Research Article

Synthesis of Various-Shaped Gold Nanoparticles by Using Extracts of Cascara and Coffee Flowers and Checking the Possibility of Being Applied as a Chemical Sensor

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Abstract
This study investigates the green synthesis of morphology-controlled gold nanoparticles by using the extracts of cascara, Arabica and Robusta coffee flowers. These nanoparticles, along with chemically synthesized CTAB-capped gold nanorods, were subsequently evaluated as sensors for NH_4OH. The extracts were prepared by immersing 2 g of powders in 100 mL of water at 80 °C. It was found that the reducing power and total phenolic content of Robusta samples were the highest, followed by those of Arabica and Cascara samples. The gold nanoparticles on the synthesis day were found spherical. For samples using the extracts with some dilution ratios and $HAuCl_4$ concentrations, the particles grew to various shapes, such as triangles and polygons, after being kept for one week in a refrigerator. The localized surface plasmon resonance (LSPR) results of the various-shaped gold nanoparticles showed two peaks. The gold nanospheres with a diameter of 20.54 ± 5.04 nm, polygonal and triangular gold nanoparticles with sizes of 53.02 ± 22.86 and 92.08 ± 39.12 nm, respectively, and a CTAB-capped gold nanorod with a length of 108.68 ± 6.8 nm were used in sensing studies. The second peak of various-shaped gold nanoparticles had a sensitivity comparable to that of CTAB-capped gold nanorods, which was higher than that of gold nanospheres. Therefore, the various-shaped gold nanoparticles synthesized from the extracts were potentially applied as a chemical sensor.

Keywords: Cascara, Coffee flowers, Gold nanoparticles, Localized surface plasmon resonance, Nanorods, Seed-mediated method

1 Introduction

Nanoparticles such as copper [1] and gold nanoparticles [2] have been utilized in many fields because of their small size, making them have different properties from those of larger-sized materials. Gold nanoparticles (AuNPs) have gained popularity in various applications. For instance, in medical fields, they are used in drug delivery [2] and chemical sensing systems [3]. When stimulated by light of various wavelengths, metallic nanoparticles exhibit phenomena called localized surface plasmon resonance (LSPR). Nanoparticles of different types of metals exhibit the absorbance peaks at different ranges of wavelength. For gold nanoparticles, the absorbance peaks were seen in the range of 520–550 nm. The nanoparticles with a larger average size show an absorbance peak at a higher wavelength [4]. The shift of the absorbance peak [5] and the change of absorbance intensity of gold nanoparticles when they are in contact with chemicals make them possible to apply as chemical sensors.

Many synthesis methods have been employed to prepare AuNPs, such as the template method, electrochemical method, photochemical method, and seed-mediated method [6]. The seed-mediated method is known for its simplicity and high-yield production of gold nanorods (AuNRs). In this method, sodium borohydride ($NaBH_4$) is used as a reducing agent and cetyltrimethylammonium bromide (CTAB) as a surfactant and capping agent to generate the seeds. The growth solution is prepared from ascorbic acid (AA) as a reducing agent and CTAB. The seed solution is then added to the growth solution and the

mixture is left for many hours for the gold nanoparticles to grow into nanorods [7]. They exhibit the unique LSPR results composed of two absorbance peaks, typically corresponding to transverse and longitudinal surface plasmon modes.

In the past, chemical substances were mainly used for synthesizing nanoparticles, especially in the seed-mediated method above. However, recently, there has been a growing trend towards green synthesis in which natural substances have been used more extensively because they are environmentally friendly and pose no harm to living organisms. Many substances have been investigated for the synthesis of AuNPs, such as sweet oil, biota, and pink morning glory [8].

One of the interesting plants for this purpose is coffee. In 2023, coffee consumption reached up to 44.2 million kilograms per year and tended to increase year after year [9]. Additionally, there is a growing variety of coffee consumption. Usually, coffee consumption mainly involves using just the coffee beans, leading to a large quantity of natural wastes, including spent coffee grounds from various brewing methods, coffee flowers from the coffee harvesting process, and coffee shells or cascara from consuming coffee beans. Previously, the extracts from coffee grounds were used to synthesize AuNPs and the effect of coating with various polymers was investigated to enhance their stability [10]. In addition, the extracts of coffee flowers have been recently used to synthesize AuNPs and their photocatalytic activity was investigated [11]. In both research works, it was observed that AuNPs mostly exhibited spherical shapes, with some having rod-shaped and triangular shapes. As for cascara, there has been no report on using it in the synthesis of nanoparticles before.

