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Perspective

Single-Atom Nanoarchitectonics for Robotics and Other Functions

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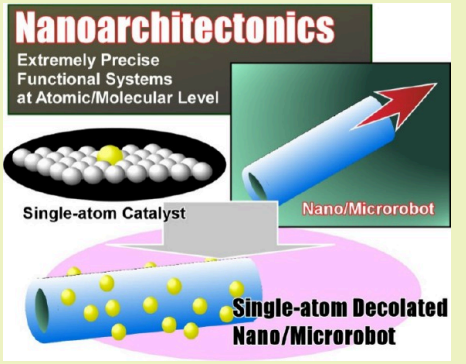
ABSTRACT: For a sustainable society, the development of materials must be carried out with the aim of maintaining good economic activity in the long term without damaging the global environment or overusing resources. Therefore, there is an increasing need for the development of advanced and sensitive materials. It has become clear that structural control at the nanoscale is particularly important for efficient and highly selective functional expression. This can be achieved with an emerging methodology of nanoarchitectonics, which is the concept of building functional materials from individual atoms, molecules, and their clusters into nanomaterials. One of the ultimate goals of this line of research is to construct extremely precise functional dynamic systems originating at the atomic and molecular levels. From this point of view, single-atom catalysts can be considered as the active components to achieve this goal. This Perspective discusses functional systems that implement single-atom catalysts as active components in the field of nano/microrobotics. Single-atom-decorated nano/microrobots are dynamic systems that exploit the catalytic activity of single atoms; they can be used to enhance propulsion abilities or to provide the catalytic capability. This Perspective consists of three main parts: (i) reviews on single-atom catalysts and others; (ii) single-atom-decorated nano/microrobots; (iii) other functions of single-atom nanoarchitectonics. Finally, the paper concludes with a discussion on the future direction and development of single-atom-decorated nano/microrobots. In particular, it is expected that the next generation of intelligent single-atom-decorated nano/microrobots will be developed using artificial intelligence. The combination of the basic story, the main story, and the side story will affect the diversity and future potential in the research fields of single-atom nanoarchitectonics for robotics and other functions.



KEYWORDS: *biomedical, energy, environment, nanoarchitectonics, nano/microrobot, nanozyme, single atom, single-atom catalyst*

INTRODUCTION

Humanity is facing many problems, such as energy production/storage,[1−5] environmental protection,[6−10] and biomedical issues.[11−15] Technological advances such as the development of devices and information equipment[16−19] can contribute, but the most important matter is the development of functional materials that can solve these problems. Accordingly, material chemistry and related engineering support human society. In fact, chemistry related to organic,[20−22] inorganic,[23−25] and various hybrid materials[26−28] has been continuously developed to create highly functional materials. Especially in these days, materials are required not only to be highly functional but also to have elements that satisfy the sustainability of society.[29−31] For a sustainable society, the development of materials will be carried out with the aim of maintaining good economic activity in the long term without damaging the global environment or overusing resources. This means that advanced and sensitive material development is increasingly required.

For the development of advanced materials, it is important to control not only the bulk properties of the materials themselves but also their fine internal structures. We have learned the importance of controlling the nanostructure throughout the history of research and development. The properties of the same material can be completely different when the nanostructure differs.[32,33] It has become clear that structural control at the nanoscale is particularly important for efficient and highly selective functional expression.[34−36] This progress in research is driven both by the proposal of an outline concept and by scientific and technological innovation. Concepts that are attracting attention in the matter of controlling nanostructures are nanotechnology and nanoarchitectonics.[37] Thanks to the concept of nanotechnology that was proposed by Richard Feynman in the mid-20th century,[38,39] it has become possible to

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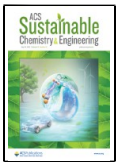

observe[40−42] and manipulate[43−45] structures at the nano level and to study specific properties[46−49] in nanoscaled fine structures. In response, Masakazu Aono proposed a new concept of nanoarchitectonics in the early 21st century.[50−53] Nanoarchitectonics is a concept that enables the construction of functional materials from individual atoms and molecules into highly ordered nanostructures and the evaluation of synergy between individual building blocks, the resulting nanostructures, and their environment.[54−56]

The development of science and technology and the establishment of concepts are two crucial and inseparable approaches toward the development of functional materials of highly organized structures for a sustainable society. The concepts of nanotechnology and nanoarchitecture have already enabled the creation of functional materials by controlling structures at the atomic, molecular, and nano levels in various scientific fields. For example, the synthesis of molecules in organic chemistry can be followed by their subsequent manipulation and organization into the resulting structures that originate at the molecular level.[57−59] In recent years, in the field of surface synthesis,[60,61] organic synthesis can be performed while observing molecular images. In addition, in local probe chemistry,[62,63] molecules can be manipulated with the tip of a probe microscope to induce a reaction. These are nice examples of fusions of nanotechnology into organic chemistry. Furthermore, in coordination chemistry, materials with regular nanopore structures, called metal−organic frameworks,[64,65] can be synthesized by designing highly ordered structures. Similarly, polymer chemistry produces regular nanopore materials, termed covalent organic frameworks.[66−68] There has been a remarkable evolution from conventional material chemistry to nanostructure control. The formation of highly organized nanostructures through self-assembly of molecules and materials is traditionally achieved in supramolecular chemistry.[69−72] For example, new classifications, such as supramolecular polymers,[73−75] have been proposed. In particular, localized assembly,[76] which is not simply a collection of molecules, but reflects locality, can lead to the expression of advanced functions, as seen in liquid−liquid phase separation in biological systems.[77,78] Template synthesis,[79,80] which uses molecular aggregates as templates, is used in inorganic chemistry to create regular nanostructures. In interfacial science, techniques such as self-assembled monolayers,[81,82] the Langmuir−Blodgett method,[83−86] and layer-by-layer assembly[87−89] have also made significant contributions in controlling the thin film structures at the nanoscale. All these areas of material chemistry are currently working on the element of nanostructure control and can be integrated into nanoarchitectonics, a paradigm for creating materials from the nanoscale.[90,91]

One of the ultimate goals in the nanoarchitectonics of functional material structures is to assemble complex functional systems with a variety of harmonious functions; similarly as observed in living organisms.[92,93] In living organisms, each individual function has a mechanism controlled at the molecular level, and these functions are harmonized to produce diverse and versatile functions. The ideal of material chemistry based on nanotechnology and nanoarchitecture can artificially create such highly organized functional material systems.

The opposite direction is to construct extremely precise functional systems at the atomic and molecular levels.[94,95] This Perspective presents research approaches in line with the latter goal. One of the ultimate forms for the latter goal is the single-atom catalysis,[96−98] where a single metallic atom acts as an active site that is supported in the overall structure of the catalyst. Because the bonds around the atom are unsaturated, it has much higher catalytic activity than bulk metals. It also has an extremely high metal atom utilization efficiency. Therefore, the ultrahigh atomic efficiency and unique reaction properties have attracted attention.

Another ultimate form for the latter goal is a molecular machine comprised of molecules and/or supramolecules working in synchronization.[99−101] Research began by measuring the behavior of molecular machines in solution, for which a Nobel Prize was awarded.[102−104] Since then, molecular machines were operated at liquid interfaces,[105−107] or on solid surfaces.[108,109] As an even more advanced example, the driving motion of molecular cars or nanoscale cars, called nanocars, has been observed.[110,111] An event using these ultimately small cars, a nanocar race has also been held.[112,113] In addition to single molecules and supramolecules, DNA machines[114,115] using DNA origami[116,117] are attracting attention as microscopic machines and robots with biological components. As distinct developments of nanomaterials, nano/microrobots have been actively researched.[118,119] A nano/microrobot is a very small robotic system that is designed to be autonomously propelled in a desired environment and to perform a specific given task, such as sensing, transport, collection, degradation, conversion of chemical substances, etc.

Based on this background, this Perspective discusses functional systems that combine single-atom catalysts and nano/microrobots (Figure 1), which are the two ultimate objects of

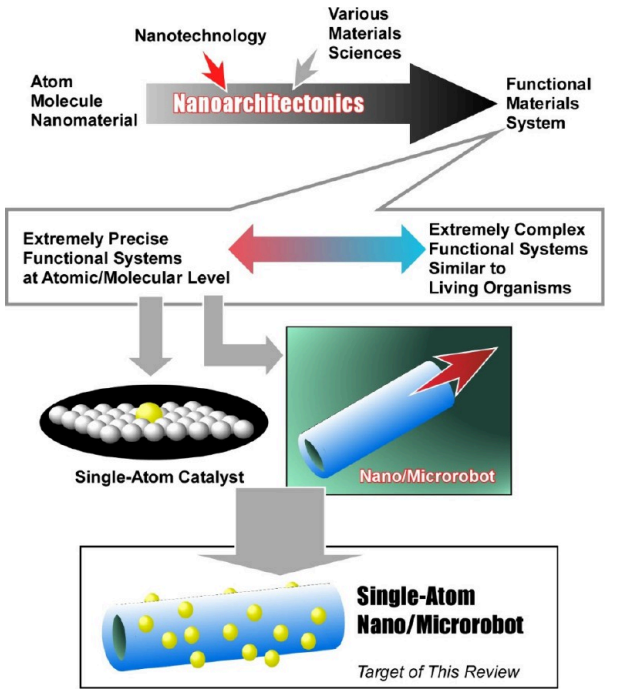

Figure 1. Background and target of this Perspective.

