



Interactions of zeatin with gold ions and biomimetic formation of gold complexes and nanoparticles

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ABSTRACT

We report here a simple one-pot synthesis for the preparation of gold nanoparticles biomimetically using zeatin nanostructures. Zeatin, a plant phytohormone was self-assembled into nanospheres. Those nanospheres transformed into nanoribbons over a period of time upon formation of zeatin–gold (III) complexes in the presence of hydrogen tetrachloroaurate. Further, upon heating, gold nanoparticles were formed due to mineralization in the presence of zeatin nanofibers. The effect of pH on the self-assembly of zeatin and the formation of gold nanoparticles, was investigated. We also compared the preparation of gold nanoparticles in the presence of zeatin nanoribbons, using a known reducing agent such as hydrazine, which resulted in loss of morphology control and alignment of the gold nanoparticles. Thus zeatin nanoribbons act as templates which allow for size as well as alignment control for the gold nanoparticles. The materials obtained were analyzed using FTIR, absorbance spectroscopy as well as by transmission electron microscopy, EDX, SEM and AFM. The method involved here is a mild, green-synthetic process, which could be used for facile preparation of morphology controlled gold nanoparticles and may open up new avenues for device fabrications for a wide range of applications, particularly in optoelectronics and sensors.

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1. Introduction

Hybrid organic–inorganic nanocomposite materials are becoming increasingly significant scientifically and technologically due to their wide range of applications, which include optoelectronics, magnetic materials, solid electrolytes, catalysis, sensors, molecular adsorption and separation science [1–7]. Several synthetic approaches have been used to prepare such nanocomposites, in order to enhance the physical properties of metal nanoparticles including stabilization of nanoparticles with micelles, polymers, and organic ligands [8,9]. For example, gold nanoparticles have been incorporated into polymer matrices, organic gels and dendrimers [10,11]. It was recently found that poly(ethyleneoxide diamine) can form nanoparticles directly under mild conditions within the polymer matrix, allowing for the efficient deposition of size-controlled nanoparticles, by implementing both the water-soluble polymer matrix and the well-developed thin polymer layer deposition technology [12]. In those studies, the supramolecular structure formed often acts as a template for the organization of nanoparticles [13]. For instance, polyvinyl pyrrolidone matrices (PVP) have been used as templates for the preparation of shape controlled silver nanocubes and gold nanoboxes [14].

Recently, biomineralization methods have been gaining considerable importance due to their efficiency in controlling the morphologies of the nanoparticles [15–19]. Using biomimetic methods to prepare nanoparticles is more advantageous because it is mostly grown under mild conditions of temperature, pH and pressure, requiring a less energy intensive process and without the use of harsh chemicals. There are various microbial systems that could successfully synthesize nanoparticles including bacteria that have been known to extracellularly synthesize iron-based magnetic nanoparticles to form iron oxide for applications in magnetic recording and magnetic storage devices, ferrofluids, or contrast enhancers for MRI [20,21]. Biomineralization using bacteria, fungus and actinomycete species has also been carried out for the preparation of morphology controlled gold and silver nanoparticles [22]. Researchers have also applied enzymes as nanoreactors, and other peptide-based synthetic strategies for the synthesis of nanoparticles in which biological interactions control the nucleation of nanoparticles [23]. Virus particle assemblies from capsid subunits and nucleic acid aggregates have also been used to incorporate gold nanoparticles within an icosahedral virus in order to render single viruses optically detectable to further study self-assembly pathways and structural dynamics of the virus capsid [24].

It was also found that gold nanoparticles can be formed in plant tissues [25]. For example alfalfa biomass upon binding to gold (III) ions formed gold nanoparticles, with uniform particle size distribution [26]. In another study, when tetrachloroaurate solutions

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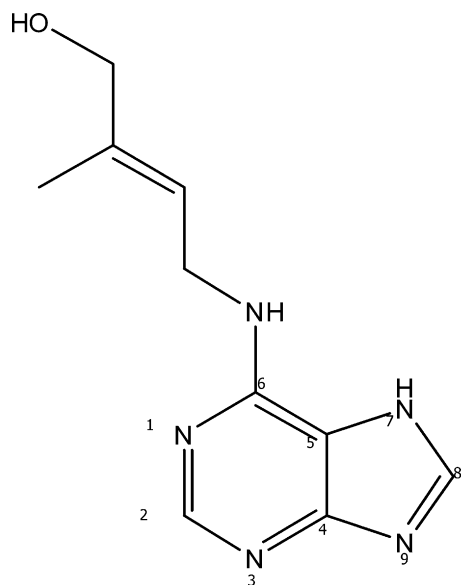


Fig. 1. Chemical structure of trans-zeatin.

