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Research Article

Green Synthesis of Gold Nanoparticles Using Aqueous Extract of *Garcinia mangostana* Fruit Peels

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The synthesis of gold nanoparticles (Au-NPs) is performed by the reduction of aqueous gold metal ions in contact with the aqueous peel extract of plant, *Garcinia mangostana* (*G. mangostana*). An absorption peak of the gold nanoparticles is observed at the range of 540–550 nm using UV-visible spectroscopy. All the diffraction peaks at $2\theta = 38.48^{\circ}$, 44.85°, 66.05°, and 78.00° that index to (111), (200), (220), and (311) planes confirm the successful synthesis of Au-NPs. Mostly spherical shape particles with size range of 32.96 ± 5.25 nm are measured using transmission electron microscopy (TEM). From the FTIR results, the peaks obtained are closely related to phenols, flavonoids, benzophenones, and anthocyanins which suggest that they may act as the reducing agent. This method is environmentally safe without the usage of synthetic materials which is highly potential in biomedical applications.

1. Introduction

Nanotechnology is technology that deals with nanoscale materials range from 1 to 100 nm and their applications [1]. Among different type of nanomaterials, noble metal nanoparticles gained considerable attention due to their special catalytic, electronic, and optical properties [2]. Gold nanoparticles (Au-NPs) have been widely investigated due to their uniqueness especially in biomedication [3]. The multiple surface functionality of Au-NPs ease nanobiological attachment of Au-NPs with drug [4], oligonucleotides [5], antibodies [6], and protein [7]. The optical property of Au-NPs also enables them to play a role in bioimaging by acting as marking agent [8].

Despite the popularity and development of synthesizing nanoparticles using chemical and physical methods, the need to establish environmental friendly methods that do not involve the usage of toxic chemicals is crucial especially in medical purpose [9]. Synthesis method that uses natural products as reducing agents need more focus to reduce the hazards on environment and human. Greener substrates such as enzyme [10], fungus [11], and algae [12, 13] were reported successfully in the production of Au-NPs. However, as compared to the difficulties faced in microbe assisted synthesis [14], plant mediated synthesis is developing due to the ease to handle and to control the size and shape of nanoparticles. Plant-based synthesis is relatively fast, safe, and light and works under room condition without the needs of high physical requirements [15]. Every part of the plant is proved to be useful especially the leaves [16–19], but few reports are targeted on the fruit peels [20].

Garcinia mangostana (*G. mangostana*) which is commonly known as mangosteen belong to the family of Guttiferae. It can grow up to 6–25 m in height and is mainly cultivated in Southeast Asian countries such as Indonesia, Malaysia, Thailand, Philippines, and Sri Lanka. Traditionally, mangosteen has been used as medicine to treat abdominal pain, dysentery, diarrhoea, infected wound, and chronic ulcer [21]. Secondary metabolites such as phenolic acid, flavonoids, alkaloids, and terpenoids that are contained in plant crude extract are involved in the reduction of nanoparticles [15]. *G. mangostana* here contains high level of phenolic compound, namely, xanthone, especially in its pericarp (peels) [22]. There are more than 30 xanthones isolated from *G. mangostana* where the major constituents are α-mangostin and γ-mangostin [23]. This phenolic compound possesses

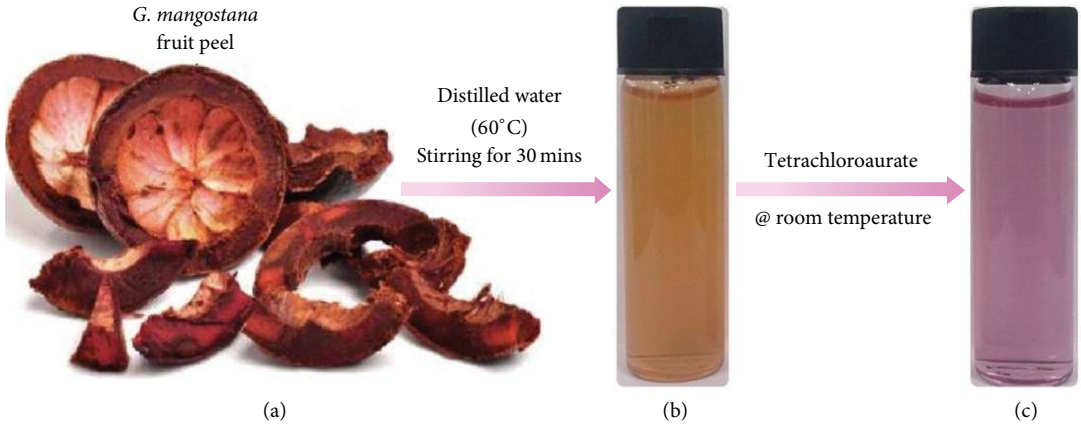

FIGURE 1: Photos of the colour changes after the addition of tetrachloroaurate where (a) is the mangosteen peels, (b) pure mangosteen extract, and (c) Au-NPs solution after the reaction with dilution.