nanoarchitectonics. Single-atom-decorated nano/microrobots are dynamic systems with controllable propulsion abilities. They use the catalytic activity of single-atom sites to assist with a specific chemical conversion that can be a driving force for propulsion abilities and other functionalities. The history of single-atom-decorated nano/microrobots is not long.[120] Therefore, this Perspective includes the background of single-atom catalysts and related recent examples to introduce the field of

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ACS Sustainable Chem. Eng. 2025, 13, 6900−6917

single-atom catalysis in a broader context. Because we cannot select all the typical examples, recent related examples with features of nanoarchitectonics (nanoconstructions) are especially described in this Perspective. This Perspective is organized as follows. It consists of three main parts: (i) reviews on single-atom catalysts and others; (ii) single-atom-decorated nano/microrobots; (iii) other functions of single-atom nanoarchitectonics. This Perspective follows the logical flow, starting with single-atom catalysts as basics, discussing the main topics of single-atom-decorated nano/microrobotics, and expanding to other related functions. Finally, the future direction and further development of single-atom-decorated nano/microrobots is discussed. In addition, although not limited to single-atom catalysts and nano/microrobots, this Perspective also includes traditional single-atom-related examples such as the creation of structures with molecular recognition capabilities at the single-atom level and the control of spin and catalytic functions by modifying the carbon backbone at the single-atom level. The combination of the basic story, main story, and side story will affect varieties and future potentials in the research fields of single-atom nanoarchitectonics for robotics and other dynamic systems. This Perspective provides actual importance of combination of single-atom-level science, nano/microscopic dynamic functions, and possible relation to realistic application, as a novel emphasis point among nanoarchitectonics science and technology.

REVIEWS ON SINGLE-ATOM CATALYSTS AND OTHERS

Single-atom-decorated nano/microrobots operate using reactions over the single-atom sites to provide a specific functionality. Research on single-atom catalysts has generally been progressed more than that on single-atom-decorated nano/microrobots. Looking at the research overview and trends in single-atom catalysis in a larger concept is beneficial for transferring the principles and findings to the field of nano/microrobotics. Instead of focusing on a large number of papers, we selected several review papers from the field of single-atom catalysts and projected the main findings and functionalities into the field of nano/microrobotics.

The review article by Varma, Zbořil, and co-workers[121] discussed the application of earth-abundant single-atom catalysts with regard to electrochemical energy storage and electrocatalytic conversion of chemicals to fuels or high-energy products. The review summarized the activity of single atoms as active sites for electrochemical water splitting from the point of view of hydrogen evolution reaction (HER) and oxygen evolution reaction (OER). Simultaneously, nitrogen reduction reaction as a pathway toward the electrocatalytic generation of ammonia was highlighted. Last but not least, the applicability of single-atom catalysts to fuel cell technologies and metal−air batteries was discussed (Figure 2). The review also pointed out the need to implement computer-aided design in future directions to make the development of new single-atom catalysts for energy production and storage more efficient. Traditional approaches toward the design of functional materials are largely experimental and empirical, and they often require long and economically challenging optimization processes. Therefore, the integration of machine learning and artificial intelligence with computational methodologies such as density functional theory (DFT) will guide the scientists toward a better understanding of electronic and atomic-level interactions in catalytic materials. It will also help to assess how the local environment of a single

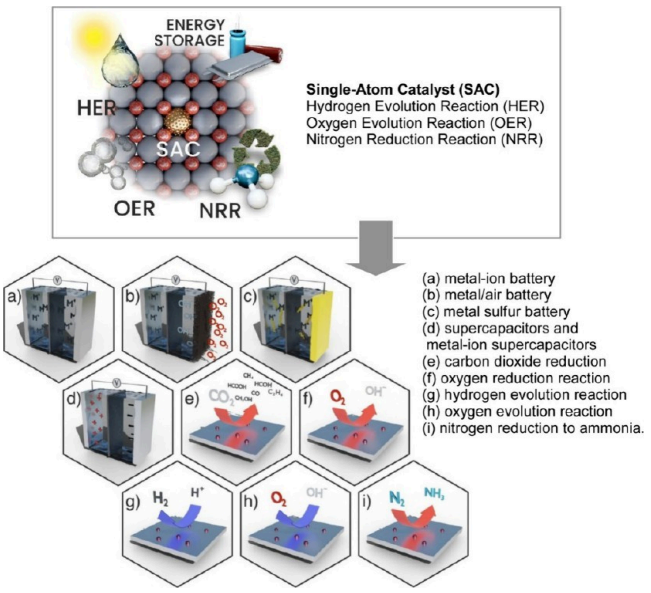

Figure 2. Application of single-atom catalysts in electrochemical energy storage and electrocatalytic conversion of chemicals to fuels and high-energy products. Reproduced under terms of the CC-BY license from ref 121. Copyright 2024 American Chemical Society.

atom affects its catalytic activity and stability. This approach to highly optimized design will be useful in targeting robust, selective, and sustainable materials for green energy applications.

The review article by Duan, Xu, and co-workers[122] explored heterogeneous metal-based catalysts of Fenton-like reactions with the focus on water purification processes. The review systematically explored the use of nanoparticles, atomic clusters, and single-atom catalysts for advanced oxidation processes with regard to their morphology, electronic structure, and oxidation state. Moreover, technical aspects of wastewater treatment were scrutinized; in particular, the production of high-quality single-atom catalysts, reducing the cost of catalysts and limiting the use of excessive oxidants, and designing complex reactors were considered. Specific challenges, such as the treatment of residual sulfates in the effluent and avoiding the complexity of the effluent matrix, were discussed. The review implied the need for further technological advances in the larger-scale technical applications with respect to the economic feasibility of Fenton-like processes and technologies based on advanced metal nanoparticles or single-atom catalysts (Figure 3).

Ammonia plays a vital role worldwide and has a significant impact on human activities, particularly in agriculture, chemical production, and the textile sector. Electrochemical conversion

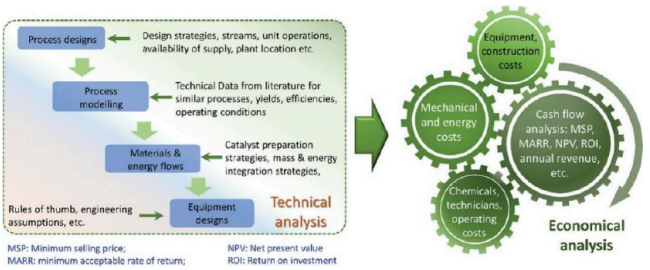

Figure 3. Technoeconomic assessment of single-atom-catalyst-based systems. Reproduced under terms of the CC-BY license from ref 122. Copyright 2024 Wiley-VCH.

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over single-atom catalysts offers a sustainable and long-term solution for ammonia synthesis[123,124] due to its environmentally friendly character. A review article by Pumera and co-workers[125] summarized the fabrication and characterization of Fe-single-atom-modified materials with the primary focus on the application for electrochemical ammonia synthesis. Mechanisms of electrochemical synthesis of ammonia over Fe-single-atom catalysts were explained with respect to current status. For example, the emerging field of nitrate reduction using Fe-single-atom catalysts was initiated less than several years ago, and it has already achieved notable milestones, reaching a 100% faradaic efficiency and an impressive ammonia yield. This progress presents a promising avenue for challenging the conventional Haber−Bosch process. However, maintaining high efficiency and yield concurrently remains a significant challenge in the realm of nitrate reduction. To fully harness the transformative potential of Fe-single-atom catalysts, it is essential to overcome the competing HER along with focusing on ammonia yield. Through advanced characterization, theoretical modelling, and a comprehensive understanding of the catalyst-electrolyte system, these gaps can be bridged. The full potential of single-atom Fe catalysts can be realized in order to promote their industrial application in sustainable ammonia production. Exceeding the Haber−Bosch efficiency will pave the way for more sustainable and energy-efficient ammonia production.

Another review article by Subhadarshini and Pumera[126] dealt with electrochemical ammonia production using single-atom catalysts. This review not only focused on the basic mechanisms of ammonia production over single-atom catalysts but also addressed the stability, selectivity, and efficiency of the applied catalysts. Moreover, advanced concepts, such as dual-atom catalysts and single-atom alloys, were discussed with respect to the sustainability of ammonia production. There are several factors that influence the overall ammonia generation activity from nitrates of single-atom transition-metal catalysts; these include the nature of the central transition metal, the d-orbital electron distribution, the type and heterogeneity of stabilizing ligands, the polarity around the transition metal, the properties of the base matrix, the influence of neighboring single-atom transition-metal catalysts, the effect of stability, and the effect of intermetallic single-atom/diatom alloy formation. These factors can be evaluated by computational studies and DFT simulations, etc. Therefore, this review considers the salient parameters that affect the activity of single-atom catalysts and provides guidance for the design of highly efficient and selective single-atom transition-metal catalysts for nitrate reduction.

