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Environmentally Friendly Silver Nanoparticle Synthesis Using Gandaria (*Bouea macrophylla*) Leaf Waste Extract for Mercury Detection in Facial Cream Products

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ABSTRACT: This study aimed to synthesize and characterize silver nanoparticles (AgNPs) using the stem bark extract of Gandaria (*Bouea macrophylla*) and to evaluate their selectivity for mercury (Hg) detection. The Gandaria stem bark was extracted using methanol as the solvent through the maceration method. Silver nanoparticles were synthesized using extract concentrations of 1%, 2%, and 4%. Characterization of the AgNPs using UV–Visible spectrophotometry showed that the highest absorbance was observed at the 4% extract concentration, whereas the lowest absorbance was recorded at the 1% concentration. The selectivity test demonstrated that the synthesized AgNPs were highly selective toward Hg in $HgCl_2$ solution compared with Pb in $PbCl_2$ solution and Ca in $CaCl_2$ solution. AgNPs synthesized using the 4% extract concentration could be used as a preliminary qualitative colorimetric detection material for identifying mercury in facial whitening creams available on the market. Based on Particle Size Analyzer (PSA) and Scanning electron microscopy with energy dispersive X-ray spectrometry (SEM–EDX) analyses, the AgNPs synthesized using the 4% Gandaria stem bark extract were confirmed to fall within the nanoparticle size range.

KEYWORDS: Environmentally friendly synthesis; gandaria; facial cream

1. Introduction

The practice of skin care had evolved from a mere cosmetic preference into a necessity for a significant proportion of the female population. The prevailing perception among Indonesian women regarding ideal skin characteristics was smooth, bright skin that was free from brown or dark spots. Whitening creams were reported to contain potentially hazardous substances, including mercury, hydroquinone, retinoic acid, and rhodamine B [1]. The Indonesian National Agency of Drug and Food Control (BPOM RI) reported that 1,541 illegal cosmetic products were circulating in the market throughout 2022, with the majority found to contain mercury. Mercury was classified as a prohibited substance in cosmetic and skincare products because of its potential to cause a wide range of adverse health effects [2].

According to the Regulation of the Indonesian National Agency of Drug and Food Control (BPOM RI) No. 12 of 2019, the permissible limit of heavy metal contamination in cosmetics was 1 mg/kg or 1 mg/L (1 ppm). Mercury was one of the active substances that could be present in whitening creams because it acted as a skin-lightening agent with a strong bleaching effect. It was hypothesized that mercury ions inhibited melanin synthesis in melanocyte cells [3]. The presence of mercury in whitening creams was reported to cause various adverse effects, including skin discoloration, allergic reactions, and skin irritation. Short-term exposure to high doses of mercury was also associated with vomiting, diarrhea, and lung damage. Furthermore, mercury was classified as a carcinogenic substance in humans, as reported by Sari et al. [4]. Conventional techniques for mercury detection, such as atomic absorption spectroscopy (AAS) and inductively coupled plasma mass spectrometry (ICP-MS), offered high sensitivity and accuracy. However, these methods required sophisticated instrumentation, skilled operators, and relatively high operational costs, which limited their widespread application, particularly in resource-limited settings. Consequently, increasing attention was directed toward alternative detection methods that were simple, cost-effective, and environmentally friendly [5].

In response to these challenges, there was a need to develop a qualitative and quantitative method for mercury detection that was rapid, reliable, and environmentally sustainable. One promising approach involved the use of silver nanoparticles (AgNPs), which could be synthesized from gandaria stem bark extract through a green chemistry approach. The stem bark extract of gandaria (*Bouea macrophylla* Griff.) possessed the potential to act as a bioreducing agent because of its high phenolic and flavonoid contents [6, 7]. Rudiana et al. reported that the methanol extract of gandaria stem bark contained total phenolics of 19.35 mg gallic acid equivalent per gram and total flavonoids of 31.18 mg quercetin equivalent per gram [8]. Furthermore, Andina and Musfirah reported that phytochemical screening of the methanol extract revealed the presence of flavonoids, tannins, quinones, saponins, and steroids [9]. The flavonoid compounds present in gandaria stem bark were reported to reduce Ag^{+} ions into Ag^{0} nanoparticles, representing the fundamental mechanism underlying plant-mediated reduction during the green synthesis of silver nanoparticles [10]. Silver nanoparticles had also been employed as colorimetric indicators for the qualitative and quantitative determination of mercury in facial care cream samples [11].