In this work, various-shaped gold nanoparticles were of interest and they should depend mainly on the growth step, like in the chemical synthesis using CTAB. However, in the synthesis of gold nanoparticles using the extracts of cascara, Arabica and Robusta coffee flowers, the seed preparation step was done in a microwave and then the mixture was immediately taken to a refrigerator and kept for a week for the growth step, in which various shaped-AuNPs should be evidently developed. They are called AuNPs throughout this article, whereas nanoparticles with spherical shapes are called gold nanospheres (AuNSs). In addition, this work later compared the sensing ability of these various-shaped nanoparticles with that of the gold nanorods (AuNRs) from the CTAB method.

2 Materials and Methods

2.1 *Materials*

Dried cascara, dried Arabica coffee flowers and dried Robusta coffee flowers were purchased from online stores, namely HONEY HOME, Khun Lao Coffee, and AU2519, respectively. Gold (III) chloride trihydrate ($HAuCl_4 \bullet 3H_2O$), as a precursor of the gold ion, polyethylene glycols with MW of 6000 Da were supplied by Sigma Aldrich CO., USA. Cetyltrimethyl ammonium Bromide (CTAB), and sodium borohydride ($NaBH_4$) were supplied from Loba Chemie Pvt Ltd., India. Silver nitrate ($AgNO_3$) and hydrochloric acid were supplied by Merck Ltd., Germany. L-ascorbic acid was supplied by ChemSupply, Australia. Phosphate buffer saline (PBS) was supplied by AMRESCO Inc., USA. Potassium ferricyanide and sodium carbonate were purchased from Kemaus Pty Ltd., Australia. Trichloroacetic acid was supplied by ITW Reagents Inc., Germany. Ferric chloride anhydrous and Folin & Ciocalteu Reagent were supplied by Sisco Research Laboratories Pvt. Ltd., India. Sodium chloride was purchased from Ajax Finechem Pty Ltd., Australia. Ammonium hydroxide was supplied by J.T. Baker.

2.2 *Instruments*

The absorbance spectra of the synthesized gold nanoparticles were measured using UV-Vis spectroscopy (model UV-5100, Metash). The collection of gold nanoparticles was performed with a microcentrifuge (Wise spin CF-10). The mixing of substances was done with an analog vortex mixer (Model VXMNAL, OHAUS). The mixing of the solution in the seed and growth steps was done with a hot plate stirrer (IKA C-MAG HS7). The sensor applications were studied using an ultrasonic bath (GT Sonic). Furthermore, the morphology of the synthesized gold nanoparticles was characterized by transmission electron microscopy (JEOL, JEM-2010).

2.3 *Preparation of the extract solutions of cascara and coffee flowers*

2 g of dried cascara, Arabica coffee flowers, and Robusta coffee flowers were separately immersed in

100 mL of hot water at 80 ºC for 10 min. The extracts were then diluted with RO water at various extract to water volume ratios of 60:40, 50:50, 30:70, and 20:80.

2.4 *Determination of total phenolic content (TPC)*

0.75 mL of Folin-Ciocalteu reagent was diluted with RO water and mixed with 0.1 mL of the extract solution at various ratios. The mixture was rigorously mixed using a vortex mixer and left standing for 5 min. After that, 0.75 mL of 10% sodium carbonate solution and 10 mL of RO water were added. It was mixed thoroughly and allowed to stand at room temperature for 90 min and then the absorbance was recorded at a wavelength of 725 nm, and compared with the calibration curve of gallic acid solution for the determination of TPC in mg/L gallic acid equivalence (GAE).

2.5 *Determination of reducing power*

1 mL of the extract solution at various dilution ratios was mixed with 2.5 mL of sodium phosphate buffer and 2.5 mL of 1% potassium ferricyanide solution. The sample was incubated at 50 ºC for 30 min and 2.5 mL of 10% trichloroacetic acid was added. The mixture was centrifuged for 10 min. 5 mL of supernatant was sampled and mixed with 5 mL of RO water and 1 mL of 0.1% ferric chloride solution. The sample was left in the dark at room temperature for 10 min and absorbance was measured at 700 nm, and then compared with the calibration curve of gallic acid solution for the determination of reducing power, also in mg/L GAE.

2.6 *Synthesis of gold nanoparticles using the extracts*

2.5 mL of the extract solution at various concentrations upon dilution with water was mixed with 2.5 mL of $HAuCl_4$ solutions at different concentrations (0.5, 1.0, 1.5, 2.0, and 2.5 mM). Each mixture was heated in a microwave oven at 800 W for 7 s until the color changed to purple or reddish-purple. After that, all samples were kept in a refrigerator for a week. Both the freshly synthesized gold nanoparticles colloids and those after being cooled for a week were checked with a UV-Vis spectrophotometer using the scanning range from 340 to 940 nm, which is the maximal limit of the spectrophotometer and they were collected after centrifugation at 10,500 rpm for 10 min. A chosen sample of gold nanoparticles was also investigated with a transmission electron microscope (TEM). The statistics were obtained from 100 sampled particles from many TEM figures.

