Synthesis of Silver Nanoparticles using Gandaria Seeds Bioreductor

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Synthesis of Silver Nanoparticles using Gandaria Seeds Bioreductor

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Abstract

The silver nanoparticles (NPP) are synthesized with the chemical reduction method by using a water extract bioreductor of gandaria seed (Bouea macrophylla G.) which acts as a reducing precursor, in this case, $AgNO_3$ Ag^+ is reduced to AgO. The concentration of $AgNO_3$ is made between 0.5 mm and 1 mm. The characteristic of NPP is unstable, so a modification is needed with and without the addition of PVA 1%. The process of NPP shaping was monitored by observing the uptaken of UV-Vis when the color changes occur. The high NPP concentration of $AgNO_3$ has higher absorbance and is wider if compared to the lower $AgNO_3$ concentration. The result of this research based on the absorbance value and the wavelength showed the NPP synthesized without the addition of PVA 1% (b/v) is wider. The addition of PVA 1% (b/v) provides better stability and maintains the absorbance of spectrum changes from day to day. The maximum uptaken of UV-Vis from NPP $AgNO_3$ 0.5 mm by using green synthesis and 1 mm without adding PVA are 0.946 and 0.980, respectively. However, NPP with the addition of PVA has 0.968 and 0.978 absorbance. The best concentration of NPP produced was 1 mm $AgNO_3$.

Keywords: Gandaria, silver nanoparticles, bioreductor

Introduction

In general, nanotechnology is the science and engineering of the creation of materials, functional structures, and devices on the nanometer scale. Nanometer-sized materials have several chemical and physical properties that are superior when compared to bulk materials (Abdullah et al., 2008). A material can be categorized as a nanoparticle if it is 1 - 100 nm in size. Silver nanoparticles are one of the most intensively studied metals in the field of nanotechnology (Jiang et al., 2004).

Silver nanoparticles are chosen as the output product of green synthesis based on their broad potential to be developed in various fields of application. In addition, silver is one of the precious metals that has a fairly good optical quality after gold at a more affordable price (Loiseau et al., 2019; Pala et al., 2018; Prabhu & Poulose, 2012; Caro et al., 2010).

Synthesis of nanoparticles using plant extracts is currently widely used. The advantages of this method include being easily available, safe to handle, regarded as environmentally friendly nanoparticles, low production cost, more efficient with energy and temperature pressures and doesn't require toxic chemicals so that plants become an alternative in nanoparticle bioreduction (Nafia, 2012).

The use of plants for the synthesis of nanoparticles involves secondary metabolites from plants, such as flavonoids and triterpenoids (Shankar et al., 2004). The antioxidant activity of gandaria seed extract shows an IC_{50} value of 2.43 μg/ml which is comparable to vitamin C, with an IC_{50} value of 2.25 μg/mL. Antioxidant activity in gandaria seeds is related to the content of secondary metabolites it contains, namely compounds belonging to the flavonoid group. Flavonoid compounds in plants are distributed in the form of anthocyanins or pigments that give color to plants (Londo', 2015; Hanifa & Susilawati, 2017; Rudiana et al., 2018).

Nanoparticles tend to experience aggregation (clumping). Efforts to prevent the aggregates to form between nanoparticles can be done by adding material or particle-coating molecules (Lembang, 2013). Polyvinyl alcohol has been known to be used as a stabilizer. PVA also functions to maintain the aggregation that occurs when the indicator solution is tested with certain analytes (Lee & Lee, 2002). The resulting stable silver nanoparticles are

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characterized by the formation of brownish-yellow colloidal silver, but there is not always a correlation between the intensity of the color and the increase in absorbance (Aftrid, 2018; Zulaikha et al., 2021).

The purpose of this study was to synthesize silver nanoparticles using gandaria seed extract as a bioreductor. A study was conducted by testing various concentrations of the $AgNO_3$ precursor solution. This research also studied the effect of adding PVA to silver nanoparticle products.

Methods

Equipment and material

The tools used in this study were glassware that was commonly used, Whatman No. 42 filter paper, Buchner funnel, vacuum pump, UV–Vis - 2600 spectrophotometer, FTIR Shimadzu 820 IPC, magnetic stirrer, analytical balance, 100 mL and 1000 mL Erlenmeyer, 100 mL and 1000 mL volumetric flask, volume pipette, micropipette, silver nitrate ($AgNO_3$), polyvinyl alcohol (PVA), aquabides, distilled water, ethanol (*Merck*), 1 kg of gandaria seeds (Bouea macrophylla Griffith).

