

## Green Synthesis of Gold Nanoparticles Using *Solanus Lycopersicum* (TOMATO) Aqueous Extract

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**Abstract:** In the present study we explore the reducing and capping potential of aqueous extract from tomato juice for the synthesis of gold nanoparticles. The extract with different concentration reduced with  $\text{HAuCl}_4$  aqueous solution at room temperature. The color change, pH change and UV-visible spectroscopic analysis reveal the Surface Plasmon Resonance (SPR) of the final reaction product which confirms the reduction of  $\text{Au}^{3+}$  ion to gold nanoparticles. XRD, particle size analysis results represent strong reducing potential of tomato aqueous extract which can also be tested in the green synthesis of other metallic nanoparticles.

**Key words:** Aqueous extract • Gold nanoparticles • Surface Plasmon Resonance (SPR) • Capping, Tomato

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### INTRODUCTION

In recent decades there is growing interest in the synthesis of metal nanoparticles due to their unique properties, which are significantly different from the behavior of the respective bulk material (H. Xiaohua, I. H. El-Sayed 2006 [1], M. C. Daniel and D. Astruc 2004) [2],. Gold nanoparticles (Au NPs) have wide attraction because of their electronic, biosensing, plasmonic, photonics, catalysis, bio-medical and surface-enhanced Raman scattering (SERS) properties (V. Tjoa, W. Jun, V. Dravid 2011 [3], L. Dykmana and N. Khlebtsov 2012 [4], L. Rodriguez-Lorenzo 2012 [5], S. Y. Li and M. Wang 2012 [6], S. Kapoor, R. Joshi 2002 [7]). Most of the chemical methods reported in the literature for the synthesis of Au NPs often involve use of toxic reducing agents (such as sodium borohydride, hydrazine, etc.) and harsh reaction parameters like high temperatures in the polyol method (S. Kapoor, R. Joshi 2002. [8], M. Vaseem, K. M. Lee 2010 [9], K. S. N. Kamarudin 2010 [10]). In our earlier reports, synthesis of gold nanoparticles have been shown by the reduction of aqueous  $\text{AuCl}_4$  ions using extracts from *Embllica officinalis* (Indian Gooseberry) fruit (Ankamwar B, Damle C 2005 [11]), and *Tamarindus indica* (Ankamwar B, Chaudhary M 2005 [12]). leaf. Recently, we

had demonstrated the biological synthesis of triangular gold nanoprisms by a single step, room temperature reduction of aqueous chloroaurate ions by the extract of the plant, lemongrass (Shiv Shankar S, Rai A 2004 [13]). There is still much scope for improvement in bio-based methods for metal nanoparticle synthesis, particularly in relation to improving the monodispersity of the nanoparticles and modulating their size and shape, as well as in reducing the time required for nanoparticle synthesis. On a more fundamental level, it would be interesting to study the nature of nanoparticles formed using extracts from different parts of a plant.

The tomato is the edible, often red fruit/berry of the nightshade *Solanum lycopersicum*, commonly known as a tomato plant. The species originated in the South American Andes<sup>1</sup> and its use as a food originated in Mexico and spread throughout the world following the Spanish colonization of the Americas. Its many varieties are now widely grown, sometimes in greenhouses in cooler climates. The tomato is consumed in diverse ways, including raw, as an ingredient in many dishes, sauces, salads and drinks. While it is botanically a fruit, it is considered a vegetable for culinary purposes (as well as under U.S. customs regulations, see *Nix v. Hedden*), which has caused some confusion. The fruit is rich in

lycopene, which may have beneficial health effects. The tomato belongs to the nightshade family, Solanaceae. The plants typically grow to 1-3 meters (3-10 ft) in height and have a weak stem that often sprawls over the ground and vines over other plants. It is a perennial in its native habitat, although often grown outdoors in temperate climates as an annual. An average common tomato weighs approximately 100 grams (4 oz).