Single-atom catalysts can also play a key role in the resolution of environmental problems such as the treatment and detection of various pollutants. A review article by Luo and co-workers[127] indicated that single-atom catalysts possess great applicability in technologies for air and water treatment and energy conversion. For example, single-atom catalysts can be used to promote the activation of persulfates to degrade bisphenol A and other pollutants, thereby achieving the objective of environmental control. This strategy is also useful for controlling the emission of volatile organic compounds, activating peroxymonosulfate to degrade pollutants, and green energy conversion. In such environmental applications, particular attention should be paid to maintaining and prolonging the stability and efficacy of single-atom catalysts. However, the effects of surface area, shape, size, and crystallinity of single-atom catalysts on these properties are not fully understood yet. Therefore, there is still room for technological development to bring single-atom catalysts as a next-generation sustainable solution in environmental applications on an industrial scale. For this problem, methodology to bridge atom-level science and macroscopic production is indispensable. Concepts and approaches of nanoarchitectonics would have meaningful contributions to these problems.

In addition to energy conversion and environmental remediation, single-atom catalysts have been intensively studied as nanozymes. Compared to natural enzymes, which are expensive and unstable, nanozymes[128−130] are usually easy to produce and have high stability, which shows great potential, especially in antibacterial research. From nanoparticles to single atoms, the geometric arrangement, electronic structure, and surface defects of the catalyst change accordingly, accompanied by changes in their catalytic activities. In principle, the catalytic activity can be efficiently improved by decreasing the particle size, which increases the availability of unsaturated coordination atoms. Therefore, single-atom nanozymes have great potential due to enhanced catalytic activity.[131,132] Accordingly, single-atom nanozymes show promising applicability as alternative antibiotics due their high atomic utilization, availability of active centers, and similarity with natural enzymes. The antibacterial applications of single-atom nanozymes and their combination with photothermal and sonodynamic therapies were reviewed in the article by Zhang et al.,[133] who discussed catalytic properties from the aspects of active sites, regulation of coordination environment, and carrier selection. Although the optimization of single-atom nanozymes has been greatly facilitated by theoretical simulations and many experimental investigations, there are still challenges that need to be addressed. First, possible intermediates and reaction pathways have not necessarily been discovered through the advanced characterization, leaving room for debates and in-depth studies. In addition, it is necessary to search for universal biocompatible supports for biorelated applications. Next, it is important to enrich the model database to better predict and evaluate the activity of single-atom nanozymes. In summary, the research on single-atom nanozymes and their antibacterial properties is still in an early stage. Further developments are expected to provide valuable insights into areas such as clinical treatment, food safety, and biosensors.

With the rapid development of characterization techniques in recent years, it has become possible to accurately and conveniently analyze single-atom catalysts. A review article by Liu and Zhang[134] reported on the analytical methods used for the characterization of single-atom catalysts. Although the review focused primarily on Pt-single-atom catalysts, the main findings can be extended to the characterization of other single-atom catalysts. The review listed crucial characterization techniques, such as high-resolution scanning tunneling microscopy, high-angle annular dark-field scanning transmission electron microscopy, extended X-ray absorption fine structure, X-ray absorption near-edge structure, diffuse-reflectance infrared Fourier transform spectroscopy (DRIFTS), X-ray photoelectron spectroscopy (XPS), and others, to accurately evaluate the nature and state of single-atom catalysts. As a result, the main drawback of the listed characterization techniques lies in their ex situ character. In the next steps, it is crucial to develop in situ characterization techniques for future investigations to address the formation process, thermal stability, and catalytic mechanism of single-atom catalysts.

Single-atom catalysts, in which the particles that make up the active reaction sites are reduced to the single-atomic level, can maximize the efficiency of active site utilization. Single-atom catalysts are promising because of their excellent catalytic

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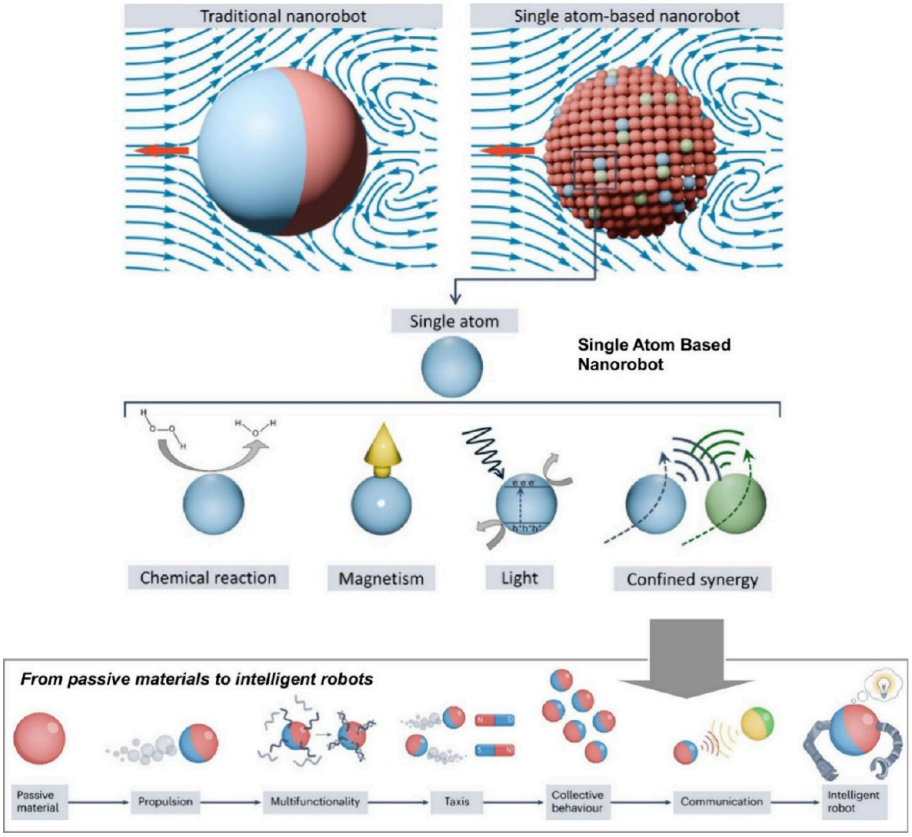

Figure 4. Single-atom-decorated nanorobots and precise manipulation at the atomic scale (top) and a development from passive materials to intelligent robots (bottom). Reproduced under terms of the CC-BY license from ref 135. Copyright 2024 American Chemical Society.

activity, selectivity, and cost performance. The examples presented in the above reviews showed a wide range of applications, such as electrochemical water splitting for green hydrogen fuel, HER, and OER. In addition, the production of clean ammonia by electrocatalytic nitrogen reduction reactions is attracting attention. Applications of single-atom catalysts in environmental catalysis include the control of emissions of volatile organic compounds and the degradation of pollutants. Single-atom nanozymes, which use single-atom catalysts as artificial enzymes, not only have high atom utilization, but also abundant active centers, and are excellent mimics of natural enzymes. They can provide an alternative to antibiotics in antibacterial applications. There is a clear relationship between physical properties such as size, degree of unsaturation, electronic structure and oxidation state, and catalytic behavior and efficacy. State-of-the-art characterization techniques and theoretical calculations are also being developed to analyze the complex electronic and structural properties of active sites. In the next section, we will discuss single-atom-decorated nano/microrobots that bring these single-atom catalysts in motion.

SINGLE-ATOM-DECORATED NANO/MICROROBOT

In this section, we focus on the combination of nano/microrobotics with single-atom catalysts to define highly organized and functional dynamic systems. Single-atom-decorated nano/microrobots utilize individual atoms to enable precise functionalization at the atomic scale (Figure 4), providing unprecedented advantages in biological, environmental, and food applications.

First, we present a recent perspective article in this field that was published by Ju and Pumera,[135] who focused on the implications of single-atom decoration in the field of nano/microrobotics. The perspective evaluated the contribution of single-atom-decorated nano/microrobots across biological, environmental, and food applications. The enhancement of desired efficiency per unit mass was highlighted across all applications as well as environmental impact and biological toxicity. At the current stage, nanoarchitectonics is incorporated at the level of single atoms to provide catalytic abilities such as catalytic decomposition of fuels and the generation of reactive oxygen species (ROS) toward catalytic degradation. Atomic precision increases catalytic efficiency per unit mass, and improves energy efficiency and effectiveness in tasks such as pollution remediation in environmental applications. It could also reduce the burden of catalytic materials, minimizing environmental impact and biological toxicity. The applications of single-atom functionality are limitless, and their integration can provide multiple functions within a single nanorobot configuration to improve the overall versatility and applicability in various fields. For example, nanorobots decorated with single atoms could even replace antibiotics in the treatment of infections. These tiny swarms could be coordinated to perform nanosurgery. On a more realistic level, we could consider the dreamy prospect of controlling the degradation of nanoplastics. This is an exciting frontier for advanced medical and environmental interventions.

