Dimension Engineering in Noble-Metal-Based Nanocatalysts

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Catalysts play a pivotal role in modern industries, such as energy, pharmaceuticals, and petrochemicals, serving as cornerstone of high-tech production. Noble metals, such as gold, silver, and platinum group elements, possess the superb catalytic characteristics of high-temperature oxidation resistance, corrosion resistance, stable electrochemical performance, high catalytic activity, and so on. These characteristics offer excellent prospects for applications in catalysis.

Keywords: catalysts ; nanomaterials ; noble metals ; dimension engineering

1. Introduction

Catalysts play a vital role in industrial production by altering the reaction rate without affecting the total standard Gibbs free energy of the reaction. The use of microorganisms and biological enzymes as catalysts to brew wine and pickles represents one of the earliest and simplest applications of this concept. The earliest industrial catalysis can be traced back to the lead chamber method for sulfuric acid. Since the onset of the industrial revolution, catalytic technology has developed rapidly and plays a crucial role in contemporary industrial production and scientific research.

The noble metals encompass eight metal elements: gold, silver, platinum, ruthenium, rhodium, palladium, osmium, and iridium. Owing to their chemical stability, corrosion resistance, and unique electronic structure, they are considered the most suitable choice for synthesizing high-quality and efficient catalysts. In recent years, a growing number of researchers have directed their attention toward noble metal nanomaterials (NMNs). With the continuous innovation in nanostructures and the ongoing progress in synthesis technology, the specific surface area of NMNs can be further increased, leading to a higher density of active sites.

From the perspective of structure, NMNs include zero-dimensional (0D) NMNs, one-dimensional (1D) NMNs, twodimensional (2D) NMNs, and three-dimensional (3D) NMNs. These variously structured NMNs exhibit distinctive and exceptional physical and chemical properties. However, synthesizing efficient and cost-effective NMNs necessitates identifying appropriate structures and preparation methods in a challenging but essential pursuit.

2. NMNs with Different Dimensions

2.1. 0D NMNs

It can define 0D nanostructures as materials at the nanoscale (approximately 100 nm), with 3D spatial dimensions, including quantum dots, nanoclusters, and nanoparticles (NPs). A nanocluster is a molecular-like structure formed by a certain number of atoms, molecules, or ions, with a size that does not generally exceed 10 nm.

Due to the unique structure of 0D nanomaterials, they usually have significant quantum size effects, small size effects, surface effects, and macroscopic quantum tunneling effects. Additionally, their energy level structure and electronic structure are fundamentally different from other multi-dimensional structures, often showing different activity and selectivity from multidimensional nanomaterials.

In essence, the strategies for synthesizing NMNs are mainly divided into two types. The top-down strategy involves the use of complex macroscopic materials and costly technologies to continuously reduce the size of bulk materials or incorporate nanoscale features, including lithography, electron beam lithography, immersion lithography, molecular beam epitaxy, etc. On the other hand, the bottom-up strategy involves aggregating and nucleating molecular or atomic-scale primitives, which then grow to form nanomaterials, including water/solvent chemical synthesis, chemical vapor deposition (CVD), etc. The bottom-up strategy has attracted more attention, and a large number of NMNs are synthesized using this

strategy, since they are usually cheaper and easily controlled during the synthesis process through the manipulation of many experimental parameters, including the temperature, precursor properties, stabilizers, solvents, reducing agents, their molar ratio, concentration, etc., and there is the ability to reach an industrial scale. It is worth noting that, unlike 2D NMNs, NMNs in other dimensions rarely use the top-down synthesis strategy.

Noble-metal single-atom catalysis (NMSAC) has become a frontier of catalysis due to its extremely high atomic utilization and corresponding high catalytic activity. NMSAC refers to noble metals atomically dispersed on solid supports. Zhang's group fabricated Pt_1/FeO_x SAC by anchoring single Pt atoms on the surface of FeO nanocrystals ^[1]. The Pt_1/FeO_x SAC exhibited excellent stability and activity for both CO oxidation and the preferential oxidation of CO in H₂. In addition, NMSAC has a higher catalytic performance than noble metal nanoparticle catalysts in some catalytic systems.