antioxidant, antitumour, antiallergic, and antiviral properties where there are researches that shows that α-mangostin and γ-mangostin are high potential antioxidants [24] which are believed to take part in the synthesis reaction of Au-NPs [25]. Also, different kind of flavonoids, benzophenones, and anthocyanins present in the plant may be involved closely in the reduction of nanoparticles [26].

To the best of our knowledge, there is no work reported on adopting *G. mangostana* in the synthesis of Au-NPs or any other metal nanoparticles. Here, we demonstrate the biosynthesis and characterization of Au-NPs by using tetrachloroaurate and aqueous extract of *G. mangostana* fruit peel.

2. Methods

2.1. Chemicals. *G. mangostana* fruits were collected from Terengganu, Malaysia. Analytical grade tetrachloroaurate salt ($HAuCl_4$, 99.98%) was purchased from Sigma-Aldrich, USA, and used as gold precursor. All reagents used were of analytical grade. All aqueous solutions were prepared using distilled water. All glassware used was cleaned and washed with distilled water and dried before used.

2.2. Preparation of Aqueous G. mangostana Fruit Peel Extract. The peels were washed thoroughly with tap water to remove dirt and washed again with distilled water before being dried in oven (Esco Isotherm Forced Convection Laboratory Oven) at 40°C. All the peels were ground into fine powder using an electric blender (Panasonic) and stored at room temperature for further use. The extract was prepared by taking 0.50 g of the fine powder with 20 mL distilled water and boiled at 60°C for 30 mins. The crude extract was filtered with Filtres Fioroni 601 filter paper.

2.3. Synthesis of Au-NPs. In a conical flask, 20 mL of the peel extract was reacted with 10 mM of tetrachloroaurate at room temperature under static conditions. The colour change of the reaction was observed and the time taken for the changes was noted. The solution colour changes immediately from pale brownish to purple colour indicating the formation of [Au/*G. mangostana*]. The Au-NPs nanoparticles emulsion obtained was kept at 4°C.

2.4. Characterization of Au-NPs. The reduction of Au-NPs was confirmed by using UV-vis spectroscopy at regular intervals in the range of 300 to 1000 nm (Shimadzu, UV-1800 UV-VIS Spectrometer). The nanoparticles emulsion was oven dried at 40°C for one day. The dried sample was collected and examined for the structure and composition using powder X-ray diffraction spectroscopy. The data was recorded using PANalyticXPert Pro (λ = 0.15406 nm) at 45 kV and 20 mA. The dried sample was scanned in the range of 2θ = 10–80° with 2°/min. Transmission electron microscopy (TECNAI, G^2 F20) was used to investigate the size and morphology of the Au-NPs using SC1000 Orius CCD camera. The stability of Au-NPs was measured using Particulate Systems Nano-Plus Zeta/Nano Particle Analyser, Japan. The bioreduction compounds that are responsible for the reaction were determined using Fourier Transform Infrared spectroscopy. The spectrum was obtained by Thermo Scientific Nicolet 6700 system with 16 scans per sample at the range of 550–4000 cm^{-1}.

3. Results and Discussion

G. mangostana peel extract (0.50 g, 20 mL) acts as both the reducing and stabilizing agent and $HAuCl_4$ (10 mM) acts as the gold precursor. The reduction of $HAuCl_4$ was indicated by the colour changes of *G. mangostana* extract as shown in Figure 1. The reaction was rapid as the pale brownish colour of the *G. mangostana* peels extract turns into purple colour within 3 min indicating formation of Au-NPs [27].