This research topic was also aligned with the Sustainable Development Goals (SDGs), particularly Goal 3 (Good Health and Well-Being), Goal 14 (Life Below Water), and Goal 15 (Life on Land), which collectively promoted the reduction of hazardous pollutants and the protection of human health and ecosystems [12]. A review of the available literature indicated that studies on the green synthesis of silver nanoparticles (AgNPs) using *Bouea macrophylla* (gandaria) remained very limited. Existing publications had primarily utilized seed extracts or aqueous leaf extracts as bioreducing agents for antibacterial and photocatalytic degradation applications, whereas the use of methanolic stem bark extract of *B. macrophylla* for the green synthesis of AgNPs had rarely been reported. Moreover, previous studies had focused predominantly on nanoparticle synthesis and antimicrobial applications, while their application for mercury ion detection, particularly in facial care creams, had received little attention. Therefore, this study was conducted to synthesize silver nanoparticles using methanolic gandaria stem bark extract as a bioreducing agent and to evaluate their potential as a colorimetric sensor for mercury ion detection in facial care cream samples.

2. Materials and Methods

2.1. Materials.

The plant material used in this study was the stem bark of *Bouea macrophylla*, which was collected from Mancak District, Serang Regency, Banten Province, Indonesia. The chemicals and reagents used included 96% methanol, distilled water (aquades), silver nitrate ($AgNO_3$), mercury(II) chloride ($HgCl_2$), lead(II) chloride ($PbCl_2$), calcium chloride ($CaCl_2$), and two commercially available facial whitening cream samples.

2.2. Preparation of Plant Material.

A total of 1.5 kg of fresh *B. macrophylla* stem bark was collected and used as the plant material. The stem bark was thoroughly washed to remove adhering impurities, cut into small pieces, and air-dried at room temperature for 14 days. The dried material was subsequently extracted using the maceration method with methanol as the extraction solvent. Maceration was carried out for 3 × 24 h with intermittent stirring every 12 h to enhance extraction efficiency. After the extraction process was completed, the filtrate was collected and concentrated using a rotary evaporator to obtain the crude methanolic extract of *B. macrophylla* stem bark.

2.3. Preparation of Extract Concentrations.

Methanolic extracts of *B. macrophylla* stem bark were prepared at concentrations of 1%, 2%, and 4% (w/v), each with a final volume of 250 mL. For the 1% (w/v) solution, 2.50 g of the crude extract was accurately weighed, dissolved in methanol, and transferred into a 250 mL volumetric flask. The solution was then diluted to volume with methanol and thoroughly mixed to obtain a homogeneous solution. Similarly, 5.00 g and 10.00 g of the crude extract were used to prepare the 2% and 4% (w/v) solutions, respectively, following the same procedure. All extract solutions were homogenized before being used in the biosynthesis of silver nanoparticles [5].

2.4. Green Synthesis of Silver Nanoparticles (AgNPs).

Silver nanoparticles (AgNPs) were synthesized using *B. macrophylla* stem bark extract at concentrations of 1%, 2%, and 4% (w/v). The synthesis was performed by reacting the extract with 0.01 M silver nitrate ($AgNO_3$) solution at a volume ratio of 1:9. Briefly, 20 mL of the extract solution was mixed with 180 mL of 0.01 M $AgNO_3$ solution. The reaction mixture was stirred using a magnetic stirrer for 1 h to ensure complete mixing and initiate the reduction process. Subsequently, the mixture was exposed to indirect sunlight for 30 min to facilitate nanoparticle formation and then incubated at room temperature for 24 h to allow complete reduction of silver ions. The formation of AgNPs was initially indicated by a visible color change of the reaction mixture and was subsequently confirmed using UV–Visible spectrophotometry [13, 14].

2.5. Selectivity Test of AgNPs.

The selectivity of the synthesized AgNPs toward different metal ions was evaluated using mercury(II), lead(II), and calcium ions. Briefly, 2 mL of the AgNP colloidal solution was mixed with 0.2 mL of $HgCl_2$, $PbCl_2$, or $CaCl_2$ solution. The resulting color changes were

visually observed, and the absorption spectra of each mixture were recorded using a UV–Visible spectrophotometer over the wavelength range of 400–800 nm.

