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Original Research Article

Green Synthesis of Gold Nanoparticles using Extract of *Vitis vinifera,* Buchananialanzan, Juglandaceae, Phoenix Dactylifera Plants, and Evaluation of Antimicrobial Activity

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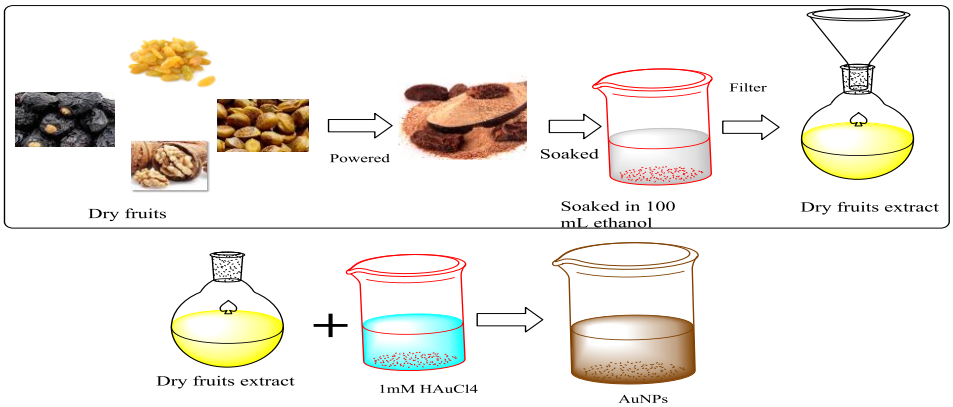
ABSTRACT

The establishment of nanotechnology, nanoparticles from gold (Au-NPs) is one of the extensive attention due to its essential properties. In this study, the green synthesis of gold NPs from the extract of Raisins, Charoli (Buchananialanzan), Walnut (Juglandaceae), and Black dates (Phoenix dactylifera) plants is reported. While all the (bark)show FCC face-centered cube structure as FCC as permitted reflection is (111), (200), (220), (311), the gold particles either show the cubic structure or face center cubic structure. The FT-IR spectra show the presence of natural products with functional groups hydroxyl, amine, amide, acid, and ester, respectively. The antimicrobial activity shows that the NPD3 shows well to moderate antimicrobial activity with *S. aureus, E. coli, B. subtilis, and P. aeruginosa organisms. The* NPD3 at 50 mg/mL concentration shows the 12mm, 11mm, 14m, and 14mm zone of inhibition.

GRAPHICAL ABSTRACT



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Introduction

The development of new nanomaterials in nanotechnology and nanoscience has emerged in recent years [1]. These technologies have attracted a range of fields, such as medicinal chemistry, pharmaceutical, material physics, nanochemistry, and sense, owing to their unique properties [2, 3]. The nonmaterial size ranges from 1–100 nm [4]. The application of nanotechnology is widely divided into all science research areas, including photochemistry, fluorescence technology, drug discovery, energy science, and optical flow applications [5]. The unique application of nanotechnology and its distinctive properties, such as extreme surface-to-volume proportion and elevated surface energy, especially predominantly mechanical, electrical, magnetic, thermal, and optical performance [6]. The application of gold nanoparticles is a broad research area, including sensing material, novel catalysis, electronic material, and medicinal products [7, 8]. The metallic nanoparticles have more importance and have a wide range of applications in broad areas [9]. In the last decade, considerable attention has been paid to nanotechnology, and the synthesis of metal nanoparticles, particularly AuNPs, has 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 conditions than other materials. In the literature, numerous techniques have been reported for preparing gold nanoparticles to activate their surface and upgrade their utilization.

The most common method for the preparation of Au nanoparticles is chemical reduction. However, the chemical reduction process usually requires additional 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. For the greener approaches synthesis, the functional group's hydroxyl, carbonyl, and aldehyde groups play an important role in reducing and stabilizing metal nanoparticles. In the past decade, numerous protocols have been reported that include the mechanical grinding method [11], microwave irradiation [12], and heat reduction [13].

The microwave radiation at 80 °C for 60 min synthesis of Lignin-AuNPs liquid marble to detecte Pb^{2+} [14]. The hemicellulose/lignin at 100 °C was used to synthesize Au-NPs [15]. Moreover, the greener lignin-based nanoparticles (LNPs) have been prepared to have LNPs acting as a reducing agent, stabilizing agent, and template properties [16, 17].

P. Elia et al. have reported the green synthesis of nanoparticles from the extract of *Salvia officinalis, Lippia citriodora, Pelargonium graveolens, and Punica granatum* [18].

