

Original Article

Green Synthesis of Gold Nanoparticles using extract of Ginger, Neem, Apta, Umber plants and Their Characterization using XRD, UV-Vis spectrophotometer

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ABSTRACT

The establishment of nanotechnology, nanoparticles from gold (Au NPs) is one of the extensive attentions due to the presence of important properties. In this study, the green synthesis of gold NPs from the extract of ginger, Neem, Apta, Umber (GAuNPs, NAuNPs, AAuNPs, and UAuNPs) plants is reported. The obtained AuNPs were characterized by UV-Vis spectrum and X-ray diffraction analysis (XRD). Since all the plants (bark) showed FCC face centered cube structure as FCC as permitted reflection were (111), (200), (220), (311), and gold nanoparticles indicated the maximum wavelength in the range of 520 nm to 680 nm.

Introduction

The development of new nanomaterial in the nanotechnology and nanoscience has been emerged in recent years [1]. These technologies have attracted in a range of fields, such as medicinal chemistry, pharmaceutical,

material physics, nanochemistry, and sensing, owing to their unique properties [2, 3]. The size of nonmaterial is ranging from 1–100 nm in size [4]. The application of nanotechnology is widely divided in the all research area of science that includes photochemistry, fluorescence technology, drug discovery, energy science, and optical flow applications

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[5]. The unique application of nanotechnology and their distinctive properties such as the extreme surface-to-volume proportion, elevated surface energy, especial mechanical, electrical, magnetic, thermal, and optical performance [6]. The application of gold nanoparticles are very broad research area includes sensing material, novel catalysis, electronic material, and medicinal products [7,8]. The metallic nanoparticles has more important and have a wide range of applications in broad area [9]. Since from the last decade, notable attention has been made in the field of nanotechnology, the synthesis of metal nanoparticles particularly AuNPs have received appreciable awareness due to their biological activity [10]. The principal superiority of gold NPs is that they were simple to prepare by organic transformation with less hazardous condition as compared with other material. In the literature, numerous techniques have been reported for the preparation of gold nanoparticles to activate their surface to upgrade their utilization.

The most common method for the preparation of Au nanoparticles is chemical reduction. However, the chemical reduction process usually requires a chemical addition (reducing agents, stabilizing agents, and surfactants) and harsh conditions which violate the basic principles of green chemistry [10]. Thus, a more environmentally friendly procedure is needed to prepare the Au NPs. The greener approaches synthesis of functional groups hydroxyl, carbonyl, and aldehyde groups play an important role in reducing and stabilizing metal nanoparticles. Since from the past decade, numerous protocols have been reported that includes mechanical grinding method [11], microwave irradiation [12], and heat reduction [13].

The microwave radiation was done at 80 °C for 60 min synthesis of Lignin-AuNPs liquid

marble for the Pb^{2+} detection [14]. The hemicellulose/lignin at 100 °C used for the AuNPs synthesis [15]. Moreover, the greener lignin-based nanoparticles (LNPs) have been prepared having LNPs acted as a reducing agent, stabilizing agent, and template properties [16–17].

P. Elia et al. reported the green synthesis of nanoparticles from the extract of *Salvia officinalis*, *Lippiacitriodora*, *Pelargonium graveolens*, and *Punicagranatum* [18].

KarXin Lee *et al.* reported the extract of *Garciniamangostana* fruit peel for the Au nanoparticles [19]. The ethanol-water of *Mimosa tenuiflora* (Mt) bark was used for the AuNPs preparation at various metallic concentrations [20].

In this work, AuNPs were synthesized by using an extract of Neem, Apta, and Umber plants. The total green synthesis of AuNPs conducted at room temperature was achieved by principles of green chemistry. Our results were compared with similar works of catalysis with AuNPs synthesized by “green” methods.

