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Evolving top-down approaches for single-atom catalysis: synthesis and specialized applications

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Single-atom catalysts (SACs) exhibit exceptional performance across diverse catalytic reactions and can be synthesized through both bottom-up and top-down strategies. Among these, the top-down strategy, where bulk metals or nanoparticles are converted into isolated metal atoms, offers distinct advantages, including enhanced metal–support interactions, intrinsic cost-effectiveness and catalyst upcycling. This review summarizes recent breakthrough in top-down synthesis methodologies, covering fundamental synthetic techniques (*e.g.*, thermal treatment, abrasion, light/electric energy-driven methods) and their specific applications in various catalytic systems. Notably, top-down strategies show strong adaptability under practical reaction environments, enabling functions such as the regeneration of spent catalysts and kinetic restructuring in response to reaction conditions. Overall, this review aims to promote a deeper understanding of top-down synthetic principles, accelerate their industrial implementation, and unlock the full potential of SACs.

1. Introduction

Single-atom catalysts (SACs), defined by atomically dispersed metal sites stabilized on supports such as metal oxides, carbon matrices, and zeolites, represent a transformative milestone in heterogeneous catalysis.[1,2] By overcoming the inherent limitations of nanoparticle-based catalysts, SACs attain nearly 100% metal atom utilization and deliver remarkable improvements in catalytic activity and selectivity.[3] This shift from the "nanoscale" to the "atomic precision" regime has reshaped the fundamental understanding of active-site engineering and reaction mechanisms.[4,5] As a result, SACs are emerging as powerful platforms for addressing pressing challenges in energy conversion and environmental remediation.[6–8]

The exceptional performance of SACs arises from their unique physicochemical characteristics endowed by atomic dispersion, which can be summarized into six core aspects: (1) maximized atomic utilization and cost efficiency: by eliminating inactive bulk atoms, SACs drastically reduce precious metal loadings while maintaining high catalytic activity.[9] (2) Precisely tunable electronic structure and coordination environment: strong metal–support interactions (MSIs) and well-defined coordination geometries induce ligand and quantum-confinement effects, enabling fine control over adsorption energies and activation barriers.[10] (3) The uniformity of single-atom sites suppresses undesired pathways and mitigates poisoning by avoiding multi-atom ensemble sites.[11] (4) Superior intrinsic activity and theoretical predictability: isolated metal

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centers typically exhibit unique electronic states that deliver high intrinsic activity,[12] while their structural simplicity enables accurate density functional theory (DFT) modeling and rational catalyst design.[13] (5) Robust stabilization against aggregation: strong anchoring at defects, heteroatom ligands, or vacancy sites—combined with high atomic migration barriers—imparts excellent thermal stability and mitigates sintering.[14] (6) Synergistic bifunctionality: SACs can integrate single-atom sites with redox-active supports to cooperatively activate multiple reactants, enabling efficient cascade and tandem reactions.[15]

The synthesis of SACs can be broadly categorized into two strategies according to the construction scale: bottom-up and top-down (Fig. 1). In the bottom-up strategy, atomically dispersed metal precursors, typically ions or complexes, serve as the starting units.[16,17] Its central mechanism relies on kinetic trapping and stabilization through rational chemical design, including defect engineering, coordination chemistry, and spatial confinement. These strategies effectively suppress metal aggregation during synthesis, enabling the precise anchoring of individual atoms onto the support. The goal is the direct construction of isolated single-atom sites. In contrast, the top-down strategy starts from pre-existing metallic aggregates, such as nanoparticles or bulk metals.[18] This strategy depends on the controlled disassembly of these metal structures through thermochemical, electrochemical or mechanochemical processes.[19] The generated atomic or sub-nanometer metal fragments are subsequently captured and stabilized by anchoring sites on the support.[20–23] This approach thus achieves the deconstruction of bulk or nanoscale metals into isolated atomic species. In summary, the bottom-up strategy constructs isolated single atoms through precise chemical design and controlled assembly from molecular precursors, whereas the top-down approach generates atomically dispersed species by disintegrating bulk or nanoparticulate metals. To ensure that SACs operate stably and efficiently across diverse catalytic reactions, maintaining their superior structural stability is crucial, which fundamentally depends on strengthening the interactions between the isolated metal atoms and the support. Strategies such as chemical modification of the support or heteroatom doping into the support can substantially enhance the MSIs, thereby improving the overall stability and durability of SACs. Moreover, the top-down strategy typically drives single atoms to anchor at the most thermodynamically stable sites, providing a significant advantage for enhancing the structural stability of SACs.