A comprehensive study by Jancik-Prochazkova et al. reported on the propulsion abilities of TiO_2 nanorobots decorated with Pt single atoms and evaluated the efficiency in microplastics capture.[136] As shown in Figure 5, three different tubular TiO_2 nanorobots were tested: (i) TiO_2 nanorobots, (ii) TiO_2 nanorobots with point surface defects represented by oxygen

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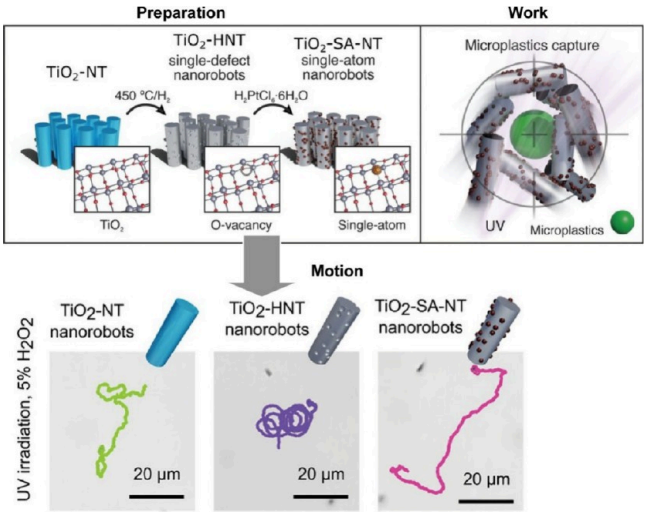

Figure 5. Propulsion abilities of Pt-single-atom-decorated TiO_2 nanorobots for microplastics capture, in which three different tubular TiO_2 nanorobots were tested: TiO_2 nanorobots, single-defect nanorobots, and single-atom-decorated nanorobots. Reproduced under terms of the CC-BY license from ref 136. Copyright 2024 Wiley-VCH.

vacancies, and (iii) Pt-single-atom-decorated nanorobots. As a result, single-atomic-scale nanoarchitectonics on the surface of TiO_2 nanorobots showed a significant influence on propulsion abilities, resulting trajectory, and interaction with microplastics. It was observed that surface point defects supported schooling behavior of nanorobots leading to efficient microplastics capture, whereas the presence of Pt single atoms enhanced the propulsion in 3D due to the negative photogravitaxis phenomena suggesting bigger degrees of freedom when propelled. Under UV irradiation, the nanorobots induced clustering, resulting in efficient and irreversible capture of microplastics. The proof-of-concept results pave the way for efficient microplastic remediation from aqueous environments and related environmental technologies using single-atom-decorated nanorobots.

Integration of a synergistic approach of Pt single atoms and Pt atomic clusters toward photocatalytic performance in environmental applications, namely in pollutant photodegradation applications, was reported by Ying, Ji, Wang and co-workers.[137] While single atoms have the hightest efficiency per unit mass, atomic clusters possess metallic characteristics, including conductivity and charge transfer ability; representing a great synergy in the resulting applications. The study was performed using bowl-shaped TiO_2 microstructures that were decorated with Pt single atoms and Pt atomic clusters through wet deposition impregnation (Figure 6). The metallic character of the Pt atomic clusters was crucial in achieving propulsion abilities in the mode of the bubble propulsion mechanism by catalytically decomposing H_2O_2 fuel. However, the presence of single Pt atoms and atomic clusters contributed to the degradation efficiency of model pollutants due to efficient electron transfer from Pt single atoms to TiO_2 and facilitated charge separation at the TiO_2 and Pt atomic cluster interface. Briefly, single Pt atoms and Pt clusters on the TiO_2 surface increased the number of active sites, which, in turn, enhanced charge separation during photocatalysis and extended the hole lifetime. The surface holes oxidized the hydroxyl groups to form $^{\bullet}OH$ radicals and the electrons captured by the single Pt atoms or clusters reacted with O_2 to form $^{\bullet}O_2^-$. Both $^{\bullet}OH$ and $^{\bullet}O_2^-$ promoted the decomposition of pollutants simultaneously. The photocatalytic abilities of the micromotors were demonstrated using model contaminants, methyl orange and tetracycline hydrochloride. This work represents a promising platform for environmental applications of single-atom-based catalytic micromotors, which is expected to open new avenues for large-scale applications in environmental and catalytic fields.

The development of nano/microrobotics has shown great applicability in biomedicine, it has been demonstrated for drug delivery, local manipulation, and biosensing, among others. One of the biggest challenges in the application of nano/micro-

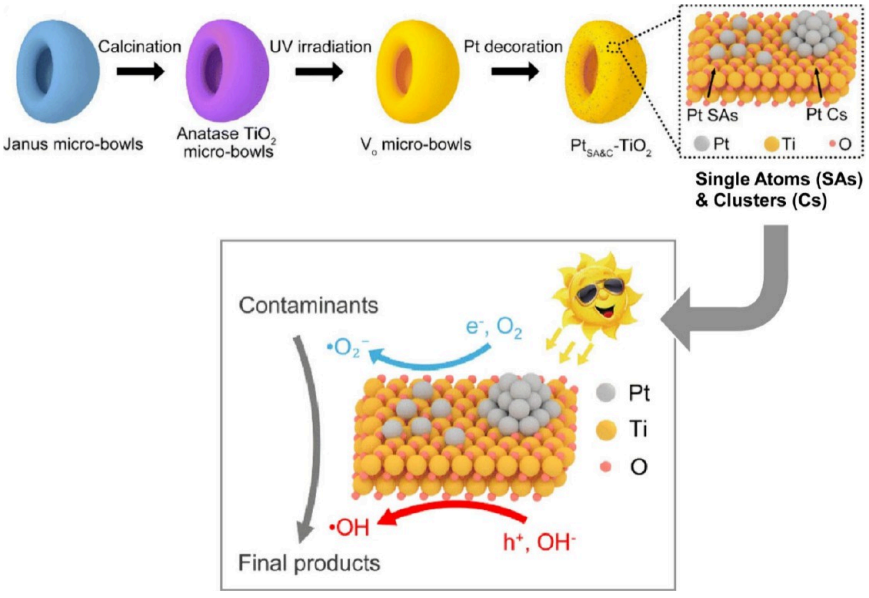

Figure 6. Bowl-shaped TiO_2 microstructures decorated with Pt single atoms and Pt atomic clusters via wet deposition impregnation, where the micromotors were employed for photodegradation of model contaminants. Reproduced with permission from ref 137. Copyright 2025 Royal Society of Chemistry.

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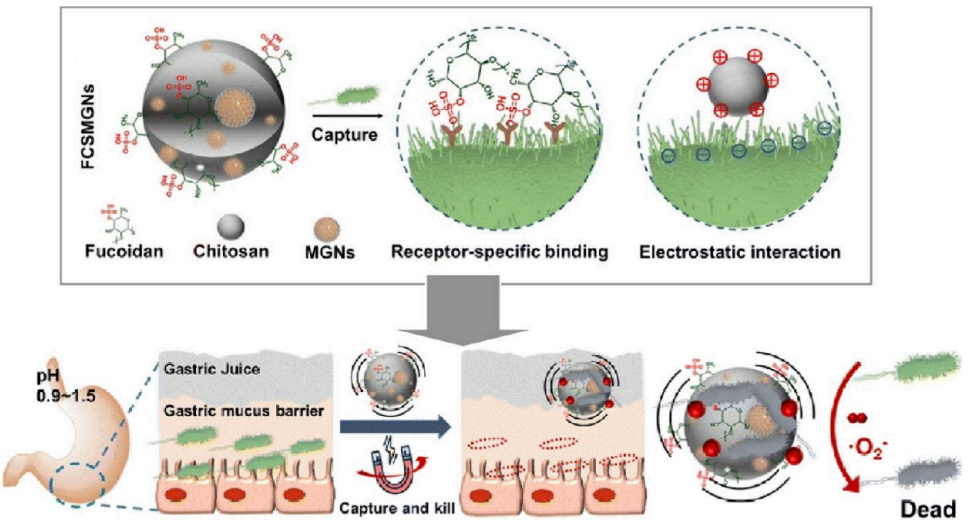

Figure 7. Single-atom-anchored microsweepers for *H. pylori* inhibition to generate ROS under acidic conditions as the next-generation bacterial infection treatment (beyond resistance). Reproduced with permission from ref 138. Copyright 2023 Royal Society of Chemistry.

robotics in medical applications is the limitation of their propulsion abilities in the environment of living organisms. For example, the propulsion abilities of nano/microrobots accelerate the development of drug delivery. However, traditional designs of the propulsion abilities face challenges in the stomach environment due to the barriers of acid, pepsin, and viscous mucus in the gastrointestinal tract. From this point of view, localized acid-driven/bubble-driven micromotors are expected to be able to target the gastrointestinal wall and improve drug retention. As shown in Figure 7, Tan, Chen, and co-workers developed single-atom-anchored microsweepers to inhibit *Helicobacter pylori* in the gastrointestinal tract.[138] The microsweepers were fabricated by coupling magnetic graphitic nanocapsules with fucoidan and chitosan during electrostatic spraying, and they were subsequently decorated with Fe single atoms by attachment of iron-containing porphyrin and subsequent high-temperature pyrolysis. The nanoarchitectonics in the design of microrobots enabled efficient capture of *H. pylori* in simulated gastric fluid: (i) the presence of magnetic graphitic capsules enabled precise navigation in gastric media using external magnetic field, (ii) fucoidan and chitosan provided the ability to efficiently capture *H. pylori* in gastric mucus, and (iii) single Fe atoms displayed oxidase-like activity resulting in the generation of ROS under acidic conditions. The synergy between all three aforementioned functionalities led to an efficient activation of the microrobots under acidic conditions that provided antibacterial performance in the defined localization of the gastrointestinal tract. Microsweepers could also be integrated with other therapeutic molecules for other gastric diseases, demonstrating an efficient pathway for next-generation detection and treatment approaches to various gastric diseases.