2.7 *Synthesis of gold nanorods using chemicals and CTAB*

The samples were prepared using the seed-mediated method. For the synthesis of seed solution, 5 mL of CTAB solution at different concentrations (0.05, 0.075, 0.1, 0.2, and 0.3 M) was mixed with 5 mL of $HAuCl_4$ at a fixed concentration of 0.5 mM and then stirred for 15 min. 0.6 mL of ice-cold $NaBH_4$ solution at 0.01 M was added to the mixture. The solution was stirred for 2 min until it appeared pale brown-yellow in color, and then it was wrapped in a piece of aluminum foil and kept at room temperature for 3 h. This synthesized seed solution was used afterwards.

The growth solution was prepared by mixing 0.2 mL of $HAuCl_4$ solution at 0.05 M and 0.2 mL of $AgNO_3$ solution at 0.01 M, followed by the addition of 0.16 mL of freshly prepared L-ascorbic acid solution at 0.1 M. The mixture was mixed gently until it became colorless, where Au^{3+} ions were reduced to Au^{+} ions. After that, 0.4 mL of HCl solution at 1 M was added. The solution was stirred using a magnetic stirrer for 2 min, and then 25 µL of the seed solution was added. Subsequently, the mixture was stirred with a magnetic stirrer for 30 s and then wrapped in a piece of aluminum foil and kept at room temperature for 18 h.

These gold nanoparticles, synthesized using chemicals and CTAB, were characterized by the UV-Vis spectrophotometer, using the scanning range from 340-940 nm. The gold nanoparticles were collected after centrifugation at 10,500 rpm for 10 min. The chosen samples were investigated with TEM. The statistics were obtained from 100 sampled particles from many TEM figures.

2.8 *Coating gold nanoparticles synthesized using the extracts with polyethylene glycol for sensing applications*

It was reported that nanoparticles had to be well dispersed and stabilized when applied for effective sensing applications based on the localized surface plasmon resonance (LSPR) phenomena [4]. Because in chemical synthesis, CTAB acted as an efficient stabilizer, it is not necessary to enhance the stability of gold nanorods for sensing applications, while in the green synthesis using cascara and coffee flower extracts, the increase in stability has to be done.

The coating polymer was chosen to be polyethylene glycol (PEG) with a molecular weight of 6000 Da (PEG6000). The PEG6000 solution at 5%w/v was mixed vigorously with the gold nanoparticles in a microcentrifuge tube using the vortex mixer. The coated gold nanoparticles were then collected after centrifugation at 10,500 rpm for 10 min.

2.9 *Dilution of gold nanocolloids for sensing applications*

Both the coated gold nanoparticles synthesized using the extracts and those synthesized using chemicals were diluted with RO water to achieve an absorbance of around 0.3 as starting materials for sensing applications. The LSPR phenomenon depends on the concentration of nanocolloids, so dilution can affect LSPR. In addition, it can reflect how sensitive the LSPR of nanoparticles is during dilution.

2.10 *Ammonium hydroxide (NH_4OH) sensing*

After each sample was diluted with RO water in Section 2.9, it had an absorbance of around 0.3 before adding the NH_4OH solution. 1 mL of NH_4OH solution at various concentrations (0, 100, 200, 300, 400, and 500 µM) was mixed with 2 mL of diluted gold nanocolloids. The solution was mixed with the vortex mixer and then kept in an ultrasonic bath at 80ºC with a soft vibration mode for 1 h.

3 Results and Discussion

3.1 *Characterization of the extract solutions*

The total phenolic content (TPC) and reducing power of the extract solution after dilution with water at various ratios were determined and the results are shown in Figure 1.

It was reported that cascara extract contained 85 mg/mL protocatechuic acid and 69.6 mg/L chlorogenic acid [12]. Arabica coffee flower extract contained 5.28 mg/g trigonelline, 0.734 mg/g chlorogenic acid, 4.987 mg/g caffeine and 0.568 mg/g gallic acid [13] while Robusta coffee flower extract contained 0.775 mg/g protocatechuic acid, 10.93 mg/g trigonelline, 10.71 mg/g caffeine and 0.287 mg/g gallic acid [14]. The corresponding TPCs of cascara, Arabica coffee flower and Robusta coffee flower extracts were 19.9–24.8 mg GAE/g [9], 8.34 mg GAE/g [13], and 22.6–42.4 mg GAE/g [14], respectively. Therefore, these extracts contain reducing and stabilizing agents that are useful for the synthesis of gold nanoparticles.

