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Innovative and sustainable approaches to gold nanoparticle synthesis: beyond the Turkevich method

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ABSTRACT

Gold nanoparticles (AuNPs) have attracted considerable interest due to their unique physicochemical properties and potential applications in diverse fields such as optoelectronics and biomedicine. The conventional Turkevich method, while simple, has limitations in terms of nanoparticle size and shape control, scalability, and energy efficiency. Alternative synthesis methods include plant-mediated, microorganism-mediated, microwave-assisted, photochemical, and electrochemical approaches. Each of these methods has its own advantages, including eco-friendliness, scalability, speed, and precision. However, each method also presents its own challenges and requires process optimization to fully replace existing methods. This article reviews these synthesis methods for AuNPs with a focus on recent sustainable approaches. The potential for sustainable synthesis of AuNPs to serve various industries and applications lies in the collaborative efforts of researchers across different disciplines.

KEYWORDS

Gold nanoparticles; Synthesis; Sustainable; Green synthesis; Turkevich

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Introduction

Gold nanoparticles (AuNPs) possess unique optical and hemical properties such as tunable reactivity, colorimetric chemistry, and high surface area [1]. Their surface plasmonic characteristics are governed by factors such as size, shape, and composition [2]. Precisely modulating these parameters is critical for specific applications, particularly in the field of optoelectronics, where they effectively harness the visible and near-infrared regions of the electromagnetic spectrum. The properties of AuNPs are influenced by factors such as nanoparticle stabilization, surface functionality, interparticle spacing, size, and structure [3]. A range of synthesis techniques, including chemical reduction, sonochemical reduction, photochemical processes, mechanical approaches, and environmentally friendly extraction methods, can be utilized to manipulate the attributes of gold nanoparticles. These methods offer distinct advantages and challenges in terms of precision, scalability, and eco-friendliness. Researchers are continually working to optimize these processes to achieve the desired properties of gold nanoparticles [4]. The selection of the appropriate synthesis method is pivotal in attaining specific physicochemical and optoelectronic properties. The Turkevich method is one of the pioneering methods for AuNP synthesis and has been in practice since its discovery [5]. However, this method has several limitations, and there is a need for more efficient and eco-friendly substitutive methods. This article explores recent approaches that are being selectively used to synthesize AuNPs.

Turkevich Method and its Limitations

The Turkevich method, developed by John Turkevich in the 1950s, is one of the earliest and most commonly used approaches for synthesizing AuNPs [6]. This method is relatively straightforward and relies on reducing gold ions in solution to form AuNPs. In this method, a solution of a gold salt, typically chloroauric acid ($HAuCl_4$), is heated until it reaches its boiling point. A reducing agent, often sodium citrate, is added as the solution boils. This reduces the gold ions (Au^{3+}) to form metallic gold (Au^0) nanoparticles. The nanoparticles typically have a spherical shape and a size that can be controlled by varying the relative concentration of the gold salt and the reducing agent. The Turkevich method has several advantages contributing to its long-standing use in AuNP synthesis. It is a relatively simple and cost-effective technique, making it accessible for researchers with limited resources. Since its discovery, the method has been modified in a vast number of ways to accommodate the desired AuNPs [7].

The Turkevich method also has several limitations [8]. One of its primary drawbacks is the difficulty of precisely controlling the size and shape of the resulting nanoparticles. The method can produce nanoparticles with a relatively narrow size distribution, but controlling the exact size and shape of the nanoparticles can be challenging. The size of the nanoparticles produced is typically around 12 nm, but achieving sizes significantly smaller or larger than this can be difficult. While the Turkevich method is straightforward and reliable, it may not be easily scalable for large-scale production of gold nanoparticles. It requires elevated temperatures, which can lead to higher energy consumption compared to other methods. Furthermore, the mechanism of the Turkevich method involves two consecutive reduction steps ($Au^{3+} \rightarrow Au^+ \rightarrow Au^0$) rather than a reduction followed by the disproportionation reaction as conventionally believed. This complexity can make the process more difficult to control and optimize. There can be variability from batch to batch, which can affect the reproducibility of the synthesis.