This review provides a comprehensive overview of recent advances in top-down strategies for synthesizing SACs and their unique applications. By integrating the latest progress in this field, we aim to stimulate further innovation and promote broader adoption of top-down methodologies. We begin by outlining the fundamental characteristics of the top-down approach and summarizing key breakthroughs in synthetic techniques. We then critically examine specific application domains in which top-down derived SACs exhibit unique advantages. By highlighting the inherent properties and potential advantages of top-down strategies in both catalyst synthesis and practical deployment, we underscore their significant promise for the synthesis of high-performance heterogeneous catalysts for industrially relevant reactions. Finally, we offer an outlook on the major challenges and future opportunities in this rapidly evolving area, providing guidance for the continued development of top-down strategies.

2. Characteristics of the top-down strategy

2.1 How to realize the top-down synthetic strategy

The core principle of the top-down strategy lies in disrupting the metallic bonding network in metal particles or bulk materials

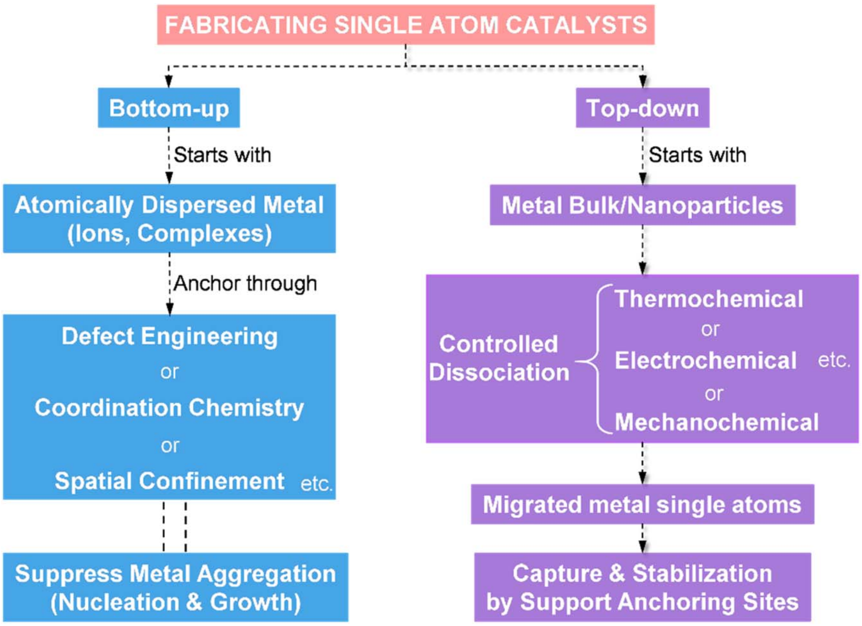

Fig. 1 The synthetic scheme of the bottom-up strategy and top-down strategy for synthesizing single atom catalysts.

through physical or chemical interventions. This destabilization breaks the metal lattice and releases individual atoms or small clusters, which are subsequently captured and stabilized by high-affinity sites on the support (*e.g.*, defects, functional groups, lattice vacancies), ultimately yielding atomically dispersed metal species. Conceptually, this process follows a three-step sequence of "depolymerization–capture–stabilization". Step 1: external energy input, provided by thermal activation,[24,25] mechanical milling, electrochemical potential,[26–28] or chemical etching/oxidation, overcomes the cohesive energy of the metal lattice, breaks M–M bonds and liberates atoms. The primary thermodynamic driving forces include the reduction of surface/interfacial energy from large particles and the entropic gain associated with atomic dispersion. Step 2: the liberated atoms diffuse across the support surface, with kinetics governed by temperature and support morphology. High-affinity anchoring sites (*e.g.*, oxygen vacancies in oxides,[29] N/P/S-dopant sites in carbons or Brønsted/Lewis acid sites in zeolites) thermodynamically favor capture *via* strong adsorption, thereby preventing re-aggregation. Step 3: the captured metal atoms are stabilized by electronic interaction, geometric confinement or covalent/ionic bonding, forming strong MSIs.

2.2 Advantages of the top-down synthetic strategy

The strategic advantages of the top-down strategy for synthesizing SACs arise from their ability to strengthen MSIs, enable intrinsic cost-effectiveness and allow catalyst upcycling. A landmark study by Datye *et al.* (2016) demonstrated the formation of thermally stable single-atom Pt on ceria *via* high-temperature atom trapping. An appropriately selected support captures volatile metal species released under reaction conditions, and effectively reverses nanoparticle sintering. This atom-trapping process generates highly robust single-atom configurations through strong MSIs, markedly improving the structural stability of the catalyst.[21] Li *et al.* further provided the direct observations of the transformation from metal nanoparticles into thermally stable single atoms using *in situ* environmental transmission electron microscopy.[30] In general, high-temperature treatments, characteristic of the top-down strategy (*e.g.*, thermal migration, carbothermal shock), promote strong covalent bonding and electronic coupling between the atomically dispersed metal and the support. Such reinforced MSIs are essential for preventing sintering and sustaining the structural integrity of SACs under practical operating conditions.