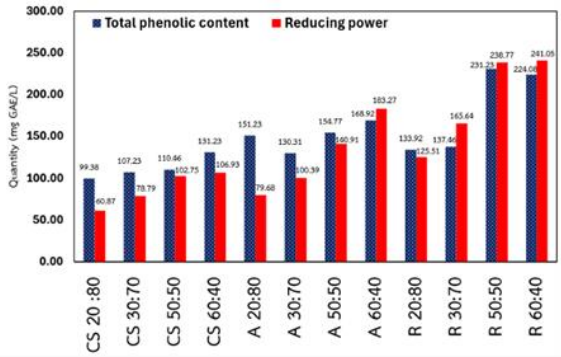


Figure 1: The total phenolic content and reducing power of the extract solutions of cascara (CS), Arabica coffee flowers (A) and Robusta coffee flowers (R) at different extract-to-water ratios.

Figure 1 shows that when compared at the same dilution ratio, TPCs and reducing powers of Robusta coffee flower extract (R) are greater than that of Arabica coffee flower extract (A), which is greater than that of cascara extracts (CS). The highest values of TPCs at 60:40 dilution ratios are 224.68, 168.92 and 131.23 mg/L GAE, respectively. These correspond to 11.23, 8.45 and 6.56 mg/g GAE. The values were smaller than those reported in the literature [12]–[14] discussed above because of the dilution.

Polyphenol compounds have antioxidant activity, which can act as a reducing agent in the synthesis of nanoparticles, so the TPC and reducing power show the same decreasing trend when diluted with a larger amount of water.

In our preliminary work, if the extracts were used undiluted, the gold nanoparticles grew so rapidly that they precipitated and nanoparticles could not be obtained, so the dilution of water was necessary in the synthesis. The range of reducing power reported here should be useful in the synthesis of gold nanoparticles in future research.

3.2 *Characterization of gold nanoparticles synthesized using the extract*

The results of localized surface plasmon resonance (LSPR) of gold nanoparticles were used to confirm their emergence. Figure 2(a) and (b) show the spectra of some gold colloids freshly prepared and kept for one week in the refrigerator, respectively. Only one

peak was seen on the synthesis day around 530–540 nm. The intensity of the first peak increased after the sample had been cooled for a week, indicating the growth of gold nanoparticles. In addition, for some samples listed as shown in the indices of Figure 2(b), the second peak appeared at a larger wavelength (690 nm onwards), implying that during the growth step, some spherical gold nanoparticles were transformed to those with non-isotropic shapes.

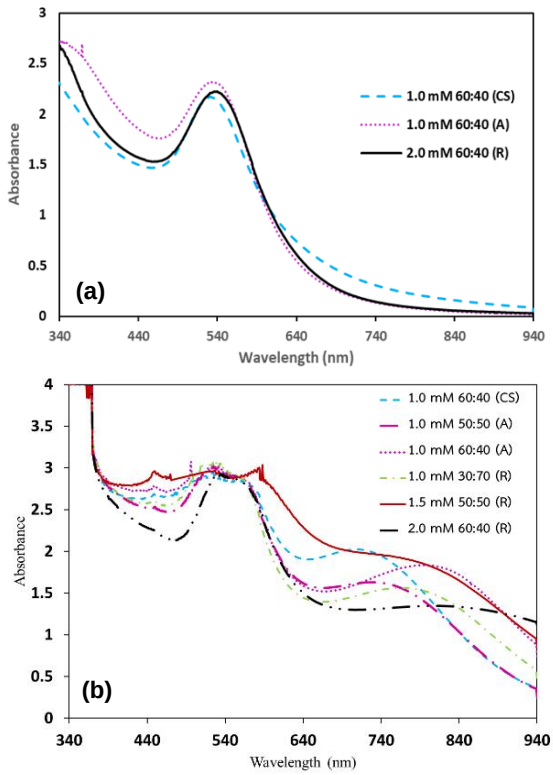


Figure 2: UV-Vis absorbance spectra of gold nanoparticle samples (a) freshly prepared and (b) kept for one week in the refrigerator.

Not all variations of gold precursor concentrations (from 1.0 to 2.5 mM) and extract concentrations (dilution ratio ranging from 20:80 to 60:40) yielded the gold nanoparticles. The summary of the conditions yielding the seed generation (the first absorbance peak) and growth to various shapes (the second absorbance peak) was reported in Figure 3.

Comparing the results of CS extracts and A extracts with R extracts, it can be seen in Figure 3(c) that gold nanoparticles can be formed when using the concentration of HAuCl4 at 2.5 mM and the dilution ratios down to 50:50. This is because the R extracts have higher values of reducing power and TPC. In this work, the second absorbance peaks of LSPR results are of interest because they imply that other shapes than nanospheres might be formed during the growth step. The second absorbance peaks were reported to have higher sensitivity towards the presence of some chemicals, so they can be applied in chemical sensing purposes.