Researchers are actively exploring alternative methods and engaged in the optimization of existing ones to address

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the limitations of the Turkevich method. For example, proposed modifications to the Turkevich method involve the use of a pre-passivated precursor, which offers improved control over the initial reaction stage. However, these modifications result in a slight increase in reaction time. In response to these challenges, more sustainable approaches to AuNP synthesis are being sought that aim to reduce energy consumption, minimize the use of harmful chemicals, and enhance the eco-friendliness of AuNP production. The following sections explore innovative and sustainable methods and their potential to address the challenges associated with traditional synthesis techniques.

Plant-Mediated Synthesis

Plants contain various natural compounds like alkaloids, flavonoids, and tannins, which act as both reducing and stabilizing agents. The biosynthesis of AuNPs from plant sources is a straightforward one-pot process. It involves washing and preparing different plant parts (leaves, fruits, bark, etc.), boiling them in water to extract bioactive compounds, purifying the extract, and mixing it with a gold salt solution. The gold salt is then reduced into AuNPs, which can take just minutes to a few hours. The reaction progress is monitored by a color change, and the synthesized AuNPs are purified for further use. This eco-friendly process can be easily scaled up. The use of different plant parts leads to variations in the size and shape of the nanoparticles. Several studies have successfully used plant extracts for AuNPs synthesis. For example, extracts of Salix alba [9], coriander [10], Camellia Sinensis [11], Curcumae [12], Amaranthus [13], Aloe [14], Limonia [15] and many other plants have been reported recently for the synthesis of AuNPs. The synthesis process is generally affected by factors like temperature and pH, which influence the size and shape of AuNPs. Bogireddy et al. showed that sundried Coffea arabica seed extract produced tunable AuNPs based on pH [16]. Similarly, Salvia officinalis extracts led to small and large AuNPs in different pH conditions due to the differential activity of its polyphenols at different pH [17].

The synthesized AuNPs have shown promise in various applications. Many phytogenic AuNPs show antibacterial properties [18]. AuNPs from different plant extracts also exhibit biocompatibility and stability [19,20]. The use of plant extracts for AuNPs synthesis is a green and versatile method with potential applications in biomedicine and beyond. The choice of plant source and control of synthesis conditions can yield AuNPs with tailored properties for specific applications.

Microorganism-Mediated Synthesis

Microorganism-mediated synthesis involves using microorganisms such as bacteria, fungi, and yeast to produce AuNPs. Like plant extract, microbes and microbial extract serve as reducing and stabilizing agents, offering a sustainable and biocompatible approach. Certain strains of bacteria, like Escherichia coli, can intracellularly or extracellularly reduce Au^{3+} ions to AuNPs [21]. Similarly, fungal and yeast species, including Candida [22] and Fusarium oxysporum, have also shown potential for AuNP synthesis [23]. Their secreted enzymes and metabolites can reduce gold ions and produce stable AuNPs. Microbes can synthesize gold AuNPs intracellularly and extracellularly [24]. In intracellular synthesis, microbes actively remove gold ions from their surroundings and enzymatically reduce them to form AuNPs within their cellular compartments. This process often results in the production of uniformly sized and highly stable nanoparticles. On the other hand, extracellular synthesis occurs when microbes secrete enzymes or metabolites that mediate the reduction of gold ions in their immediate environment, forming AuNPs outside their cells. This extracellular method is advantageous for large-scale production and can be more easily controlled.

Microorganism-mediated synthesis is advantageous for its scalability and ability to synthesize AuNPs under mild conditions. These green synthesis methods represent a promising shift towards more sustainable AuNP production. They minimize the need for harsh chemicals, reduce energy consumption, and offer opportunities for tailoring the properties of AuNPs for specific applications. It is essential to explore these green methods further and address challenges related to scalability, standardization, and application-specific requirements to realize their potential in diverse fields.

Microwave-Assisted Synthesis

Microwave-assisted synthesis is a rapidly growing and innovative approach to AuNP synthesis [25]. Microwave radiation initiates and accelerates the chemical reactions involved in AuNP synthesis. The microwave energy generates localized heating within the reaction mixture, leading to rapid nucleation and growth of nanoparticles. Arshi et al. showed a simple method for microwave-synthesizing AuNPs using citric acid and CTAB. They demonstrated the effect of irradiation time on the types of nanoparticles produced [26]. In another study, Rawat et al. synthesized tyrosine-protected AuNPs in a one-step process using microwave irradiation [27]. Bayazit et al. explored a novel approach for the continuous and reproducible synthesis of AuNPs in aqueous solutions by combining single-mode microwave irradiation with microflow chemistry, allowing for precise nanoparticle size control [28].