**Gold Nanoparticles:** Elemental gold has many unique properties which have attracted and fascinated mankind since its discovery. Being very unreactive, gold does not tarnish in the atmosphere and so keeps its attractive colour forever (Hutchings *et al.* 2008) [14]. That is one of the main reasons why gold has been used in shaping jewellery. It has been used for many colourful, decorative, ceremonial and religious artifacts and has been a metal with a high monetary value. Colourful aqueous solutions of gold colloids date back to Roman times and were known to medieval alchemists as aurum potable (Mellor, 1923) [15]. A Roman cup, called the Lycurgus cup, used nanosized (ca 50 nm) gold and silver alloys, with some Cu clusters to create different colours depending on whether it was illuminated from the front or the back. The cause of this effect was not known to those who exploited it. Michael Faraday was the first to recognise that the colour was due to the minute size of the gold particles (Faraday, 1857) [16]. On February 5, 1857, Michael Faraday delivered the Bakerian Lecture of the Royal Society in London entitled "Experimental Relations of Gold (and other metals) to Light. In his speech, he mentioned that known phenomena (the nature of the ruby glass) appeared to indicate that a mere variation in the size of its particles gave rise to a variety of resultant colours. Nearly a century later, electron microscope investigations on Faraday's rubycoloured gold colloids have revealed that Faraday's fluid preparations contain particles of gold of average diameter ( $6 \pm 2$  nm) (Turkevich, 1951) [17]. Although some scientists see the Faraday's experiment as a landmark in the history of nanoscience and nanotechnology (Peter and John Meurig, 2007) the chemical inertness of gold as a bulk metal appeared to provide very little opportunities to open up new and exciting chemistries (Hutchings *et al.* 2008). The new field of nanotechnology made it possible to discover the unique properties of matter when subdivided to the nanoscale. Gold at nanoscale manifests a number of interesting physico-chemical properties that have

fascinated many disciplines of science including: material scientists, catalysts, biologists, surface and synthetic chemists and theoreticians in great number. Today, in the 21st century, gold chemistry is based on solid ground regarding the preparation and characterisation of a wide variety of fundamental compounds with gold atoms and gold clusters as core units (Murray [18], 2000, Peter, 2000 [19], Gagotsi, 2006 [20]). The fact that gold NPs have been studied in many different scientific fields has led not only to a deep understanding of many of the physico-chemical features that determine the characteristic behaviour of these nanoscale gold nanoparticles but also to invent, test and validate reliable novel procedures for the preparation, synthesis and characterisation of gold nanoparticles of basically any desired size and shape.

The bottom up process by far more common and effective (Sardar *et al.* 2009) [21] and has become a popular method in current nano-science and nanoengineering. It has a number of potentially very attractive advantages. These include experimental simplicity down to the atomic size scale, the possibility of three-dimensional assembly and the potential for inexpensive mass fabrication (Brust and Kiely, 2002) [22]. The simplest and most common bottom up method employed for the production of the gold nanoparticles of different sizes is the reduction of Au (III) salt (usually HAuCl<sub>4</sub>) by sodium citrate in water. In this method, pioneered by Turkevich and co-workers in 1951 (Turkevich, 1951) [23] and later refined by Frens in the 1970s (Frens, 1973) [24] and more recently further developed by Kumar (Kumar *et al.* 2006) [25]. It is generally accepted that the AuCl<sub>4</sub><sup>-</sup> ions are first reduced to atomic gold (Au), the concentration of which rises quickly to the supersaturation level. Collision of the Au atoms leads to a sudden burst of nuclei formation which marks the start of the nucleation step. It is the attachment and coalescence of those nuclei which results in the growth and formation of desired nanoparticles (Pong *et al.* 2007) [27]. Figure 1 illustrates the reduction, nucleation and growth steps during the formation of the nanoparticles. It shows that the reduction and nucleation are fast (>200 ms) while growth step is the rate determining step since it is much slower than the antecedent nucleation step. Many times, difficulty in controlling the nucleation and growth steps, which are intermediate stages of particle formation process may result in a broad particles size distribution (Belloni, 1996) [27]. In the presence of various reactive polymers in the

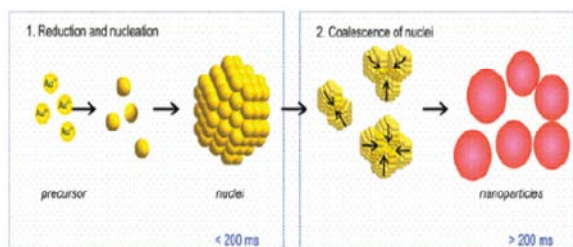


Fig. 1: Schematic illustration for the deduced process of gold nanoparticles formation, reduction and nucleation are faster process than coalescence of nuclei (Polte *et al.*, 2010). Reprinted with the permission from copying 2010 American chemical society.

reaction medium, that is, polymers having various functional groups, the growing metallic particles are stabilized by the adsorption of the polymer chains onto the surface of the growing metal fragments, thus lowering their surface energy and creating a barrier to further aggregation (King *et al.* 2003) [28].