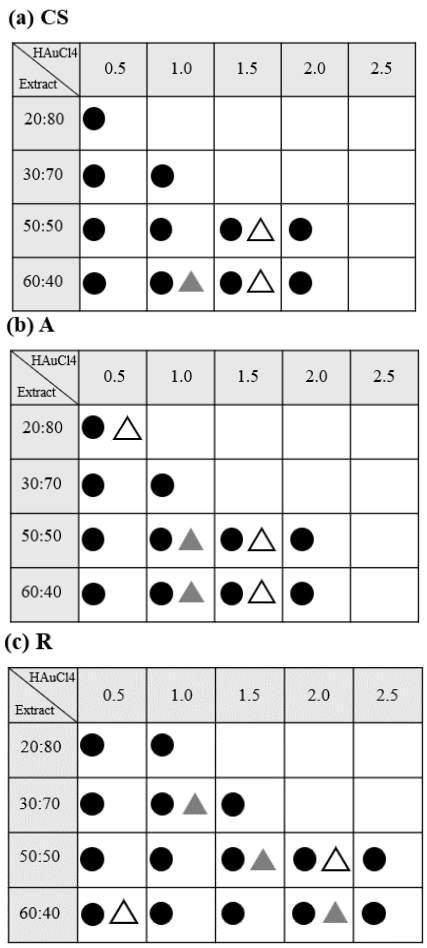


Figure 3: Summary of the synthesis of gold nanoparticles when varying the dilution ratio of the extracts from (a) Cascara, (b) Robusta and (c) Arabica coffee flowers, and varying precursor concentration in mM. Black circles = 1st peak (seed formation), grey triangles = 2nd peak (growth to various shapes), white triangles = peak probably beyond 940 nm.

It can be seen from Figure 3 that only a few conditions can yield the second absorbance peak. For the CS extracts, the dilution ratio has to be 60:40 at an $HAuCl_4$ concentration of 1.0 mM. The second peak appears at 712 nm. For the A extracts, the dilution

ratios have to be 50:50 and 60:40 at $HAuCl_4$ concentration of 1.0 mM and the second peaks are seen at 732 and 797 nm, respectively. When the dilution ratio is 20:80, the $HAuCl_4$ concentration has to be 0.5 mM and the small second peak is observed. In Figure 3(c), for the R extracts, at the $HAuCl_4$ concentration of 1.0 mM and the dilution ratio of 30:70, the second peak is seen at 765 nm. At the $HAuCl_4$ concentration of 1.5 mM and the dilution ratio of 50:50, the second peak is seen at 778 nm and at the $HAuCl_4$ concentration of 2.0 mM and the dilution ratio of 60:40, the second peak is seen at 805 nm.

Overall, it seems that to obtain the second absorbance peak, the concentration of $HAuCl_4$ has to be low enough to slow the growth rate to form other shapes of gold nanoparticles. In case of using a higher $HAuCl_4$ concentration, the dilution ratio has to be higher in order to yield a larger quantity of phenolic compounds, which can act as stabilizing agents for a slow growth rate. In addition, the second absorbance peak might appear beyond 940 nm when the concentrations of $HAuCl_4$ and the extracts are higher.

The morphology of the gold nanoparticles was examined using transmission electron microscopy (TEM) and the size distributions were obtained for the gold nanoparticles on the synthesis day and after growing for a week. Figure 4 shows that the synthesized gold nanoparticles using the extracts were mostly spherical in shape (gold nanospheres or AuNSs) on the synthesis day. The average diameter is 20.54 ± 5.04 nm, as shown in Figure 4(a).

After being left for one week in the refrigerator for the nanoparticles to grow, it is observed that they predominantly have triangular and polygonal shapes, such as pentagons and hexagons, as well as rod shapes, together with round shapes. The average particle size of the polygonal gold nanoparticles is 53.02 ± 22.86 nm. To measure the average size of the triangular gold nanoparticles, the length from the vertex of the triangle to the base (height) and the length of the base of the triangle (base length) were measured. The average size of the triangular-shaped AuNPs is 92.08 ± 39.12 nm and 103.39 ± 41.87 nm, respectively, so they are equilateral triangles, as can be observed in Figure 4(b).

The emergence of two LSPR peaks should be related to the shape of the gold nanoparticles. It was known that gold nanorods (AuNRs) that they exhibited two LSPR peaks, which correspond to the transverse and longitudinal surface plasmon modes and the position of the second peaks was observed to shift to a higher wavelength as the aspect ratio of the gold nanorod increased [7]. For various shaped gold nanoparticles, the two peaks should also correspond to different two projected areas of light projections (or even more than two, but the results are from the average of all projected areas). But at this stage, it has not been clear how a different shape of gold nanoparticles is correlated with the position of the second peak.