Kar Xin Lee *et al*. reported the Extract of Garcinia mangostana Fruit Peel for the Au nanoparticles [19]. The ethanol-water of *Mimosa tenuiflora* (Mt) bark was used for the preparation of AuNPs at various metallic concentrations [20].

In this work, AuNPs were synthesized using an extract of Resins, Charoli (Buchananialanzan), Walnut (Juglandaceae), and Black dates (Phoenix dactylifera) plants. The green synthesis of Au NPs was conducted at room temperature and achieved by green chemistry principles. Our results were compared concerning similar works of catalysis with AuNPs synthesized by “green” methods.

Materials and Methods

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

of the Gold(III) chloride solution (0.001 M) with continuous stirring. The spectrum was scanned from 200 to 800 nm wavelengths. X-ray diffraction (XRD) measurements were carried out. 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 the material was subjected to analysis.

General method for synthesis of gold nanoparticles using dry fruits extract

Extract preparation

The plant Dry fruits, extract solution was prepared by taking 10 g of thoroughly washed and dried leaves. After accurately weighing the leaves, sample take extract by using 100 mL of ethanol and filter after 5 days. This solution or extract is used for further procedures.

Synthesis of gold nanoparticles

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

Flow sheet of steps involved in the synthesis

Resins, Charoli (Buchananialanzan), Walnut (Juglandaceae), and Black dates (Phoenix dactylifera) were used for extract preparation (Figure 1).

Preparation of extract from Resins NPD1

Dry fruit powder was added into a beaker containing 100 mL ethanol for extraction. The extract was filtered with the Whatman filter paper no. 41. This filtrate was used for further procedures.

Synthesis of gold nanoparticles

Different volumes (0.25-1.0 mL) of the dry fruit extract were added to 3 mL 1 mM solution of $HAuCl_4$ separately. After 6 hr, the color change was observed due to the formation of nanoparticles. UV-visible spectroscopy was done to confirm synthesized gold nanoparticles, and λ_{max} was calculated. Further advanced characterization was done with XRD and SEM.

Marathi name: Bedane

Dry fruit extract was prepared by taking resin into a beaker containing 100 mL ethanol for extraction. The extract was filtered with the Whatman filter paper no. 41. This filtrate was used for further procedure (Figure 2 and 3).

Synthesis of gold nanoparticles from resins extract

0.5 mL of the resin extract was added to 3 mL solution 1 mM of $HAuCl_4$ separately. After 6 hr, the color change was observed due to the formation of nanoparticles. UV-visible spectroscopy was done to confirm synthesized gold nanoparticles, and λ_{max} was calculated. Further advanced characterization was done with XRD and SEM (Figure 4).