One important factor for understanding the behaviour of the natural particles in the environment and the bioavailability of heavy metals loaded on them is their interaction with microorganisms associated with biomass population. The nanoparticles could possibly be immobilised, absorbed, reacted or retarded by biomass in the environment.

## MATERIALS AND METHODS

**Reagents and Chemicals:** Tetrachloroauric acid ( $\text{HAuCl}_4 \cdot \text{xH}_2\text{O}$ ) was obtained from Sigma Aldrich Chemicals. Freshly prepared triple distilled water was used throughout the experimental work [30-57].

**Preparation of Tomato Aqueous Extract:** In our synthesis procedure, tomato aqueous extract were used as reducing and capping agent. Extract was prepared by soaking 2 gm of tomato juice in 20 ml deionized water for overnight and crush it with mortar and pestle, the mixture was boiled for 10-15 minute at 70-80°C. The extract was followed by centrifuge for 15 minute at 5000 rpm; collected supernatant was then filtered by standard sterilized filtration method. Extract was then stored at 4°C for further use.

**Synthesis of Gold Nanoparticles:** In a typical experiment, AuNPs synthesis protocol was optimized by stirring a mixture of tomato aqueous extract at three different

Table 1: Indication of Colour change in green synthesis of Gold nanoparticles

Nanoparticle Solution	Colour change			Time
	Before	After	Colour intensity	
TOMATO	Pale Yellow	Cocoa Brown	+++	24 Hours

Table 2: Indication of change in  $\text{P}^{\text{H}}$  during green synthesis of Gold nanoparticles

Nanoparticle Solution	$\text{P}^{\text{H}}$ change			Result
	Before Reduction	After Reduction		
Tomato	3.91	2.69		+

concentrations with 1mM  $\text{HAuCl}_4$  aqueous solution (1;1, 5;1, 10;1) at 200 rpm at room temperature for 1 hour. Within a particular time change in color was observed indicating nanoparticle synthesized.

**UV-Vis Spectra Analysis:** The reduction of pure  $\text{Au}^{3+}$  to nanoparticle was monitored by measuring the UV-vis spectrum the most confirmatory tool for the detection of surface Plasmon resonance property (SPR) of AuNPs, by diluting a small aliquot of the sample in distilled water. UV- Vis spectral analysis was done by using UV-Vis spectrophotometer Systronics 118 within the range of 350-650 nm.

**X-ray Diffraction (XRD) Analysis:** XRD measurement of biologically synthesized AuNPs from tetrachloroauric acid, AuNPs solution drop-coated on glass were done on a Bruker axs- D8 Advance instrument operating at a voltage of 40 KV and current of 20 mA with  $\text{Cu K}\alpha$  radiation.

**Particle Size Analysis:** Size analysis of gold nanoparticles were carried out on Brookhaven 90 Plus Nanoparticle Size Analyzer with following measurement parameter, Refractive index fluid-1.330, Angle-15.00, Average count rate-5.2kcps with run completed 3 times.

## RESULTS AND DISCUSSION

**Image of Tomato Shown below :**



TAMATO VEGETABLE

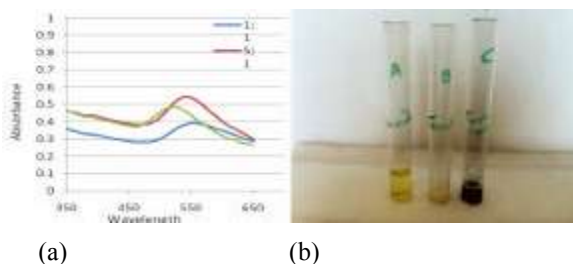


Fig. 2: A UV-Vis spectra of AuNPs synthesized by reacting different concentration of Solanum Lycopersium extract with 1Mm HAuCl<sub>4</sub> aqueous solution (5;1, 10;1, 1;1) at room temperature. B Tube A- contain yellow color gold solution, Tube B- contain pale yellow color Solanum Lycopersium extract, Tube C- contain cocoa brown gold nanoparticles solution.