were incubated with *Sesbania drummondii* seedlings, gold nanoparticles were formed [27]. Furthermore, those nanoparticles bearing biomatrices could directly reduce toxic pollutants. Several other plants have also been successfully used for efficient extracellular synthesis of silver and gold nanoparticles including leaf extracts of henna, geranium (*Pelargonium graveolens*), lemongrass (*Cymbopogon flexuosus*), Cinnamomum camphora, neem (*Azadirachta indica*), Aloe vera, tamarind (*Tamarindus indica*) fruit extract of *Emblca officinalis*, biomasses of wheat (*Triticum aestivum*) and oat (*Avena sativa*), alfalfa (*Medicago sativa*), native and chemically modified hop biomass and remnant water collected from soaked Bengal gram bean (*Cicer arietinum*) [28–38]. Researchers have also successfully conducted the biosynthesis of the akaganeite nanocrystals using *Acidithiobacillus ferrooxidans* cells [39]. It was found that plant cell walls also can be efficiently utilized as templates for the growth of ZnO nanostructures [40]. Thus, various plant systems can provide a matrix for the formation of nanoparticles. In this work, we explored the plausibility of formation of gold nanoparticles using the plant cytokinin zeatin. In general, plant cytokinins regulate cell division and tissue differentiation as well as promote growth and play a role in controlling plant development and aging [41]. However, to our knowledge, the possibility of using plant cytokinins to form nanoparticles has not yet been explored.

Trans-zeatin, is a plant phytohormone, which was the first plant cytokinin to be isolated in a state of purity [42,43]. As shown in Fig. 1, the chemical structure of zeatin consists of a purine ring system connected to an aliphatic 2-methyl butenol group. The allylic hydroxyl group in zeatin is considered a structural feature associated with high cytokinin activity [42].

Because of the presence of the basic purine system similar to adenine, as well as the aliphatic butene moiety terminated by a hydroxyl group, we hypothesized that zeatin may be capable of self-assembly due to π – π stacking interactions as well as hydrogen bonding and intra and intermolecular interactions. We therefore, studied the self-assembly of zeatin under aqueous conditions at varying pH to investigate the formation of assemblies. Further, we explored the efficiency of formation of gold nanoparticles using the self-assembled zeatin nanostructures as templates. Samples were analyzed using absorbance spectroscopy, atomic force microscopy, transmission electron microscopy, scanning electron microscopy and FTIR spectroscopy. The scheme for preparation of gold nanoparticles in the presence of self-assembled zeatin nanostructures is shown in Fig. 2. This simple biomimetic method allows for the synthesis of morphology controlled growth of gold nanoparticles in an entirely “green” and cost-effective process. Further, the investigation of organized nanostructures of plant phytohormones such as zeatin, with structural resemblance to nucleic acid bases such as adenine might help in further comprehending the assembly mechanism of higher organized biological structures.

2. Experimental

2.1. Materials

Trans-zeatin hydrochloride (min 97%), and potassium bromide (99+%, metals basis), gold chloride hydrate (~50% Au) and hydrazine hydrate were purchased from Sigma–Aldrich. Buffer solutions of various pH values were purchased from Fisher Scientific. All chemicals were used as received.

2.2. Self-assembly of trans-zeatin

Trans-zeatin was self-assembled in aqueous solutions over a three-week period. Solutions of trans-zeatin (1 mM) were prepared in buffer solutions at a pH range of 2 through 10. The solutions of the self-assembled zeatin were washed and centrifuged twice at 4000 rpm for two hours before analysis.

2.3. Formation of gold complexes with self-assembled zeatin nanostructures

Tetrachloroaurate (200 μ L, 0.1 mM) was added to 200 μ L of the self-assembled zeatin nanostructures at a pH range of 4 through 8. The samples were agitated slowly over a period of two weeks in a shaker. The samples were then washed twice with distilled water and centrifuged at 4000 rpm for two hours before analysis.

2.4. Formation of gold nanoparticles using self-assembled zeatin nanostructures as templates

In some cases, the zeatin nanostructures at various pH were heated after incubation with the tetrachloroaurate solution for an

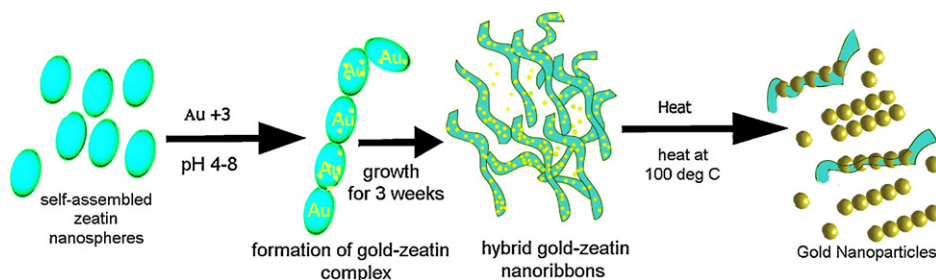


Fig. 2. Scheme for formation of hybrid zeatin–gold nanoribbons and subsequent formation of gold nanoparticles.

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