2.6. Characterization of AgNPs.

The synthesized AgNPs were characterized using UV–Visible spectrophotometry to determine their optical properties, Particle Size Analysis (PSA) to measure particle size distribution, and Scanning Electron Microscopy coupled with Energy Dispersive X-ray Spectroscopy (SEM–EDX) to examine particle morphology and elemental composition.

2.7. Application of AgNPs for Mercury Detection in Facial Whitening Creams.

The practical applicability of the synthesized AgNPs was evaluated using two commercially available facial whitening cream brands purchased from Keranggot Market, Cilegon City, Indonesia. A colorimetric assay was performed by comparing the color responses of the cream samples with those of a negative control (cream mixed with distilled water) and positive controls consisting of 0.01 M $HgCl_2$ solution and the synthesized AgNP colloidal solution. The resulting color changes were used as preliminary qualitative indicators of mercury contamination in the cream samples.

3. Results and Discussion

3.1. Extraction of Bouea macrophylla Stem Bark.

From 250 g of dried stem bark, 18.19 g of concentrated methanolic extract was obtained, corresponding to an extraction yield of 7.28%. This extraction yield suggested that methanol was an effective solvent for extracting bioactive compounds from *Bouea macrophylla* stem bark [15]. Methanol, a polar solvent, has been widely used for the extraction of secondary metabolites, particularly phenolic and flavonoid compounds, which have been reported to be abundant in *B. macrophylla* stem bark [16]. The concentrated extract was subsequently used to prepare extract solutions at concentrations of 1%, 2%, and 4% (w/v) for the biosynthesis of silver nanoparticles.

3.2. Green Synthesis of Silver Nanoparticles (AgNPs).

Silver nanoparticles (AgNPs) were synthesized using *B. macrophylla* stem bark extract at concentrations of 1%, 2%, and 4% (w/v). The extract solutions were reacted with 0.01 M $AgNO_3$ solution at a volume ratio of 1:9. The ratio between the extract and silver nitrate solution played an important role in determining the efficiency of the reduction process and the successful formation of AgNPs. The reaction mixtures were stirred for 1 h to ensure complete homogenization before being exposed to indirect sunlight for 30 min. Sunlight irradiation accelerated the formation of silver nanoparticles by enhancing the photochemical reduction of silver ions [17]. Ultraviolet radiation in sunlight promoted the generation of reactive species from phytochemicals present in the extract, particularly phenolic and flavonoid compounds, which acted as natural reducing agents. These bioactive compounds reduced Ag^+ ions to metallic silver (Ag^0), resulting in the formation of silver nanoparticles. The visual color changes observed during the synthesis process are presented in Figure 1.

(a) (b)

Figure 1. The observed color change of the silver nanoparticle (AgNPs) mixture is shown as follows: (a) at 0 minutes of reaction, and (b) after 1 hour of reaction at room temperature followed by 30 minutes of exposure to sunlight.

Based on Figure 1, the reaction mixture initially exhibited a pale pink color before the reduction process occurred. After stirring for 1 h and subsequent exposure to indirect sunlight for 30 min, the solution gradually changed to a reddish-brown color. This color transformation indicated the reduction of silver ions and the formation of silver nanoparticles. The reddish-brown color was attributed to the surface plasmon resonance (SPR) phenomenon, which is characteristic of metallic silver nanoparticles. Although the observed color change provided preliminary evidence of AgNP formation, visual observation alone was insufficient to confirm successful nanoparticle synthesis. Therefore, further characterization using UV–Visible spectrophotometry, Particle Size Analysis (PSA), and Scanning Electron Microscopy coupled with Energy Dispersive X-ray Spectroscopy (SEM–EDX) was conducted to verify the formation, particle size distribution, morphology, and elemental composition of the synthesized AgNPs.

3.3. Characterization of AgNPS using UV-Visible spectrophotometry.

The characterization of AgNPs was carried out using a UV–Visible spectrophotometer in the wavelength range of 400–800 nm. The analysis was performed by measuring distilled water as a blank, 0.01 M $AgNO_3$ solution, and silver nanoparticle colloidal samples at concentration variations of 1%, 2%, and 4%. The results of the UV–Visible characterization are presented in Figure 2.