Experimental

Materials and methods

The AuNPs synthesis was carried out from various plants, which are collected from different areas like college campus, farms, and gold (III) chloride [Sigma-Aldrich, USA; 99.99% pure]. Mature plant parts like leaves, fruits, stem, etc. were weighed, cleaned, and cut into small pieces or sometimes made powder. This fine pieces or powder is then added to 100 mL of ethanol and filtered through Whatman filter paper no. 41. The filtrate was used as reducing agent and stabilizer. The production and stabilization of the reduced AuNPs in the solution was monitored by the UV-Vis spectrophotometer analysis. Thereafter, the plant extract (0.5 mL) was added to 3 mL of the

gold (III) chloride solution (0.001 M) with continuous stirring. The spectrum was scanned from 200 to 800 nm wavelengths. The X-ray diffraction (XRD) measurements were carried. The samples were characterized morphologically by doing SEM. A pinch of dried AuNPs was coated on Silicon Wafer in an auto fine coater, and then material was subjected to analysis.

Experimental methods

Extract preparation

The plant (bark, stem), dry fruits, and spices extract solution was prepared by taking 10 g of thoroughly washed and dried leaves. After accurately weighing the leaves sample, the extract was taken by using 100 mL of ethanol

and filtered after 5 days. This solution or extract is used for further procedure.

Synthesis of gold nanoparticles

Different volumes of 0.25 mL, 0.5 mL, 0.75 mL, and 1 mL of the leaf, fruit pulp, and spices extract were added separately to 3 mL solution of 1 mM HAuCl_4 in different test tubes. After 6 hours, this solution was observed by using the UV-Visible Spectroscopy, to know the effect of amount of extract on the synthesis of gold nanoparticles. For further characterization like XRD, SEM, 0.5 mL: 3 mL ratio is selected by observing the graph of the UV-Visible Spectroscopy for all leaves, fruits, and spices extracts.

Flow sheet of steps involved in synthesis

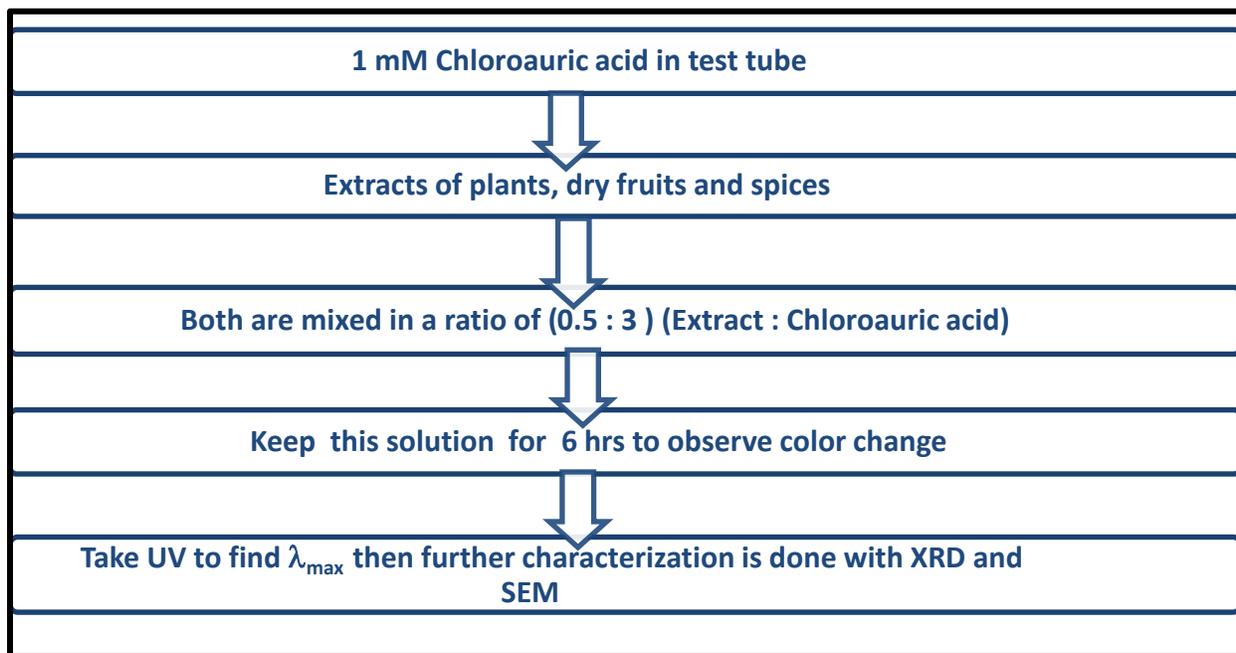


Figure 1. Synthesis Route

Selected plants (Bark/Stem)

Four plant leaves or stem or bark were collected and dried for one week at room

temperature. They were Ginger (*Zingiberofficinale*), Neem (*Azadirachta indica*), Apta (*Bauhinia racemosa*), and Umber (*Ficus racemosa*).