In the development of nano/microrobots, it is crucial to achieve high mobility, which can be achieved through the decoration with Fe single atoms. One possible approach is to form a high density of Fe active sites on the support. However, conventional pyrolysis methods of single-atomic Fe catalysts require high temperatures and long annealing times, leading to the formation of Fe-based aggregates. Therefore, a pyrolysis approach that improves the distribution of single Fe atoms is needed. Cao, Müllen, Zhou, and co-workers have developed a versatile low-energy approach of pulsed hydrogen pyrolysis to synthesize Fe-single-atom catalysts (Figure 8).[139] Briefly, Fe

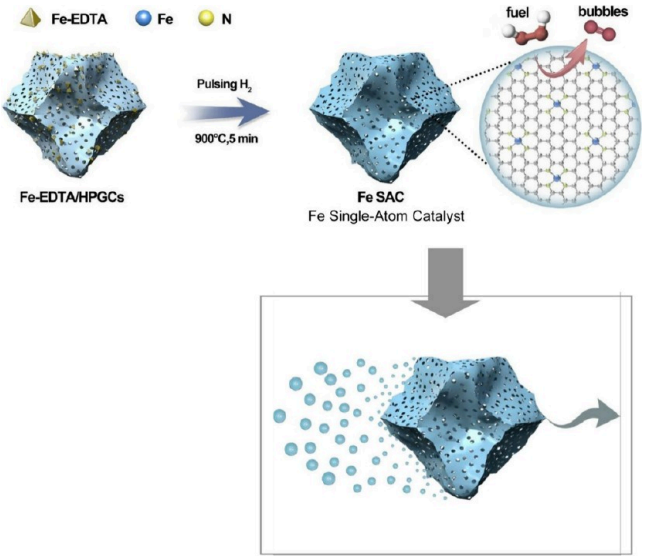

Figure 8. Pulsed hydrogen pyrolysis for synthesis of Fe-single-atom-decorated catalytic nanomotors that were propelled by catalytic decomposition of hydrogen peroxide fuel over Fe single atoms. Reproduced with permission from ref 139. Copyright 2024 Elsevier.

single atoms were introduced to the structure via pulsing hydrogen pyrolysis of carbonized zeolitic imidazole frameworks decorated with ferric ethylenediaminetetraacetic acid. As a result, Fe single atoms catalytically decomposed hydrogen peroxide, thus enabling the propulsion abilities. The resulting Fe-single-atom catalytic nanomotors exhibited active locomotion at H_2O_2 fuel concentrations as low as 10 mM. In addition, a great biocompatibility of nanomotors was demonstrated with breast cancer cells (4T1) and mouse embryonic fibroblast cells (NIH 3T3); the viability tests showed 90% and 100% viability, respectively, suggesting suitability for biomedical-oriented applications. The use of catalytically active single atoms as engines for the propulsion of nano/microrobots would be beneficial for further miniaturization that could lead to effective navigation and penetration into biological tissues. The excellent biocompatibility of Fe-single-atom catalytic nanomotors motivates their development for tasks such as cargo loading/

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ACS Sustainable Chem. Eng. 2025, 13, 6900−6917

release and catalytic repair of prodrugs in the biomedical field. Single-atom-decorated catalytic nanomotors with porous features could accommodate various cargos, such as molecular anticancer drugs and biosynthetic enzymes.

As a pioneering study on nano/microrobots decoration with Cu single atoms and their application in tumor therapy was presented by Du, Ma, Zhang, and co-workers (Figure 9).[140] N-doped jellyfish-like mesoporous carbon nanomotors coordinated with single-atom copper catalysts were prepared by an emulsion-induced interfacial anisotropic assembly strategy. The nanorobots were propelled under near-infrared (NIR)-light irradiation via self-thermophoresis, avoiding the necessity of using chemical fuels. The generation of •OH radicals catalyzed by single Cu atoms provided promising in vivo performance in tumor growth inhibition that was enhanced in the thermophoresis-driven propulsion mode. The study demonstrated cancer cell inhibition using a breast cancer cell line (MCF-7). NIR-light-driven propulsion increased the penetration depth of jellyfish-like mesoporous carbon nanomotors coupled with single-atom copper catalysts. More importantly, in in vivo experiments, the combination of single-atom chemodynamic therapy and NIR-light propulsion achieved a tumor inhibition rate of over 85% without the need for additional photothermal therapy. This study provided a rational design and fabrication strategy for integrating single-atom chemodynamic therapy and nanomotor self-propulsion to realize active nanomedicine.

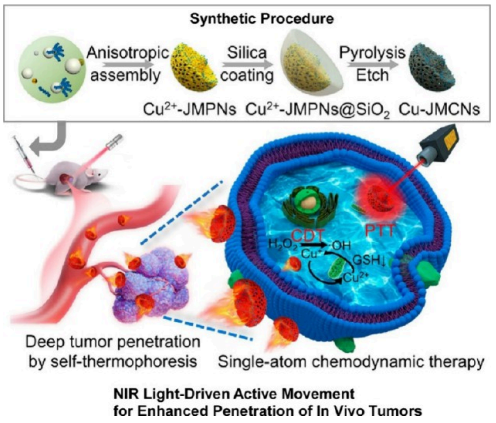

Figure 9. Cu-single-atom jellyfish-like nanomotors (Cu-JMCN) with NIR-light-driven propulsion for enhanced penetration of tumors in vivo. Reproduced with permission from ref 140. Copyright 2023 American Chemical Society.

OTHER SINGLE-ATOM NANOARCITECTONICS FUNCTIONS

Not limited to single-atom-decorated nano/microrobots, various dynamic functional systems based on single-atom catalytic abilities have been reported. Designs and functions of these systems will give important clues for the development of single-atom-decorated nano/microrobots. In this section, several recent examples of single-atom catalyst related research activities are discussed together with other related examples including single-atom-level molecular discriminations and single-atom-level carbon framework controls.

Single-Atom Nanozymes and Catalysts for Biology. The development of efficient catalysts to enhance interfacial redox reactions is essential for the construction of high-performance electrochemical biosensors.[141−143] Single-atom

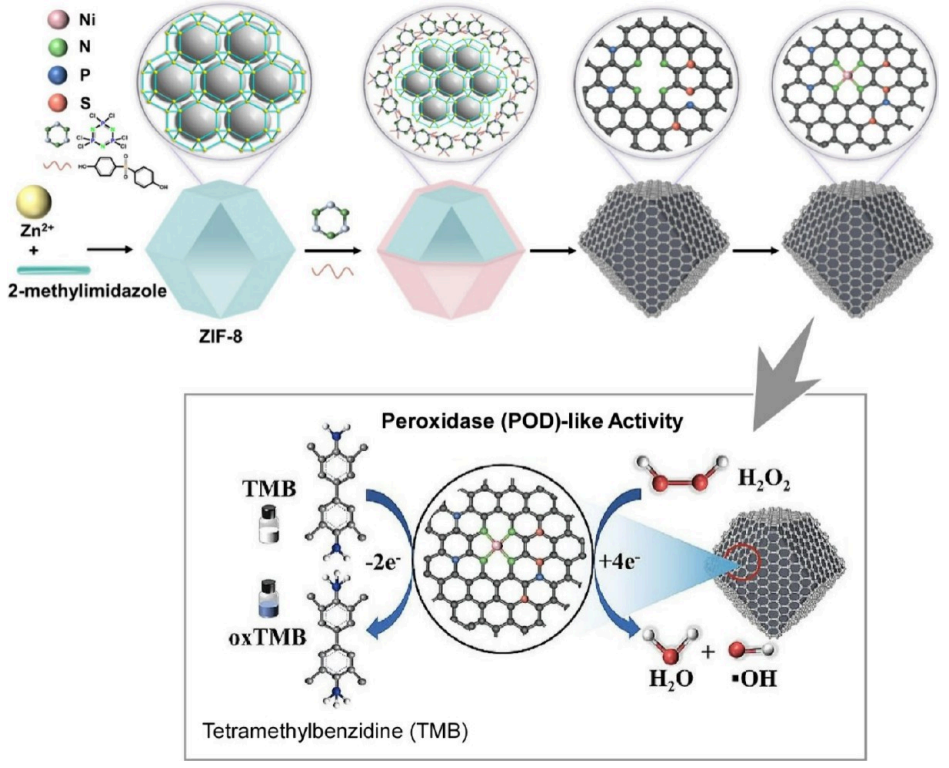

Figure 10. Ni-based single-atom nanozymes with heteroatom doping: (top) synthesis using a domain-restricted strategy for high-temperature pyrolysis of porous materials, ZIF-8; (bottom) excellent peroxidase-like activity and kinetics. Reproduced with permission from ref 144. Copyright 2025 Elsevier.

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nanozymes have attracted great attention for their excellent catalytic activity, predetermining them as promising materials for biosensing applications. Zhu, He, Cao, and co-workers took advantage of the fact that the enzyme-like activity of Ni-based single-atom nanozymes can be enhanced by heteroatom doping (Figure 10).[144] The Ni-single-atom-decorated S-, P-, and N-codoped porous carbon nanostructures (SPNC single-atom nanozyme) were synthesized using a domain-restricted strategy for high-temperature pyrolysis of porous materials. ZIF-8 was used as the carbon framework due to its abundant N species and its porous structure with a high surface area. The developed Ni-SPNC single-atom nanozyme exhibited excellent peroxidase-like activity; they outperformed the representative controls of Ni-based single-atom nanozymes with different atomic configurations. The change in the monatomic Ni coordination, driven by the heteroatoms P and S, provided the nanozyme with the peroxidase-like activity that was attributed to its Fenton-like reaction with H_2O_2. This process was also shown to generate a large amount of $^{\bullet}OH$, which reacted with tetramethylbenzidine to produce oxidized tetramethylbenzidine. Furthermore, the single Ni atom Ni-SPNC nanozyme was innovatively integrated with the motor-assisted CRISPR/Cas12a adaptamer DNAzyme. It was constructed as a new biosensor for the sensitive detection of atrazine. This sensing system showed low detection limit, remarkable selectivity, and good stability. This single-atom nanozyme-based electrochemical sensing platform opened a new avenue for the herbicide biosensing strategy. In general, the concept of a single-atom biosensor will greatly expand the applications of various sensing platforms.