Leveraging established synthesis and processing techniques for bulk and nanomaterials provides a more scalable route for SAC production. Wu *et al.* demonstrated a gas-phase diffusion method capable of directly converting bulk transition metals (*e.g.*, Cu, Co, Ni) into thermodynamically stable single-atom sites anchored on carbon supports.[31–34] This approach circumvents the need for complex chemical precursors and multi-step wet-chemistry procedures, enabling continuous and large-scale fabrication of SACs—an essential step toward industrial implementation. A particularly notable advantage of this strategy is its versatility and economic efficiency. Beyond transition metals, it can also transform precious metal bulk sources (*e.g.*, Pt, Pd bulk or oxides) into the corresponding single-atom forms, thereby eliminating reliance on costly molecular precursors such as H_2PtCl_6.[35] This not only reduces raw material costs but also minimizes chemical waste.

Furthermore, Wu *et al.* extended the top-down strategy to develop a regenerative nitrogen-doped carbon trapping strategy.[36] This approach enables the reactivation of sintered supported catalysts—specifically those deactivated by metal particle growth through Ostwald ripening or sintering—by converting the aggregated nanoparticles back into thermodynamically stable single atoms anchored at defect sites on N-doped carbon. This regenerative atom-trapping process not only restores catalytic performance but also illustrates the broader potential of top-down methodologies for catalyst recycling.

3. Different synthetic methods of the top-down strategy

Over the past three years, significant progress has been achieved in the synthesis of SACs using the top-down strategy. These approaches can be systematically classified according to the primary external energy input that drives atomization, including heat treatment, abrasion and light/electrical energy.

3.1 Heat treatment

High-temperature treatment represents one of the most prominent top-down approaches for converting bulk metals into isolated single-atom sites on functional supports. Recent breakthroughs in this field have enabled the precise construction of high-performance SACs with enhanced activity, selectivity, and stability.[37–44] The major developments and underlying mechanistic insights are discussed in detail in the following section.

The top-down strategy offers distinct advantages for anchoring isolated metal single atoms onto support surfaces, enabling the construction of synergistic catalytic architectures. Tilley *et al.* successful stabilized Pt single atoms on Ru nanoparticle surfaces by transforming Pt islands into Pt single atoms.[45] This transformation is thermodynamically driven by the formation of strong Pt–Ru bonds and the reduction of the surface energy of the Pt islands. *In situ* environmental transmission electron microscopy (ETEM) captured the dynamic process: under thermal treatment in a H_2 atmosphere, the Pt islands gradually spread across the Ru surface until fully atomized dispersion was achieved (Fig. 2a). The resulting single-Pt-atom-on-Ru catalyst exhibits exceptional activity toward methanol oxidation reaction (MOR) and remarkable CO tolerance, both of which arise from the synergistic electronic modulation at the Pt–Ru interfacial sites.

The top-down strategy also facilitates the construction of autonomous, self-regenerating heterogeneous catalysts through *in situ* structural reconfiguration, in which reversible MSIs dynamically reverse sintering during repeated reaction–regeneration cycles. Recently, Hong *et al.* employed Ge-MFI zeolites to