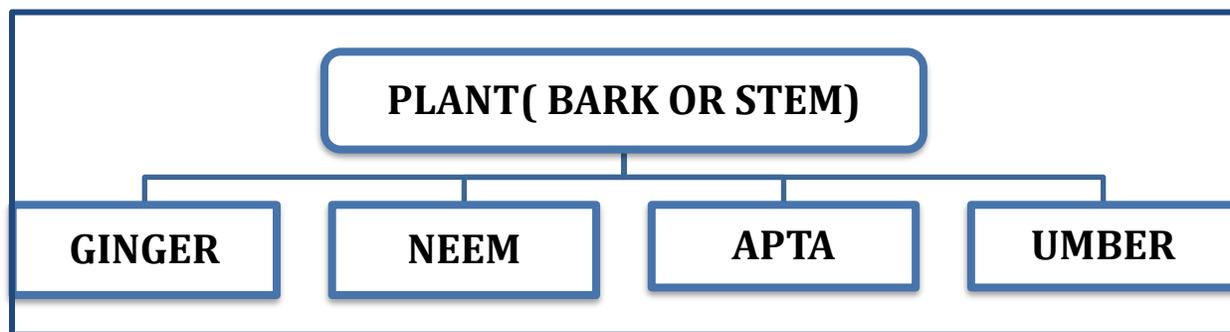


Figure 3. Plants Used for Gold Nanoparticles Synthesis

Extract preparation

The plant (bark or stem) extract was prepared by taking 10 g of thoroughly washed and dried leaves. This weighed leaves were taken into the beaker containing 100 mL ethanol, and then this mixture was kept at room temperature and filtered after 5 days. This filtrate was used for further procedure.

Synthesis of gold nanoparticles

0.5 mL of leaf extract was added to 3 mL, 1 mM solution of HAuCl_4 . After 6 hours, the color change was observed due to formation of nanoparticles. The UV-visible spectroscopy was done for confirmation of synthesized gold nanoparticles and λ_{max} was calculated. Furthermore, advance characterization was done by using XRD and SEM (Figure 2).

