REVIEW PAPER

Green synthesis of gold nanoparticles using plant extract: Mini-review

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ARTICLE INFO

Article History:
Received 20 November 2016
Accepted 1 January 2017
Published 15 January 2017

Keywords:
Gold Nano particle (AuNPs)
Green synthesis
Nano metals
plant extract

ABSTRACT

In this review, we examine the greenest nanoparticles of zero-valent metals, metal oxides and metal salts, with emphasis on recent developments routes. Products from nature or those derived from natural products, such as extracts of several plants or parts of plants, tea, coffee, banana, simple amino acids, as well as wine, table sugar and glucose, have been used as reductants and as capping agents during the present synthesis method. Polyphenols found in plant material often play a key role in the processes mentioned here. The techniques involved are generally one-pot processes, environmentally friendly and simple. Green synthesis of gold nanoparticles using several extracts and spices extracts was conducted, in which aqueous extracts $HAuCl_4.3H_2O$ reduce to Au° has establishing themselves in specific crystal phase. Synthesized nanoparticles were confirmed by the color change of auric chloride which is yellow. The growth of nanoparticles was monitored by the behavior of surface Plasmon using UV-Vis spectroscopy; also the pH was determined meanwhile. Moreover, this approach is not only of a green rapid synthesis kind and considered as a better alternative to chemical synthesis, but also found to be effective for large scale synthesis of gold nanoparticles.

How to cite this article
Teimuri-Mofrad R, Hadi R, Tahmasebi B, Farhoudian S, Mehravar M, Nasiri R. Green synthesis of gold nanoparticles using plant extract: Mini-review. Nanochem Res, 2017; 2(1):8-19. DOI: 10.22036/ncr.2017.01.002

INTRODUCTION

As a result of the global problems related to environmental pollution, the most "green" eco-environmentally technologies and chemicals are becoming increasingly popular.

in the last two years, several books on the subject of 'green chemistry' have been published describing ecological processes [1–4] and their general specialized aspects, including ultrasound, microwaves, and other methods in synthesis [5,6], green analytical chemistry [7], green tribology [8], polymers and green polymerization [9,10], green engineering and manufacturing [11,12], food [13], textiles [14], hydrogen and syngas production and their uses [15,16], wastewater treatment [17] particle technology [18], biofuels, biomass and biocomposites [19–21], and other fields of green chemistry [22,23].

The 12 principles of green chemistry [24] have become a typical guideline for chemical technologists and chemists in developing fewer perilous chemical syntheses. Although UV irradiation, laser ablation, lithography, ultrasonic fields, aerosol technologies and photochemical reduction have been successfully used to produce nanoparticles that remain expensive and involve the use of hazardous chemicals that becomes a compelling reason to carry a lot of interest in the development of organic and ecological [25].

At the same time, despite intensive developments in nanotechnology, the adverse effects of nanomaterials

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are still relatively unknown. The toxicity of the resulting materials and environmental effects of the products might decrease when using biocompatible reagents in their production procedure [26, 27]. Non-toxic solvents (preferably water), closed reactors, 'green' techniques without contacting reaction media and air (ultrasound, microwave, hydrothermal, magnetic, biological methods, among others), and low temperatures for all of the right things achieve this goal.

Representing the methods mentioned as relatively greener routes to nanoparticles and a nanomaterial including plant extracts [28] and other natural products. In this brief review, we focus on the green methods

The methods of obtaining nanoparticles by means of naturally occurring reagents such as plant extracts, vitamins, biodegradable polymers, sugars, and microorganisms as reductants and capping agents could be considered as an attractive field for nanotechnology application. Only a bonded number of inorganic nanoparticles have been fabricated through different syntheses. Large-scale biosynthesis of nanoparticles is a main factor in green syntheses in which suitability of the reagents plays an important role. Plant based materials are the best candidates among the mentioned reagents [29] Parts of plant such as leaf, root, latex, seed and stem are used in the synthesis of metal nanoparticles. Recently, silver nanoparticles were synthesized using the plant extract [30-34]. Ghaffari-Moghaddam and Hadi-Dabanlou reported the synthesis of Silver nanoparticles by the use of Crataegus douglasii fruit extract [35].