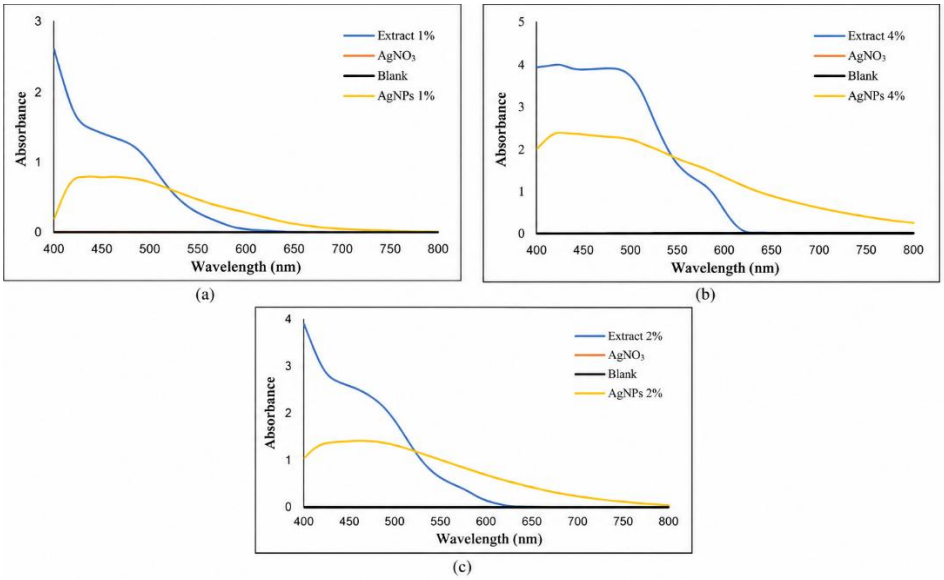


Figure 2. Characterization of AgNPs using a UV–Visible spectrophotometer at a wavelength range of 400–800 nm. Extract variation 1% (a), extract variation 2% (b), and extract variation 4% (c).

From Figure 2, it was observed that no characteristic absorption peaks were detected for either the gandaria extract or the $AgNO_3$ solution before the synthesis reaction. After the extract was reacted with $AgNO_3$ for 30 min under indirect sunlight at room temperature, a distinct absorption peak appeared within the wavelength range of 440–460 nm. This absorption peak indicated the formation of silver nanoparticles (AgNPs) [18]. Silver nanoparticles typically exhibit surface plasmon resonance (SPR) absorption within the wavelength range of 420–455 nm [19]. The highest absorbance was observed for AgNPs synthesized using the 4% gandaria extract, whereas the lowest absorbance was recorded for the 1% extract. Higher absorbance values indicated the formation of a greater number of silver nanoparticles [20]. In contrast, shifts in the absorption wavelength were associated with differences in particle size, with changes in the SPR peak reflecting variations in nanoparticle dimensions and distribution.

3.4. Selectivity test of AgNPS toward $HgCl_2$, $PbCl_2$, and $CaCl_2$.

The selectivity of the synthesized AgNPs toward $HgCl_2$, $PbCl_2$, and $CaCl_2$ was evaluated using 0.01 M solutions of each metal salt. The experiment was conducted using AgNPs synthesized from *Bouea macrophylla* (gandaria) stem bark extract at concentrations of 1%, 2%, and 4% (w/v). Briefly, 2 mL of each metal salt solution ($HgCl_2$, $PbCl_2$, and $CaCl_2$) was transferred into separate reaction vials, followed by the addition of 1 mL of the AgNP colloidal solution. The reaction mixtures were allowed to stand for 15 min before visual observations were made to evaluate the color changes resulting from the interaction between the AgNPs and the respective metal ions. The results of the selectivity test toward $HgCl_2$, $PbCl_2$, and $CaCl_2$ are shown in Figure 3..