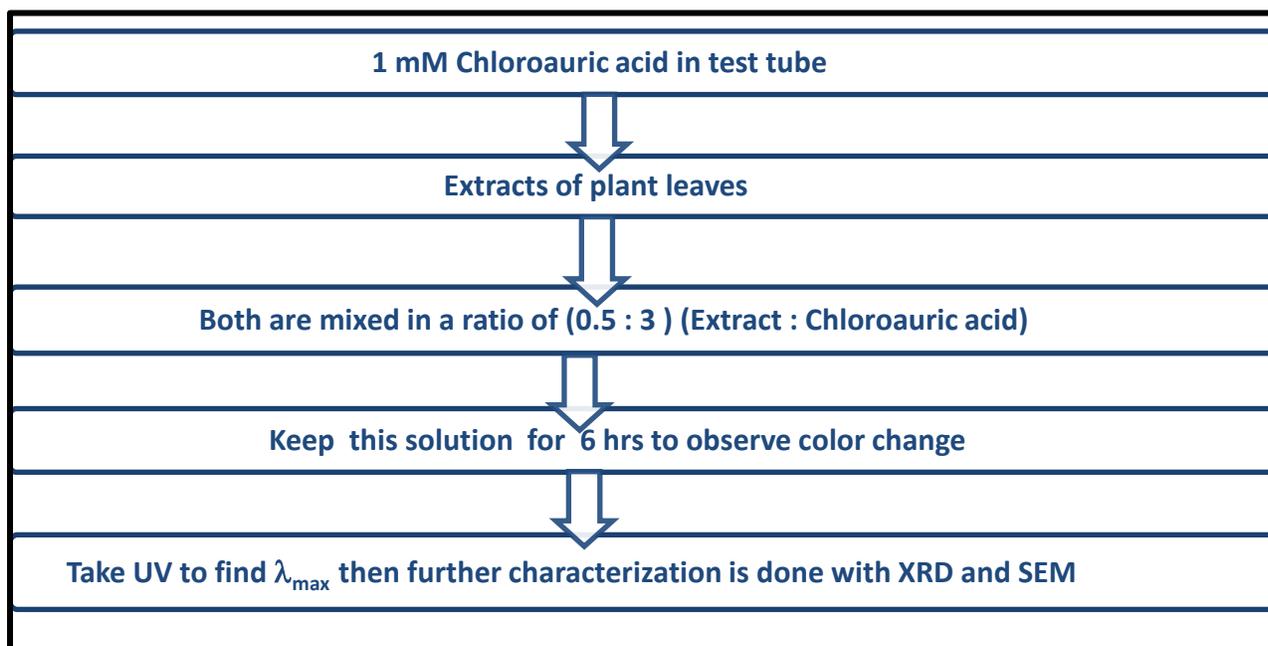


Figure 2. Biochemical Method of Synthesis of Gold Nanoparticles

Extract and preparation of AuNPs from Ginger (GAuNPs)

Ginger is one of the important naturally occurring food, which is having large amount of phenolic and alkaloids compounds, the *Zingiberofficinale* is an botanical name of ginger [21], Ala is an local Marathi languages in the state Maharashtra (Figure 3). Ginger, the rhizome of the *Zingiberofficinale*, has shown therapeutic role in the health management since the ancient time and has been considered as a potential chemopreventive agent.



Figure 3.Ginger

Extract preparation

The Ginger (*Zingiberofficinale*) stem extract was prepared by taking 10 g of thoroughly washed and dried stem pieces. These weighed pieces were taken into the beaker containing 100 mL ethanol, and then this mixture was kept at room temperature and filtered after 5 days. This filtrate was used for further procedure.

Synthesis of gold nanoparticles

0.5 mL of the Ginger (*Zingiberofficinale*) stem extract was added to 3 mL solution 1 mM solution of HAuCl_4 separately. After 6 hours, the color change was observed due to formation of (GAuNPs)nanoparticles. The UV-visible spectroscopy was done for confirmation of the synthesized gold nanoparticles and λ_{max} was calculated. Furthermore, the advance characterization was done by using XRD and SEM.

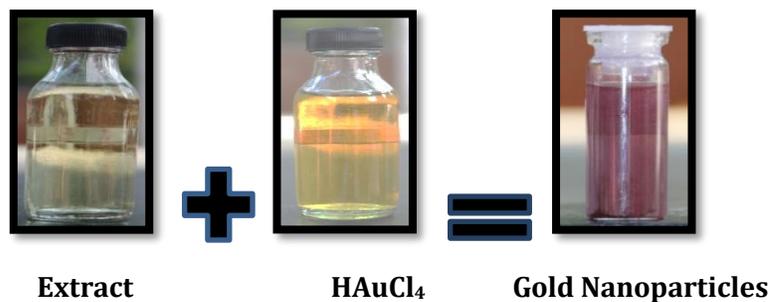


Figure 4.Images for GAuNPs synthesis

Figure 4 displays that the extract solution of ginger was light colour less in nature. Upon the addition of a HAuCl_4 solution to the extract solution, after 10 minutes, the colour of the solution changes from light brown to purple at RT; such changes indicate the AuNPs formation [11].

Extract and preparation of AuNPs from Ginger Neem (NAuNPs)

The neem of leaves, bark, and steam contains organic molecules includes, ascorbic acid, n-hexacosanol as nimbin, nimbanene, nimbolide, and amino acid, 7-desacetyl-7-benzoylazadiradione, 7-desacetyl-7-benzoylgedunin, 17-hydroxyazadiradione, 6-desacetylnimbinene, nimbandiol, and nimbiol. The botanical name of the neem is *Azadirachta indica* (neem) belonging

to *Meliaceae* family [22]. Traditionally, neem has had medicinal importance and has been used for various diseases [22].



Figure 5.Neem

Extract preparation

The plant bark extract was prepared by taking 10 g of thoroughly washed and dried bark of Neem (*Azadirachta indica*). This weighed bark pieces were taken into the beaker containing 100 mL ethanol, and then this mixture was kept at room temperature and filtered after 5 days. This filtrate was used for further procedure.