Double-atom catalysts (DACs), also known as dual-atom catalysts, perfectly inherit the advantages of SACs' 100% active atom utilization. Meanwhile, there are internuclear electron interactions between dinuclear sites, and this results in dinuclear co-adsorption and catalytic cooperative interactions, which can overcome the limitation of the intermediate adsorption correlation of mononuclear sites, greatly improving the catalytic activity and selectivity of DACs. A typical example is Pt_2 DACs for the selective hydrogenation of p-nitrobenzene to aniline ^[2]. Compared with Pt_1 SAC and Pt NP catalysts, Pt_2 DACs exhibit better catalytic activity for the reaction, which can be attributed to the fact that Pt_2 not only makes the H_2 reactant easy to activate and dissociate, but also makes the desorption of aniline products easier.

2.2. 1D NMNs

A 1D nanostructure refers to a structure with two of the three dimensions between 1 and 100 nm ^[3]. The AR (defined as the ratio of length to diameter) of a 1D metal nanostructure is the basic parameter. According to their different AR values, 1D nanostructures can be divided into nanorods (NRs) and nanowires (NWs). NRs usually refer to 1D nanostructures with an AR less than 30, while the AR of NWs is usually greater than 100. Additionally, 1D nanostructures could exhibit other variations, including branching (or dendritic) structures, hollow structures, core–sheath structures, core–shell structures, alloy structures, and spatially segmented structures. Branched or dendritic nanostructures can be obtained via the additional growth of NRs or NWs from the surface of the pre-synthesized 1D nanostructures. If the 1D nanostructure has a hollow interior, when the AR is relatively large, nanotubes (NTs) will be obtained. When the second metal is uniformly applied to the entire surface of the 1D nanostructure, depending on the AR, a structure with a core–sheath or core–shell morphology can be obtained. The position-selective overgrowth of the second metal at the end of 1D metal nanostructures can form a structure with segmented morphology.

Unlike their 0D and 2D counterparts, 1D nanostructures provide a unique platform to tailor and utilize the electronic, plasmonic, electrical, mechanical, and thermal properties of metals to suit or achieve various types of applications ^[4].

2.3. 2D NMNs

A 2D nanostructure refers to a structure with only one of the three dimensions between 1 and 100 nm, including NSs, nanoplates, and ultrathin 2D structures. It can divide 2D nanomaterials into two types: layered structural nanomaterials and non-layered structural nanomaterials. For layered nanomaterials, the in-plane atoms in each layer are interconnected by strong chemical bonds, and these layers are stacked together by weak van der Waals interactions to form bulk crystals. Non-layer structural nanomaterials crystallize in the 3D direction through atoms or chemical bonds to form bulk crystals. In particular, they can be crystallized into various crystalline phases according to the coordination mode between atoms, the arrangement of atoms, or the stacking sequence between layers. It is worth noting that 2D NMNs are non-layered structures due to metal bonds.

Due to the high aspect ratio, unique surface chemistry, and quantum size effect, 2D nanomaterials usually have rich active sites, significant electronic effects, and strain effects.

Unlike intrinsic layered materials, most metals have highly symmetrical lattices ^[5]. This indicates that the 2D morphology is not thermodynamically favorable during the growth of noble metal nanocrystals ^[6]. Therefore, in order to promote 2D anisotropic growth, the growth path is usually dynamically controlled by slowing down the atom addition process or reducing the total free energy of the metal nanostructure ^[7]. The synthesis methods of 2D noble metal nanomaterials are mainly divided into top-down and bottom-up strategies. On this basis, further distinctions have been made according to the types of key reactants or key experimental conditions. The methods based on the bottom-up synthesis strategy include organic ligand-assisted growth, small-molecule- and ion-mediated synthesis, 2D template-confined growth, the polyol method, seed growth, photochemical synthesis, the water/solvothermal method, crystal phase transition, etc. The

methods based on the top-down synthesis strategy include the mechanical compression method, the exfoliating method, nanolithography, etc.

2.4. 3D NMNs

A 3D nanomaterial is composed of one or several types of 0D, 1D, and 2D nanomaterials as basic units, which are arranged or combined in 3D space. The 3D NMs mainly include nanonetworks, nanoaerogels, hollow nanospheres, and so on. A 3D NMN integrates different components into a hybrid nanosystem, using synergistic effects to improve material properties. A 3D NMN often exhibits better structural stability and tunability than other low-dimensional NMNs due to its larger size and more complex solid structure. A 3D NMN still has the characteristics of quantum effect, size effect, and surface effect given by nanostructures and can realize the synergistic modulation of physical properties such as electron transport and coupling, spin polarization, excitation behavior, and wavefront control, and can obtain new characteristics that low-dimensional nanostructures do not have.