**UV-Vis Spectroscopic Analysis of Au Nanoparticles:**

The appearance of violet color evident that the formation gold nanoparticles in the reaction mixture and the efficient reduction of the Au<sup>3+</sup> to Au<sup>0</sup> (Fig.2B), the formed color solution allowed to measure the absorbance against distinct wave length to conform the formation of gold nanoparticles. The corresponding UV-vis absorption spectra are shown in Fig. 2A. The change in pH of aqueous gold solution 2.95 and tomato extract 3.91 to 2.69 of tomato gold nanoparticles solution in 24hour. In the present work, AuNPs synthesis with three different concentration of tomato extract with fixed concentration of gold solution as ratio 1; 1, 5; 1, 10; 1. UV-vis scanning of reaction product showed SPR absorption band and peaks (Fig. 2a). Reaction mixture with 1;1 ratio, in which reduction of Au<sup>3+</sup> ions just to occurred and SPR band intensities was less and peak is broad which suggest partial reduction of Au<sup>3+</sup> ion and formation of larger AuNPs with SPR at 550 nm. And in reaction mixture ratio 1;10 the observed intensity of SPR peak is more with small sharpness in the peak compare to the reaction mixture 1;1 with SPR at 530 nm. Where as in reaction mixture 1; 5 the SPR band intensity and peak is highest indicating complete reduction of gold ions with SPR at 540 nm. Thus maximum yield of reduced sized AuNPs at reaction ratio 5; 1 suggested as optimum reaction condition under room temperature condition.

**XRD Analysis:** The crystalline structure of biologically synthesized AuNPs using tomato extract were analysed by XRD measurements. A typical XRD pattern of the Au was found by Bragg reflections corresponding to (111), (200) and (220) sets of lattice planes are observed that

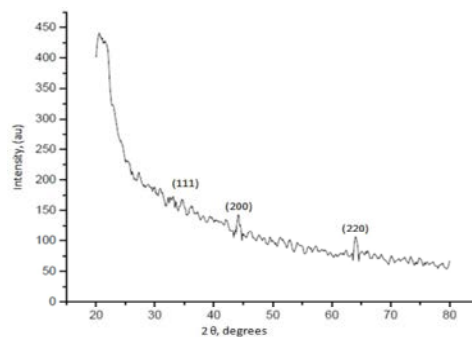


Fig. 3: XRD of gold nanoparticles.

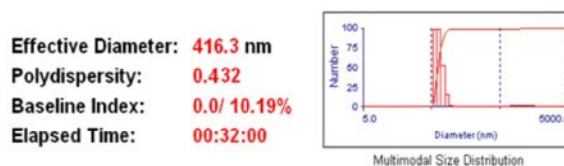


Fig. 4: Particle size analysis.

may be indexed on the bases of the fcc structure of gold. The characteristic peaks corresponding to (111), (200) and (220) are located at 2θ= 38.80°, 44.13° and 64.82° respectively and the weak intensities of peaks indicates that gold nanocrystals are embedded in the film, shown in Figure 3.

**Particle Size Analysis:** Laser diffraction particle size analyzer provides the detail about the particle nature, such as monodispersed, didispersed and polydispersed. Our investigation revealed that nanoparticles show polydispersity at 0.432 indexing and various sizes of nanoparticles ranging with effective diameter around 416.3 nanometer, lognormal summary given below in Figure 4.

**CONCLUSION**

The study demonstrates the rapid synthesis of gold nanoparticles with small sized and high crystallinity. The reduction of the metal ions and stabilization of the gold nanoparticles is believed to occur by the proton releasing hydroxyl group, containing α-terpineol, citronellol, borneol, trans-nerolidol, cis/trans-linalol oxides, α-sitosterol, phytol, geraniol, stigma sterol or any other secondary metabolites and various acids present in extract. The concentration of tomato extract and metal ions plays a crucial role for the synthesis of gold nanoparticles of desired size with reaction conditions. The spectroscopic characterizations using UV-vis, XRD and Particle size analysis were useful in providing the

formation of nanoparticles and also to confirm their characteristic. From literature study proposed that hydroxyl and amine group containing components are responsible as an active reductant and capping agent, but further FTIR analysis can give evidence to understand the appropriate chemical and molecular interactions which could be responsible for the gold salt reduction. As, the appearance of single peak in UV-Vis spectrum represents spherical shape of generated nanoparticles which can be further confirmed by representing the Scanning electron microscopy (SEM) and Transmission electron microscopy (TEM) images.

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