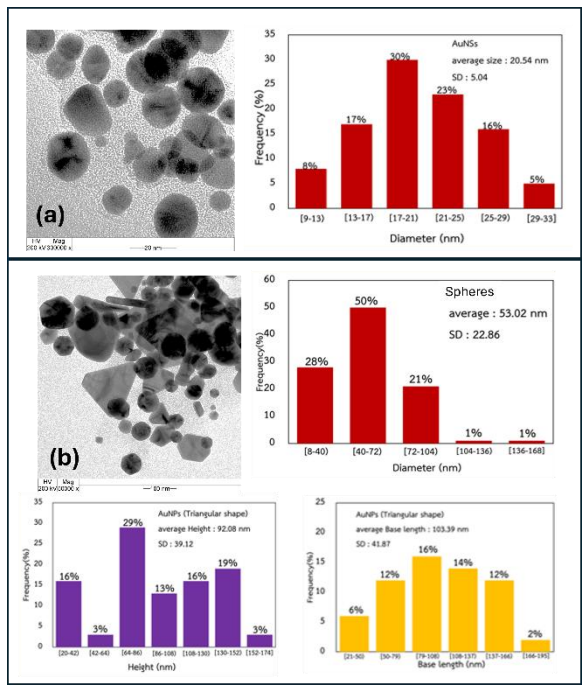


Figure 4: TEM micrographs and size distributions of the chosen samples, (a) nanospheres on the synthesis day (300000x magnification with the scale bar of 20 nm) and (b) various-shaped nanoparticles after growth for a week (50000x magnification with the scale bar of 100 nm).

3.3 *Characterization of AuNRs synthesized using chemicals and CTAB*

The seed solution was prepared according to the method of Sheenam *et al*. [15], while the growth solution was prepared by following the method of Tianyu *et al*. [16]. In both literatures, the concentrations of CTAB were designed the same in the seed steps and growth steps. However, in our study using the mixed methods of both literatures, the concentrations of CTAB in the growth step were

greater than that in the seed step. It is interesting to investigate the effect of CTAB concentration in the study on the shapes and sizes of gold nanorods (AuNRs).

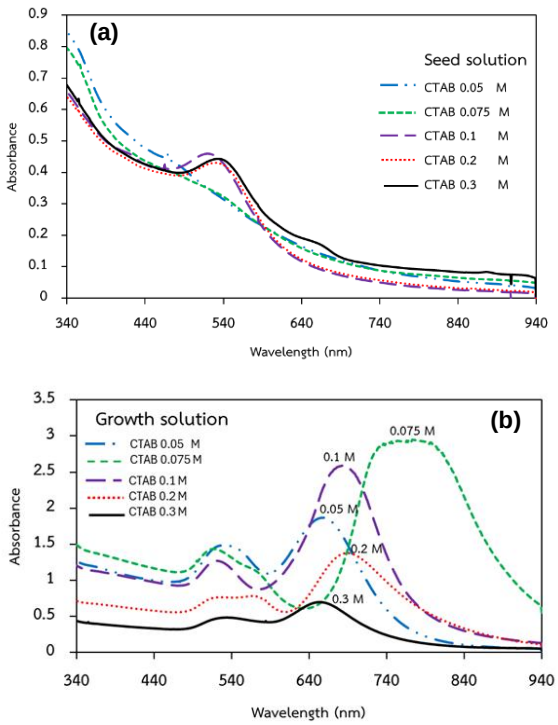


Figure 5: UV-Vis absorbance spectra of (a) seed solution, and (b) growth solution when varying the CTAB concentration.

In the seed step shown in Figure 5(a), due to using ice-cold sodium borohydride ($NaBH_4$) solution as a reducing agent, the gold seeds were formed and grew slowly. No peak is observed for seeds using CTAB concentrations at 0.05 and 0.075 mM. This may be due to the particles being too small, making it difficult for the particles to absorb light, and thus no peak appears.

In the growth step shown in Figure 5(b), the positions and absorbances of the second peaks depend on the CTAB concentration. The results are also reported in Table 1 for easy comparison. It can be seen that when varying the CTAB concentration from 0.05 to 0.3 mM, there is an optimal CTAB concentration to give the largest quantity of AuNRs, which in this case is 0.075 mM.

In other literature [16], [17], where the concentration of $HAuCl_4$ was greater in the growth step than in the seed step and the concentration of CTAB was kept similar in both the seed and growth steps. According to this, when the CTAB concentration increased, the second peak shifted to a higher wavelength and its absorbance increased. The results in Figure 5 show that increasing the CTAB concentration from 0.5 to 0.075 mM, the results are consistent with the literature, but when increasing from 0.075 to 0.3 mM, the results show the opposite trend.

Table 1: λ_{max} and the absorbance of the second peak of AuNRs when varying the CTAB concentration.