3. The Structure–Activity Relationship of NMNs

In different catalytic systems, due to the differences in lattice planes, edges, and angles exposed by nanomaterials of different dimensions, the thermodynamic and kinetic processes of reaction substrates, active intermediates, and products on the catalyst surface are different. Thus, dimension engineering plays various roles in different catalytic reactions. In addition, the microscopic factors that determine the performances of nanocatalysts in different dimensions are very complicated and diverse.

In some catalytic systems, when a certain structural characteristic of the 0D nanocatalyst is changed, its catalytic activity will change accordingly. Crystal orientation is an important factor affecting 0D nanocatalysts. Chen et al. loaded Ru nanoparticles with an fcc structure and hcp structure on γ -Al₂O₃^[S]. The results show that the hcp Ru/ γ -Al₂O₃ had better catalytic activity than the fcc Ru/ γ -Al₂O₃ under the condition of similar Ru size. By changing the crystal orientation of the catalyst, the crystal plane exposed in the catalytic system contained more active sites, thereby improving the catalytic activity of the catalyst. The grain size is also an important factor affecting 0D nanocatalysts. As the particle size of the catalyst decreases, the active surface area of the material exposed to the catalytic reaction increases, which, in turn, increases the catalytic activity. This phenomenon is called the size effect, and it is most pronounced in 0D nanocatalysts. Tanabe et al. used Au nanospheres of different sizes (5–60 nm) to modify TiO₂ photocatalysts ^[9]. The attenuated total reflection spectroscopy measurement showed that the Au nanospheres with smaller particle size aroused greater spectral changes, which meant greater electronic state changes and higher charge separation efficiency enhancement. In fact, TiO₂ modified by smaller Au nanospheres showed higher photocatalytic activity. Moreover, Au NRs or Au NCs with different aspect ratios are deposited on TiO₂, but their spectra did not change significantly.

The 1D nanocatalysts provide a unique platform to tailor and utilize distinct plasmonic, electrical, mechanical, and thermal properties to regulate various catalytic systems. Si et al. successfully synthesized a NWs/C catalyst and Pt NPs/C catalyst ^[10]. Electrochemical tests demonstrate that compared with Pt NPs nanocatalysts, Pt NWs nanocatalysts have a higher electrochemical surface area, higher CO resistance, and higher electron transfer coefficient. Due to the larger continuous surface and more active sites of Pt NWs, they have a larger hydrogen adsorption/desorption peak area and stronger adsorption capacity in dimethyl ether electrooxidation reaction. AR is one of the most important factors affecting the 1D nanocatalysts. Zhou et al. fabricated three kinds of Au NRs with different AR (AR = 2.8, 3.3, 3.8) ^[11]. The catalytic results show that the Au NRs with larger AR had higher catalytic activity. Similar conclusions can also be drawn from the literature $\frac{[9][12][13][14]}{[14]}$. The facet shape is also one of the most important factors affecting the 1D nanocatalysts. The facets, such as cuboid facets, convex cube facets, and concave cube facets, have richer optical extinction spectra, a wider plasma tuning range, and enhanced catalytic tunability compared to the traditional circular facets ^[15].

Due to their large specific surface area and unique surface chemistry, 2D nanocatalysts generally exhibit rich active sites, significant electronic effects, and strain effects, which could be applied in specific catalytic reactions. Sadeghi et al. fabricated spherical Ag NPs and Ag NSs with different sizes on the surface of zinc oxide tin (ZTO) ^[16]. Ascorbic acid oxidation tests are carried out on the two electrodes. It was observed that the peak current of the ZTO electrode modified by Ag NSs was almost two to three times higher than that of the ZTO electrode modified by spherical Ag NPs. Compared with the ZTO electrode modified by spherical Ag NPs, the ZTO modified by Ag nanosheets has higher catalytic activity. Ag NSs have more sharp edges, which exhibit higher catalytic activity than flat areas.