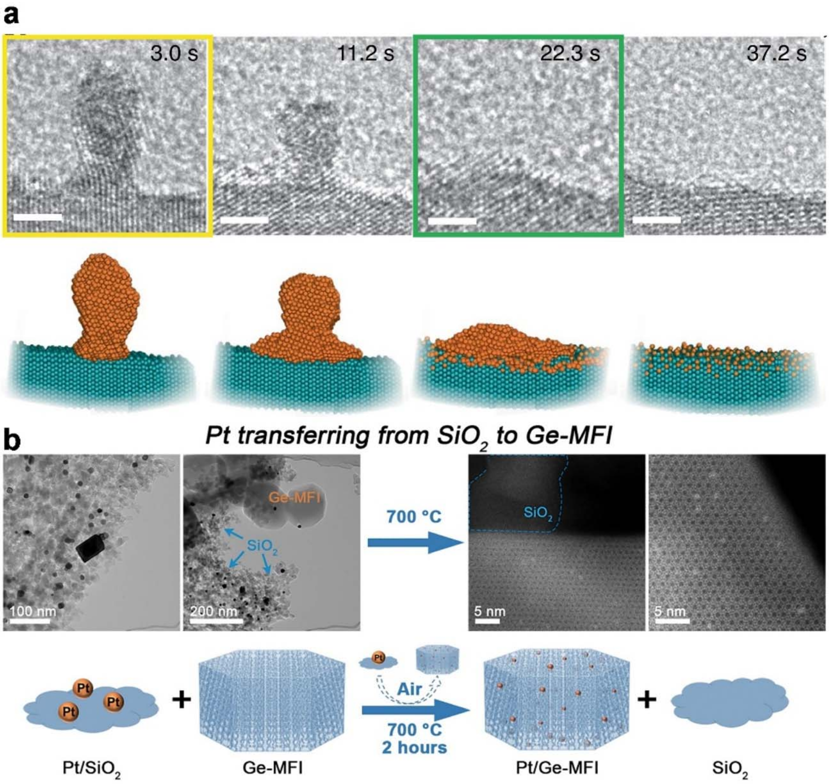

Fig. 2 (a) ETEM images and corresponding cartoons of the Pt island as it transforms during *in situ* heating under a flow of H_2. Scale bars, 2.5 nm. Reproduced from ref. 45 with permission from Springer Nature, copyright 2022. (b) Synthesis of Pt/Ge-MFI catalysts through spontaneous dispersion of Pt species. Reproduced from ref. 46 with permission from The American Association for the Advancement of Science, copyright 2025.

capture and atomize various Pt precursors (such as Pt and H_2PtCl_6 particles), yielding self-regenerating Pt/Ge-MFI catalysts (Fig. 2b).[46] Pt species undergo spontaneous thermal redispersion within the Ge-MFI framework, effectively suppressing Ostwald ripening. A key feature of this system is that Ge atoms in the zeolite framework act as the exclusive anchoring sites for Pt atomization—an ability not observed in Sn-, Zn-, In-, Ga-MFI or pure MFI analogues—due to their unique electron-deficient character and favorable coordination environment. As a result, the Pt/Ge-MFI catalyst exhibits exceptional cyclicity in propane dehydrogenation (PDH), retaining full activity over more than 50 consecutive reaction–regeneration cycles. Mechanistic insights from DFT and *Ab initio* molecular dynamic (AIMD) simulations reveal that Ge sites cooperate with the micropores zeolite environment to confine mononuclear PtO_2 species formed after cluster fragmentation. Moreover, this approach enables kilogram-scale production of Pt/Ge-MFI and readily extends to Rh/Ge-MFI, Pt/Ge-BEA, and Rh/Ge-BEA systems, highlighting its industrial potential for scalable SAC manufacturing.

Increasing the metal site density in SACs is essential for advancing their practical deployment in functional devices.[47] However, conventional direct pyrolysis routes for synthesizing high-loading SACs often lead to metal atom aggregation. To overcome this challenge, Lu *et al.* proposed a universal top-down strategy for fabricating ultrahigh-density (UHD) SACs through a metal-sulfide-mediated atomization process (Fig. 3a).[48] In this approach, nitrogen-doping-assisted atomic dispersion converts metal sulfide nanoparticles into UHD-SACs. DFT calculations indicate that the transformation from metal sulfides to single atoms is thermodynamically favorable, with abundant N-doping sites providing critical anchoring centers that stabilize high concentrations of metal atoms. This method enables the synthesis of 23 distinct UHD-SACs with metal loading exceeding 10 wt%, including 17 systems exceeding 20 wt% and 6 systems surpassing 30 wt% (Fig. 3b). Remarkably, this approach is compatible with multicomponent systems, yielding quinary SACs with 25 wt% metal loading and vicenary SACs with 18.1 wt% metal loading. In oxygen evolution reaction (OER) catalysis, Ni–N–C UHD-SACs exhibit pronounced loading-dependent activity enhancement, attributed to synergistic interatomic electron coupling among adjacent Ni single-atom sites.

Placing single atoms at the interface between different supports allows one to leverage the complementary surface characteristics of each component, thereby simultaneously activating adsorbed molecules and increasing the frequency of effective collisions. Wu *et al.* reported a thermal printing strategy to transform metal nanoparticles into single atoms at the interface of different interfaces (Fig. 3c).[49] This strategy relies on the controlled interfacial migration of metal nanoparticles and the concurrent release of metal atoms, which are