Single-atom catalysts have also been targeted as active materials for the treatment of certain serious diseases. Neurodegenerative diseases such as Parkinson's disease are closely related to oxidative stress due to the excess of highly ROS, leading to damage of dopaminergic neurons. Wang et al. developed a Co−Cu diatomic nanozyme (CoCu-DAzyme) by uniformly immobilizing active Co and Cu sites on an AlO(OH) substrate for efficient ROS scavenging (Figure 11).[145] The

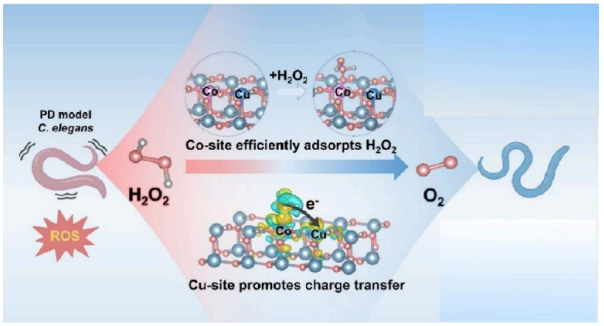

Figure 11. Co−Cu diatomic nanozyme with uniformly immobilized Co and Cu active sites on an AlO(OH) substrate for efficient ROS scavenging, where therapeutics are tested using the nematode (*C. elegans*) model of Parkinson's disease. Reproduced with permission from ref 145. Copyright 2024 American Chemical Society.

catalytic efficiency of the Co−Cu diatomic nanozyme far surpassed that of its single-atom counterparts of Co or Cu. The Co sites were found to efficiently enable H_2O_2 adsorption, while the Cu sites promoted charge transfer, which synergistically promoted the catalytic decomposition of H_2O_2 to H_2O and O_2. The developed Co−Cu diatomic nanozyme significantly improved α-synuclein aggregation by substantially scavenging ROS in the body. Therapeutic results showed a significant reduction in α-synuclein aggregation and an improvement in motor dysfunction in the nematode model (*Caenorhabditis elegans*) of Parkinson's disease. This study demonstrates a new therapeutic strategy for oxidative stress-related neurodegenerative diseases through the development of well-designed nanozymes. These findings highlight the potential of diatomic nanozymes as effective antioxidant drugs and provide new perspectives for the treatment of neurodegenerative diseases.

Wang, Zhao, and co-workers elucidated the effects of trace dopants, particularly metal elements, on biomass-derived carbon materials.[146] This work demonstrated transformation of metalloproteins into single-atom sites implemented in microbe-derived carbon materials toward electrochemical production of H_2O_2. The microbe-derived carbon materials were obtained by pyrolysis of selected microorganisms and, depending on the presence of metalloproteins, different atomic sites were formed during the fabrication process, such as Mg, Mn, Fe, Yn, Cu, Ni, and Co. The Fe-single-atom sites generated by pyrolysis of *Bacillus pumilus* were evaluated as the most suitable active sites for electrochemical H_2O_2 production, as they achieved a high H_2O_2 selectivity and remarkable yield. The results indicated that the precise control of heteroatom ligands within microbial Fe-dependent proteins is a sustainable and cost-effective approach for the fabrication of Fe-single-atom catalysts. The adjacent O coordination regulated the charge distribution of the $FeN_{5-x}O_x$ sites and shifted the reactive site from the Fe atom to the O-adjacent C atom. When catalyzed by FeN_3O_2 sites, the O-adjacent C atom effectively bound the intermediate $^{\bullet}OOH$, allowing significant H_2O_2 production. In addition to adjacent O coordination, axial O ligands in the various coordination configurations of $FeN_{5-x}O_x$ sites provided steric obstacles and prevented $^{\bullet}OOH$ from binding excessively to the Fe atom, thus improving the performance of the oxygen reduction reaction (ORR) for H_2O_2 production. By rational design of the coordination configurations, the highly active sites could improve the catalytic performance of biomass-derived carbon materials under milder conditions. In addition, this approach could reduce the risks associated with accidents and eliminate the need for hazardous chemicals.

Antibiotic resistant bacteria pose a threat to global health, and there is an urgent need to develop powerful antibacterial agents that are not susceptible to the development of bacterial resistance. To solve this problem, Bakandritsos, Zbořil, and co-workers developed single-atom decorated material to suppress resistance in bacterial treatments.[147] N-doped graphene acid was modified with Mn single atoms (Figure 12) and processed into a next generation of antibiotics showing its

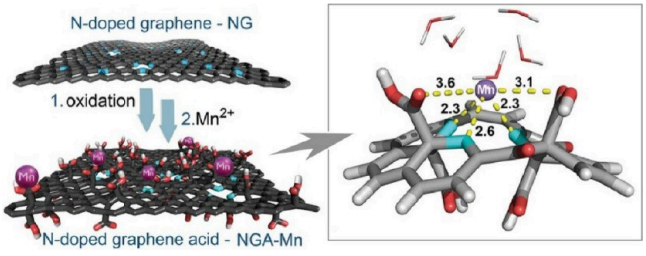

Figure 12. N-doped graphene acid modified with Mn single atoms as a next generation of antibiotics showing its potential against a broad spectrum of multidrug-resistant bacteria. Reproduced under terms of the CC-BY license from ref 147. Copyright 2024 Wiley-VCH.

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potential against a broad spectrum of multidrug-resistant bacteria. Interestingly, neither standalone N-doped graphene acid nor manganese ions showed any antibacterial activity. However, in synergy they possessed strong antibacterial activity, high cytocompatibility with human cells, and did not support the development of bacterial resistance even after 30 bacterial generation passages that overcame commonly used Ag nanoparticles. This approach developed potent, durable, and broad-spectrum antibacterial agents without the use of molecular antibiotics, which are often related to the development of bacterial resistance. In particular, the strong coordination of manganese cations on the functionalized graphene derivative (termed NGA-Mn) completely inhibited the growth of both Gram-positive and Gram-negative multidrug-resistant bacteria.

Single-Atom Catalysts for Sensing. Single-atom catalysts and nano/microrobots can also be used in advanced biosensing. In particular, self-adaptability is highly anticipated for artificial devices such as chemical-sniffing robots. To this end, the development of catalysts with multiple modularizable reaction pathways is a promising approach. However, they are often hampered by inconsistent reaction conditions and negative internal perturbations. To overcome this, Shen, Zhang, and co-workers reported a Cu-single-atom-decorated graphitic catalyst based on C_6N_6, an adaptable two-dimensional material (Figure 13).[148] It promoted the base oxidation of peroxidase substrates

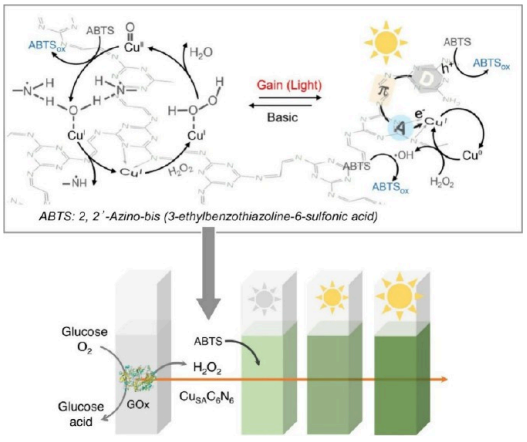

Figure 13. Proposed mechanism for dual peroxidase-like and photocatalytic pathways mimicking the basic activity and gain effect using a Cu-single-atom catalyst based on C_6N_6, an adaptable two-dimensional material, which can further be applied to glucose biosensors that can intelligently switch between sensitivity and linear detection range in vitro. Reproduced under terms of the CC-BY license from ref 148. Copyright 2023 Springer Nature.

via a coupled copper−oxo pathway and performed a second, light-triggered amplification reaction via a free hydroxyl radical pathway. This diversity of reactive oxygen-related intermediates for the same oxidation reaction allowed the reaction conditions to be the same. In addition, the unique topological structure of the single Cu atoms implemented on C_6N_6 and the special donor−π−acceptor linker facilitated the separation and transfer of intramolecular charge, suppressing the negative interference of the two reaction pathways. As a result, superior gains of up to 3.6 times were observed even under household lighting, outperforming the controls containing peroxidase-like catalysts, photocatalysts, or their mixtures. The single Cu atoms dispersed on C_6N_6 can also be applied to glucose biosensors that can intelligently switch between sensitivity and linear detection range in vitro.