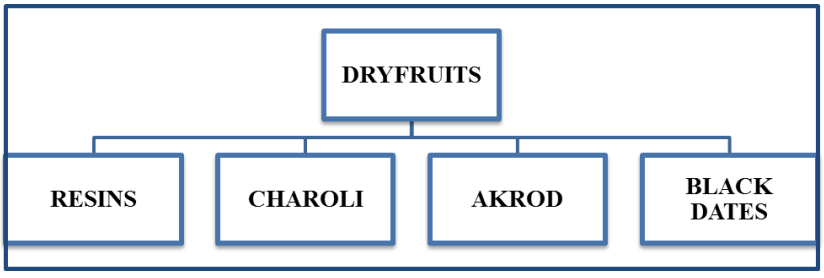

Figure 1: Dry fruits used for gold nanoparticle synthesis

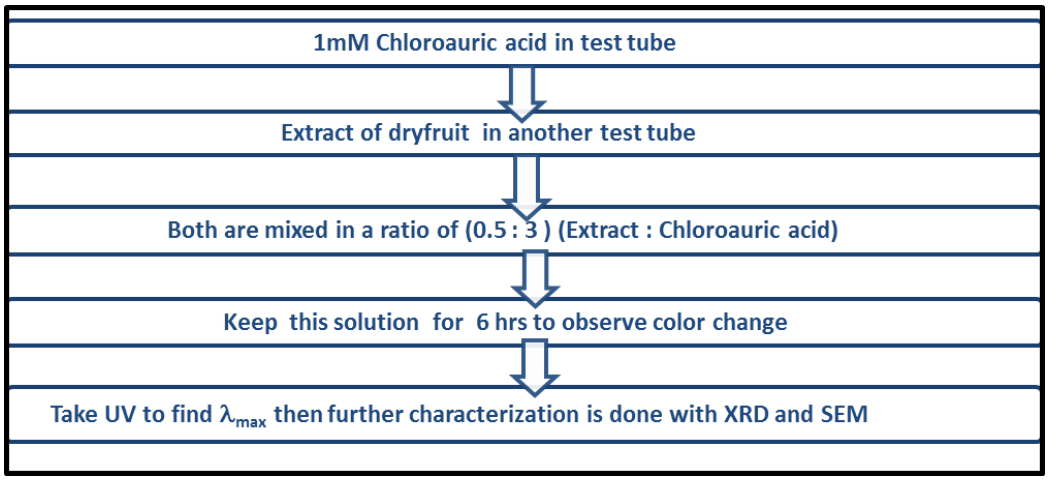

Figure 2: Synthesis route

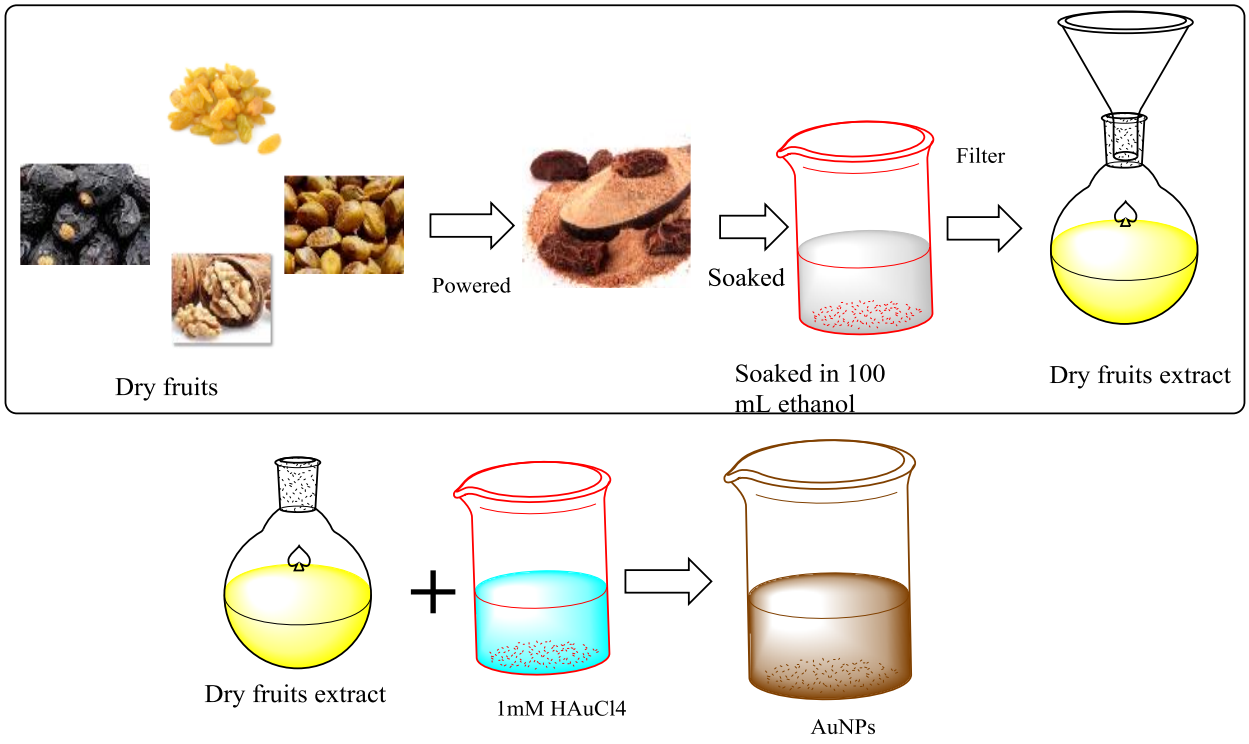

Figure 3: Schematic diagram for green synthesis of AuNPs using fruit extract

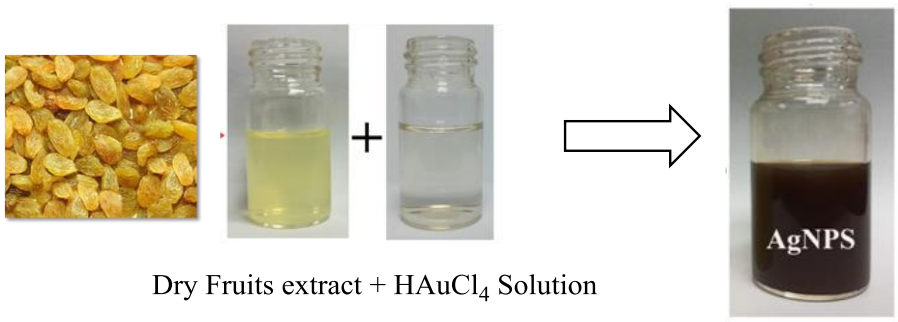

Figure 4: Images for the synthesis of NPs

Preparation of extract from Charoli NPD2

10 g Charoli (Buchananialanzan) dry fruit powder was added into 100 mL ethanol for extraction. The extract was filtered with the Whatman filter paper no. 41. This filtrate was used for further procedure.

Synthesis of gold nanoparticles

0.5 mL of the Charoli (Buchananialanzan) dry fruit extract was added to 3 mL solution 1 mM of $HAuCl_4$ separately. After 6 hr, the color change was observed due to the formation of nanoparticles. UV-visible spectroscopy was done to confirm synthesized gold nanoparticles, and λ_{max} was calculated. Further advanced characterization was done with XRD and SEM (Figure 5).

Preparation of extract from Walnut NPD3

Preparation of extract