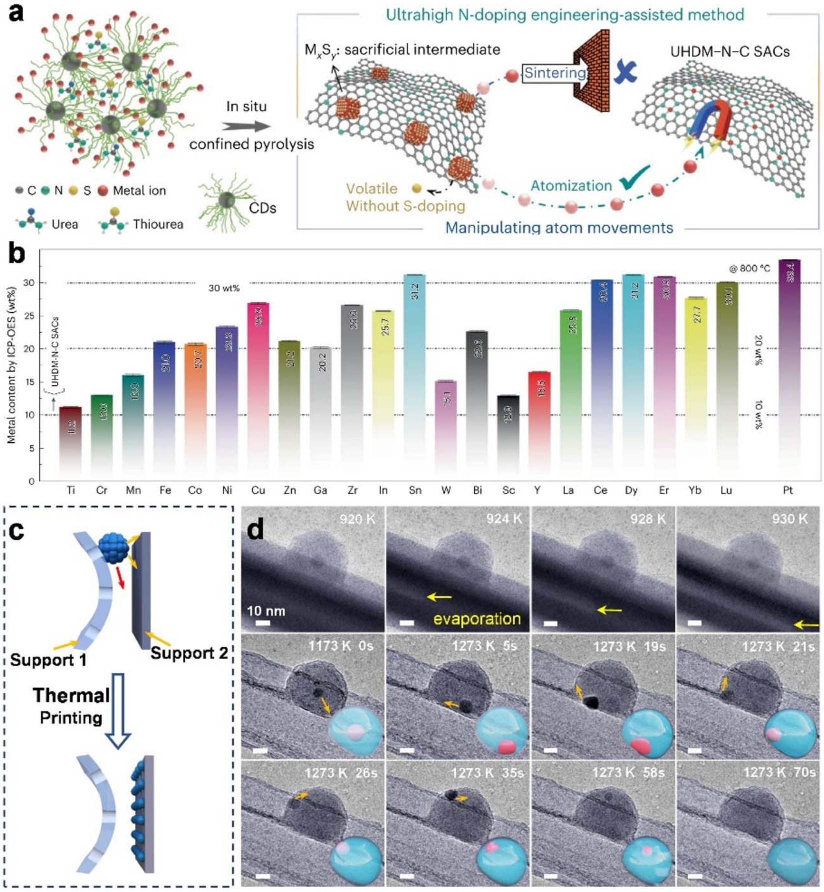

Fig. 3 (a) The proposed ultrahigh-N-doping engineering-assisted method, which provides sufficient anchoring sites to accommodate the released metal atoms, thereby manipulating the metal-atom migration pathway to form highly dispersed single metal atoms. (b) Ultrahigh metal loadings (by ICP-OES) achieved in this work for 23 different UHD SACs. Data are presented as mean metal loading values. Error bars indicate s.d. of five independent experimental results. Reproduced from ref. 48 with permission from Springer Nature, copyright 2024. (c) The proposed thermal printing strategy; (d) *In situ* transmission electron microscopy images at different temperatures and durations. Reproduced from ref. 49 with permission from the National Academy of Sciences, copyright 2023.

subsequently *in situ* trapped by the surrounding carbon support. *In situ* transmission electron microscopy revealed the dynamic evolution of Fe_3O_4 nanoparticles during the process: the nanoparticles flowed toward the SiO_2–carbon interface at 1173 K, migrated along the interface at 1273 K, and gradually decreased in size (Fig. 3d). DFT calculations further indicate that the released metal atoms preferentially anchor onto the adjacent defective support. This thermal-printing method is broadly applicable for preparing various SACs (Fe, Co, Mn, Pt, Au, Pd) at the interface, showing significant potential for diverse catalytic applications.

3.2 Abrasion

Baek *et al.* developed a top-down abrasion method in which bulk metals (Fe, Co, Ni, Cu) are directly atomized onto carbon-based, oxide, or nitride supports (Fig. 4a).[50] In this approach, mechanochemical energy serves as the driving force. This process requires only benign and readily available precursors, bulk metals, N_2, and graphite, without the need for hazardous chemicals or solvents, rendering this method both simple and environmentally friendly. Moreover, this method enables potential utilization of naturally occurring energy sources. During abrasion, the metal balls function as kinetic energy transmitters that generate abundant defects on the support surface (Fig. 4b). Simultaneously, N_2 not only provides a protective atmosphere but also dissociates under high-energy impacts, incorporating nitrogen atoms into the support. Furthermore, metal atom loading can be precisely tuned by regulating the abrasion rate.

3.3 Light/electrical energy

Light energy offers an alternative route for transforming metal nanoparticles into single atoms. Li *et al.* developed a laser ablation strategy that enables the precise conversion of Pt nanoparticles into Pt single atoms on CeO_2 (Fig. 4c).[51] By systematically tuning the laser power and irradiation time, the energy input can be accurately controlled, allowing fine adjustment of the Pt single-atom fraction relative to the total Pt loading on CeO_2. Catalytic CO oxidation tests revealed that Pt/CeO_2 catalysts with Pt single-atom proportions ranging from 1% to 100% exhibit different activities, with optimal performance achieved at a single-atom fraction of 19%. Similar to thermal