CTAB Concentration (M)	The second peak	
	λ_{max} (nm)	Absorbance
0.050	659	1.8721
0.075	787	2.9292
0.1	683	2.5894
0.2	689	1.3813
0.3	654	0.6986

It is possible that at low concentrations of CTAB (0.05 and 0.075 mM), the seed can grow bigger and with a low concentration of CTAB, it minimally controls the growth rate of gold nanoparticles, which finally transform into gold nanorods with a higher aspect ratio, which is defined as the length ratio of the major axis (longer) to the minor one (shorter). When the CTAB concentration increases to 0.1, 0.2 and 0.3 mM, where the true concentration of CTAB in the growth solution was twice that in the seed solution, the growth rate of gold nanoparticles is controlled more pronouncedly at the higher concentration of CTAB. With very small seeds in the seed step, the quantity of the resulting nanorods in the growth step is then small and the nanorods should have a smaller aspect ratio.

The morphology of AuNRs was examined using transmission electron microscopy (TEM). Figure 6 shows the shapes and sizes of AuNRs synthesized using different CTAB concentrations. For CTAB concentration at 0.1 M, the average lengths of the major axis and minor axis are 108.68 ± 6.8 and 47.98 ± 11.05 nm, respectively. The average aspect ratio is then 2.39 ± 0.65, as shown in Figure 6(a).

For CTAB concentration at 0.075 M, as shown in Fig. 6(b), the resulting gold nanorods are more elongated compared to those synthesized using 0.1 M CTAB. The average lengths of major axis and minor axis are 102.12 ± 31.03 and 35.84 ± 9.40 nm, respectively, so the average aspect ratio is 2.96 ± 0.84. The gold nanorods synthesized using the CTAB concentration of 0.075 M were then chosen to be

utilized as sensors it gave the highest quantity of gold nanorods while minimizing the use of CTAB.

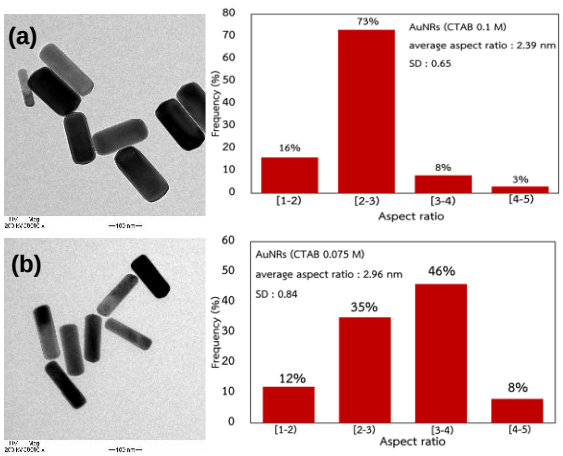


Figure 6: TEM micrographs and size distributions of the chosen AuNRs samples using the CTAB concentrations at (a) 0.1 and (b) 0.075 M.

3.4 *Application of gold nanoparticles for NH_4OH sensing*

3.4.1 Effect of diluting AuNPs with water

Dilution affects the dispersion of gold nanoparticles, thereby affecting the LSPR results. Table 2 shows the amounts of water used to lower the absorbances of different gold nanoparticle samples from the initial values before dilution to the final values after dilution. The samples are gold nanospheres (AuNSs), various-shaped gold nanoparticles (AuNPs) synthesized from the extracts and gold nanorods (AuNRs) synthesized using chemicals and CTAB. The absorbances of the first peak of AuNSs before and after dilution are reported, while those of the second peaks of AuNPs and AuNRs are reported in Table 2.

Table 2: The volume of water added to change the absorbances of the nanocolloids to around the same initial and final values.

Sample	Before dilution	After dilution	Water added (mL)
AuNSs	0.3248	0.2896	6
AuNPs	0.3077	0.2687	2
AuNRs	0.3458	0.2840	1

It can be seen that the change of absorbance values per volume of water added is found to be 5.87×10^{-3}, 0.0195 and 0.0618 mL^{-1} for AuNSs, AuNPs and AuNRs, respectively. Therefore, dilution affects the LSPR of AuNRs the most, followed by those of AuNPs and AuNSs. This finding can imply that the degree of sensitivity of AuNRs samples is the highest, followed by those of AuNPs and AuNSs when being applied as chemical sensors.

In addition, the decrease in absorbance of the first and the second peaks of AuNRs upon dilution with water was compared. The results are shown in Figure 7. Water was added incrementally until the total volume reached 13 mL while reducing the absorbances of both peaks to reach a similar value. It is evidently seen that the absorbance of the second peak decreases more rapidly with water volume than the first peak. This again shows the implication of using the second peak of the LSPR result to detect chemicals at different concentrations because it shows a higher degree of sensitivity.