One of the primary advantages of microwave-assisted synthesis is its speed [29]. Reactions that might take hours or even days using conventional methods can be completed in minutes or seconds with microwave irradiation. This is particularly advantageous in industrial and research settings where time efficiency is crucial. Microwave irradiation allows precise control over the size and morphology of the AuNPs. By adjusting parameters like the microwave power, reaction time, and concentration of reactants, researchers can tailor the characteristics of the synthesized nanoparticles to meet specific application requirements. This level of control is essential in fields like catalysis and nanomedicine. The microwave-assisted method shows high energy efficiency. It minimizes heat loss to the surroundings, reducing energy consumption compared to traditional heating methods. This aligns well with sustainability goals and contributes to a reduced environmental footprint. The method is adaptable to various scales, from small laboratory batches to large-scale industrial production, making it versatile. It can be easily integrated into existing manufacturing processes, paving the way for the commercial production of AuNPs with minimal adaptation.

Photochemical Synthesis

TIn this approach, gold precursors in solution are exposed to visible or ultraviolet (UV) light, reducing gold ions and subsequent AuNP formation [30]. Unlike traditional chemical

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methods, photochemical synthesis avoids toxic compounds, expensive equipment, and specialized personnel, allowing it to be carried out at room temperature and atmospheric pressure. To achieve controlled AuNP synthesis, stabilizers and capping agents are crucial. Various polymers, such as poly (vinyl pyrrolidone) (PVP), polymethacrylate (PMA), and natural rubber latex, have been studied for their impact on AuNP characteristics [31].

UV light, with its distinct wavelength ranges (UV-A, UV-B, and UV-C), is an energy source in the photochemical synthesis process [32]. Several factors, including pH, precursor concentration, and exposure time, influence AuNP size and shape during UV photochemical synthesis [33]. The pH of the reaction medium plays a critical role, with basic conditions favoring the formation of nanospheres and acidic conditions leading to anisotropic AuNP shapes like nanorods. Precursor concentration, such as tetrachloroauric acid ($HAuCl_4$), affects AuNP morphology, with higher concentrations promoting spherical nanoparticles. Stabilizing and reducing agents, like citric acid and polyethyleneimine (PEI), influence NP size.

Increasing their concentration results in smaller, more uniform NPs. Surfactants, which also act as stabilizers, are essential in AuNP synthesis. Cationic surfactants favor the formation of larger nanoparticles, and anionic surfactants produce smaller ones.

Electrochemical synthesis

In electrochemical synthesis, an electrode immersed in a solution containing gold ions serves as both the source of electrons for reducing gold ions and the substrate on which AuNPs nucleate and grow. Gold electrodeposition techniques on substrates such as indium tin oxide (ITO), silicon, glassy carbon, carbon nanotubes (CNTs), and graphene have been reported in the literature [34]. Glassy carbon electrodes are an excellent platform for studying the Volmer-Weber growth mode during gold electrodeposition [35]. In contrast, carbon nanotubes provide a unique substrate-enhanced electroless deposition method for supporting the nanoparticles [36]. These approaches allow for precise control over AuNP size and distribution and have shown promise in the development of electrochemical sensors. The self-assembly of AuNPs on graphene sheets allows electrostatic interactions, enabling the loading of AuNPs with high precision [37]. High-density AuNP arrays can be synthesized on silicon substrates through electrochemical deposition, using high overpotentials to promote nucleation [38]. These composite materials exhibit exceptional electrocatalytic activity and have found applications in various sensing technologies.

Electrochemical synthesis enables precise control over AuNP size and morphology. The properties of the synthesized nanoparticles can be controlled by adjusting parameters such as the electrode potential, current density, and reaction time [39]. Furthermore, the electrode surface can be modified with various molecules, such as surfactants or ligands, to impart specific surface functionalities to the synthesized AuNPs [40]. This method also offers the advantage of real-time monitoring and control over the synthesis process [41].

