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Advances in Supported Gold-Based Catalysts for Low-Temperature CO Oxidation

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Abstract. Catalytic oxidation of CO is an important technology in environmental catalysis and energy application. Supported Au catalysts have become a research focus due to their excellent low-temperature catalytic performance. This paper reviews the research progress of supported Au catalysts for CO oxidation, including typical preparation methods such as impregnation, coprecipitation, deposition-precipitation and colloidal deposition. The size effect of Au nanoparticles and its influence on catalytic activity are discussed, indicating that small and highly dispersed Au nanoparticles contribute to high activity. In addition, the reaction pathways of CO oxidation, such as Langmuir-Hinshelwood mechanism, metal-support interface activation mechanism, and Mars-van Krevelen mechanism, are outlined, indicating that the metal-support interface serves as the dominant active site and the reaction mechanism varies dynamically with support properties and temperature conditions. Finally, the future development of high-performance Au catalysts is prospected, providing theoretical support for the design and development of high-performance Au-based catalysts for CO oxidation.

Keywords: Heterogeneous catalysis, Au, nanoparticles, CO oxidation

1. Introduction

The rapid development of global industrialization and transportation has caused excessive emissions of carbon monoxide (CO), which poses severe threats to the environment and human health. Given its wide sources and invisible characteristics, developing efficient and low-cost CO removal technologies is highly urgent. Catalytic oxidation is regarded as the most effective method for CO purification, which can convert CO into non-toxic CO_2 at near-room temperature with a strong thermodynamic driving force. ($\Delta H = -284.09$ kJ/mol) ($\Delta H = -284.09$ kJ/mol) [1]. Compared with traditional high-temperature combustion, this technology greatly lowers energy consumption and exhibits wider application prospects, from enclosed space air purification to fuel cells.

Catalysts for low-temperature CO oxidation are generally divided into non-noble and noble metal types based on active components. Non-noble metal catalysts, mainly transition-metal oxides (e.g., $CuO–CeO_2$, Co_3O_4, MnO_x) and their composites, feature low cost and abundant resources. Their performance can be improved via mesoporous structure construction or metal-ion doping to enhance specific surface area and oxygen vacancies. However, they suffer from high activation temperatures,

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insufficient low-temperature activity, and humidity-induced deactivation, limiting harsh-environment applications [1]. Noble metal catalysts represented by Pt, Pd, Rh, Ru, Ag and Au show excellent low-temperature catalytic activity due to their unique d-electron structure and strong adsorption ability for CO and O_2. Among them, Pt and Pd-based catalysts are the earliest and most widely used as key components in three-way catalysts. However, their high cost, scarcity and easy poisoning under high CO concentration have prompted researchers to explore alternative catalysts and optimization strategies.

The exploration of noble metal catalysts includes gold. Gold's catalytic potential has undergone a remarkable transition. This transition occurred over several decades, a labyrinthine journey. Bulk gold was long thought to possess little catalytic value. Its chemical inertness stems from high ionization energy. Fully filled d orbitals contribute to this inertness, a perdurable belief. Japanese scientist Masatake Haruta overturned this perception in 1987. He discovered that gold nanoparticles smaller than 5 nm on transitionmetal oxide supports (Fe_2O_3, TiO_2, Co_3O_4) catalyze CO oxidation. This occurs at −70°C [2,3], a perspicacious breakthrough. The nanoscale size effect and metalsupport interactions play decisive roles. Gold catalysts push to the forefront of catalysis. They exhibit exceptional lowtemperature activity below zero, an anfractuous achievement. Gold is more abundant, coststable, and nontoxic than platinumgroup metals. Mild reaction conditions fit green chemistry principles. Gold shows unique value in environmental protection, energy conversion, and chemical synthesis [4], a sere demonstration. Gold catalysts have become one of the most promising research directions. They are prominent in heterogeneous catalysis. This marks a laconic conclusion.

In summary, the severity of CO pollution and drawbacks of traditional purification technologies have promoted the development of low-temperature CO catalytic oxidation. Au-based catalysts have become a research focus due to their unique advantages. This paper will discuss preparation methods of supported gold catalysts, the size effect of Au nanoparticles, and CO oxidation reaction mechanisms, aiming to provide theoretical support and technical guidance for the design and application of high-performance gold catalysts.