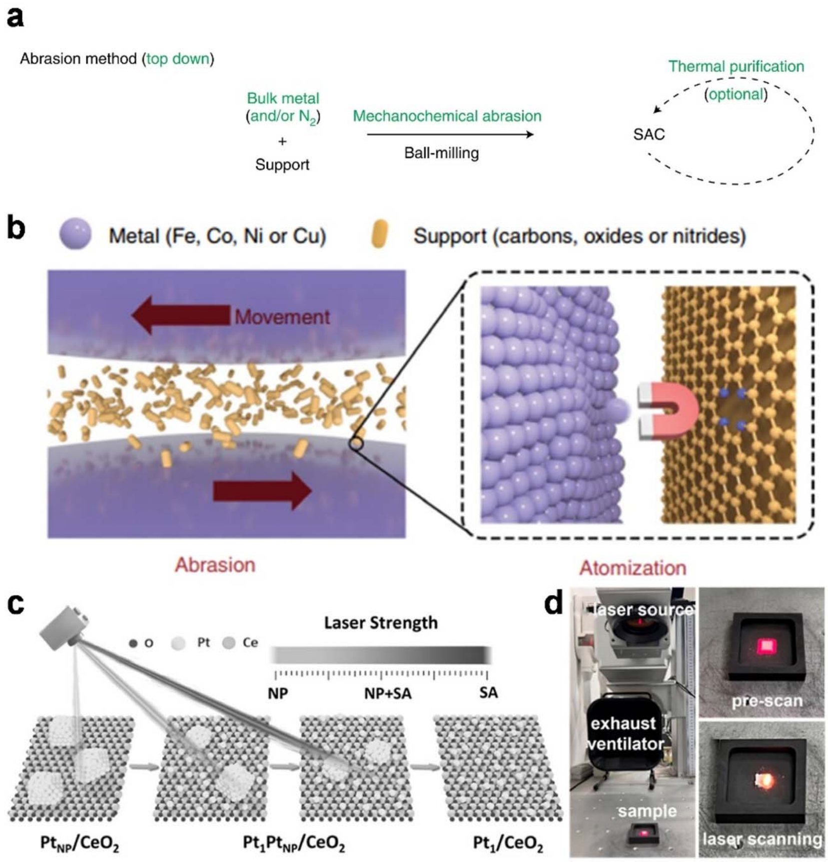

Fig. 4 (a) Abrasion method; (b) schematic of the mechanism for forming a SAC using the abrasion method. Reproduced from ref. 50 with permission from Springer Nature, copyright 2022. (c) Schematic illustration of the transformation of nanoparticles into single atoms by laser ablation; (d) photo of the laser ablation system and the laser ablation process. In a typical laser ablation process, the sample was first placed in a pre-scan square and then irradiated by the laser. Reproduced from ref. 51 with permission from American Chemical Society, copyright 2023.

activation, laser irradiation provides sufficient energy to overcome dispersed energy barriers while simultaneously heating the sample. Owing to its exceptional tunability, this top-down method enables precise manipulation of single-atom/nanoparticle ratios, offering valuable insights into synergistic effects in complex catalysis systems.

Zhao *et al.* further demonstrated a photoinduced route for transforming Au nanoparticles into single atoms.[52] In their approach, Au_{11} nanoclusters were first synthesized and electrostatically assembled on graphene oxide (GO) nanosheets. Subsequent light irradiation dispersed these clusters into Au single atoms, with the conversion efficiency governed by the irradiation wavelength. Comparative studies of R-Auc@GO exposed to infrared ($\lambda > 800$ nm), visible (400 nm $< \lambda <$ 800 nm), and ultraviolet ($\lambda < 400$ nm) light—along a dark control—revealed a wavelength dependent dispersion behavior, in which shorter wavelengths promoted more extensive cluster atomization. The resulting mixtures of Au clusters and single atoms exhibited synergistic catalytic activity in the enantioselective hydrogenation of α-methylbenzoylformate to chiral methyl mandelate.

Electrochemical etching has also emerged as a general top-down strategy for converting metal nanoparticles into SACs. Luo *et al.* reported a cathodic corrosion method in which a high cathodic potential disintegrates metal nanoparticles (Pt, Pd, Ir and Cu) into migrating single atoms, which subsequently diffuse and become immobilized on the N doped carbon support.[26,53]

4. Special application scenarios of the top-down strategy

The top-down strategy holds substantial promise for the practical synthesis of SACs, with the resulting SACs exhibiting exceptional catalytic performance across diverse reactions. By leveraging the intrinsic advantages of top-down synthesis, particularly its compatibility with realistic reaction environments, the full utility of this strategy can be effectively harnessed.[29,54] This section highlights several distinctive applications in which the top-down strategy directly addresses key challenges in real-world catalyst deployment.