Single-atom catalysts have recently attracted considerable research interest in the field of electrochemiluminescence because of their excellent catalytic activity.[149,150] Hu, Chen, and co-workers applied the Ni-single-atom catalyst to a graphitic carbon nitride (g-C_3N_4)−H_2O_2 electrochemiluminescence system and successfully improved its cathodoluminescence.[151] In particular, g-C_3N_4 acted not only as an electrochemiluminescent luminophore but also as a support for immobilizing the Ni-single-atom catalysts. This system incorporated an entropy-driven DNA walking machine-assisted CRISPR-Cas12a amplification strategy (Figure 14). This led to the development of a Ni-single-atom catalyst@g-C_3N_4−H_2O_2 electrochemiluminescence system for the detection of hepatitis B virus (HBV) DNA as low as 17 aM. The biosensor was constructed by modifying the Ni-single-atom catalyst@g-C_3N_4 on a glassy carbon electrode (GCE). An initial electrochemiluminescence signal (ON state) was obtained by adding H_2O_2 as a coreactant to the detection solution. Subsequently, hemin/G-quadruplex was introduced to the GCE surface to consume H_2O_2, which quenched the electrochemiluminescence signal (OFF state). The Cas12a-CrRNA complex was then incubated to activate the trans-cleavage activity of Cas12a. Finally, activated CRISPR/Cas12a cleaved the G-quadruplex and restored the electrochemiluminescence signal (ON state). This work extends the application of single-atom catalysts in electrochemiluminescence systems and provides new ideas for improving the electrochemiluminescence of g-C_3N_4.

Single-Atom-Level Molecular Discrimination. It has been shown that bottom-up synthesis of structures can distinguish differences in the structure of molecules at the level of single atoms, rather than functionalizing single atoms themselves. Song et al. reported the bottom-up preparation of a new material, molecularly thin N-doped 2D fullerphene (Figure 15).[152] A hybrid molecular thin film of fullerene C_{60} and ethylenediamine was first prepared in bottom-up procedures using a liquid interface technique. The thin film was thermally annealed at 700 °C to produce a N-doped ultrathin carbon film, fullerphene, with a hierarchical micro/mesoporous surface structure.

The N doping of fullerphene was achieved employing by pyrrole and quaternary nitrogen atoms. This allowed selective and repeated adsorption/desorption of low molecular weight carboxylic acid vapors through noncovalent interactions. The N-doped ultrafine porous nanostructure showed a better sensitivity to formic acid vapors than other common low molecular weight carboxylic acid molecules. The sensitivity of fullerphene to formic acid over acetic acid in the gas phase was much better, indicating that the new 2D fullerphene could discriminate carboxylic acids at the C-single-atom level. The hierarchical micro- and mesoporous structure of fullerphene C_{60} (half-pore width ≈ 0.27 nm; mesopore diameter ≈ 3.67 nm) provided highly efficient entry pathways for the diffusion of formic acid with a maximum molecular dimension of 0.28 nm. In addition, because of the low entropy loss, the spatially confined formic acid molecules enhanced intermolecular interactions, resulting in a highly cooperative adsorption phenomenon. The pore structure with C-single-atom molecular discrimination ability of the fullerphene film may be an attractive platform for selective immobilization of single-atom catalysts and single-atom-decorated nanorobots.

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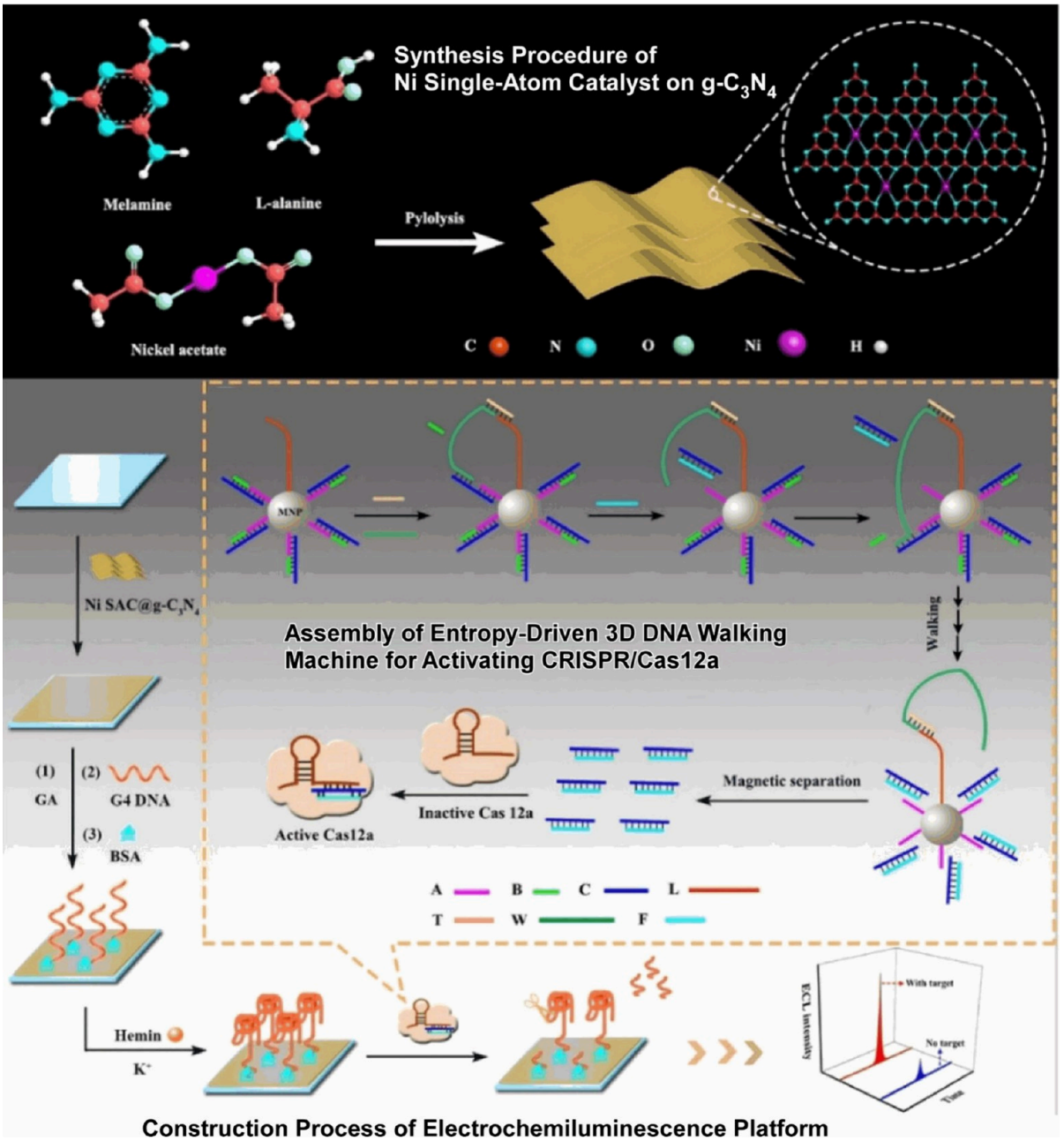

Figure 14. Implementation of Ni-single-atom catalyst to a g-C_3N_4−H_2O_2 electrochemiluminescence system together with an entropy-driven DNA walking machine-assisted CRISPR-Cas12a amplification strategy. Reproduced with permission from ref 151. Copyright 2023 American Chemical Society.

Single-Atom-Level Carbon Framework Modulation. The effect of single atoms in the catalytic function is not only limited to metal atoms, as in traditional single-atom catalysts; it can also be related to the structural changes in the modifications of a carbon atom in the carbon skeleton, which can also control the catalytic activity. The spin state is changed by deliberately changing the normal hexagonal structure of the skeleton of carbon materials to a pentagonal structure. This can improve catalytic activity in the ORR and other reactions. The interaction between electron spin and oxygen molecules in nonplatinum catalysts, especially carbon catalysts, greatly affects the catalytic performance of the ORR. The introduction of a five-membered ring structure with spin into graphitic carbon is a promising approach to develop high performance catalysts.

Chen, Nakamura, and co-workers successfully synthesized a cage-like cubic carbon catalyst rich in pentagonal structures using C_{60} precursors with NaCl template.[153] As shown in Figure 16, N doping caused structure deformation during pyrolysis, resulting in pentagonal defects. N doping is an important key to create a high density of active pentagonal sites. As the pentagon frameworks in the structure become rich, the number of electron spins increases. Accordingly, the catalytic activity of the ORR has been improved. The high concentration of pentagons results in a unique spin configuration, which further influences the catalytic ability and electronic properties of the material. The electron spin resonance spectra showed signal broadening in the samples, indicating the presence of spin. Signal broadening occurs when radicals are not uniformly distributed in the bulk.