2. Preparation methods of supported gold catalysts

2.1. Impregnation method

Impregnation method is one of the earliest technologies used to prepare loaded metal catalysts. This method is simple to operate, widely applicable to various carriers, and easy to industrial amplify [5-7]. In this method, chloric acid ($HAuCl_4$) is often used as a gold precursor [8]. The catalyst is made by mixing the precursor solution with carriers such as titanium dioxide (TiO_2) or aluminum trioxide (Al_2O_3) before drying and calcination. However, the traditional impregnation method has obvious disadvantages: chlorine ions will induce sintering of gold particles, resulting in larger particle size and reduced catalytic activity. Through the research, it has been found that chlorine ions can promote the migration and aggregation of Au species during heat treatment [9].

People have developed improved impregnation processes to solve these problems. For example, after mixing $HAuCl_4$ with the carrier, the material can be placed in an ammonia atmosphere. After passing the interaction between ammonia water and tetrachloroquinate radical ions, it can be thoroughly washed, which can effectively remove chlorine ions. After using this improved method, the catalyst has been significantly improved in the dispersion of gold particles. The characteristics of transmission electron microscopy (TEM) show that the Au particles loaded on TiO_2 prepared in this way are uniformly distributed and the size is usually 3 ~ 6 nm. Further analysis of the X-ray

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diffraction (XRD) spectrum (Figure 1) is consistent with the TEM observation results, and it is confirmed that the average grain size of Au particles loaded on TiO_2 is about 4.6 nm.

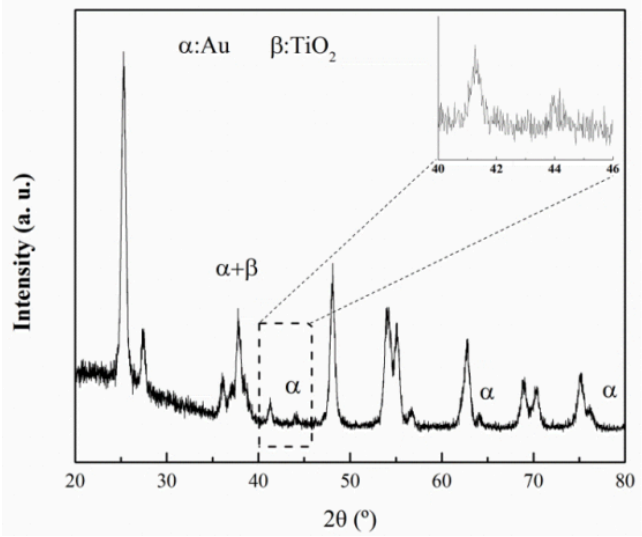

Figure 1. XRD diffraction peaks of 2% Au/TiO_2-200 [9]

As shown in Figure 2, the continuous cycle test further verifies the stability and thermal stability of the catalyst. Another point worth noting is that the calcined sample 2% Au / TiO_2-200 (calcined at 200℃) showed stronger structural stability than the fresh sample (2% Au / TiO_2-f) in the first and second cycle tests. But high temperature will destroy this highly dispersed structure. When the calcination temperature rises to 600℃, Au particles undergo severe sintering and growth, which is the reason why the T50 value rises significantly to about 150℃ (Figure 3). In addition, the load of gold must also be optimized. In the Au/TiO_2 system, the load of 2 wt% usually provides the best balance between the number of active sites and the particle size, which is the reason for the lowest T50. Lower load (1 wt %) or higher load (3 wt %) will reduce catalytic activity [9].