The possible chemical equations for synthesizing the Au-NPs are

$$HAuCl_4\ (aq) + G.\,mangostana \xrightarrow{\text{Stirring at Room Temp.}} [\text{Au}/G.\,mangostana] \quad (1)$$

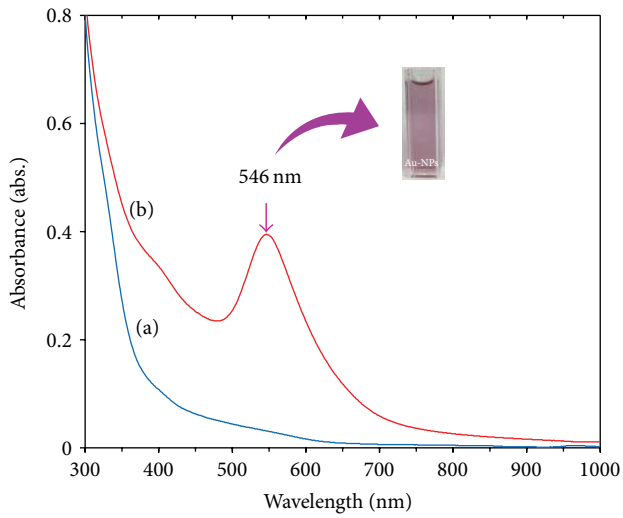

FIGURE 2: UV-vis absorbance bands for (a) pure *G. mangostana* peels extract and (b) Au-NPs forms using *G. mangostana* peels extract.

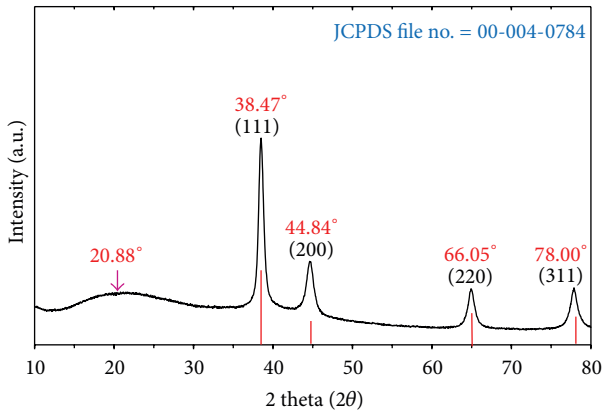

FIGURE 3: XRD spectra for Au-NPs forms using *G. mangostana* peels extract and the intensity peak of the reference peak.

After dispersion of $HAuCl_4$ in the *G. mangostana* aqueous solution matrix, the extract was reacted with the functional groups of *G. mangostana* components to form [Au/*G. mangostana*] [28].

3.1. UV-Visible Spectroscopy Study. The presence of Au-NPs is confirmed by UV-vis spectra in Figure 2. The results showed that there is no obvious peak for *G. mangostana* peel extract. However, after the addition of tetrachloroaurate, a sharp peak appears at the range of 540–550 nm [29]. It is further confirmed by other characterizations that this peak indicates the formation of monodispersed spherical shape Au-NPs. The reaction takes place within 3 minutes with obvious colour change.

3.2. X-Ray Diffraction Analysis. Powder X-ray diffraction pattern in Figure 3 shows that the Au-NPs synthesized is in crystalline structure. The spectrum gives an intense peak at $2\theta = 38.47^\circ$, 44.84°, 66.05°, and 78.00° which correspond to the (111), (200), (220), and (311) plane proving the structure of Au-NPs to be face center cubic (fcc). The crystallinity of Au-NPs is pure by comparing its XRD pattern with the database; JCPDS file number 00-004-0784 [30]. However, there is shifting of the peaks with database where crystal structure of pure metallic Au-NPs is present [9]. The particle size of Au-NPs can be estimated using the Debye-Scherrer equation,

$$d = \frac{k\lambda}{(\beta \cdot \cos\theta)}, \tag{2}$$

where d is the average crystallite size, k is the Scherrer constant (0.9), λ is the X-ray wavelength (0.154 nm), β is the line broadening in radians, and θ is the Bragg angle [31]. By using the Scherrer equation, 16 nm is calculated to be the average crystallite size of the Au-NPs.

3.3. Transmission Electron Microscopy and Field Emission Scanning Electron Microscopy Study. The size and the morphology of the Au-NPs synthesized was investigated using the TEM which is represented by Figure 4. The Au-NPs is well dispersed with *G. mangostana* matrix surrounding it indicating that *G. mangostana* matrix acts as the capping agent to separate the Au-NPs from aggregation. The average size of the Au-NPs synthesized is 32.96 ± 5.25 nm with mostly spherical and some hexagonal and triangular shape. The dispersity of Au-NPs is further supported by the FESEM where no aggregation occurs and also the nanoparticles produced are encapsulated by the matrix of *G. mangostana*.