10 g fine pieces of Walnut (Juglandaceae) dry fruit was added into 100 mL ethanol for extraction. This mixture is kept at room temperature and filtered after five days. This filtrate was used for further procedure. The extract is filtered with the Whatman filter paper no. 41. This filtrate is used for further procedures.

Synthesis of gold nanoparticles

0.5 mL of the Walnut (Juglandaceae) dry fruit extract was added to 3 mL solution 1 mM of $HAuCl_4$ separately. After 6 hr, the color change was observed due to the formation of nanoparticles. UV-visible spectroscopy was done to confirm synthesized gold nanoparticles, and λ_{max} was calculated. Further advanced characterization was done with XRD and SEM (Figure 6).

Preparation of extract from black dates NPD4

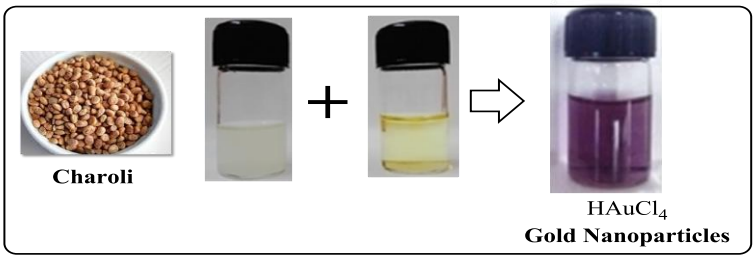

Figure 5: Images for synthesis of NPs

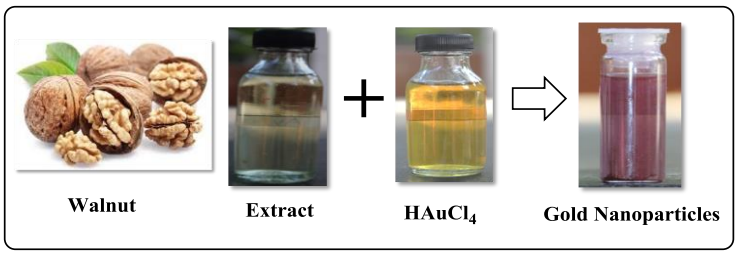

Figure 6: Walnut

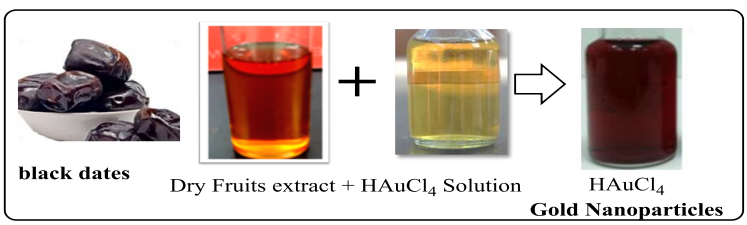

Figure 7: Black date

Preparation of extract

Black Dates (Phoenix dactylifera) fruit was added into 100 mL ethanol for extraction. The extract was filtered with the Whatman filter paper no. 41. This filtrate was used for further procedure.