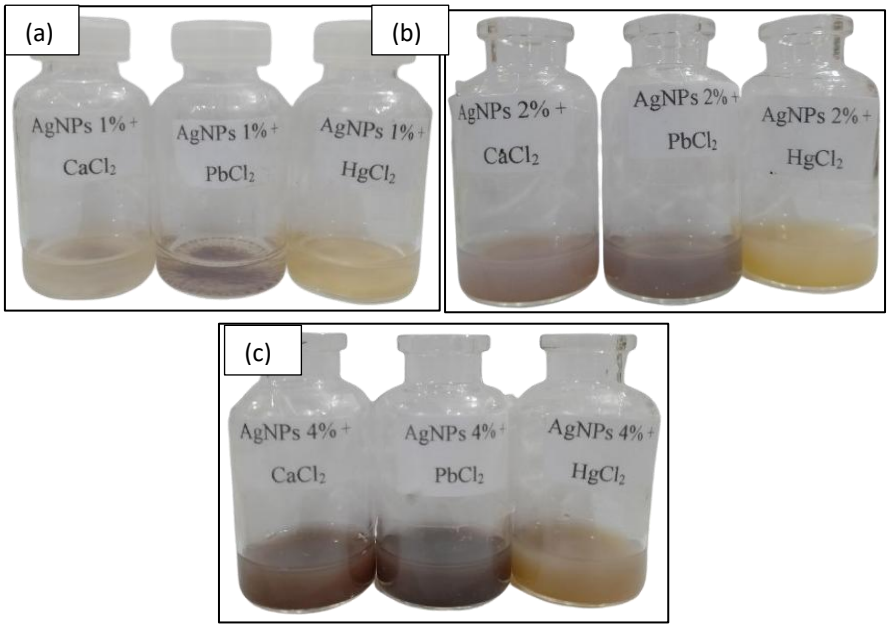


Figure 3. Selectivity test of silver nanoparticles (AgNPs) toward $HgCl_2$, $PbCl_2$, and $CaCl_2$. AgNPs synthesized using extract concentrations of 1% (a), 2% (b), and 4% (c).

The selectivity test demonstrated that the synthesized silver nanoparticles (AgNPs) were highly selective toward mercury (Hg^{2+}) ions. A rapid color change was observed immediately after the AgNP colloidal solution was added to the $HgCl_2$ solution. As shown in Figure 3, AgNPs synthesized using the 4% stem bark extract exhibited the most pronounced color change compared with those synthesized using the 1% and 2% extract concentrations. This finding

suggested that the 4% extract concentration provided superior selectivity toward Hg^{2+} ions, indicating its greater potential as a colorimetric sensing material.

Previous studies primarily employed aqueous leaf or seed extracts from various plant species as reducing and stabilizing agents, resulting in AgNPs with different physicochemical properties and sensing performances. In contrast, the present study utilized the methanolic stem bark extract of *Bouea macrophylla*, a plant material that had rarely been investigated for the green synthesis of AgNPs. The higher concentrations of phenolic and flavonoid compounds present in the methanolic stem bark extract were expected to enhance both the reduction of Ag^{+} ions and the stabilization of the synthesized nanoparticles. Consequently, the resulting AgNPs exhibited improved nanoparticle formation and demonstrated promising selectivity toward Hg^{2+} detection. These findings highlighted the potential of *B. macrophylla* stem bark extract as an effective and sustainable bioreducing agent for the green synthesis of AgNPs and their application as a colorimetric sensor for mercury detection.

3.5. Characterization of AgNPS using SEM-EDX.

The results of the morphological characterization of AgNPs derived from gandaria (*Bouea macrophylla*) stem bark using SEM are shown in Figure 4. Based on Figure 4, the grain size and surface morphology of the synthesized AgNPs can be clearly observed. The silver nanoparticles appeared agglomerated (clustered), forming aggregates distributed across the surface of the synthesized material, while the individual particles exhibited relatively uniform sizes [11]. The observed agglomeration may be attributed to the high surface energy of nanoparticles and strong interparticle interactions during the drying process for SEM sample preparation. In addition, phytochemical compounds present in the gandaria stem bark extract, such as phenolics and flavonoids, may have acted as capping agents that influenced particle growth and aggregation behavior. Despite the formation of aggregates, the nanoparticles maintained a relatively homogeneous morphology, indicating that the green synthesis approach successfully produced nanoscale AgNPs with a fairly uniform particle distribution [11].