It is a common viewpoint that polyphenols are the major active agent's playing important roles in green synthesis. Green synthesis of nanoparticles is more advanced than other methods because it is easy, relatively reproducible, and cost-effective and often results in more stable materials [36].

Microorganisms are one of the other candidates for the production of nanoparticles. But the challenge is the low speed of the synthesis which finally gives a limited number of sizes and shapes for the produced NPs compared to the ones prepared from routes including plant baled materials.

Nowadays in recent experiments fungi are gaining global reputation as Nano-factories for the synthesis of nanoparticles [37].

Gold Nanoparticles

Because of their remarkable biocompatibility, Au particles are extensively and particularly exploited in organisms [38]. Gold nanoparticles possess chemical thermal or photo functionality if designed but they are considered biologically inert in natural form. On near infrared (NIR) irradiation Au-based nanomaterials, Au Nanocages, Au Nanorods and Au Nanospheres possessing the ability to absorb NIR can destroy cancer cells and bacteria via photo thermal heating. Au-based nanoparticles can combine to photosensitizers for photodynamic antimicrobial chemotherapy. Au Nano rods conjugated with photo sensitizers can kill MRSA by photodynamic antimicrobial chemotherapy and NIR photo thermal radiation [39-40]. Existence of aggregated forms of nanoparticles like gold Nano triangles have been

Table 1. Biosynthesis of nanoparticles using some plant extracts

Plant	Reference
Hibiscus sabdariffa	[56]
Mimusops elengi	[115]
Nepenthes khasiana	[116]
Gnidia glauca	[117]
Ampelopsis grossedentata	[118]
Momordica cochinchinensis	[119]
Piper longum	[64]
Couroupita guianensis	[120]
Limonia acidissima	[121]
Pogestemon benghalensis	[122]
Stevia rebaudiana	[123]
Lantana camara	[124]
bean (Cicer arietinum)	[125]
Terminalia catappa	[126]
Pyrus sp (pear fruit extract)	[127]
Psidium guajava	[128]
Mucuna pruriens	[129]
Geranium leaf	[130]
Chenopodium album	[112]
Sorbus aucuparia	[131]
Mangifera indica	[132]
Anacardium occidentale	[133]
lemongrass	[134]
Ocimum sanctum	[135]
Cinnamomum camphora	[136]
Emblica officianalis	[137]
Tamarindus indica	[41]
Coriandrum sativum	[68]
Magnolia kobus	[83]
mushroom	[138]
Honey	[139]
Cashew	[133]
Horsegram	[140]
Tulsi	[135]
Emblicaofficianalis	[137]
alfalfa	[141]
Cinnamomum camphora (Alliumcepa)	[136]
Coleus amboinicus	[142]
Cinnamon zeylanicum	[143]
Garcinia combogia	[144]
Morinda citrifolia	[145]
Saraca indica	[146]
Cocos nucifera	[147]

reported in *lemon grass* extracts and *tamarind* leaf extracts [41], also dead biomass of *Humulus lupulus* produces gold nanoparticles as well [42].

Various optical, thermal, catalytic and physical properties of Gold nanoparticles (AuNPs) which depend on their size and shape have drawn attention towards the synthesis of AuNPs. Recently much attention has been paid to the use of biologic synthesis processes without the need for toxic chemicals in synthesis protocols to avoid adverse effects on biomedical applications to synthesize biocompatible metal. Biotechnology, cosmetics, electronics, DNA labeling, biological and chemical sensors, coatings and packaging are all counted as susceptible fields for the application of metallic nanoparticles [43-44]. Application of biological organisms such as microorganisms, plant extracts or plant biomass could be regarded as an alternative to the chemical and physical methods to produce nanoparticles in a sustainable manner [45-46].