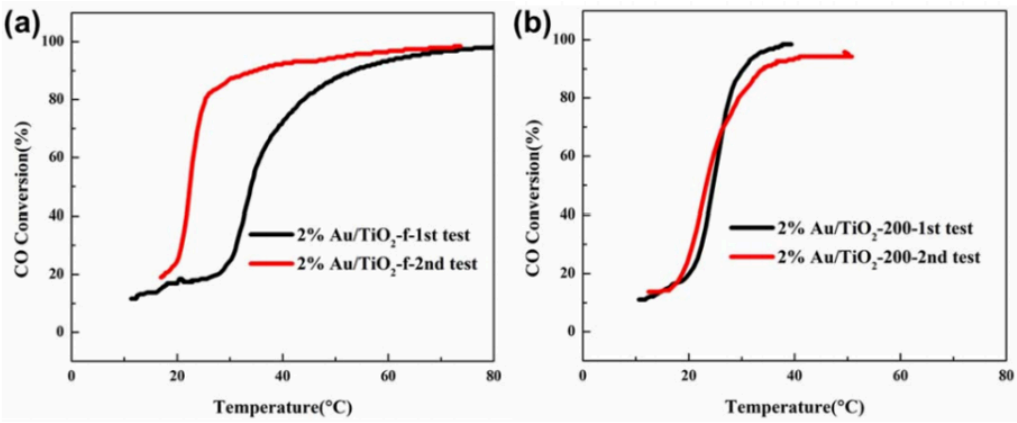

Figure 2. CO conversion curves of (a) 2%Au/TiO_2-f and (b) 2%Au/TiO_2-200 during consecutive cycling tests

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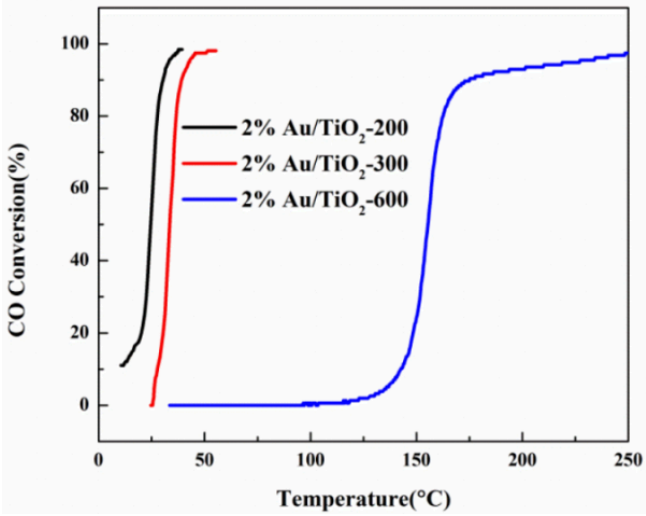

Figure 3. CO conversion curves of samples calcined at different temperatures (200℃, 300 ℃, and 600℃)

Although process optimization can partially control the particle size of gold and enable the preparation of highly active catalysts using the impregnation method, this approach still has limitations in achieving uniform particle size distribution and precise control of nanoparticle dimensions.

2.2. Co-precipitation method

The co-precipitation method was successfully used for the first time in the preparation of highly active gold catalysts by the Masatake Haruta project group in 1987. The coprecipitation method is to mix $HAuCl_4$ solution with transition metal salts ,for example,nitrates of Fe, Co and Ni. Under alkaline conditions, the precursor will be co-precipitated into a hydroxide composite, and then washed, dried and calcined. Carriers such as MgO, Fe_2O_3, Co_3O_4 and NiO are all suitable for this method, and finally prepare gold nanoparticles with good dispersion and excellent low-temperature CO oxidation activity (average < 10nm).

However, the method has practical limitations, especially in achieving high gold loading and uniform dispersion. For instance, in preparing Au/α-Fe_2O_3 catalysts, although a well-defined hexagonal crystalline support can be obtained [1], gold loading is often low. TEM observations reveal that only a small number of gold nanoparticles deposit on the rod-like α-Fe_2O_3 support, with uneven distribution and relatively large particle sizes (around 15 nm, Figure 4).

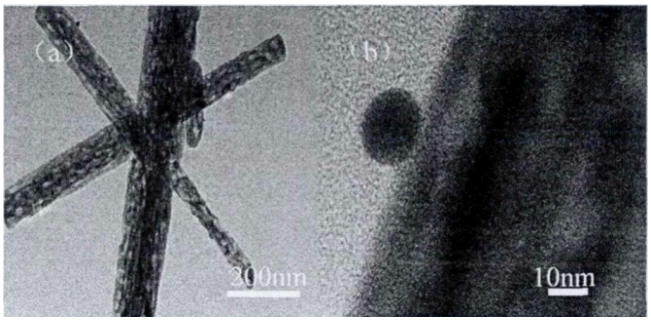

Figure 4. TEM image of the Au/α-Fe_2O_3 composite catalyst prepared by the co-precipitation method [1]