Synthesis of gold nanoparticles

0.5 mL of the Black Dates (Phoenix dactylifera) dry fruit extract was added to 3 mL solution 1mM of $HAuCl_4$ separately. After 6 hr, the color change was observed due to the formation of nanoparticles. UV-visible spectroscopy was done to confirm synthesized gold nanoparticles, and λ_{max} was calculated. Further advanced characterization was done with XRD and SEM (Figure 7).

Results and Discussion

The four different medicinal plant extracts of the dried *Raisins (Vitis vinifera)*, Charoli (*Buchananialanzan*), Walnut (*Juglandaceae*), and Black dates (*Phoenix dactylifera*) were concentrated to obtain the paste and were subjected to a qualitative photochemical test to the standard procedures [26]. The dry fruits raisins are important and popular in all parts of the area in India as well as all around the country. The raisin has a high source of bioactive product that includes epicatechin, catechins, gallic acids, and procyanidins; these bioactive scaffolds are very beneficial for human health. The presence of glucose and fructose in the raisin makes it sweeter than grapes [27]. The second dry fruit *Buchanania lanzan* Spreng has a wide scope of bioactive compounds. *Buchanania lanzan* Marathi, name Charoli has important bioactive compounds which are useful for human health. *Buchanania lanzan* (Charoli) has saponins, coumarins, alkaloids, phenols, flavones, glycosides, and tannins present [28]. Walnut (*Juglandaceae)* has much more attention in human life because of the availability of bioactive components, including fatty acids like stearic, palmitic, and oleic acid), triglycerides, and sterols. Walnut (*Juglandaceae) is rich in* phenolic compounds and flavonoids such as quercetin, guaiacyl derivatives, catechin, and pinobanksin [29]. The date palm fruit (*Phoenix dactylifera*)'s the main function is to reduce human's triglyceride levels; this fruit is a source of fiber, vitamins, carbohydrates, minerals, and polyphenols compounds. Some species of Walnut has a rich source of important chemical component and useful bioactive compounds like ferulic acid, sinapic, 5-*O*-caffeoyl shikimic acid, and cinnamic acid analogs [30]. A large amount of natural presence in the selected dry fruits makes them important for preparing AuNps from their extract. During the synthesis of AuNPs, the extract of each dry fruit contains its respective natural product. In the presence of natural products in the extract of dry fruits, there are changes in the formation of adducts and coordination bonds between Au and bioactive compounds during the preparation of AuNPs. The spectral study and literature report indicate that the Au nano is always in the vicinity of the respective alkaloids. While the FT-IR spectra of the AuNPs show the particular band at 3253, 3093, 2933, 1708, 1634, 1545, 1355,1233, 1171 and 859 cm^{-1} for novel AuNPs NPD1, while the peak at 3282, 3093, 2933, 2854, 1712, 1634, 1545, 1400, 1212, 1169, 1063 and 836 for NPD_2. The FT-IR peak of 3457 and 3425 cm^{-1} belong to the free OH stretching vibrations of alcohol [29] (Figure 8 and 9). The AuNpPs revealed the presence of functional group carboxylate and phenolic (alcoholic) groups, which are responsible for the binding with the AuNPs.

The NPD2 which is synthesized from the Walnut (Juglandaceae) 3451, 3171, 3130, 3085, 2948,2883, 1631, 1509, 1467, 1434, 104, 886 cm-1 respectively. In this, the peak of 3451 cm^{-1} and 3171 cm^{-1} indicates the availability of OH and N-H functional groups (Figure 10). While the stretching frequency 1631 cm^{-1} belongs to

aromatic carbonyl frequency and 1509 cm^{-1} aromatic C-H stretching. While the NPD4 Black dates (Phoenix dactylifera) display the stretching peak at 3476, 3446, 3296,3276, 3107, 3057, 2957, 2871, 1615, 1518, 1475, 839 cm^{-1}. The stretching peak at 3476, 3446, 3296, and 3276 cm^{-1} indicate the presence of functional group N-H, OH, C-Halkyl, and COOH groups [30].