Different synthesis methods via using organisms, both unicellular and multicellular such as yeast, fungi and bacteria came into existence and used to synthesize inorganic materials either extracellularly nor intracellularly [47]. Some plants called 'hyper accumulators' can absorb and accumulate metals from water and soil in which they are grown. *Alfalfa* can accumulate gold and store it in their leaves and stems biomasses as discrete nanoparticles of pure metal [48]. In recent years, various plants have successfully been used and reported for effective and rapid extracellular synthesis of gold, silver and copper nanoparticles such as broth extracts of neem [49], Aloe Vera [50-51], Arena sativa [52], wheat [53], alfalfa [54], geranium [55], Hibiscus sabdariffa [56] and lemongrass [57] It shows nanoscale gold novel features and has various activities that are suitable for therapeutic use and broad applications in nanobiotechnology [58-59]. Barrier of cytotoxicity could be crossed by other plants and spices which mediate stabilized or capped AuNPs that is considered as a main factor for biomedical application of AuNPs. Improved contrast agents for molecular imaging cancer diagnosis require highly biocompatible nanoparticles in their composition to function with a good quality in imaging instruments among which gold nanoparticles derived from phytochemicals could display a considerable biocompatibility [60-62].

Green synthesis of gold nanoparticles using plant extracts

This review focuses on the synthesis of AuNPs in which plant extracts are used. The reaction is generally completed in a short time. Gold nanoparticles and many other nanoparticles could be produced by this method.

Recently, AuNPs were synthesized using *Piper longum* extract. The average size of the AuNPs was 56 nm as confirmed by the DLS particle size analyzer [63].

Yu *et al.* [64] reported the synthesis of gold nanoparticles using aqueous extracts of *Citrus maxima* (C. maxima).

AuNPs were synthesized using aqueous extracts of neem (*Azadirachta indica*) by Anuradha *et al.* [65] At a fairly wide various stoichiometric ratios, the nanoparticle formation began as soon as the neem extract and the Au (III) solution were added to each other. The synthesis process was completed in 24 hours and during this period the characteristic Plasmon vibration peak of gold nanoparticles was stabilized at 557±1 nm. Using energy dispersive, X-ray spectroscopy (EDAX), (Fig. 1) and scanning electron microscopy (SEM) techniques, the synthesis process and the associated studies were described.

Synthesis of AuNPs with an average particle size of 100 nm mediated by an extract of *Allium cepa* [66].

A dilute extract containing *phyllanthin* which

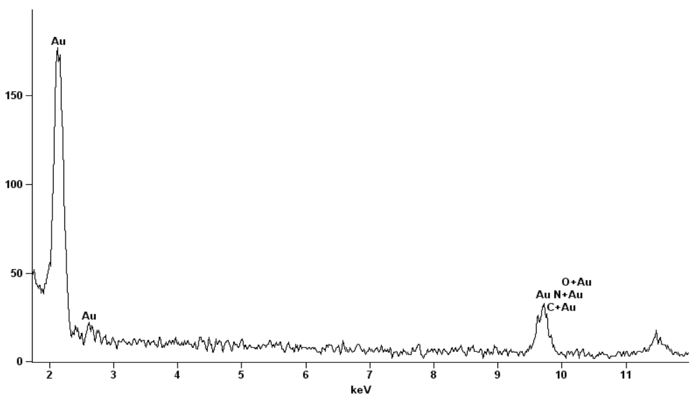

Fig. 1. EDAX profile of gold nanoparticles synthesized by A. indicia extract.

is derived from the plant *Phyllanthus amarus* was used in the experiments done by Kasthuri *et al.* [67] To generate triangular and hexagonal gold nanoparticles from $HAuCl_4$. For the attainment of spherical nanoparticles increasing the concentration of the extract could produce the proper solution. Narayanan and Sakthive [68] produced diverse shaped nanoparticles of gold (spherical, triangular, decahedral) from about 7 to 58 nm in size using a leaf extract of *Corian drum sativum* (coriander). In another study by Edison and Sethuraman [69] an aqueous extract of *Terminalia chebula* was consumed for the production of gold nanoparticles with sizes between 6 to 60 nm that are active against both *S. aureus* and *E. coli*.