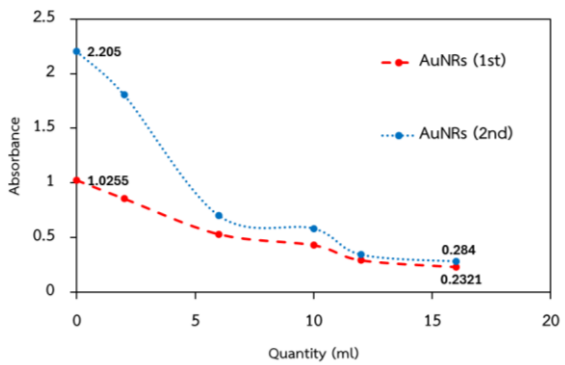


Figure 7: The decrease of absorbances of the 1st and the 2nd peaks of AuNRs colloids upon dilution with water.

3.4.2 NH_4OH sensing

The synthesized gold nanoparticles of various shapes were tested as sensors for ammonium hydroxide (NH_4OH) solution at 100, 200, 300, 400, and 500 μM. The results were compared among AuNSs, AuNPs and AuNRs. The results are shown in Figure 8.

For all samples, when the concentration of NH_4OH increases, the absorbances of gold nano-colloids increase. The NH_4OH solution is a basic solution having hydroxide ions. It is possible that at a very low concentration of NH_4OH, hydroxide ions can be adsorbed on the gold nanoparticles and neutralized by positive charges on gold nanoparticles, leading to a lower degree of repulsive interactions among nanoparticles and some degree of aggregation is possible. This results in higher values of absorbance. This result aligns with that reported in the literature

[18], which showed a linear relationship between absorbance and NH_4OH concentration.

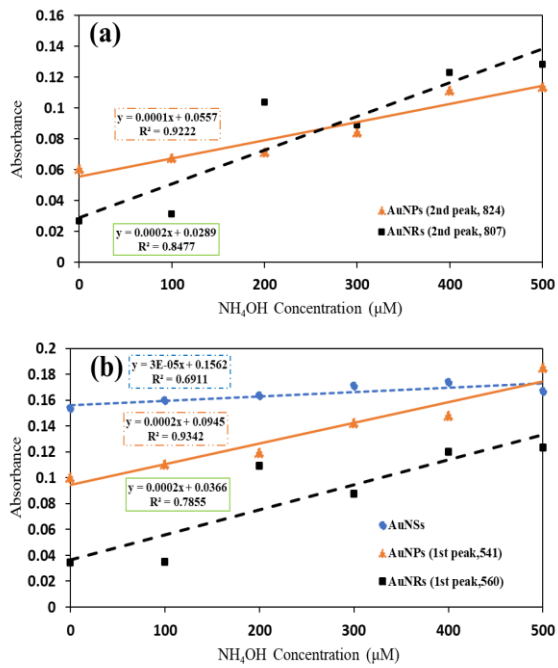


Figure 8: Effect of NH_4OH concentration on (a) the 2nd, and (b) the 1st peaks of the LSPR of various-shaped gold nanocolloids.

Comparing the sensitivity of the gold samples, the slope of the linear relation can be considered. Figure 8(a) shows the results using the second peaks of AuNPs and AuNRs, while Figure 8(b) shows those using their first peaks compared with the peaks of AuNSs. It can be seen in Figure 8(a) and Figure 8(b) that the slopes of the AuNRs samples and AuNPs samples are comparable, which are a lot higher than the slope of the AuNS samples. This finding shows that the various-shaped AuNPs can be used as sensors for NH_4OH as effectively as the AuNRs.

4 *Conclusions*

The gold nanoparticles were successfully synthesized using the extracts from cascara, Arabica, and Robusta coffee flowers. On the synthesis day, the gold nanoparticles had a spherical shape and they grew into various shapes, mostly polygons and triangles, while being kept in the refrigerator for one week. In a chemical synthesis using CTAB, the gold nanorods were obtained and the aspect ratio was dependent on the concentration of CTAB.

According to the LSPR results, only one peak was observed for the gold nanospheres, while two peaks were seen for the gold nanorods and various-shaped gold nanoparticles. These gold nano-colloid samples were tested as sensors for NH_4OH. It was found in the dilution test that the second peak was more sensitive than the first peak. Upon testing with NH_4OH, the absorbances of all samples show linear relations with NH_4OH concentration. The various-shaped AuNPs synthesized from the extract showed the sensitivity comparable to the CTAB-capped gold nanorods, which was evidently greater than that of gold nanospheres. The investigations with more numbers of chemicals should be performed in the future.

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Author Contributions

A.S.: investigation, methodology, writing an original draft; S.K.: investigation, methodology, writing an original draft; P.D.: research design, data analysis; writing—reviewing and editing, funding acquisition, project administration. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

Declaration of generative AI and AI-assisted technologies in the writing process

The authors did not utilize the ChatGPT tool or other AI tools to enhance the language and readability of the manuscript.

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