four-electron oxygen evolution reaction is very important for improving the performance and efficiency of water electrolysis [5]. The creation of this extra overpotential (η) is affected by many things, including activation energy barriers, solution resistance, and contact resistance.

2.2 Performance evaluation indicators of the oxygen evolution reaction

Just like the HER, the performance of OER catalysts is mainly judged based on important measures such as Tafel slope, Faraday efficiency, turnover frequency, and stability. For the early check of electrode efficiency, Linear Sweep Voltammetry (LSV) and Cyclic Voltammetry (CV) are often used. These tests can be done in two different ways that work together: measuring the overpotential under steady electric current conditions or finding the stable electric current at a given overpotential [5].

The Tafel graph taken from the LSV curve shows the connection between current density (j) and overpotential (η). Its straight part fits the Tafel equation ($\eta = b \log j + a$). The Tafel slope (b) acts amount of O_2 made in the OER experiment with the amount of O_2 calculated in theory.

Turnover frequency (TOF) is an important sign for judging the ability of a single active site to speed up the OER reaction. Stability over time is the key to the real use of catalysts. Two common ways are used for judging: current-time curves and cyclic LSV/CV tests. Generally, a catalyst that keeps its original reaction-speeds a sign of how fast the OER happens. When the overpotential is 0, the exchange current density (j_0) always used to evaluate the intrinsic catalytic efficiency of the catalytic material at the reversible potential. For OER electrocatalysts, a smaller Tafel slope coupled with a larger exchange current density corresponds to more excellent catalytic performance.

Faraday efficiency is a sign to measure how well electrons are used in electrochemical reactions. Its value is found by comparing the real ading ability after more than 5000 cycles shows great ability to last.

3 The application of transition metal-based catalysts in the oxygen evolution reaction

3.1 Transition metal phosphides catalysts

Transition Metal Phosphides (TMPs) have got a lot of attention because they have high catalytic activity, are easy to make, and have good ability to resist rust. The catalytic activity of TMPs comes from the way transition metals and phosphorus atoms work well together: phosphorus has a higher electronegativity (2.19) than most transition metals. This can change the electron density of metal active sites through electron transfer, make the adsorption energy of intermediates better, and thus speed up how fast the OER reaction happens. However, common TMPs (like Ni_2P , CoP) have obvious problems: they're not good at breaking water molecules apart on their surface. Also, under alkaline conditions (usually used in commercial alkaline water electrolysis systems), their active sites are likely to become less

active because of hydroxyl groups. This leads to not the best OER catalytic performance (for example, needing an overpotential > 300 mV to get 10 mA/cm^2). To fix these problems, researchers have come up with different ways to make them better, including heteroatom doping and making porous/core-shell shapes. By adding different metal elements (like Fe, Mn, V) or non-metallic elements (like O, S), the electronic and crystal structures of TMPs can be adjusted slightly. Besides, making porous or core-shell structures increases the specific surface area of TMPs, shows more active sites, and makes contact between the electrolyte and active sites work better.

As illustrated in Fig. 2, The stability of the TMP pre-catalyst was evaluated by chronopotentiometry (CP) at a constant current density. The results showed that it exhibited excellent long-term stability, which was attributed to the in-situ dephosphatization/oxidation of the electrochemical process, leading to the exposure of more electrocatalytic active sites and thus promoting the OER performance.

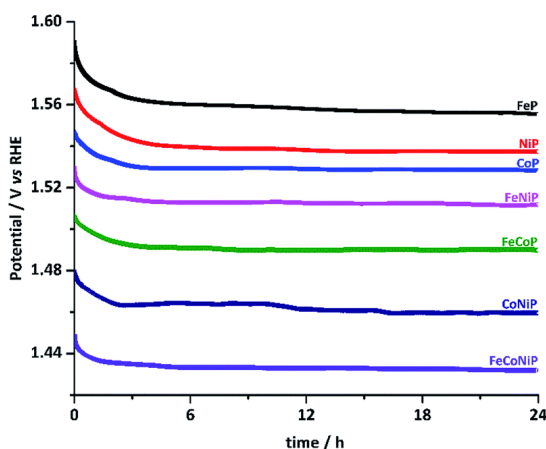


Fig. 2. Chronopotentiometric curves for TMP pre-catalysts recorded at a constant current density of 10 mA cm^{-2} [6].