The 3D nanocatalysts based on combination of different structures could significantly magnify the synergistic effects and, hence, improve the catalytic performance in specific catalytic systems. Bukhtiyarov et al. found that the Au-shell-Pd-core

nanocatalysts exhibited higher pairwise selectivity than the PdAu alloy nanocatalysts ^[17]. The reason is that the core–shell structure enhances the dilution effects of Au atoms on Pd sites, and the dilution effects are positively correlated with catalytic performance.

4. Applications

4.1. Direct Alcohol Fuel Cells (DAFCs)

Due to the empty d orbital, noble metals can efficiently form coordination bonds to participate in the reaction. Noble metal nanocatalysts have the advantages of easy adsorption of reactants on the surface, good chemical activity, and good stability, and have a very broad application prospect in the field of fuel cells.

The AOR refers to the electro-oxidation reaction of methanol, ethanol, ethylene glycol, and other alcohol fuels, which constitutes the anode reaction in DAFCs. Pt-based nanomaterials are commonly used in the production of DAFCs due to the good catalytic activity of Pt for the alcohol oxidation reaction. Moreover, Pd- and Ru-based nanomaterials have good electronic and proton conductivity, and have a high catalytic efficiency for the alcohol oxidation reaction in alkaline media. Therefore, Pd and Ru-based nanomaterials are also commonly used in the production of DAFCs. Tan Qiang prepares a core–shell Au@Pd nanocatalyst using a three-phase phase transfer method. The catalyst has higher catalytic activity and stability for the oxidation of methanol in alkaline medium. When the ratio of shell Pd to core Au is 5:1, the catalytic activity of the catalyst is the highest. The Au@Pd catalyst showed higher stability than the traditional pure Pd catalyst. After 1000 cyclic voltammetry aging tests, the active area decreased by only 5.9%, while the active area loss rate of the pure Pd catalyst in the same medium was 70.8%. The results of XPS and computational chemistry show that the electronic interaction between the Au@Pd shell and the core leads to the shift of the d-band center of the Pd shell, which is the reason for the high activity and stability of the catalyst.

4.2. Hydrogen Fuel Cells

With the increasing global warming and air pollution, traditional fuel vehicles no longer meet the conditions of green development. It urgently need to develop a new type of fuel cell technology. The main reaction involved in hydrogen fuel cells is the redox reaction (HOR) of hydrogen, which is the reverse reaction of the electrolysis of water to produce hydrogen and oxygen. Since the product is only water, the hydrogen fuel cell is an environmentally friendly battery. In general, hydrogen fuel cells use a proton-exchange membrane fuel cell (PEMFC) and an anion-exchange membrane fuel cell (AEMFC) technologies. Catalysts are the core components of proton-exchange membranes and anion-exchange membranes. Pt and Pd have been widely used in the production of commercial hydrogen fuel cells. PEMFC often uses Pt-based catalysts, and AEMFC often uses Rh-based and Pd-based catalysts. They have high catalytic activity for HOR and high anti-toxicity. This ensures the long-term stable operation of hydrogen fuel cells.

Elbert at el. synthesized Ru@Pt core-shell and single-crystalline NPs, with a sharp and ordered interface ^[18]. By coating Pt on Ru cores, they prepared the Ru₁@Pt_{0.5}(1ML)/C and Ru₁@Pt₁(2ML)/C catalysts, respectively, with dominant oneand two-monolayer Pt shells. The HOR-HER activities of both Ru@Pt core-shell/C catalysts were better than those of commercial Pt/C and RuPt/C catalysts. The core-shell catalysts exhibited enhancement factors of Pt mass HOR-HER activities of 4.0 for Ru₁@Pt₁(2ML) and 3.7 for Ru₁@Pt_{0.5}(1ML)/C, compared with Pt catalysts. Bellini et al. synthesized a Pd-CeO₂/C catalyst with engineered Pd-to-CeO₂ interfacial contact ^[19]. The Pd-CeO₂ interfacial contact resulted in better HOR activity in AEMFC tests (>1.4 W cm⁻² peak power densities). Mao et al. synthesized a novel HOR catalyst, RuNi₁, in which Ni was atomically dispersed on the Ru nanocrystals ^[20]. The RuNi₁ catalyst exhibited excellent catalytic activity and stability for HOR in alkaline media, better than Ru-Ni bimetallic nanocrystals, pristine Ru, and commercial Pt/C catalysts. Alia et al. synthesize Pt/Cu NWs with a diameter of 100 nm and a length of 25~40 µm via the partial galvanic displacement of Cu NWs ^[21]. The area and mass exchange current density of Pt/Cu NWs are 3.5 times and 1.9 times better than that of carbon-supported Pt, respectively.