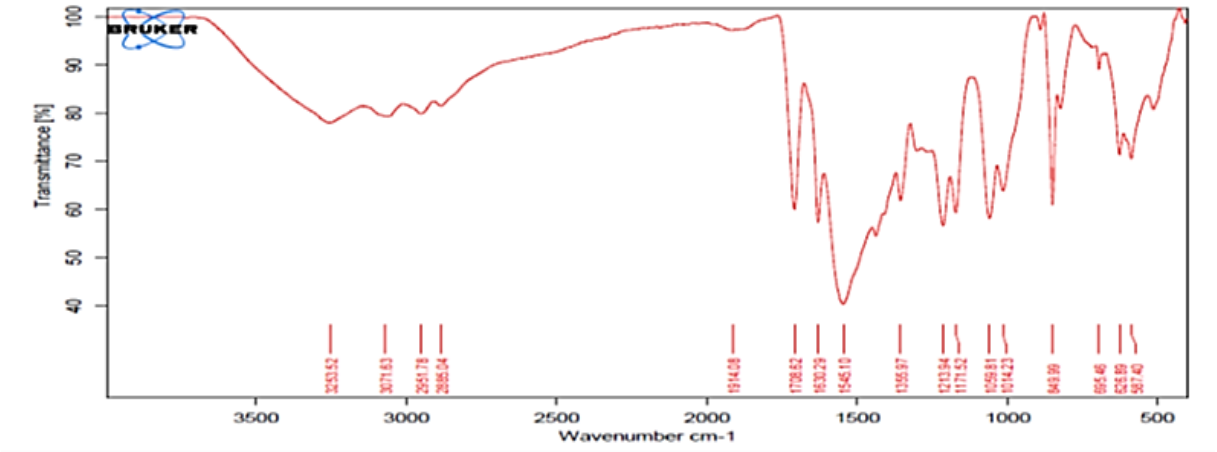

Figure 8: FT-IR spectra for novel AuNPs

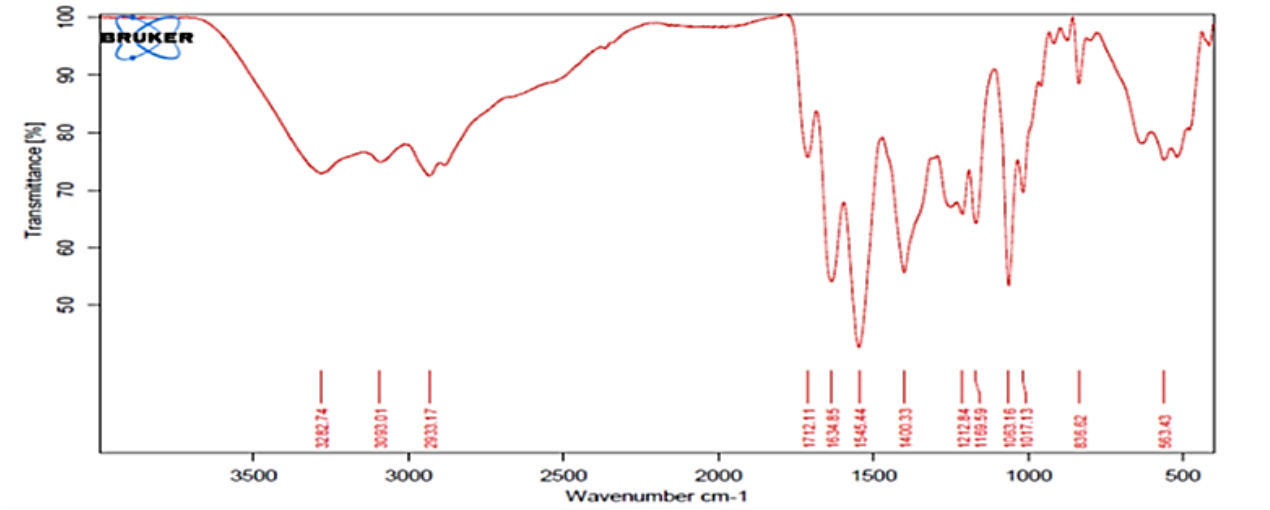

Figure 9: FT-IR spectra for NPD1

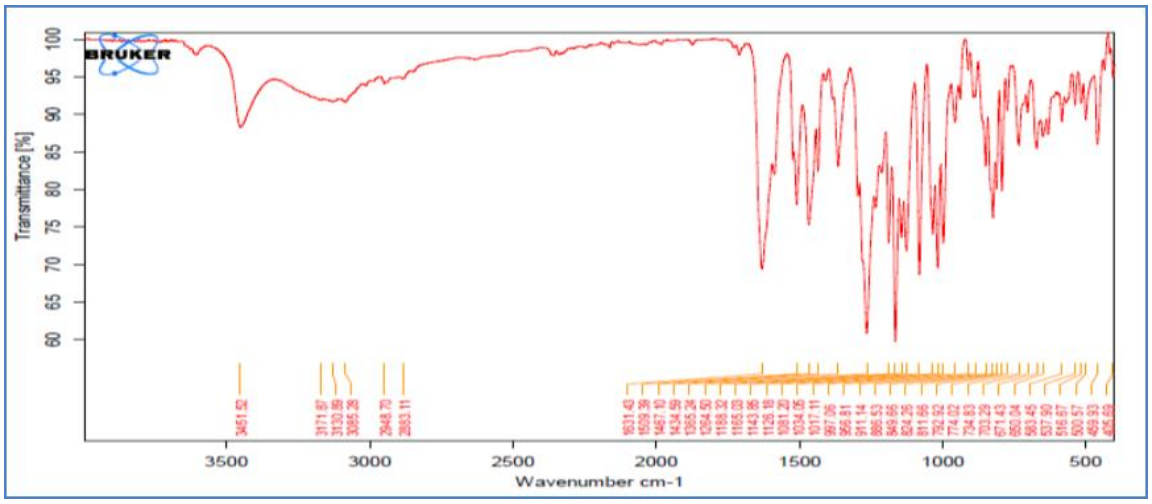

Figure 10: FT-IR spectra for NPD2

UV-VisSpectrophotometer Dry fruits UV-Visible spectrophotometer gave us the maximum wavelength at which gold nanoparticles can absorb radiations. In general, gold nanoparticles show maximum wavelength of 520 nm to 680 nm (Figure 11). The extracts of various plant barks used for the project show maximum wavelength in the range given above. It simply shows that colloidal solution formed after reducingHAuCl4 with the help of extracts showing the formation of gold nanoparticles. From much citation, it is observed that gold nanoparticles show a band gap in between the range of 1.82 eV to 2.4 eV (Table 1).