Gandaria seed extract preparation

As much as 50 g of gandaria seeds (Bouea macrophylla G.) are washed, dried, then chopped, and mashed with a blender. The fine powder of gandaria seeds was weighed as much as 4 g and put into Erlemeyer then added with 400 mL of aquabides. The mixture is then boiled at 100 °C while being stirred until it's completely extracted (indicated by color change) and then filtered. Water obtained from the process has 0.01% gandaria seeds concentration (w/v).

Sample A (without PVA addition)

As much as 3 mL of 0.01% (w/v) gandaria seed boiled water was put into a glass beaker that has been labeled with 0.5 and 1 mm $AgNO_3$, then 10 mL of $AgNO_3$ solution was added to it, and each concentration of $AgNO_3$ drop was added using a burette while the mixture stirred until it changes color. Characterization of the mixed solution in the form of color UV-Vis absorption spectrum after mixing was observed at 0, 2, 3, 4, 5, and 6 days.

Sample B (with PVA addition) (Matutu et al., 2016)

As much as 5 mL of 1% PVA solution was added to a glass beaker, then add 3 mL of gandaria seed boiled water. The mixed solution was then stirred using a magnetic stirrer while drops of 0.5 mm $AgNO_3$ were added using a burette until the color changed. The same procedure was also carried out for 1 mM $AgNO_3$ at the same time. The volume ratio between $AgNO_3$: water from gandaria seeds: PVA is 10:3:5 mL.

Results and Discussion

Silver nanoparticle (NPP) biosynthesis

Based on the results of the study of synthesis using boiled water of gandaria seeds (ARBG) (Bouea macrophylla G.) which reacted $AgNO_3$ solution as NPP precursor with a volume ratio of ARBG: $AgNO_3$, namely 3:10 mL and ARBG: $AgNO_3$:PVA 1% (w/v), namely 3:10:5 mL at each concentration of 0.5 and 1 mm $AgNO_3$ until a color change occurs on ARBG and several characteristics are obtained related to several parameters, namely solution color change, UV-Vis spectrum, the effect of contact time on UV-Vis spectrum, and the effect of adding 1% (w/v) PVA. The working principle is the ability of functional groups to reduce Ag^+ to AgO (NPP) (Fabiani et al., 2019). The color change occurred in the ARBG solution from clear to clear yellow (12 minutes) to reddish brown (17 minutes). The color change that occurs is an indication that a reaction has occurred in the formation of NPP.

Silver nanoparticle (NPP) UV - Vis spectrophotometer analysis

In general, the color change occurs due to the silver ion reduction process using a bioreductor contained in the gandaria seed-boiled water. However, this color change cannot be used as the main indication of the formation of nanoparticles. Tests are needed to confirm the formation of nanoparticles, one of which is using UV-Vis spectrophotometry. The Surface Plasmon Resonance (SPR) value of the silver nanoparticles itself has a peak in the wavelength range of 400–450 nm (Anandalakshmi et al., 2016). The significant value of the absorbance peak spectrum of NPP shows the character of the surface plasmon resonance (SPR) of nanometer (nm) sized particles. SPR itself is the result of the excitation of surface plasmon vibration by light on a nanometer-sized structure

Synthesis of nanoparticles without the addition of PVA

Based on the results obtained, colloidal NPP of 0.5 $AgNO_3$ without the addition of 1% (w/v) PVA underwent a color change after 12 minutes of reaction time from clear to yellowish clear which indicated that a reaction had occurred in the formation of NPP. The color change became brownish yellow after 7 minutes which indicated that Ag^+ had been reduced to NPP by the gandaria seed boiled water bioreductor. The absorbance of this color change was measured using UV-Vis spectroscopy to determine the maximum wavelength and the absorbance spectral pattern formed at H0 to H6. The maximum wavelength and absorbance spectrum pattern of NPP at a concentration of 0.5 mm can be seen in **Figure 1.** The maximum wavelength reached at H0 is 420 nm with an absorbance value of 0.662. The highest absorbance of 0.5 mM concentration was achieved on the sixth day (H6) which was 0.948.

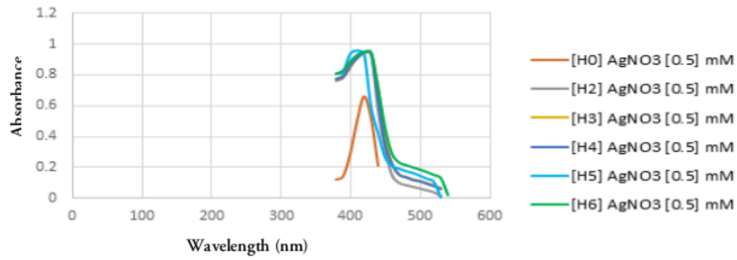

Figure 1. Maximum wavelength graph of 0.5 mM AgNO3 NPP at H0, H2, H3, H4, H5 and H6.