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This issue directly results in poor catalytic performance. CO oxidation tests indicate that the Au/α-Fe2O3 catalyst prepared by this method has an ignition temperature as high as 100 ℃, and complete CO conversion is achieved only when the temperature increases to about 450℃. The root cause of this poor performance lies in the inherent limitations of the method: during the co-precipitation process, the gold precursor is easily embedded within the bulk phase of the support rather than ideally deposited on its surface. Consequently, the actual gold loading (e.g., only about 0.75% measured by ICP) is far lower than the theoretical design value (3%), resulting in very low gold utilization efficiency. The absence of characteristic gold peaks in the X-ray diffraction (XRD) pattern further suggests that the gold species may exist in a highly dispersed or encapsulated state.

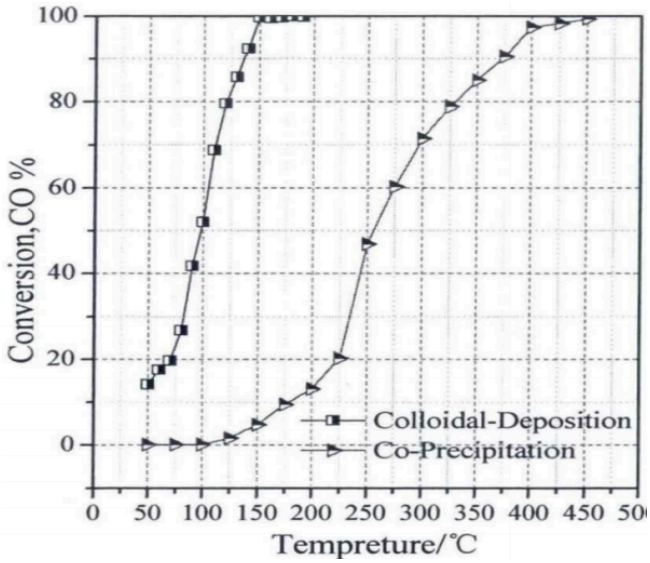

Figure 5. Catalytic activity for CO oxidation over Au/α-Fe_2O_3 composite catalysts prepared by the co-precipitation method and the colloidal deposition method [1]

Compare the two lines in the figure, and use the colloidal deposition method on the same carrier to obtain gold particles with an average size of about 4.1 nm and evenly distributed. The actual gold load is close to the theoretical value (2.01%) and the catalytic activity is significantly improved at this time, and CO is completely converted at 150°C (Figure 5). Through this comparison, it can be seen that the coprecipitation method has limitations in controlling the surface exposure and load efficiency of gold particles, and it can be seen that the applicability of this method to SiO_2, Cr_2O_3 and other carriers is also poor.

Therefore, although the co-precipitation method is relatively simple and plays a key role in the early breakthrough of gold catalysis, there are obvious shortcomings in the precise control of the size, position and load of gold particles. These limitations prompt researchers to further develop alternative preparation techniques - such as deposition-precipitation and colloidal deposition - which can better guarantee the high dispersion and surface exposure of gold nanoparticles, thus giving full play to the low-temperature catalytic potential of gold catalysts.

2.3. Deposition–precipitation method

The sedimentation-precipitation method, commonly referred to as DP, has become a widely used method for preparing highly dispersible gold nanocatalysts. It is popular because it is easy to operate and highly controllable in the actual synthesis process. The typical starting method of this method is

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to disperse the selected carrier material in $HAuCl_4$ aqueous solution. Then adjust the pH of the resulting suspension to between 7 and 10 by adding alkaline reagents, such as NaOH or Na_2CO_3. After setting the pH value, stir the mixture at a high temperature of 70-80 ℃. This heating and stirring step can promote the specific deposition of gold-containing species in the form of hydroxides on the surface of the carrier. Finally, a series of post-processing steps are carried out: including washing, drying and calcining. After this series of processes is completed, the final catalyst product is generated. A more detailed description of this process can be found in the early work [10].