R. Duan et al. showed through their research that making self-standing nitrogen-added hollow carbon tiny fiber electrodes (Co/Co₂ P-NCNFs-H, fixing Co/Co₂ P heterojunctions) using continuous concentric electrospinning method proves the synergistic effect of structure and electronic design [7, 8]. The catalyst's hollow porous structure provides plenty of active sites and mass transfer paths, while the heterojunction improves electronic setup. It has excellent overall water splitting ability in 1.0 M KOH (OER overpotential of 405.6 mV at 100 mA cm^{-2} ; HER overpotential of 247.9 mV, with no big drop in performance after 20 hours of OER use and 250 hours of HER use). Besides, it has a high specific surface area ($254 \text{ m}^2/\text{g}$), hydrophilic/superhydrophobic traits, and strong mechanical stability. The presence of the heterojunction was proven by XRD and XPS. Comparative investigations have clearly demonstrated the synergistic effect between the hollow structure and heterojunction, providing novel insights into the design of hollow carbon nanofiber catalytic materials.

3.2 Transition metal sulfide catalysts

Transition metal sulfides (TMSs) have possible catalytic activity in the oxygen evolution reaction (OER) because of their special electronic structures and good ability to carry electricity. The lone pair electrons of surface sulfur atoms can make weak connections with H^+ . Importantly, molybdenum disulfide (MoS_2), tungsten disulfide (WS_2), and nickel-

based sulfides have got attention for their ability to stay stable in both acidic and alkaline environments. Bulk TMSs have small specific surface areas, with active sites mostly exposed at the edges—which makes their catalytic activity limited. Making nanostructures, like nanosheets, nanoflowers, and nanotubes, can greatly increase the specific surface area and expose more edge active sites. Furthermore, TMSs undergo surface oxidation reconstruction during the OER process, forming transition metal oxides/hydroxides (e.g., NiS \rightarrow NiOOH), with the reconstructed oxides/hydroxides being the true active species. Based on this mechanism, researchers have accelerated the generation of active species through pre-oxidation treatments or the design of oxidizable sulfur species.

T. Yang et al. successfully synthesized CoS@NiS-80% nanosheet electrocatalysts through a facile hydrothermal method in their research [9]. The specific synthesis process of the catalyst is shown in Fig. 3. Using urea, nickel source, and sodium sulfide as raw materials, the precursor was pre-synthesized by hydrothermal reaction at 120 °C for 4 h, and then the CoS@NiS heterostructure nanosheets were finally obtained after 13 h of sulfurization treatment. Benefiting from the nanosheet architecture, interfacial effects, synergistic interaction between CoS and NiS, and surface reconstruction, the catalyst demonstrated exceptional OER performance in 1 M KOH solution: achieving a low overpotential of 280 mV at 10 mA cm⁻² with a Tafel slope of 100.87 mV dec⁻¹, and maintaining 96.2% of its initial performance after 100 hours of continuous operation at 10 mA cm⁻². Better catalytic activity was seen at 80 °C, having a lower overpotential of 201 mV at 10 mA cm⁻². What's more, the catalyst showed good potential performance in simulated alkaline seawater and real alkaline seawater environments, with overpotentials of 363 mV and 402 mV at 10 mA cm⁻² respectively. This work gives new ideas for developing effective transition metal chalcogenide catalysts through heterointerface design and surface reconstruction methods.

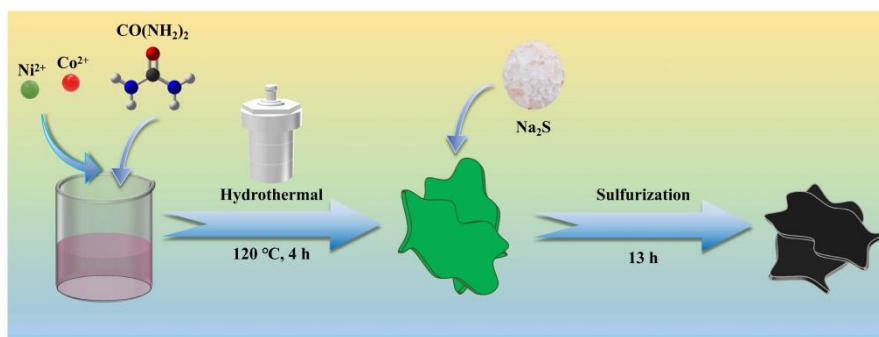


Fig. 3. Schematic diagram of the synthesis process of CoS@NiS-x [9].