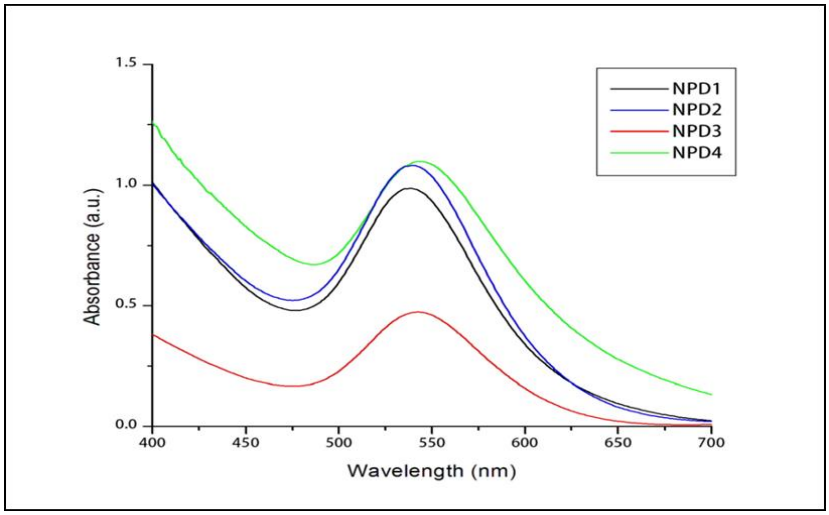

Figure 11: Results obtained for dry fruits from UV-Vis spectrophotometer

Table 1: The following table is consisting of all the results got from UV-Visible Spectrophotometer technique

Nanoparticles	Maximum wavelength (nm)	Band gap energy (eV)
NPD1	540	2.29
NPD2	540	2.29
NPD3	540	2.29
NPD4	545	2.27

X-Ray diffraction dry fruits

The unified small-angle X-ray scattering and diffraction measurement (XRD) was coupled with molecular dynamics to allow simultaneous determination of nanoparticles' shape, size, and crystallinity at the atomic scale. While all the (bark)show FCC face-centered cube structure as FCC as permitted reflection is (111), (200), (220), (311), the gold particles either show the cubic structure or face center cubic structure (Figure 12).