Extracts of *Chrysanthemum* and tea beverages are examples of other materials used in the works of Liu *et al.* to synthesize gold nanoparticles. To quantify the antioxidant properties of teas, a nanoparticle-based analysis was elaborated [70].

By the use of an aqueous *Cassia fistula* extract, Daisy and Saipriya [71] succeeded to synthesize gold nanoparticles of 55–98 nm. Hypoglycemic is a famous characteristic for the extracts of *C. fistula bark*.

Reduction of ions to gold nanoparticle is a capability found in *Apiin* which is present in the extract of *banana* leaf. The mentioned procedure that is reported by Kasthuri *et al.* [72] proves that secondary hydroxyl and carbonyl groups of *Apiin* cause the reduction explained above to obtain gold nanoparticle.

Dwivedi and Gopal [73] generated quasi-spherical shaped nanoparticles in the size range from 10 to 30 nm by the use of *Chenopodium album* leaves extracts. Nagajyothi *et al.* used an aqueous extract of *Lonicera Japonica* flower to produce gold nanoparticle which had an average diameter of 8.02 nm [74]. The spectroscopic features resulted from UV-Vis, Fourier transform infrared (FTIR), SEM and high-resolution transmission electron microscopy (HRTEM) supported the formation and the stability of the green synthesized AuNPs. Thirumurugan *et al.* reported the synthesis of gold nanoparticles using *Azadirachta indica* plant leaf extract [75].

Ali *et al.* [76] reported the synthesis of gold nanoparticles using extracts of *Mentha piperita* (peppermint). These nanoparticles showed a considerable antibacterial activity against clinically isolated human pathogens such as *E. coli* and *S. aureus*. Leaves and bark extracts of *Ficus carica* were used to produce gold nanoparticles. The results obtained by characterization of synthesized materials were compared [77].

Gold nanoparticles were also synthesized using the ethanol extract of black tea and tannin as a reducing and stabilizing agent. Ethanol extract of black tea and its free ethanol tannin extract produced gold nanoparticles in the size ranges of 2.5-27.5 and 1.25-17.5 nm with an average size of 10 and 3 nm respectively. In contrast, gold colloids which were synthesized by a free ethanol tannin extract showed no particle aggregation during short and long storage times at the same condition [78]. Nagaraj *et al* [79] reported the synthesis of AuNPs in which *Plumeria Alba* (Frangipani flower) was used as reducing agent. Two main roles are attributed to the extracts of flowers one of which is to encapsulate the gold nanoparticles and the other one is their part as the reducing agent. During the formation of gold nanoparticles in the reaction which is due to their specific properties (surface Plasmon resonance), the characteristic color change from pale yellow to dark brown was recorded while the reaction completed. The transmission electron microscopy (TEM) and UV-Vis spectroscopy were used to characterize the obtained gold nanoparticles.

The UV-Vis spectrum showed surface Plasmon peak at ~550 nm. From what TEM images reveal it is proved that samples are spherical in morphology possessing two different particle sizes of 20-30 nm for smaller particles and 80-150 nm for larger ones (Fig. 2). To obtain complementary results the