3.3 Transition metal oxide catalysts

Transition Metal Oxides (TMOs) are one of the first studied non-noble metal OER catalysts. Their main good points are in their many oxidation states and stable crystal structures, which can let electrons move through changes in the valence states of metal ions and provide good adsorption spots for OER intermediates. The iron-based, cobalt-based, nickel-based oxides and spinel oxides have become research focuses because they have high activity. The formula of spinel oxides is AB₂O₄ (A is a divalent metal ion, B is a trivalent metal ion), and their crystal structure has two coordination environments: tetrahedral and octahedral. A and B ions can work together in the OER reaction through changes in their valence states, greatly improving catalytic activity.

I. Vincent et al. studied the OER performance of Ni-Fe-Ox materials on stainless steel fiber substrates. These materials had great electrochemical traits such as the ability of resist

rust and carry electricity [10]. CV tests showed the oxidation/reduction potentials of Ni–Fe–O_x were 1.52V/1.41V, while those pure Ni were 1.45V/1.37V. At 1.52V and the current density of Ni–Fe–O_x was 12mA cm⁻², 4.8 times that pure Ni. The OER polarization curves (LSV) of Ni–Fe–O_x and pure Ni catalysts are shown in Fig. 4. In LSV tests, the OER of Ni–Fe–O_x started at 1.52V. It can be seen from Fig. 4 that at the potential of 1.55 V (vs RHE), the current density of Ni–Fe–O_x reaches 22 mA cm⁻², which is more than 5 times that of pure Ni, intuitively verifying the significant enhancement of Fe doping on the OER catalytic activity of Ni-based oxides. What's more, the Ni oxidation peak (showing the change from low-valent Ni to high-valent Ni, where high-valent Ni acts as the active site for OER) was greatly reduced before OER. This shows that the active sites were already there instead of made during use. Additionally, metallic Ni helps electrons move quickly within nanoparticles, which is important for electrocatalysis.

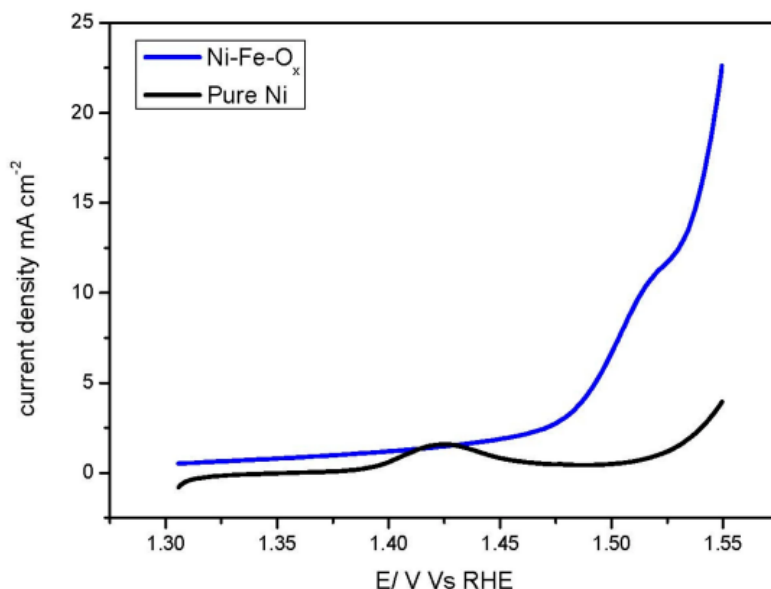


Fig. 4. LSV of Ni–Fe–O_x as an OER electrocatalyst [10].