A key advantage of the DP method lies in its ability to tune gold particle sizes within a narrow 2–5 nm range. This precise size control is a critical parameter for enhancing catalytic activity [11]. This technique exhibits particular suitability for supports with relatively high isoelectric points, including TiO_2, Al_2O_3, ZrO_2, and CeO_2. The DP method proves less effective for supports with low isoelectric points (e.g., SiO_2, WO_3) or carbonbased materials such as activated carbon, where perdurable surface charge characteristics hinder efficient gold deposition. A modified version known as deposition–precipitation with urea (DPU) has been developed. Urea serves as a slowrelease precipitating agent, generating OH^- ions gradually through an anfractuous process, which enables more homogeneous deposition of gold species and further refines catalyst performance.

2.4. Colloidal deposition method

The colloidal deposition method is an effective approach for preparing highly dispersed gold catalysts with small particle sizes. In this process, chloroauric acid is first reduced by reducing agents such as $NaBH_4$ in the presence of stabilizing agents (e.g., polyvinyl alcohol (PVA) or polyvinylpyrrolidone (PVP)), forming colloidal gold particles with sizes typically ranging from 1–5 nm. Subsequently, the support is mixed with the gold sol, allowing the gold nanoparticles to deposit onto the support surface. After centrifugation, washing, and calcination to remove the stabilizing agents, the desired catalyst is obtained [12].

Table 1. Catalysts prepared by the gold sol deposition method

Support	Stabilizing agent	Average size of gold sol（nm）	Average size of gold on the support （nm）
C	PVA	n.r.	4
C	PVA	6.0	6.9
Al_2O_3	PVA	2.2	2.6
C	THPC	2.9	4.3
Al_2O_3	THPC	3.5	3.8

This method achieves precise control of the size of gold nanoparticles and effectively avoids the problem of chlorine ion residue. For example, in Au/Al_2O_3 catalyst prepared with PVA as a stabilizer, the average size of gold particles is controlled at about 2.6 nm (see Table 1).

In the study of Au/α- Fe_2O_3 system, the colloidal deposition method shows obvious advantages. The catalyst prepared by this method shows evenly distributed Au particles, with an average size of about 4.1 nm and good dispersion. As shown in Figure 6, the actual load of Au can reach 2.01% (theoretical value: 3%), which is much higher than the 0.75% obtained by the co-precipitation method. The catalytic performance test shows that the catalyst shows excellent low-temperature activity in the CO oxidation reaction: the reaction only starts at 50 ℃, and CO can be completely

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converted at 150℃. In contrast, the catalyst prepared by co-precipitation method requires an ignition temperature of 100 ℃, and the temperature of complete conversion is as high as 450 ℃.

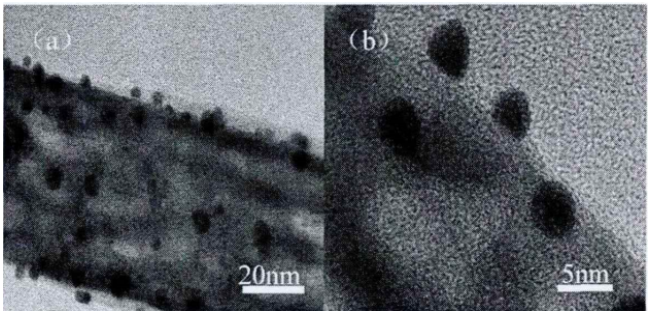

Figure 6. TEM image of the Au/α-Fe_2O_3 composite catalyst prepared by the colloidal deposition method

Despite its clear advantages in controlling particle size, the colloidal deposition method also has certain limitations. Its preparation procedure is relatively complex, and the use of organic stabilizing agents may block the pores of the support, which restricts its large-scale application.

2.5. Sol–gel method

In the sol–gel method, gold precursors such as gold alkoxides are co-hydrolyzed with support precursors to form a sol, which subsequently undergoes gelation, drying, and thermal treatment to produce the catalyst. By regulating the hydrolysis–condensation process, this method can to some extent control both the size of gold particles and the structure of the support. It is particularly suitable for preparing gold catalysts supported on mesoporous metal oxides.