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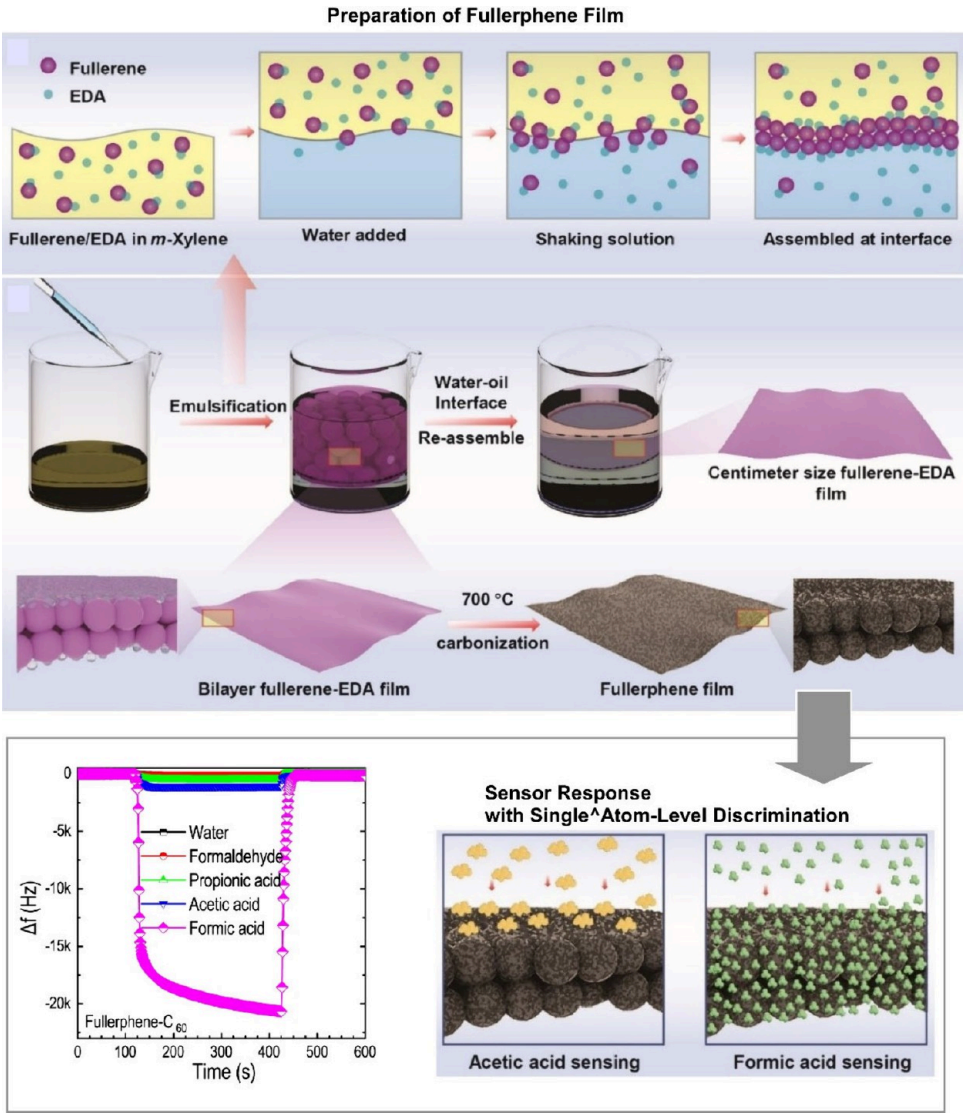

Figure 15. Bottom-up preparation of molecularly thin N-doped 2D fullerphene with C-single-atom molecular discrimination ability for formic acid detection. Reproduced with permission from ref 152. Copyright 2022 Wiley-VCH.

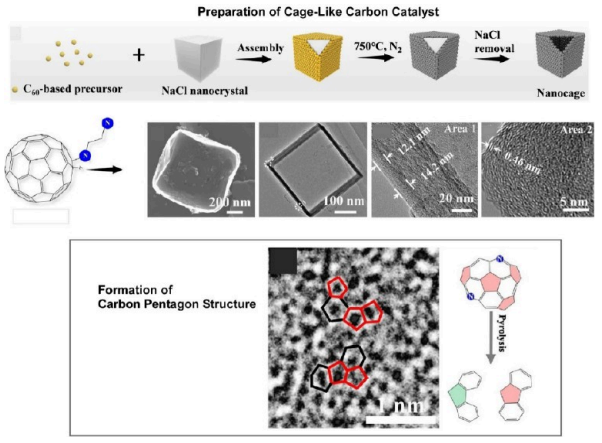

Figure 16. Preparation of a cage-like cubic carbon catalyst rich in pentagonal structures using C_{60} precursors with NaCl template. Reproduced under terms of the CC-BY license from ref 153. Copyright 2024 Wiley-VCH.

This is due to the adsorption of O_2, which occupies the spin sites, causing a shift and broadening of the peak. The cause of the paramagnetic term is found to be the movement of electrons around the double bonds adjacent to the vertices of the pentagon.

The increased activity of the ORR in acidic environments was attributed to the increased number of reactive sites in the pentagons. The high density of pentagons increased the spin density, highlighting the important role of spin−structure interactions in electrocatalytic processes. Both the experimental results and the DFT calculations concluded that the origin of the ORR activity of the pentagon-containing carbon catalysts was the spins inherently present in the pentagonal rings. The calculated free energy profile for the ORR in the pentagon-containing carbon catalysts, assuming a four-electron mechanism, showed an overall downward slope, suggesting that the ORR took place. The local spin density of the pentagons played an important role in the adsorption of O_2, which is the first step of the ORR. The overall results suggested that the local spin density of the pentagons inherent in the pentagonal ring represented one of the important factors in the activity of the

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ACS Sustainable Chem. Eng. 2025, 13, 6900−6917

ORR. Although this is not a typical single-atom catalyst, it is noteworthy that structural control at the single-atom-level contributes significantly to the control of catalytic activity.

FUTURE PERSPECTIVE

Single-atom-decorated nano/microrobots operate using the reaction over the single-atom catalysts as the driving force of their propulsion mechanisms or as an advanced functionality enabling a specific task accomplishment. Single-atom catalysts, in which the particles making up the active metal surface are reduced to the single-atom level, can maximize the efficiency of active site utilization. They are excellent in terms of catalytic activity, selectivity, and cost performance. Examples of applications in a variety of fields, including energy conversion, environmental issues, and biomedicine, have been demonstrated as shown above. There are also examples of single-atom nanozymes with enzyme-like functions and sensor applications that use them. Examples of the creation of structures with molecular recognition capabilities at the single-atom level have also been given. The discrimination and modification of single-atom-level structure brings new ideas in the design of the functionality of nano/microrobots. There are many potential applications, but more work is needed to put transfer into practice. Not limited to the presented example, huge knowledge and facts have been continuously gathered in single-atom catalysts,[154−158] nano/microrobots,[159−162] and related applications.[163−166] They also have unavoidable contributions to determine future directions. In the final section, we point out future necessity for further developments of single-atom-decorated nano/microrobots.

In future directions, the most promising use of single-atom-decorated nano/microrobots would be their applications in the field of catalysis and medical applications. In these fields, size really matters, and downsizing increases efficiency in various procedures and treatments. Not limited to these applications, several improvements and considerations on much more precise structure controls are indispensable for realistic practical usages for single-atom-decorated nano/microrobots. As shown in the last section, modifications of the structure on the level of single atoms can be projected in the nano/microrobotic technologies. Technical advancements in visualizations of single-atom-decorated nano/microrobots are also necessary. So far, expensive and elaborate methods, such as transmission electron microscopy, XPS, and synchrotron, have been used for the characterization. If the fabrication of single-atom-decorated nano/microrobots relies on these characterization methods, the practical applications from viewpoints of speeds of material developments are limited. Exploring more instantaneous and easier visualization under practical conditions would help to speed up the development processes. More considerations on stability are also necessary. In particular, under harsh conditions such as light irradiation and electric current exposure, single atoms tend to aggregate on the surface of the substrate into clusters. Strategies for stabilizations against these negative behaviors are necessary for realistic practical usages of single-atom-decorated nano/microrobots. Scalability and cost-effectiveness are also important issues for practical applications. The control of the single-atom growth is very delicate and often performed on a small scale. Moreover, techniques such as atomic layer deposition are not very scalable. The development of a technology that satisfies the two seemingly contradictory conditions of atomically precise synthesis and mass production is crucial in the next stage of the development. Indeed, such problems are realized everywhere in many research targets not limited to single-atom catalyst preparation and nano/microrobot operations. Sharing problems and solutions with the wide range of science and technology would be a practical strategy for the above-mentioned problems.

In addition, the synergy between the structure and application must be systematically investigated. Theoretical calculations with the assistance of artificial intelligence could help to understand the synergy between the structure and the functionality. In particular, when the further development of single-atom-decorated nano/microrobots was considered, the coexistence of multiple reactions could be considered to develop dynamic systems for sequential reactions. Such designs will lead to systems in which continuous material transformations, as in living organisms, are reflected in robotic functions, and to nano/microrobotic systems in which multiple systems exhibit logical behavior in response to multiple inputs. With these developments, nano/microrobots will become dynamic intelligent functional systems. Of course, designing and building such complex systems may not be easy. In such cases, the use of artificial intelligence will be beneficial. The use of machine learning in chemical and materials research is widespread.[167−170] The fusion of materials informatics and nanoarchitecture has also been proposed.[171,172] Similarly, it is expected that intelligent single-atom-decorated nano/microrobots will be developed through the design using artificial intelligence. As can be seen from the research history of single-atom-decorated nano/microrobots, mankind has overcome various difficult problems to achieve this attractive goal. One more effort with new technology would achieve a more fruitful goal with realistic practical applications.

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Author Contributions

The manuscript was written through contributions of both the authors. All authors have given approval to the final version of the manuscript.

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Notes

The authors declare no competing financial interest.

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Katsuhiko Ariga received his Ph.D. degree from the Tokyo Institute of Technology in 1990. He joined the NIMS in 2004 and is currently the leader of the Supermolecules Group and senior scientist with special missions of Research Centre for Materials Nanoarchitectonics, NIMS. He is also appointed as a professor at The University of Tokyo.

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