3.4 MXene material catalysts

MXene is a new kind of two-dimensional layered transition metal carbides/nitrides. It has excellent ability to carry electricity, high specific surface area, and changeable surface chemistry, showing special use potential in the OER area. However, the OER activity of pristine MXene remains relatively low, primarily due to the coverage of active sites by surface functional groups and insufficient oxidation stability of metal atoms. Current research focuses on enhancing the OER activity of MXene through the following strategies:

The inert functional groups on the MXene surface are removed through methods such as etching and annealing, and active functional groups (e.g., -OH) are introduced, or the surface electronic structure is modulated via heteroatom doping (e.g., N, P). Furthermore, MXene is composited with active materials such as transition metal oxides and hydroxides, leveraging MXene's high conductivity and the composite components' high catalytic activity to construct a synergistic enhanced heterostructured catalyst.

X. Li et al. successfully synthesized a NiCo-LDH/Ti₃C₂T_x/NF composite electrocatalyst for water electrolysis OER through a hydrothermal method, growing 3D flower-like NiCo-LDH on a porous Ti₃C₂T_x/NF framework [11]. This catalyst integrates

the advantages of three components: the interfacial interaction between $\text{Ti}_3\text{C}_2\text{T}_x$ and NiCo-LDH accelerates ion transfer and generates synergistic effects, while the 3D flower-like structure enhances specific surface area and double-layer capacitance to expose active sites. The OER performance of this catalyst does better than recently reported LDH/MXene composite structures, showing a low overpotential of 223 mV at 100 mA cm^{-2} and a Tafel slope of only 47.2 mV dec^{-1} . To further intuitively evaluate the comprehensive performance of this catalyst, the core performance parameters of similar OER catalysts reported in recent years are summarized in Table 1. From the data comparison in the table, compared with other LDH/MXene composite system catalysts, the NiCo-LDH/ $\text{Ti}_3\text{C}_2\text{T}_x$ /NF catalyst demonstrates superior performance in both core indicators at a high current density of 100 mA cm^{-2} , which fully proves that the 3D composite structure has both excellent reaction kinetic performance and catalytic activity under high current during the OER process, and has outstanding performance advantages and application potential in MXene-based OER catalytic materials. This progress has good potential for use in water electrolysis and provides a way of doing things for making three-part compound structures of NiCo-LDH, $\text{Ti}_3\text{C}_2\text{T}_x$, and conductive structures.

Table 1. OER catalysts that have been reported in recent years [11].

Catalyst	Current density j (mA cm^{-2})	Overpotential η (mV)	Tafel slope (mV dec^{-1})
NiCo-LDH/ $\text{Ti}_3\text{C}_2\text{T}_x$ /NF	100	223	47.2
NiFe-LDH/MXene	20	240	43
NiFe-LDH/NF	10	193	143.1
NiFe-LDH/MXene/NF	10	229	44
CoFe-LDH/MXene	10	319	50
NiFeCe-LDH/MXene	10	260	42.8
NiCo-LDH/MXene/NF	100	257.4	68
NiFeP-LDH/MXene	10	286	35
MXene/ TiO_2 /NiFeCo-LDH	10	320	98.4
NiFe LDH/NiTe/NF	50	228	51.04
NiFeAu LDH	100	267	58
CoP/MXene	10	280	95.4
Co_3O_4 /MXene	10	300	118
$\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$	10	248	51
$\text{Ni}(\text{OH})_2$ @Ni/CC	100	458	168
FeCoNi alloy	10	400	70

4 Application of non-metal catalysts in the oxygen evolution reaction

Non-metallic catalysts, such as carbon-based materials, carbon nitrides, and phosphides, have great advantages including low cost, being kind to the environment, and high stability. By changing their structure and adding different atoms to modify them, these catalysts can get an improvement in OER catalytic activity. This makes them an important complementary direction to non-precious metal catalysts. Among these, graphitic carbon nitride ($g\text{-C}_3\text{N}_4$) has got the most research in the OER field because of its graphite-like layered structure, controllable electronic properties, and plenty of nitrogen active sites.