Synthesis of silver nanoparticles (NPP) was carried out using $AgNO_3$ precursor with 1 mm concentration without the addition of PVA to determine and compare the UV-Vis spectral characteristics of colloidal NPP 0.5 mm $AgNO_3$. The color change formed at a concentration of 1 mm was slightly longer when compared to a concentration of 0.5, namely at 18 minutes. This was possible because the bioreductor had not been mechanically distributed evenly in a 1 mm silver nitrate solution. This is reinforced by the color change after the synthesis process takes place, where the level of color density at a concentration of 1 mm is higher than 0.5 mm $AgNO_3$. It is confirmed by the maximum wavelength of 420 nm and the resulting absorbance at H0 is 0.680. The highest absorbance and NPP formation time were obtained faster than the 0.5 mm concentration, namely in H3 with an absorbance value of 0.988 which can be seen in **Figure 2** (Mulfinger et al., 2007).

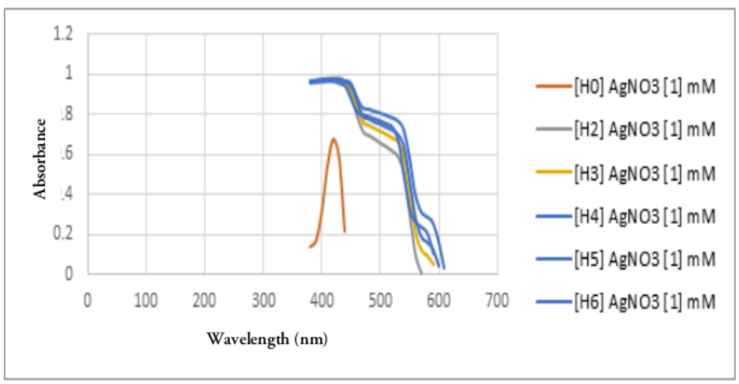

Figure 2. Wavelength graph (λ_{max}) nm vs absorbance of 1 mM $AgNO_3$ NPP at H0, H2, H3, H4, H5, and H6.

Effect of $AgNO_3$ concentration on NPP absorbance

Based on the UV - Vis absorbance test data for colloidal NPP without the addition of 1% (w/v) PVA, it was found that the λ_{max} value for each colloidal NPP at concentrations of 0.5 and 1 mm $AgNO_3$ was 420 nm with absorbance values at H0 were 0.662 and 0.680. While the highest absorbance was 0.946 and 0.980 which were obtained on the sixth and second days. So, it can be concluded that the greater the $AgNO_3$ concentration used, the greater the absorbance value and the higher the nanoparticles in the solution (Fabiani et al., 2019; Fabiani et al., 2018)Based on these data it can be concluded that the optimum concentration of $AgNO_3$ in the synthesis of NPP using the bioreductor water boiled from gandaria seeds 0.01 (w/v) obtained from each concentration of $AgNO_3$ with and without the addition of PVA was 1 mm.

Effect of 1 % (w / v) PVA addition on NPP colloids

The results of measurements using UV - Vis spectrophotometry show that the absorbance of colloidal NPP without the addition of 1% (w/v) PVA has a wider absorption range and spectral patterns that tend to change over time when compared to NPP with the addition of 1% (w/v) PVA. Acharya et al. 2018 reported that NPP aggregation can be characterized by a widened absorbance spectrum that tends to aggregate. The tendency of NPP to aggregate is due to the effects of Brownian motion and Van der Waals forces in solution. The tendency of nanoparticles to aggregate causes the size or diameter of the nanoparticles to be non-uniform.

To prevent aggregation and non-uniform NPP size distribution (instability), a stabilizing agent is needed, in this case, polymetric stabilization, or using polymers. Polyvinyl alcohol or better known as PVA is a polymer that can stabilize NPP through steric stabilization. The steric stabilization of NPP colloids is achieved by polymer molecules adhering to the surface of the particles and forming a coating, which creates repulsive forces and separates the particles from one another. The working principle of steric stabilization is that the polymer molecules work by forming a repulsive force around the nanoparticles to offset the Van der Waals forces present in the solution. If colloidal particles are brought a short distance from other particles, they are attracted to each other by van der Waals forces. If there is no counterforce, the particles will aggregate and the colloidal system will become unstable. The stability of a colloid is achieved because the repulsive forces balance the tensile forces like a stable mechanical balance (if the object is disturbed it tends to return to its original position) (Kopeliovich, 2013; Studart et al., 2007)

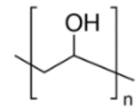

Figure 3. Structure of polyvinyl alcohol formula (source: the *Merck* index).