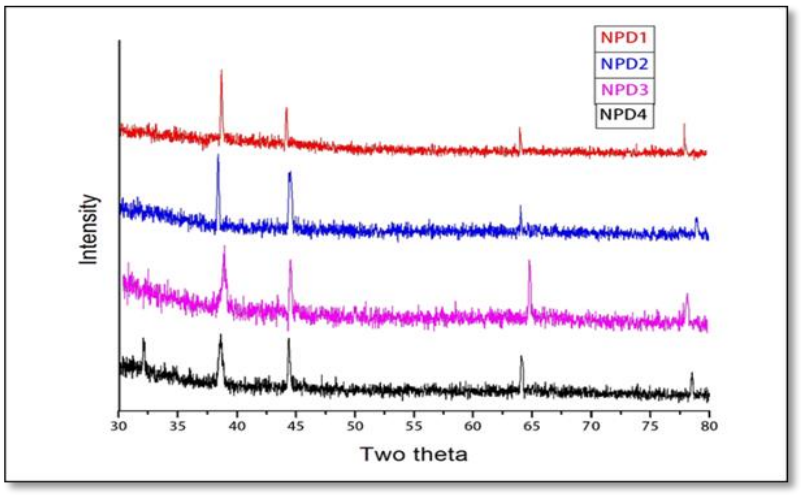

Figure 12: Results obtained for dry fruits from X-ray diffraction

Antimicrobial activity

The antimicrobial activity of GT AgNPs has been reported in several studies [30-38]. The antimicrobial activity of NPD1, NPD2, NPD3, NPD4, and AuNPs has been done with *Bacillus subtilis,* and *Staphylococcus aureus*, two-gram positive microorganisms and *Pseudomonas aeruginosa,* and *Escherichia coli, t*wo gram-negative microorganisms as waterborne pathogens. The varying concentration of test NPs Viz 10, 30, 50, 70, 100, and 150 µg/mL were applied for the research study. All the results are given in Table 2. and inhibitory concentrations are given in Table 2. as a zone of inhibitor.

The AuNPs from the NPD1, NPD2, NPD3, NPD4, and AuNPs have shown moderate antimicrobial activity as compared to the standard ciprofloxacin drug. The findings revealed that most of the compounds tested have good antibacterial activity. These bacteria were chosen because of their vast importance in the clinical field, as they cause various diseases and their various antibiotic and chemical drug resistance. Table 2 reveals that the produced compounds have biological activity against the bacteria because they may suppress the bacteria by varying the amounts of the compounds.

The series of concentrations were used for the analysis; the NPD1 shows moderate activity again the *S. aureus, E. coli, B. subtilis, and P. aeruginosa (*Table 2*).* while the nanoparticles synthesized from the Black dates (*Phoenix dactylifera*) shoes some moderate to very good antibacterial activity. The NPD3 can inhibit the growth or microorganism at 50, and 70 mg/ml concentration of NPD3 inhibits growth and show the zone of inhibitions about 12 mm for *E. coli* zone of inhibitions is 11 mm, for *B. subtilis shows* 14 mm and for P. aeruginosa 14 mm. the antibacterial activity of synthesized nanoparticles increased from the increasing concentration of dose. These results suggest that the NPS has various antibacterial activities.

Table 2: Zone inhibition of AuNPs and standard against *S. aureus, E. coli, B. subtilis and P. aeruginosa*

Test sample)	Concentration (µg/mL)	Zone of inhibition (mm)			
		S. aureus	*E. coli*	*B. subtilis*	*P. aeruginosa*
NPD1	10	-	-	5	2
	30	5	7	--	5
	50	10	8	9	8
	100	11	13	12	12

NPD2	10	2	4	3	2
	30	4	7	6	5
	50	7	10	7	8
	70	10	11	10	11
	100	14	14	13	15
NPD3	10	3	2	3	4
	30	9	6	9	8
	50	9	7	12	10
	70	12	11	14	14
	100	14	15	16	15
NPD4	10	5	4	5	5
	30	8	6	8	7
	50	11	9	11	11
	70	14	13	13	13
	100	16	14	15	15
Ciprofloxacin	50	25	26	22	23

Conclusion

Green synthesis of Au NPs with plant extract was reported to be achieved successfully. The preparation of green AuNPs was achieved by using Resin, Charoli (Buchananialanzan), Walnut (Juglandaceae), and Black dates (Phoenix dactylifera). The synthesized AuNPs NPD1, NPD2, NPD3, NPD4, and AuNPs have been characterized with FT-IR, X-Ray Diffraction, and **UV-**Vis Spectrophotometer shows the desired peak for AuNPs. The antimicrobial activity of all the AuNPs has been done with Bacillus *subtilis* and *Staphylococcus aureus*, two-gram positive microorganisms, *Pseudomonas aeruginosa,* and *Escherichia coli,* two gram-negative microorganisms as water-born pathogens. These findings revealed that the majority of the compounds show good to very good antibacterial activity.

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Authors' contributions

All authors contributed to data analysis, drafting, and revising of the paper and agreed to be responsible for all the aspects of this work.

Conflict of Interest

There are no conflicts of interest in this study.

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