Synthesis of gold nanoparticles

0.5 mL of the hibiscus leaf extract was added to 3 mL solution and 1 mM solution of HAuCl_4 separately. After 6 hours, the color change was observed due to formation of nanoparticles. The UV-visible spectroscopy was done for confirmation of the synthesized gold nanoparticles and λ_{max} was calculated. Furthermore, advance characterization was done by using XRD and SEM.

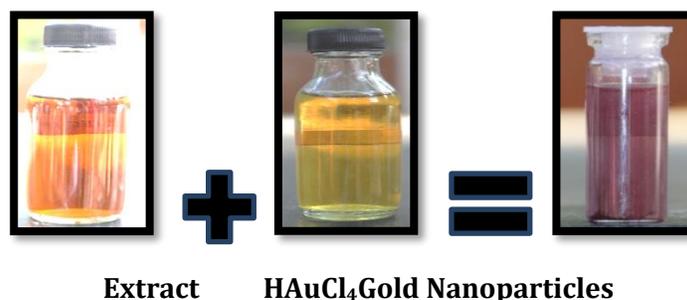


Figure 6.Images for NAuNPssynthesis

Figure 6 indicates that the extract solution of Neem was in light colour less in nature. Upon the addition of a HAuCl_4 solution to the extract solution, after 10 minutes, the colour of the solution changes from light brown to purple at RT; such changes indicate the AuNPs formation.

Extract and synthesis of gold nanoparticles from *Apta*.

Bauhinia racemosa Lam (The Sonpatta Tree) is a small, crooked, bushy, and deciduous tree with drooping branches, which can grow in

poor and very harsh climatic conditions. The deciduous tree is propagated easily from the seed. An extract of the leaves has been proved to show analgesic, anti-pyretic, anti-inflammatory, anti-spasmodic, anthelmintic, and antimicrobial activity. The tree has anti-tumor qualities and is widely used in Ayurveda to treat first stage cancer [23]. The *B. racemosa* root contains a new tetra cyclic lupeol, betulin, β -sitosterol, and tetracyclic 2, 2-dimethyl chroman [5,6]. The seed contains flavonoids, crude protein, and lipid [24,25]. The bark of the plant contains β -sitosterol and β -amyryn, and the leaves contain flavonols (kaempferol).



Figure 7.Apta

Extract preparation

The plant bark extract was prepared by taking 10 g of thoroughly washed and dried bark pieces of Apta (*Bauhinia racemosa*). This weighed bark pieces of Apta (*Bauhinia*

racemosa) were taken into the beaker containing 100 mL ethanol, and then this mixture was kept at room temperature and filtered after 5 days. This filtrate was used for further procedure.

Synthesis of gold nanoparticles from Apta (AAuNPs)

0.5 mL of the Apta (*Bauhinia racemosa*) extract was added to 3 mL solution 1 mM solution of HAuCl_4 separately. After 6 hours, the color change was observed due to the nanoparticles formation. The UV-visible spectroscopy was done for confirmation of the synthesized gold nanoparticles and λ_{max} was calculated. Furthermore, the advance characterization was done by using XRD.

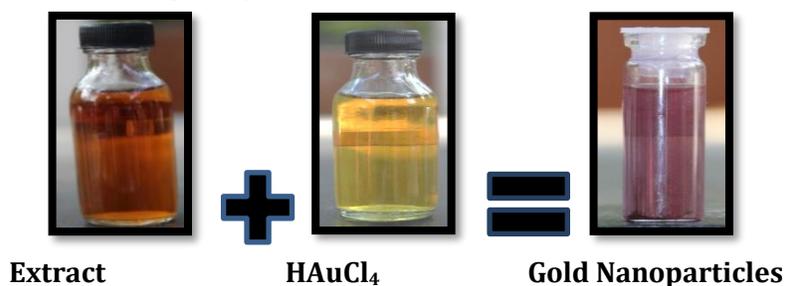


Figure 8.Images for AAuNPs synthesis

Figure 8 demonstrates that the extract solution of Apta (*Bauhinia racemosa*) was in light colour. Upon the addition of a HAuCl_4 solution to the extract solution, after 10 minutes, the colour of the solution changes from light yellow to purple at RT; such changes indicate the AuNPs formation.