In some studies, gold sols were mixed with support sols, followed by the addition of stabilizing agents such as thiols, tetrakis(hydroxymethyl)phosphonium chloride (THPC), or polyvinyl alcohol (PVA). After reduction and calcination, catalysts with gold particle sizes of about 6 nm and a loading amount of 6 wt% were successfully prepared. However, the gold particles produced by the sol–gel method are still relatively large, and the preparation procedure is complex with relatively poor reproducibility, which limits its practical catalytic applications [13].

2.6. Other preparation methods

Preparative methods discussed previously exist. Other approaches have been developed for supported gold catalyst synthesis. Chemical vapor deposition (CVD) decomposes volatile organogold precursors on the support surface via a labyrinthine process. CVD is particularly effective for acidic supports like SiO_2. Such supports remain incompatible with liquidphase deposition. The anionic adsorption method controls gold complex speciation through pH adjustment, enabling adsorption of gold species onto the support by electrostatic interactions, a quixotic yet functional route. Emerging techniques including solidstate grinding, photochemical deposition, and sonochemical methods show promise. Ultrasoundassisted Au/SiO_2 preparation yields ~5 nm gold particles uniformly distributed in the support's pores [14].

The performance of gold catalysts is governed by particle size. Dispersion also governs performance. Metal–support interaction plays a perspicacious role. Practical use requires matching preparation strategy to reaction requirements. Support properties demand matching as well. Economic constraints cannot be ignored, a sere reality. Pretreatment strategies combine with

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postmodification strategies. This combination optimizes catalyst structure. Catalyst performance sees laconic improvement.

3. Size effect of nanogold catalysts

When gold particles are reduced to less than 5 nm, their specific surface area and surface atomic codition unsaturation increase sharply, which provides highly active sites for adsorption and activation. At the same time, the reduction of particle size changes the electronic structure and state density near the Fermi energy level, and smaller Au nanoparticles form a larger metal-carrier interface as the active center of CO oxidation, promoting O_2 activation and catalytic cycle. In the research of Zhang et al., 2% Au / TiO_2 - 200 catalyst was prepared. The TEM image (Figure 7 (a)) shows that Au nanoparticles are evenly distributed on the TiO_2 carrier, and most of the particles are between 3 - 6 nm [9]. The high-resolution TEM (HRTEM) image (Figure 7 (b)) further reveals the lattice stripes of the surface of Au particles (the spacing is 0.235 nm, corresponding to the Au (111) crystal surface). In addition, it is observed that Au nanoparticles are primarily located between specific TiO_2 crystal surfaces, which is a direct evidence of strong metal-carrier interaction.

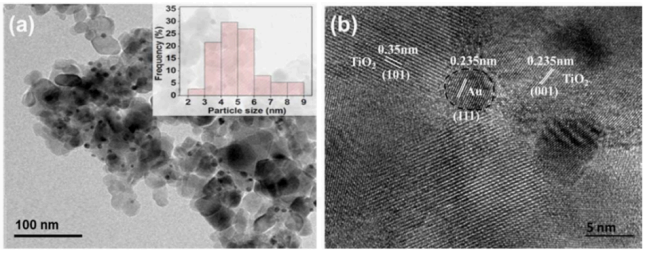

Figure 7. Low- (a) and high-magnification (b) TEM images of 2% Au/TiO_2–200

Zhang et al. visually demonstrated the strong dependence of CO oxidation turnover frequency (TOF) on Au particle size in the Au/TiO_2 system (Figure 8). When the average Au particle size exceeds 4 nm, the TOF decreases sharply, consistent with the theoretical expectation that smaller gold nanoparticles have a higher density of surface active sites.

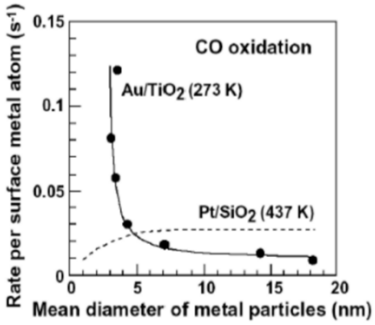

Figure 8. Relationship between CO oxidation TOF and nanoparticle size on Au/TiO_2 and Pt/SiO_2 [15]

Comparing the morphology of Au/TiO_2 catalysts calcined at different temperatures (Figure 9), it is found that there is no obvious change in the particle size and distribution of Au in uncalcined and 200°C calcined samples. This indicates that the interaction between the two Au-TiO_2 is strong and the structure is stable at low temperature. However, after calcination at 600°C, Au particles sintered to about 10 nm and the number of particles decreased, which directly confirmed the instability of

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nano gold at high temperatures: the increase in particle size reduced the number of active sites, thus having a negative impact on the catalytic performance.