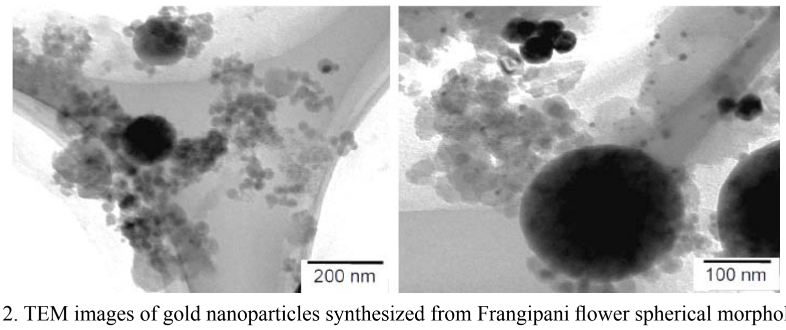

Fig. 2. TEM images of gold nanoparticles synthesized from Frangipani flower spherical morphology.

research continues with studies on antimicrobial activities of the introduced gold nanoparticles against several microorganisms (Fig. 3a, b).

Dubey *et al*. [80] reported the synthesis of AuNPs using *Rosa rugosa*. Carbonyl groups were responsible for the reduction of metal ions to generate nanoparticles; this action is proved by an FTIR study. Gold nanoparticles show various zetas potential in different ranges of pH which is in its lowest amount at strongly acidic pH. When the pH of reaction is reduced the obtained nanoparticles grow in size (50-250 nm).

AuNPs were also prepared by using the fruit peel extract of *Momordica charantia*. Best parameters for the synthesis of gold nanoparticles were pH 10, high temperature (100 °C), and 100 ppm aurochlorate salt. The results were verified using TEM, XRD and UV-Vis spectroscopy. AuNPs were monodispersed and found to be 10-100 nm in size. It was found that the AuNPs synthesized using biological protocols were much more stable than those synthesized chemically when tested using NaCl 5 M solution [81].

The extract of *Benincasa hispida* seed as both reducing and capping agents was used as another source to synthesize the AuNPs. During the reduction process (Fig. 4) carboxylic group (COOH) present in the extract changes to COO^-.

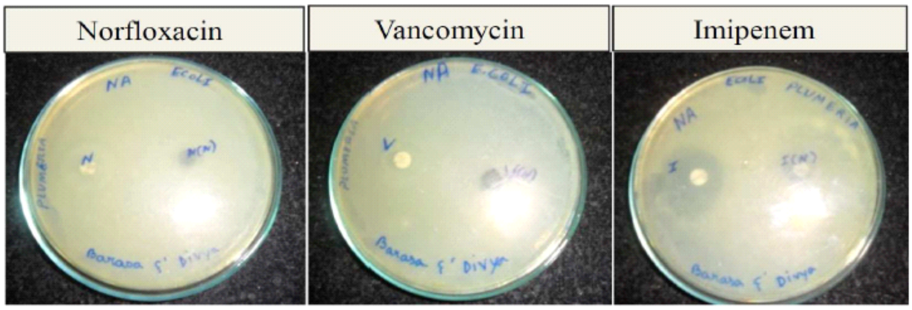

Fig. 3a. Antimicrobial activity of the gold nanoparticles synthesized from Frangipani against *E.coli.*

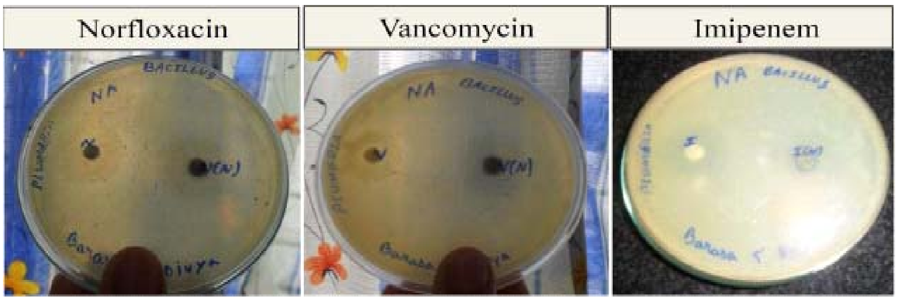

Fig. 3b. Antimicrobial activity of the gold nanoparticles synthesized from Frangipani against *Streptobacillus sp.*