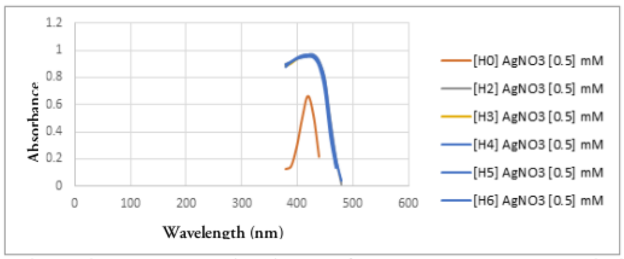

Figure 4. Wavelength graph (λ_{max}) nm vs absorbance of 0.5mm AgNO3 NPP with the addition of 1% PVA (w / v) at H0, H2, H3, H4, H5, and H6

Figure 4 shows that the 0.5 mm $AgNO_3$ NPP colloid with the addition of 1% (w / v) PVA has a relatively more stable and flatter absorbance when compared to the 0.5 mm $AgNO_3$ NPP colloid without the addition of 1% (w / v) PVA which has the spectrum that tends to fluctuate and the absorption is wider from day to day. The same change also occurred in NPP 1 mm $AgNO_3$ with the addition of 1% (w / v) PVA **Figure 5** with NPP 1 mm $AgNO_3$ without the addition of 1% (w/v) PVA. Where the colloidal spectrum of NPP produced by NPP without the addition of 1% (w/v) PVA has a broad absorption and varies from day to day. The results of this study correlate with research conducted by Wahyudi & Sugiyana (2011) which states that the stability of the colloidal NPP can be determined based on changes in absorption peaks.

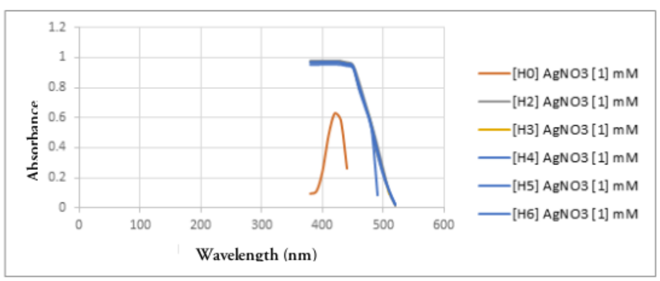

Figure 5. Wavelength graph (λ_{max}) nm vs absorbance of 1 mM $AgNO_3$ NPP with the addition of 1% PVA (w / v) at H0, H2, H3, H4, H5, and H6.

The shift of the absorption peak to a longer wavelength indicates that the stability of the NPP colloid is low. This is because there has been an agglomeration occurring in the NPP without the addition of 1% (w/v) PVA. Therefore, the addition of 1% (w/v) PVA in this study which functions as an NPP stabilizer agent as has been done by Apriandanu et al. (2013) & Rahmadani et al. (2020) in maintaining the stability and size of NPP is appropriate as it should be.

Effect of NPP UV - Vis absorbance on reaction time

Based on the UV - Vis absorbance test data for NPP without the addition of 1% (w/v) PVA, it was found that the λ_{max} value was 420 nm for NPP with $AgNO_3$ concentrations of 0.5 and 1 mm, with a concentration of 0.5 mm $AgNO_3$ reached on H6, with an absorbance value of 0.948. While in NPP with a concentration of 1 mm $AgNO_3$ without the addition of 1% (w/v) PVA it was achieved faster, namely in H2 with an absorbance of 0.980. This value is higher than the NPP concentration of 0.5 mm $AgNO_3$. The difference in formation time of this NPP is due to the quantity of $AgNO_3$ concentration contained in NPP A (0.5 mm $AgNO_3$) is smaller than NPP B (1 mm $AgNO_3$), where NPP A has a concentration of 0.5 mm $AgNO_3$, while NPP B has a concentration of 1 mM $AgNO_3$. Because the concentration of 1 mm $AgNO_3$ is higher than 0.5, the number of Ag^+ ions in 1 mm solution is greater than the amount of Ag^+ contained in a concentration of 0.5 mm. Therefore, the time needed to reduce Ag^+ ions in NPP B (1 mm $AgNO_3$) is faster than in NPP A.