Extract and preparation of gold nanoparticles from Umber (UAuNPs)

Umber has botanical name *Ficus racemosa* with an excellent source of natural product like phenolic, flavonoids, ascorbic acids, and tannins. These compounds indicate the very good biological activity, *Ficus racemosa* shows an excellent antioxidant activity. The use of

Ficus racemosa extract for the AuNPs preparation added more advantages for greener synthesis.

The plant bark extract was prepared by taking 10 g of thoroughly washed and dried bark pieces of Umber (*Ficus racemosa*). After accurately weighing, the sample extract was taken by using 100 mL of ethanol and filtered after 5 days. This solution or extract is used for further procedure.



Figure 9. Umber *Extract preparation*

Synthesis of gold nanoparticles

0.5 mL of the dried bark pieces of Umber (*Ficus racemosa*) extract was added to 3 mL solution 1 mM solution of HAuCl_4 in test tube. After 6 hours, the color change was observed due to the nanoparticles formation. The UV-visible spectroscopy was done for confirming the synthesized gold nanoparticles and λ_{max} was calculated. Furthermore, advance characterization was done by using XRD and SEM.

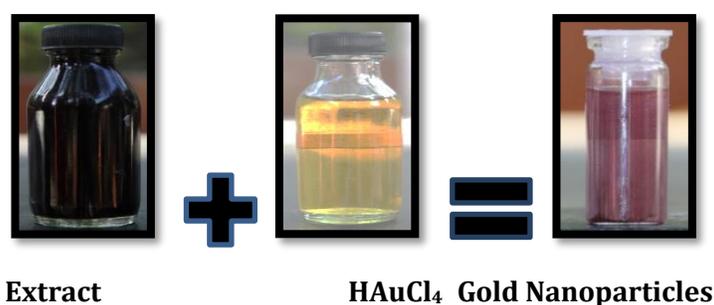


Figure 10. Images for NPs synthesis

Figure 10 indicates that the extract solution of Umber (*Ficus racemosa*) was in black colour. Upon the addition of a HAuCl_4 solution to the extract solution, after 10 minutes, the colour of the solution changes from light yellow to purple at RT; such changes indicate the AuNPs formation.

Results and Discussion

Green synthesis and characterization of AuNPs

The four different medicinal plants extracts of the *Zingiber officinale*, *Azadirachta indica*, *Bauhinia racemosa*, and *Ficus racemosa* were concentrated to obtain the paste and were subjected to a qualitative phytochemical test to the standard procedures [26]. The qualitative phytochemical illustrates the presence of

numerous natural products in the extract like fatty acids, sterols, terpenoids, chlorophyll, and alkaloids derivatives, which was removed by washing with organic solvent ethyl acetate, n-hexane, chloroform, and methanol [27]. The remaining chemicals like apoinins, polyphenols, rotenoids, glycosides, and tannins may be responsible for the AuNPs synthesis [28]. While the FTIR spectra of the AuNPs illustrates the particular band at 3458, 2989, 2886, 1629, 1331, and 982 cm^{-1} for AuNPs, while the peak at 3423, 2989, 2887, 1712, 1628, 1334, and 1068 cm^{-1} for AuNPs. The FT-IR peak of 3458 and 3423 cm^{-1} belong to the free OH stretching vibrations of alcohol [29]. The AuNPs revealed the presence of functional group carboxylate and phenolic (alcoholic) groups which is responsible for the binding with the AuNPs.