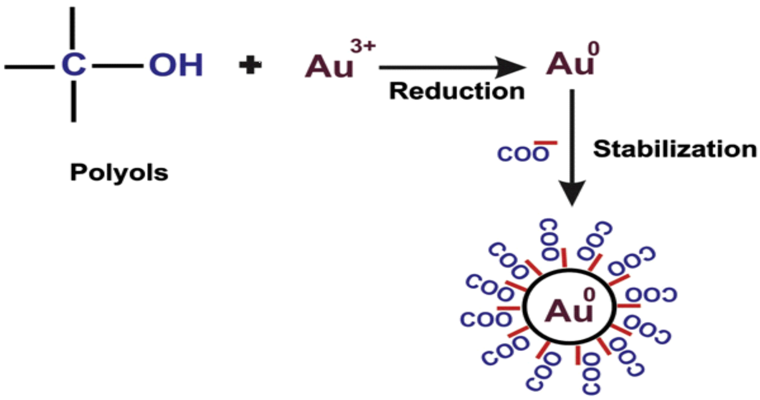

Fig. 4. Mechanism of formation of gold nanoparticles.

The carboxylate group present in proteins could act as a surfactant to attach on the surface of AuNPs and stabilize AuNPs through electrostatic stabilization. The particle size could be easily affected by the reaction conditions including quantity of the extract, temperature, and pH. Gold nanoparticles with different sizes in the range from 10 to 30 nm could be obtained by controlling the synthesis parameters. The nanoparticles were more stable at pH 6. The crystalline nature of NPs is approved by bright Bright spots in the circular pattern selected area electron diffraction (SAED), light stripes HRTEM lattice images and peaks in the pattern of X-ray diffraction.

Various functional groups are present in the biomolecule capping nanoparticles; this is clearly shown in the relevant FTIR spectrum [82].

Yong Song *et al* reported the synthesis of AuNPs using *Magnolia kobus* and *Diopyros kaki* leaf extracts. SEM and TEM images (Fig. 5 and Fig. 6) showed that a mixture of plate (triangles, pentagons, and hexagons) and spherical structures (size, 5–300 nm) were formed at lower temperatures and leaf broth concentrations, while smaller spherical shapes were obtained at higher temperatures and leaf broth concentrations [83].

Dubey *et al* used an extract of *Tansy* fruit to produce gold nanoparticles. Formation of AuNPs was confirmed by surface Plasmon spectra using UV–Vis spectrophotometer and absorbance peaks at 546 nm. Powder diffraction study showed the face-centered cubic (fcc) lattice of AuNPs. The

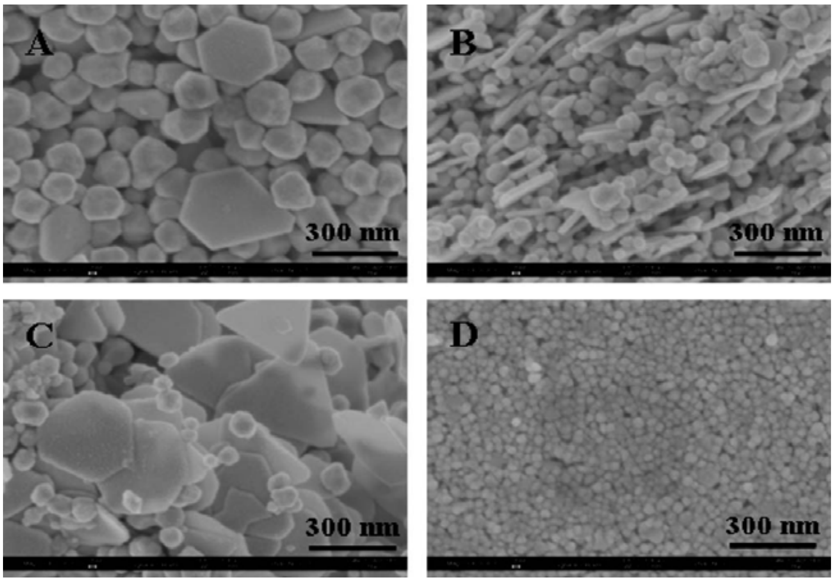

Fig. 5. SEM images of the gold nanoparticles formed by the reaction of 1 mM HAuCl4 and 5% Diopyros kaki leaf broth at different reaction temperatures: (A) 25 °C, (B) and (C) 60 °C, and (D) 95 °C.