Table 1. Effect of NPP UV–Vis absorbance on reaction time.

(H)	λ (nm)	Abs			
	NPP	NPP (0.5 mm $AgNO_3$)	NPP + PVA 1% (0.5 mm $AgNO_3$)	NPP (1 mm $AgNO_3$)	NPP + PVA (1 mm $AgNO_3$)
H0	420	0.662	0.662	0.680	0.631
H2	420	0.943	0.968	0.980	0.978
H3	420	0.943	0.963	0.973	0.973
H4	420	0.946	0.960	0.974	0.955
H5	420	0.937	0.954	0.968	0.973
H6	420	0.948	0.964	0.977	0.972

Based on **Table 1**, it can be seen that there was no change in the peak at the absorption wavelength around 420 nm for each colloidal NPP 0.5 and 1 mm $AgNO_3$ either without addition or with the addition of 1% (w/v) PVA. The absorbance at a concentration of 0.5 mm $AgNO_3$ without the addition of 1% (w/v) PVA increased from H0 to H6, namely 0.662 to 0.948. While at a concentration of 0.5 mm $AgNO_3$ with the addition of 1% (w/v) PVA, H0 has an absorbance value of 0.662 and has the highest concentration in H2, which is 0.968 and tends to decrease in H3, H4, H5 and slightly increases in H6.

The absorbance data of colloidal silver NPP with the addition of 1% (w/v) PVA with 0.5 mM $AgNO_3$, shows that the λ_{max} obtained is 420 nm, with an absorbance value of 0.968. In colloidal NPP with a concentration of 1 mM $AgNO_3$, a λ_{max} of 420 nm was obtained with an absorbance value of 0.978. In addition, the absorbance value can also show the trend in the size of the NPP. The higher the absorbance value, the more NPP is produced. Therefore, the increase in the absorbance value in the NPP synthesis process using gandaria seed boiled water does not occur spontaneously but is a result of the chemical reaction process. The results showed that the amount of NPP increased on H0, H2 and tended to decrease on H3, H4, H5, and H6 for NPP 0.5 $AgNO_3$ without the addition of 1% (w/v) PVA.

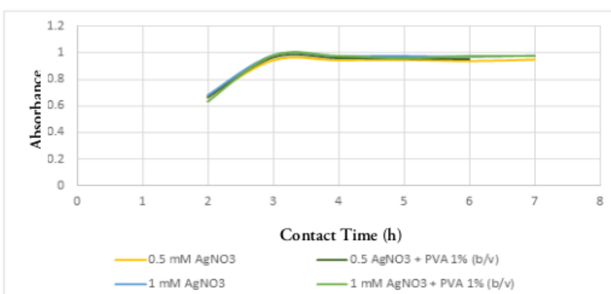

Figure 6. Relationship between contact time and absorbance graph.

The maximum UV - Vis absorption results of the samples synthesized by NPP with the addition of 1% (w / v) PVA concentration were different from those without 1% (w / v) PVA, respectively, at absorbances of 0.986 at λ = 420 nm and 0.978 at λ = 420nm. While at a concentration of 0.5 and 1 mm $AgNO_3$ without the addition of 1% (w/v) PVA 0.5 and 1 mm $AgNO_3$ was λ = 420 nm with a greater absorbance value when compared to the addition of 1% (w/v) PVA namely 0.946 in H4 and 0.980 in H2. Silver nanoparticles without the addition of PVA show a lower absorbance value, this is because the agglomerated silver nanoparticles in the agglomeration process require a lot of energy hence the smaller wavelength. As reported by Rahmadani et al. (2020) NPP with the addition of PVA is smaller than the size of NPP without the addition of PVA, this is because PVA can stabilize the size of the nanoparticles to prevent agglomeration in the formation of NPP. The higher the absorbance peak of the colloidal NPP, the greater the amount of NPP contained and the tendency to agglomerate. Meanwhile, the wider the absorption, the greater the number of NPPs experiencing agglomeration. The Spectral Bandwidth value correlates with the particle size distribution where the smaller the value, the more monodisperses the size distribution.

Conclusions

Based on the results of the research and discussion, it can be concluded that the boiled Gandaria seeds water can be used as a bioreductor in synthesizing silver nanoparticles. The optimum concentration of $AgNO_3$ precursor in synthesizing Ag nanoparticles was 1 mm and nanoparticles combined with PVA were more stable than the ones that aren't.

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