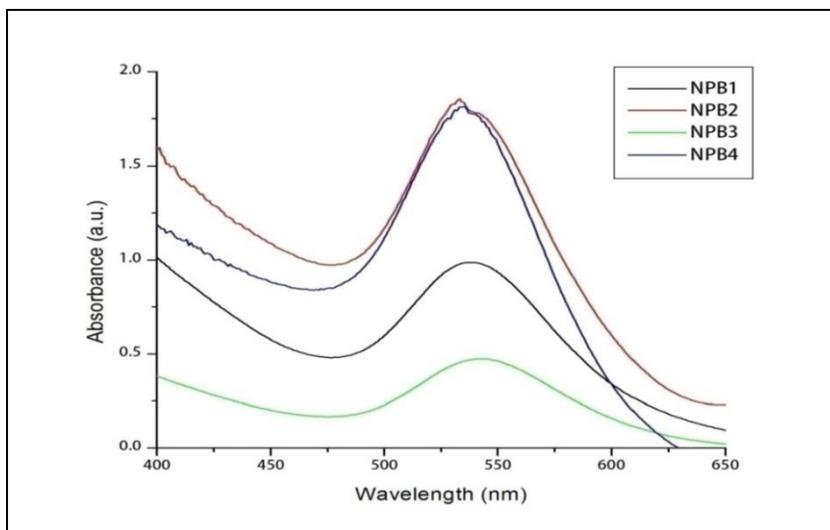


Figure 11. The obtained results for plants (bark/stem) from UV-Vis spectrophotometer

The UV-Visible spectrophotometer gave us the maximum wavelength at which gold nanoparticles can be able to absorb radiations. In general, the gold nanoparticles indicate the maximum wavelength in the range of 520 nm to 680 nm. Extracts of various barks of plants that were used for the project show the maximum wavelength in the above given range. It simply reveals that colloidal solution

formed after reducing HAuCl_4 with the help of extracts are showing formation of gold nanoparticles. From lots of citation, it is observed that the gold nanoparticles show band gap between the ranges of 1.82 eV to 2.4 eV. The following table includes all the results got from the UV-Visible Spectrophotometer technique.

Nanoparticles	Maximum wavelength (nm)	Band gap energy (eV)
NPB1	535	2.31
NPB2	530	2.33
NPB3	540	2.29
NPB4	540	2.29

Table 1. Maximum wavelength and band gap values of plants (bark/stem)

XRD-Diffraction

The X-ray diffraction (XRD) spectra were recorded by using instrument Bruker D8 Advance diffractometer with Cu/K α radiation light with a speed of scanning as (40 kV, 30 mA)

for a 6°/min in the 2 θ range of 10–85°. From various JCPDS data of gold nanoparticles, it is understood that gold nanoparticles revealed the highest intensity peaks at two theta values such as 38°, 44°, 64°, 78°, etc.

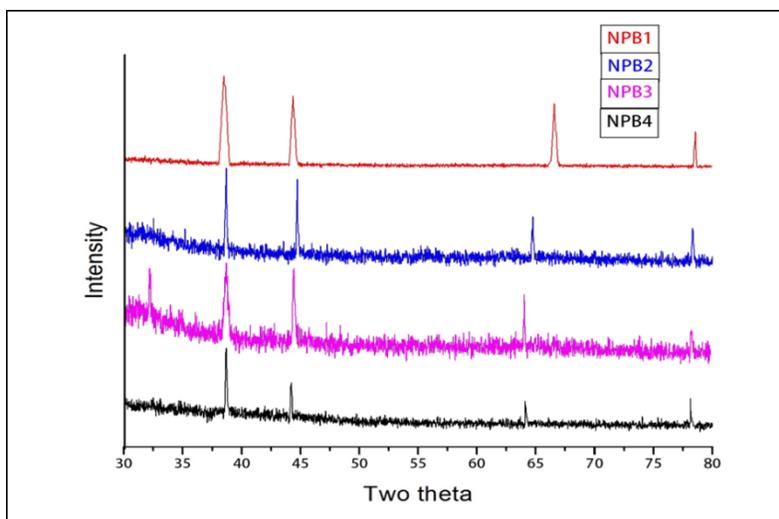


Figure 12. Results obtained for plants (bark/stem) from X-Ray Diffraction

The data obtained from XRD analysis is matched with the JCPDS file no. 86-3696 and some results are matched with the JCPDS file no. 01-1174. This data gives information about crystal structure and the nature of gold nanoparticles formed after reducing HAuCl_4 by biological extracts.

Conclusion

To achieve total green synthesis of Au NPs with plants extract were reported, the AuNPs preparation by using GAuNPs, NAuNPs, AAuNPs, and UAuNPs plants extract was achieved. Since all the plants (bark) showed FCC face centered cube structure as FCC as permitted reflection are (111), (200), (220), (311), and gold nanoparticles indicated the maximum wavelength in the range of 520 nm to 680 nm. The green synthesized AuNPs act as a good nanocatalyst and could be an attractive target for antimicrobial activity.

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Conflicts of Interest

The authors declared no conflict of interest.

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