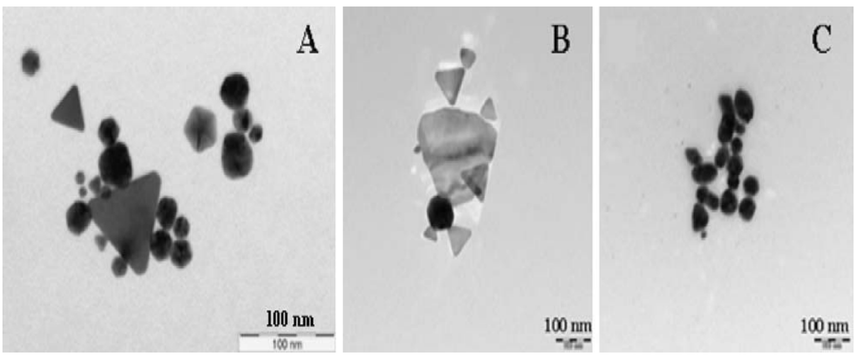

Fig. 6. TEM images of the gold nanoparticles formed by the reaction of 1 mM HAuCl4 and 5% Magnolia Kobus leaf broth at different reaction temperatures: (A) 25 °C, (B) 60 °C, and (C) 95 °C.

average crystals of AuNPs were 11 nm estimated from Scherer method [84].

In another study, *Zingiber officinale* extract was used to produce the AuNPs. The UV-Vis analysis showed a peak at 523 nm due to surface Plasmon resonance [85].

Using aqueous extract of *sugar beet pulp* as the source for the synthesis of AuNPs is one of the reported methods in which the extract as acts both reducing and capping agent. The absorption spectrum of AuNPs showed a peak at 560 nm. For a gold biorecovery optimal solution both pH and reaction time must be controlled. Bio gold sorption is carried out at low pH values Initial after 24 hours due to the presence of hydroxyl groups in biomass [86].

Smitha *et al* reported the synthesis of AuNPs using of *Cinnamomum zeylanicum* leaf broth. The optical properties of nanoparticles observed in the study correlated with the TEM observations. All the synthesized samples were found to be photo luminescent [87].

Leaf extract of *Murraya Koenigii* is the material used in the research done by Philip *et al.* for the production of AuNPs in the range of 10-20 nm diameters. The UV-Vis analysis showed a peak at 532 nm AuNPs located [88].

Synthesis of AuNPs using the extract of *Rosa hybrida* petal was reported by Noruzi *et al.* TEM, FTIR, EDAX, UV–Vis spectroscopy, dynamic light scattering (DLS) and XRD were used to characterize the gold nanoparticles. The reaction was rapid and completed within 5 min at room temperature [89].

AuNPs were prepared using aqueous extract of *cypress*. The impact of concentration and pH of extract in the size of the nanoparticles are investigated. It was found that the average size of the synthesized gold nanoparticles are mostly depends on both the extract concentration and the pH. FT-IR spectroscopy showed that molecules bioorganic covered limited to the particle surface. X-ray techniques confirmed the formation of gold nanoparticles and crystal structure [90].