Challenges and Future Perspectives

TWhile innovative and sustainable approaches to AuNP synthesis offer promising solutions to many of the limitations associated with traditional methods, several challenges still need to be addressed. Green synthesis methods hold great promise for sustainable AuNP production but are limited by scaling up for industrial applications. This method makes it difficult to consistently attain high-quality AuNPs while minimizing resource consumption and environmental impact [42]. Microwave-assisted synthesis, while efficient, struggles with ensuring reproducible outcomes across diverse microwave systems and settings. In contrast, photochemical synthesis is constrained by its reliance on specific light wavelengths, which limit the range of AuNP properties that can be obtained. Meanwhile, electrochemical synthesis offers fine-tuned control over AuNP characteristics but faces hurdles in achieving reproducibility across varying electrochemical setups and conditions. The different methods discussed in this article have been summarized in Table 1 regarding their convenience, sustainability, advantages, and disadvantages.

Optimizing scalability for green synthesis techniques, incorporating standardized protocols, and utilizing bioreactor-based production systems to bridge the gap between laboratory-scale experiments and large-scale manufacturing is necessary. Identifying suitable plant species or microorganisms with enhanced AuNP synthesis capabilities and customizing their growth conditions can enhance production efficiency and reliability. Likewise, safety protocols and containment measures for microwave-assisted synthesis should be a priority, alongside standardizing microwave parameters and creating user-friendly microwave reactors for nanoparticle synthesis. Collaborative efforts between chemists and engineers can result in safer and more precise microwave-assisted AuNP synthesis methods. Exploring innovative photosensitizers and capping agents to enable photochemical synthesis at diverse wavelengths, including infrared and terahertz regions, can unlock new possibilities for tailoring AuNP properties. While electrochemical synthesis offers precise control over AuNP characteristics, standardization is vital for widespread adoption. Thus, developing standardized electrochemical cells and protocols, alongside real-time monitoring and feedback systems, can enhance control and consistency. Exploring novel electrode materials and modifications can further expand the range of achievable AuNP properties.

Table 1. Summary of different methods of AuNP synthesis.

Method	Convenience	Sustainability	Time Consumption	Advantages	Disadvantages
Turkevich Method	High	Low	Moderate	Simple procedure, Controlled size, and shape, Can be scaled up	Requires toxic chemicals, Limited control over size, Generates hazardous waste

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Plant-Mediated Method	Moderate	High	Moderate	Uses plant extracts, Eco-friendly, Biocompatible	Limited control over size, Slower reaction kinetics, and variability due to plant source
Microorganism-Mediated Method	Moderate	High	Moderate	Biologically synthesized, Eco-friendly	Limited control over size, Longer reaction times, Contamination risks
Electrochemical Method	Low	Moderate	Low to High	Precise control of size, High purity, Scalable	Requires specialized equipment, Energy-intensive
Microwave-Assisted Method	High	Moderate	Low to Moderate	Rapid heating and synthesis, Energy-efficient, Controlled size and shape	Equipment may be expensive, and some reactions require solvents
Photochemical Method	Low to Moderate	High	Moderate to High	Precise control of size, Green and sustainable, High purity	Limited to specific materials, Requires UV light source, May need specialized equipment

Conclusions

The conventional Turkevich methodology has been instrumental in initiating the era of AuNPs. Nevertheless, its inherent limitations and energy-intensive nature have led to the emergence of alternative strategies. Plant-mediated synthesis stands out as an eco-friendly approach that allows for biocompatible AuNP production. Meanwhile, microorganism-mediated synthesis demonstrates the remarkable capacity of microbial organisms to generate AuNPs, both extracellularly and intracellularly, promising scalability and compatibility with biological systems. Microwave-assisted synthesis process ensures rapidity and precision, thereby reducing energy consumption and aligning with sustainability objectives. In parallel, photochemical synthesis leverages light to initiate AuNP formation, albeit with ongoing challenges regarding the expansion of usable wavelength ranges. Electrochemical synthesis, on the other hand, provides a means for meticulous control over AuNP properties but necessitates standardized protocols and real-time monitoring for broad adoption. The prospective evolution of AuNP synthesis depends on scalability, protocol standardization, and the advancement of materials and technologies. The critical objective is bridging the gap between laboratory-scale experimentation and large-scale production in these synthesis techniques.

Disclosure statement

No potential conflict of interest was reported by the author.

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