4.1 Recycling sulfur-poisoned Pd metal catalysts

Although Pd-based catalysts are widely applied across numerous catalytic reactions, their practical deployment is often constrained by their strong susceptibility to sulfur

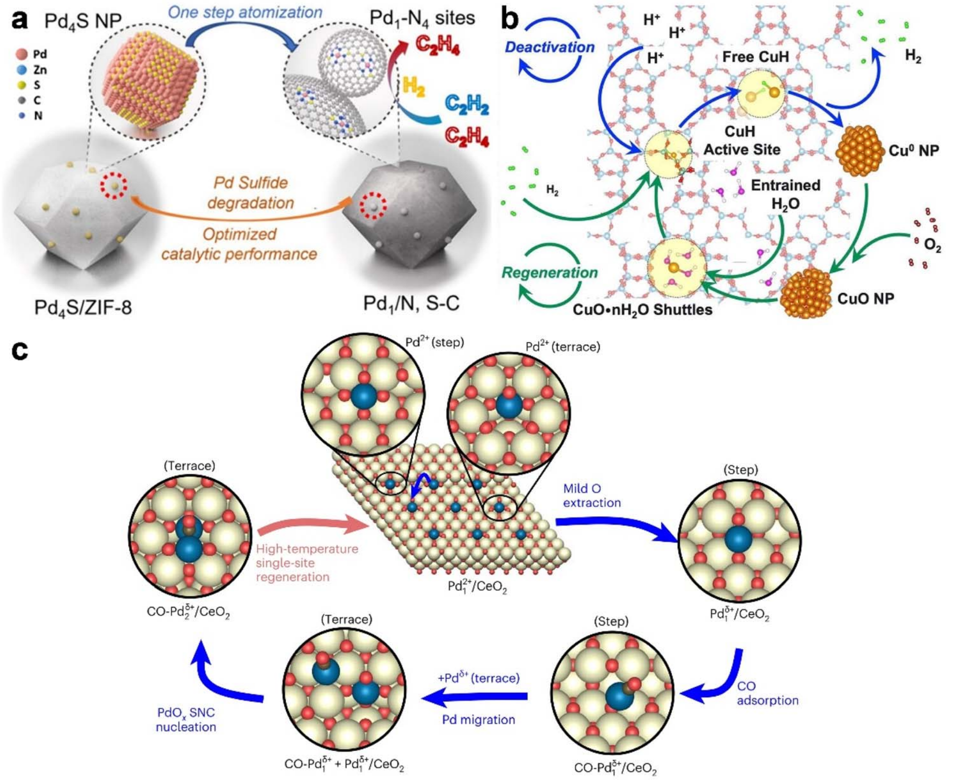

Fig. 5 (a) Scheme of the transformation of Pd sulfide nanoparticles into Pd single atoms. Reproduced from ref. 55 with permission from American Chemical Society, copyright 2025. (b) Conceptual illustration of the deactivation (blue lines/arrows) and regeneration (green lines/arrows) of Cu single atom ethanol upgrading catalysts. Reproduced from ref. 56 with permission from American Chemical Society, copyright 2024. (c) DFT computed atomic structures of the favoured Pd states on Pd-doped CeO$_2$ (111) at different operation stages, colour code: blue (Pd), red (O), brown (C) and beige (Ce). Reproduced from ref. 57 with permission from Springer Nature, copyright 2023.

poisoning. Upon exposure to sulfur-containing compounds, palladium readily forms palladium sulfide composites (Pd$_x$S$_y$), which suffer from severe deactivation and exhibit limited regenerability. To address this challenge, Li *et al.* developed a top-down strategy that converts Pd$_x$S$_y$ into high-performance Pd SACs through a one-step thermal atomization process (Fig. 5a).[55] Using Pd$_4$S as a model compound uniformly mixed with zeolitic imidazolate framework-8 (ZIF-8), high-temperature pyrolysis simultaneously decomposes Pd$_4$S and anchors atomic Pd onto the ZIF-derived N-doped carbon framework. Mechanistic investigations indicate that the atomization is driven by the abundant coordinating sites within the support. The regenerated Pd SACs exhibit exceptional activity and selectivity in acetylene semi-hydrogenation, demonstrating the effectiveness of this top-down regeneration route.

4.2 Dynamic Cu site redispersion to minimize deactivation

Zeolite-based copper catalysts are prone to irreversible sintering under reducing conditions, leading to the formation of Cu nanoparticles and consequent deactivation. To address this, Li *et al.* employed an atom-trapping strategy to achieve dynamic redispersion of Cu species in Cu–Zn–Y/deAlBeta catalysts, thereby restoring their catalytic activity (Fig. 5b).[56] In the fresh catalyst, Cu exists as atomically dispersed sites within the zeolite framework. However, after 150 hours of ethanol-to-olefin conversion, these isolated Cu sites migrate and aggregate into metallic clusters, causing deactivation. Remarkably, oxidative regeneration at 550 °C in air fully recovers catalytic activity by redispersing the aggregated Cu back into single-atom configurations. Mechanistic studies reveal that this regeneration follows a two-step pathway: (1) oxidation of Cu clusters into CuO intermediates, and (2) thermodynamically favored dissolution of CuO into mobile Cu^{2+} species, which subsequently diffuse to zeolitic anchoring sites. This top-down redispersion strategy effectively reverses sintering-induced deactivation, significantly extending catalyst operational lifetime.