3.4. Zeta Potential Study. The stability of Au-NPs was performed using zeta potential. A zeta value of ±30 mV is needed for a suspension to be physically stable while ±20 mV is necessary for a combined electrostatic and steric condition [32]. The zeta potential results for pure *G. mangostana* peel extract are −14.68 mV, whereas the reading of Au-NPs formed using the extract reduced to −20.82 mV (Figure 5). Thus, Au-NPs formed show an acceptable stability with reading not less than the required stable expression.

3.5. Fourier Transform Infrared Spectroscopy Study. FTIR spectroscopy was carried out to determine the potential functional groups that are responsible for the reduction of Au-NPs. Figure 6 shows the spectra obtained from pure *G. mangostana* peel extract and Au-NPs synthesized using the *G. mangostana* peels extract. The major stretching appearing at 3000–3500 cm^{-1} indicates the presence of O-H stretch which signifies the presence of phenols, flavonoids, benzophenones and anthocyanins [2]. A little shifting occurs here, suggesting that the carbonyl group in the peel extract capped and stabilized the Au-NPs [33]. Aside the O-H stretching, at the region of 2919 cm^{-1} and 2914 cm^{-1} the presence of C-H bond in xanthone [34] and other compounds in the peel extract is significant. Bands of C-H bond from *G. mangostana* peel extract split into two, 2914 cm^{-1} and 2845 cm^{-1}, suggest that, after the formation of Au-NPs, the transmittance changed [35], while at the region of 1700 cm^{-1} it shows the presence

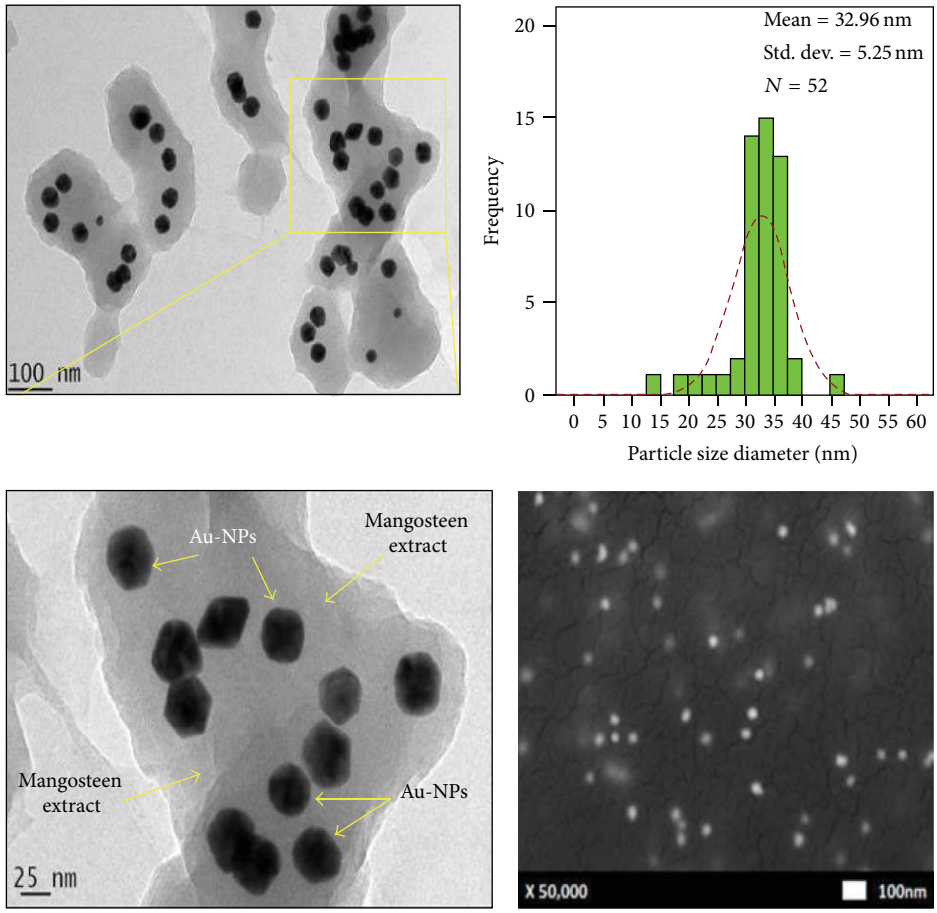

Figure 4: TEM and FESEM image of the Au-NPs forms using *G. mangostana* peel extract at magnification of 26.5kx and the histogram of the distribution of the particles size of Au-NPs.