4.3. Water Splitting

As a cathode half-reaction for water splitting to produce hydrogen, the hydrogen evolution reaction (HER) has received extensive attention. Since the $|\Delta G_H^*|$ (Gibbs free energy of hydrogen adsorbed on the metal interface) of platinum is closest to 0 among traditional metals ^[22]. Pt is one of the most efficient catalysts for HER. However, Pt also has high catalytic activity in the HOR. Moreover, for the low catalytic activity of Pt in alkaline electrolytes such as seawater ^[23], Pt-based catalysts need to be improved via structural regulation or heteroelement doping. Li et al. synthesized PtNiP MNs through a facile Ni, P co-incorporation strategy ^[24]. Benefiting from the unique mesoporous structure and the changes in

electronic structure and composition brought by metal-nonmetal ternary alloys, PtNiP MNs exhibit excellent activity and durability for the HER, the oxygen evolution reaction (OER), and overall water splitting.

Rhodium is considered to be one of the most promising candidates because its activity in alkaline media is only one order of magnitude lower than that in acidic systems. Zhang et al. synthesized Rh nanocrystals with different structures, including tetrahedron (TH), concave tetrahedron (CT), and nanosheet (NS), and further measured the catalytic activity of these Rh nanocrystals for the HER ^[25]. By measuring the HER polarization curves of the three catalysts under 0.1 M KOH, it can be found that the overpotential of Rh NS at a current density of 10 mA cm⁻² is 0.043 V lower than that of Rh/C. The overpotential of Rh/C catalyst increased sharply to 0.193 V after electrolysis for 5 h, while the overpotential of Rh NSs increased slightly to 0.065 V, which was much lower than that of the Rh/C catalyst. These results demonstrate the crucial function of structural regulation in improving catalyst performance. The preparation of Rh NP/C catalyst by combining Rh NPs with C support can also improved the performance of the catalyst ^[26].

4.4. Selective Hydrogenation Reaction

A selective hydrogenation reaction refers to the process in which the target functional group is hydrogenated and the other functional group is not affected when the reactant contains two unsaturated functional groups. The selective hydrogenation reaction has a very wide range of applications in the chemical industry, including the production of fine chemicals and petrochemicals and drug synthesis. Since the traditional nanocatalysts are prone to complete hydrogenation, it is a great challenge to obtain the target product with high selectivity.

Taking the selective hydrogenation of α , β -unsaturated aldehydes to unsaturated alcohols as an example, Pd- or Pt-based noble metal hydrogenation catalysts tend to first hydrogenate C=C to form saturated aldehydes, and then further convert the products to saturated alcohols. The Ag-based catalyst has a unique ability to first hydrogenate the C=O bond without destroying the C=C bond ^[27].

4.5. Treatment of Automobile Exhaust

Automobile exhaust has always been a major problem restricting environmental protection. Automobile exhaust contains a large number of harmful substances, including carbon monoxide (CO), hydrocarbons (HCs), nitrogen oxides (NO_X) and so on. A high-performance ternary catalyst (TWC) is a catalyst that can simultaneously purify three pollutants of carbon monoxide (CO), hydrocarbons (HCs), and nitrogen oxides (NO_X). It is the most important part of the automobile exhaust system. At present, TWC catalysts are mainly composed of Ru, Rh, and Pd as active components, and Al_2O_3 , CZ (ceria-zirconia-based oxide), and other materials as carriers to prepare composite catalysts [28].

5. Summary

Catalysts play a vital role in industrial production, which is closely related to people's lives and economic development. In recent years, the increasing global carbon dioxide emissions have significantly amplified the severity of global warming and cast great restrictions on people's lives. NMNMs have been widely used as efficient catalysts in the fabrication of DAFCs, hydrogen fuel cells, water splitting, the selective hydrogenation reaction, and the treatment of automobile exhaust. However, most commercial NMNM catalysts are currently facing difficulties that mean that they cannot be used in complex environments.

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