4.3 Reaction environment modulated dynamic and reversible transformations

The top-down strategy stands out in fabricating thermally stable single atom sites, effectively suppressing high temperature sintering of metal nanoparticles. Wang *et al.* demonstrated a dynamic and reversible transformation of Pd species on CeO$_2$, achieving both low-temperature activity and high-temperature stability for automotive exhaust aftertreatment (Fig. 5c).[57] Under low temperatures (<400 °C), PdO$_x$ clusters supported on CeO$_2$ exhibit strong activity for methane oxidation. Upon thermal activation at 650 °C, these clusters spontaneously disperse into isolated Pd single atoms. The strong MSIs between Pd$_1$ and CeO$_2$ endow the system with exceptional thermal stability up to 850 °C. Crucially, during subsequent cold–start cycles, the Pd_1/CeO_2 can regenerate catalytically active PdO_x species under CO/O_2 treatment. This reversible phase evolution aligns exceptionally well with the transient operating conditions of automotive engines, offering a practical solution for after-treatment systems. This work innovatively implements the top-down strategy in real reaction environments to address catalyst thermal degradation.

5. Summary and perspectives

The top-down strategy exhibits distinct advantages in synthesizing SACs, particularly for industrial-scale production and specialized catalytic processes. This review systematically summarizes recent advances in both synthetic methodologies and practical implementation, establishing critical structure–activity relationships to guide future development. Despite these achievements, translating laboratory success into industrial practice remains challenging, with issues such as scalability, process compatibility and long-term operational stability yet to be resolved. Herein, we critically assess the major obstacles and emerging opportunities associated with top-down synthesis under practical conditions, aiming to accelerate its deployment in energy and environment relevant applications.

5.1 Support design

During the top-down synthesis of SACs, diffusing metal atoms experience a dynamic competition between *in situ* anchoring at defect sites and agglomeration into metal clusters/nanoparticles. Anchoring is favored when the binding energy between the metal atoms and the defect site exceeds the metal–metal cohesion energy, rendering the single-atom configuration thermodynamically stable. Strengthening the electronic interactions between metal atoms and the support, or creating geometric confinement can thermodynamically reinforce anchoring and suppress sintering. Meanwhile, increasing the density of defect sites and employing rapid quenching methods can kinetically hinder atom migration, thereby delaying the aggregation pathway of metal atoms. Therefore, achieving robust anchoring of metal atoms requires rational support design, both in terms of its electronic structure and defect topology, to steer the competition decisively toward stable single-atom formation.

5.2 Data-driven synthesis

In the top-down synthesis of SACs, various methods are employed to break metal–metal bonds and liberate single metal atoms. The efficiency of this atomization process is intrinsically metal-specific, as different metals must overcome distinct activation barriers that correlate with their metal–metal binding energies. Despite recent progress, quantitative frameworks capable of predicting and tailoring the diffusion behavior of specific metals remain limited. Moreover, reaction conditions, such as temperature, atmosphere and support environment, further influence the migration kinetics of diffusing single atoms, presenting additional complexity for practical applications. Integrating advanced data-driven models with molecular dynamics simulations offers a promising route to quantitatively resolve the *in situ* diffusion pathways of single atoms and identify the governing parameters. Such mechanistic insights will enable rational control over atomization and the migration process in top-down strategies, thereby accelerating their translation to large-scale production and expanding the practical utilization of SACs.

5.3 Design of renewable catalysts that adapt to the reaction environment

The top-down strategy enables the transformation of metallic nanoparticles into metal single atoms under appropriately designed conditions. This approach provides a promising pathway for regenerating sintered catalysts recovered from industrial waste streams. Leveraging the inherent thermodynamic stability of single-atom configurations, such conversion can occur *in situ* during catalytic operation, yielding catalysts with markedly enhanced resistance to high-temperature sintering. Furthermore, periodic redox cycling endows these systems with an intrinsic capacity for atomic-scale regeneration, effectively mitigating long-standing challenges associated with carbon deposition and irreversible nanoparticle growth. Integrating top-down atomization with practical reaction environments thus offers substantial potential for extending catalyst lifetimes and broadening the industrial applicability of single-atom technologies.

Author contributions

All authors contributed to the writing and revision of the manuscript.

Conflicts of interest

There are no conflicts to declare.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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