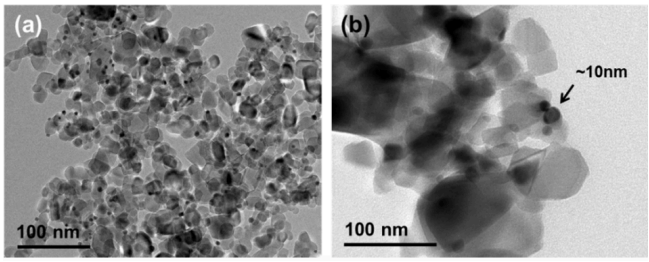

Figure 9. TEM images of (a) 2% Au/2–f and (b) 2% Au/TiO_2–600 [9]

In addition, the properties of the carrier and the influence of secondary metal doping directly affect the size and dispersion of gold particles, and the size effect together determine the catalytic behavior. The study of comparing Al_2O_3 carrier-loaded Au with different shapes (commercial, γ-phase, spherical) shows an obvious trend. TEM analysis (Figure 10) shows that on commercial Al_2O_3 with higher specific surface area and more defect sites, Au nanoparticles are the smallest (~ 2 nm), the most dispersed, and isolated point-shaped. In contrast, on the spherical Al_2O_3 with a lower specific surface area and more regular morphology, Au particles (3 ~ 5nm) are larger and mainly spherical, with a smaller contact area with the carrier, which indicates that the surface defect site on the carrier stabilizes the smaller metal particles and indirectly regulates the activity through particle size.

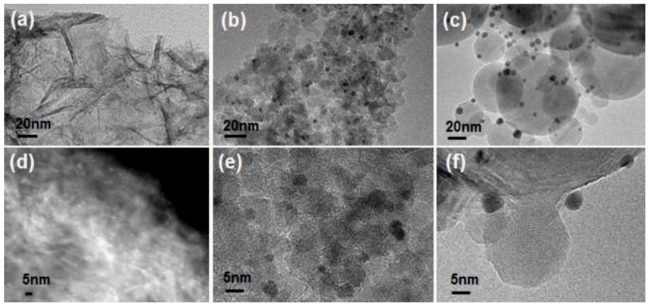

Figure 10. TEM images of 2% Au on (a, d) commercial Al_2O_3–200, (b, e) γ-Al_2O_3–200, and (c, f) spherical Al_2O_3–200 [9]

In summary, the size effect of nanogold catalysts underlies their high activity, and understanding the structure–activity relationship between particle size and catalytic performance is vital for catalyst design [16]. In practical applications, however, size alone is not the sole determining factor; the synergistic interplay of metal–support interactions, surface chemistry, and reaction environment must also be considered to precisely regulate and optimize catalyst performance.

4. Reaction mechanism of gold-catalyzed CO oxidation

The mechanism of CO oxidation reaction on research gold catalyst plays an important role in understanding its low-temperature and high-temperature activity and designing high-performance catalysts. Current research shows that the mechanism is not single, but changes according to the changes in the catalyst system and reaction conditions [1]. These mechanisms can be summarized

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into several categories, and their mutual relationships and characteristics are shown in Figure 11 [17].

4.1. Langmuir–Hinshelwood (L–H) mechanism [18]

The Mars–van Krevelen (MvK) mechanism is most commonly associated with reducible metal oxide supports. This association arises from a specific requirement. The mechanism relies on the direct participation of lattice oxygen through a labyrinthine process. A reduction step occurs first. In this step, CO reacts with active lattice oxygen to form CO_2. An oxygen vacancy is created in the support, a perdurable defect. The reoxidation step follows. Gaseous O_2 adsorbs and dissociates to refill the vacancy. This regeneration follows a perspicacious design and the process is depicted as pathway D in Figure 11 [19].