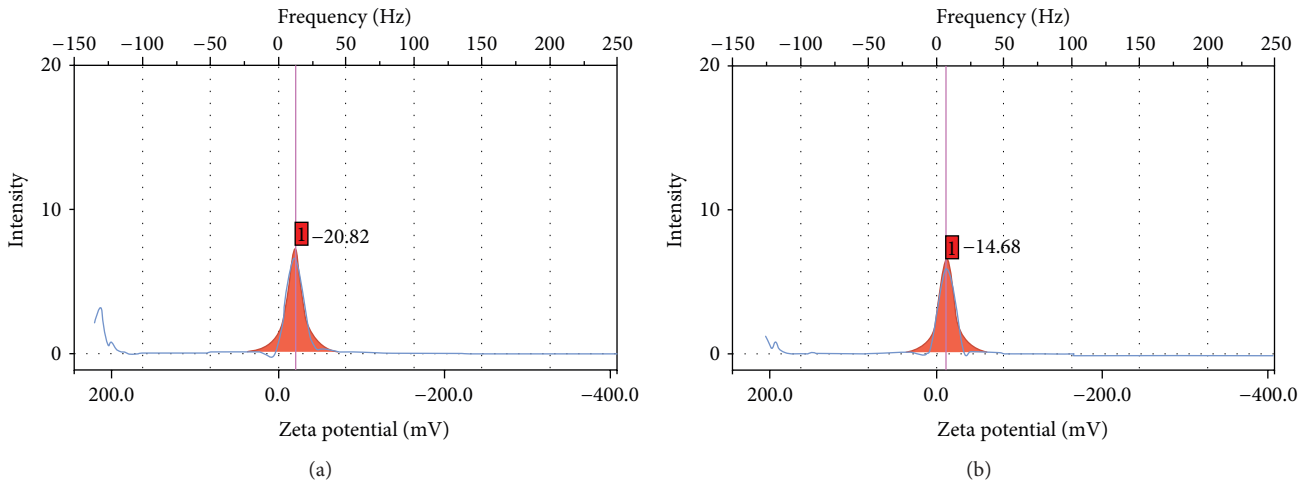

Figure 5: Zeta potential image of (a) pure *G. mangostana* peel extract and (b) Au-NPs form by using *G. mangostana* peel extract.

of C=O stretching [36]. At the region of 1600–1500 cm^{-1}, C-C in ring aromatic bond also suggests the presence of aromatics structure exists in the *G. mangostana* extract. The C-C aromatics stretch is observed for both spectra at the region of 1500–1400 cm^{-1} which is relevant to the aromatic backbone that can be found mainly in the pericarp of *G. mangostana*. Finally, C-O-C stretch can be found in the range of 1300–1000 cm^{-1} where shifting occurs from 1279 cm^{-1} to 1234 cm^{-1} after capping with Au-NPs [37]. All the above results are matching with xanthone [38], flavonoids [26],

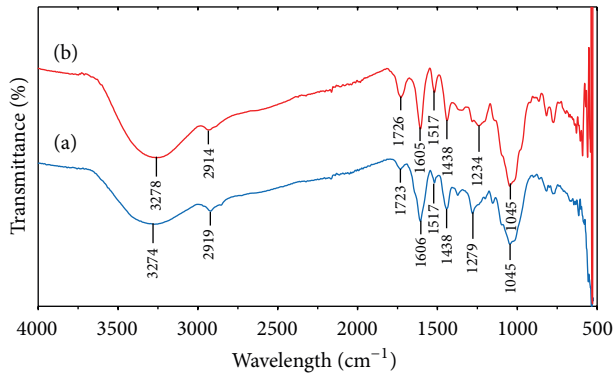

Figure 6: FT-IR graphs of (a) pure *G. mangostana* peel extract and (b) Au-NPs form by using *G. mangostana* peel extract.