Aromal and Philip synthesized the AuNPs using aqueous extract of *fenugreek* (Trigonella foenum-graecum) as reducing and protecting agent. By controlling the synthesis parameters, gold nanoparticles with sizes in the range from 15 to 25 nm achieved. The high crystallinity of nanoparticles was obviously shown in clear lattice fringes in HRTEM images, bright circular spots in the SAED pattern, and peaks in the XRD pattern. FTIR spectrum indicated the presence of different functional groups present in the capping biomolecule of the nanoparticles. The synthesized gold nanoparticles showed good catalytic activity for the reduction of 4-nitrophenol to 4-aminophenol in the presence of excess $NaBH_4$. The catalytic activity was found to be size dependent. Their activity becomes quicker as they get smaller in size (Fig. 7) [91].

Sujitha and Kannan synthesized the AuNPs from the leaf extract of *Citrus fruits*. From what TEM studies showed it was revealed that the shapes and the sizes of NPs are different from one another. The particle size ranges from 15 to 80 nm. SAED pattern confirmed the fcc phase and the crystallinity of the particles [92]. AuNPs were synthesized using the Philip. The nanoparticles are characterized by TEM, XRD, FTIR and UV–Vis spectroscopy [93].

Some examples of other studies

AuNPs were also synthesized using the extract of *Justicia glauca* [94],*Aegle marmelos* [95],*willow* tree bark [96],*Plumeria alba* [97], *Sativus* (saffron) [98], *Zingiber Officinale* [99], an olive leaf extract [100],

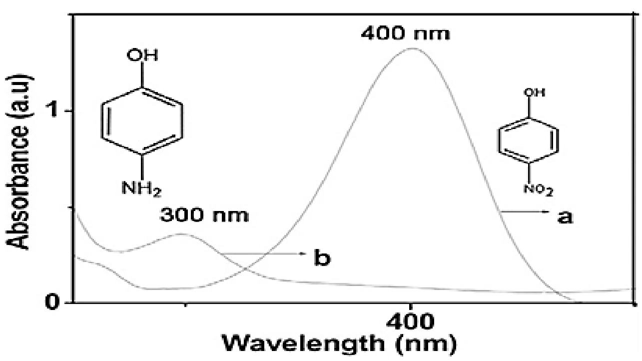

Fig. 7. UV–Visible spectrum of the (a) mixture of 4-nitrophenol and NaBH4, (b) mixture of 4-nitrophenol and NaBH4, 7 min after the addition of gold colloid (g4)

The extract of dried flowers a *Chillea Wilhelmsii* [101], an Leaf Extract of Magnolia Kobus [102], the extract of *Camellia sinensis* [103], an *Centella asiatica* leaf extract [104]. the ethanoic leaf extract of Bacopa monnieri [105], a *pomegranate* (Punica granatum) leaf extract [106], the seed aqueous extract of *Abelmoschus Esculentus* [107], leaf extract of antidiabetic potent Cassia auriculata [108], the extract of *Cissus quadrangularis* [109], Rosa damascene [110], a glucan of an *edible mushroom* [111], *Chenopodium* album leaf extract [112], *Barbated Skullcup* herb extract [113], macerated aqueous extracellular dried clove buds (Syzygium aromaticum) solution [114].

CONCLUSIONS

Gold nanoparticles have attracted great attention due to their potential applications in electrical conductivity, catalysis, optical properties and etc. AuNPs synthesized from metallic gold possess antibacterial properties which turns them to a good candidate to be used in both commercial and medical products. Green synthesis of AuNPs has numerous advantages over chemical and physical methods: profitability of being easily expanded for large scale synthesis without any need for high pressures, energy environment, temperature and toxic chemicals.

The synthesis of AuNPs using plant materials is a conventional eco-friendly method when compared to chemical and physical synthesis. Since plants are widely distributed, readily available and at the same time safe to handle there will be a lot to do to develop this method of synthesis inspired by several conventional ideas.

ACKNOWLEDGMENTS

We gratefully acknowledge the financial support for this work by the Research Council of University of Tabriz.

CONFLICT OF INTERESTS

The authors declare that there is no conflict of interests regarding the publication of this paper.

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