4.2. Metal–support interface activation mechanism

The perimeter of the metal-carrier interface is widely regarded as the key active region in the loaded gold catalyst. The mechanism mainly corresponds to the pathways B and C in Figure 11 [20-26]. A large number of studies have shown that O_2 activation is a lasting rate-determining step. However, the consensus on the exact active site still needs to be studied in depth. One view is that O_2 is directly activated on the gold-carrier interface (way B), and another view is that the oxygen vacancy on the carrier is adsorbed and activated O_2 to form an anionic species. These empty spaces are close to the interface. Anionic species, such as superoxides or peroxides, react with CO adsorbed on gold at the interface. The third idea is that the quasimolecular CO is activated on gold nanoparticles, and O_2 is reduced to O_2^- in the carrier oxygen vacancy. These anionic species migrate ("spillover") to the interface. They react with CO to form intermediates, and then produce CO_2.

4.3. Mars–van Krevelen (MvK) redox mechanism

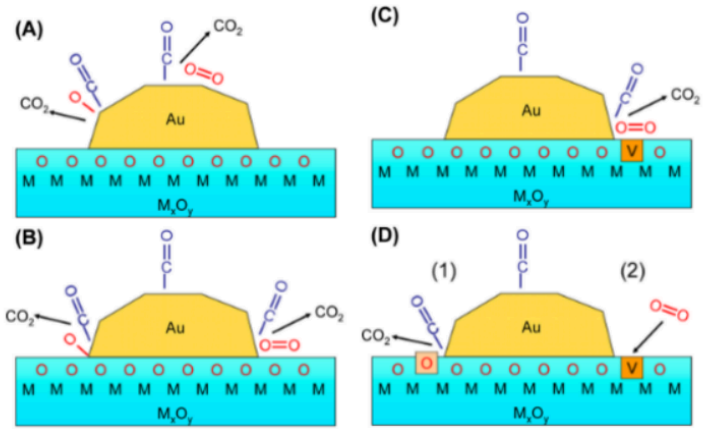

Figure 11. Schematic illustration of possible CO oxidation pathways on supported gold catalysts

The Mars-van Krevelen mechanism is usually referred to as the MvK mechanism. Because the mechanism depends on the specific characteristics supported by such, this mechanism is usually related to a carrier composed of reducible metal oxides. The direct participation of the lattice oxygen atom of the carrier material itself leads to the characteristic of the road. The whole process can be divided into two completely different steps: the first step is called the reduction step. In this process,

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carbon monoxide (CO) reacts with the active lattice oxygen species, which leads to the generation of carbon dioxide (CO_2), which ultimately leads to the production of oxygen voids in the structure of the carrier. The second step is called the reoxidation step. In the subsequent step, the gaseous molecule oxygen (O_2) is first adsorbed to the surface of the catalyst to form the adsorbed O_2 molecule, and then dissised to form the dissopated oxygen atom, and then the previously generated oxygen vacancy is refilled. Through this refilling process, the original state of the catalyst can be regenerated.This complete two-step cycle has been depicted as pathway D in Figure 11 of reference [19].

In summary, the mechanism of CO oxidation on gold catalysts mainly depends on temperature and carrier. At low temperatures, adsorption oxygen participates in the reaction, and at room temperature and above, the interface-dominated L-H and Mv K mechanisms work together. Explain that the main model of high activity of gold catalyst is the interface mechanism. Future work should clarify the active oxygen species and their formation pathways at the atomic level.

5. Conclusion

In summary, supported gold catalysts have become a research hotspot in environmental catalysis owing to their excellent low-temperature CO oxidation activity. This review systematically summarizes their preparation methods, size effects and reaction mechanisms, providing a theoretical basis for rational catalyst design and performance optimization. The key goal of various preparation strategies is to obtain small, highly dispersed Au nanoparticles with strong metal–support interactions, while support selection and modification are critical to catalytic performance. The size effect is essential for nanogold catalysis, and the overall performance is determined by the synergistic effects of particle size, interface properties, support oxygen storage capacity and reaction conditions. It is confirmed that the metal–support interface is the core active site, and the reaction mechanism switches dynamically between interface-dominated L–H [18] and MvK pathways with support and temperature changes. Future work on atomic-level preparation, precise support engineering and bimetallic or multicomponent catalysts will further improve catalytic performance and promote the industrial application of supported gold catalysts.

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