4.1 Graphitic phase carbon nitride ($g\text{-C}_3\text{N}_4$) material catalyst

The basic building block of $g\text{-C}_3\text{N}_4$ is the melamine ring, which forms a layered structure through amino connections. The nitrogen atoms on its surface (including pyridinic N, pyrrolic N, and graphitic N) have lone pair electrons. They act as active sites for the OER by holding onto reaction intermediates and taking part in electron movement. However, pure $g\text{-C}_3\text{N}_4$ has problems like bad ability to carry electricity, small specific surface area, and not many active sites—which makes its OER catalytic activity low. The electronic structure of $g\text{-C}_3\text{N}_4$ can be changed through non-metal doping (e.g., O, S, P) or single-atom metal doping. This makes it better at carrying electricity and increases the number of active sites. What's more, adding single atoms of metals (such as iron, cobalt, and nickel) can form single-atom active sites on the surface of $g\text{-C}_3\text{N}_4$. The way the metal and the carrier material work together changes the electronic structure, which helps to further improve the ability to speed up reactions. By mixing $g\text{-C}_3\text{N}_4$ with carbon materials (such as graphene and carbon nanotubes) and transition metal compounds (such as oxides and sulfides), mixed structures can be made. This uses the effect where different parts work better together, thus making it better at carrying electricity and improving its ability to speed up reactions.

M. Fiaz et al. made two types of MOFs materials called MIL-125(Ti) and UiO-66, and their heterojunctions $g\text{-C}_3\text{N}_4$ @MIL-125(Ti) and $g\text{-C}_3\text{N}_4$ @UiO-66. They used a one-step solvothermal way, adding $g\text{-C}_3\text{N}_4$ right during the research. The making of the samples was proven by XRD, FTIR, SEM, and ED-XRF tests [12]. Adding $g\text{-C}_3\text{N}_4$ did not break the crystal structures of MIL-125(Ti) and UiO-66. It let nanoparticles be added into the MOFs while keeping them whole, and made the materials better at separating light-made charges and taking in visible light. HER and OER performance of the materials was studied using cyclic voltammetry and linear sweep voltammetry. The results showed that the heterojunction $g\text{-C}_3\text{N}_4$ @MIL-125(Ti) had the best performance: to reach a current density of 10 mA cm^{-2} the overpotentials for HER and OER were only 86 mV and 173 mV, respectively. Both reactions stayed stable for 1000 seconds. The material also had good stability when exposed to visible light, showing great potential to improve the electrocatalytic activity of MOFs in water splitting uses.

5 Challenges and development

The main problems now faced in the field of water electrolysis catalysts include: precious metal-based catalysts (such as Ir and Pt) are expensive and hard to get, which limits their wide use; non-precious metal catalysts are likely to oxidize and dissolve in acidic environments, while in alkaline conditions, catalysts may have worse performance because of corrosion or structure changes; they are not stable enough at industrial-level high current

densities; and we need to make more improvements to the interface effects between catalysts and carriers, as well as the way to make them in large amounts.

The future development direction focuses on innovating the material system and developing high-performance non-noble metal catalysts. For instance, the activity of transition metal-based materials can be enhanced through defect engineering, doping modification, hetero-composition and structural design, while also improving the conductivity and stability of the materials.

6 Conclusion

This article systematically reviews the research progress of transition metal-based catalysts (phosphides, sulfides, oxides, MXenes) and non-metal catalysts ($g\text{-C}_3\text{N}_4$) in the oxygen evolution reaction of water electrolysis, and analyzes the performance characteristics, optimization strategies and catalytic mechanisms of various catalysts. Overall, significant research has been made in non-noble metal OER catalysts at present: through strategies such as structural design (e.g., nanosizing, porosification), defect engineering (e.g., oxygen vacancies, metal vacancies), doping modification (hetero-element doping, single-atom doping), and hetero-composition (multi-component synergy), the activity and stability of non-noble metal catalysts have approached or even surpassed those of noble metal catalysts (such as IrO_2), laying the foundation for the low-cost and extensive use water electrolysis hydrogen production technology. The field of water electrolysis catalysts is achieving a transformative leap from laboratory breakthroughs to industrial applications through multidimensional advancements in materials, structures, theories, and processes.

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