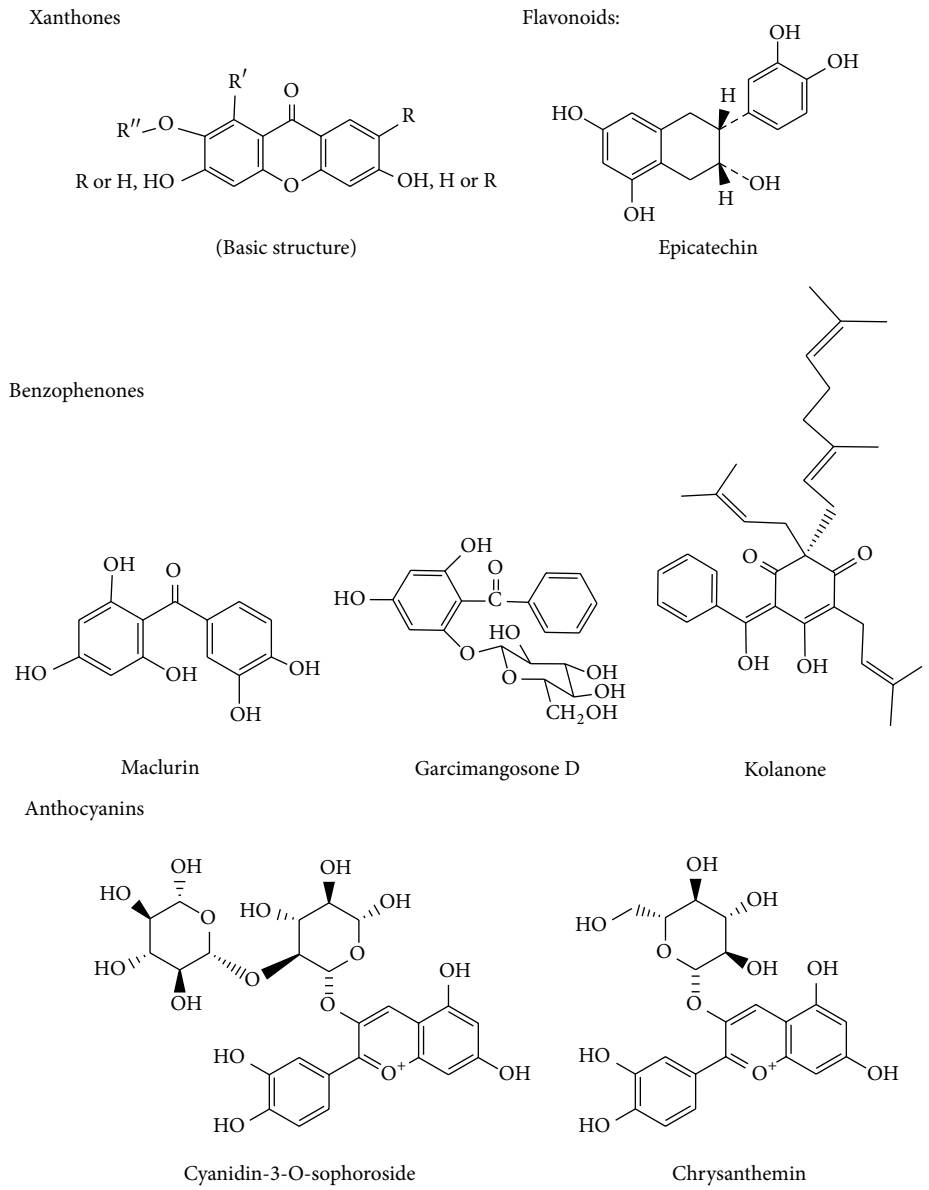

Figure 7: Compounds that *Garcinia mangostana* pericarp contains based on literature.

and other compounds that derived from the pericarp of *G. mangostana* as shown in Figure 7 suggesting that they are involved closely in the reduction and stabilization of $HAuCl_4$ to Au-NPs where the presence of oxygen atoms helped in absorption of compounds on Au-NPs [24, 39, 40].

4. Conclusion

This study gives an environmental favourable approach of the synthesis of Au-NPs using *G. mangostana* peel extract. The extract demonstrates that the properties of both reducing and stabilizing agent owe to the presence of different compounds in the pericarp of *G. mangostana*. The usage of peels from the plant takes full advantage of unwanted waste material which is economically friendly, efficient, and safe. No study is established before with the usage of *G. mangostana* for the production of Au-NPs. The synthesized Au-NPs are potential to be applied in biomedical and other applications where nontoxicity is crucial.

Competing Interests

The authors declare that they have no competing interests regarding the publication paper.

Acknowledgments

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