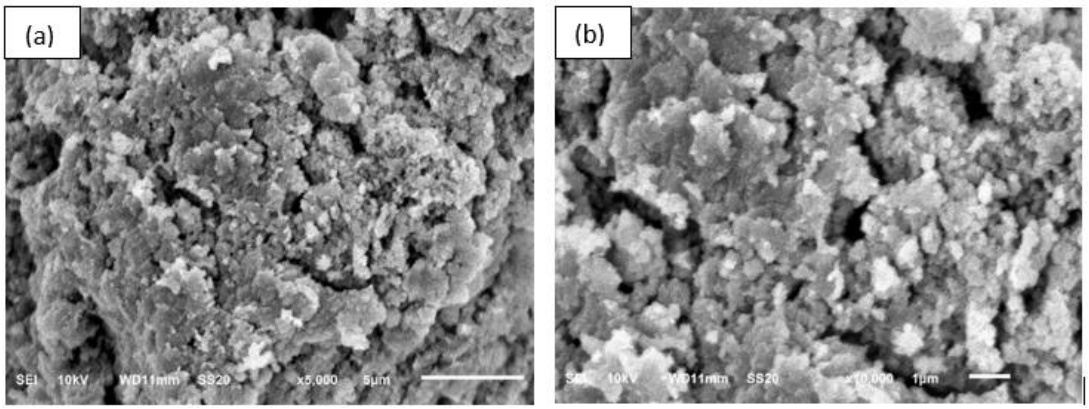
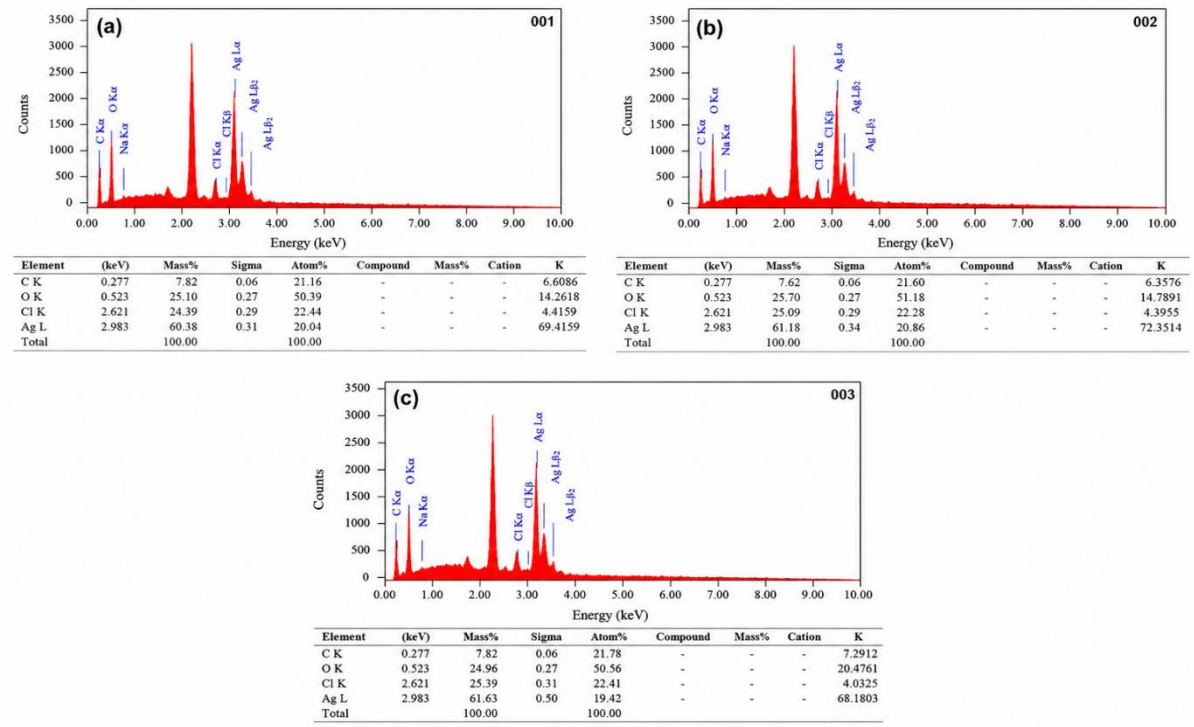


Figure 4. Morphology of AgNPs observed by SEM at a magnification of 5,000× (a) and 10,000× (b).

The results of AgNP characterization using SEM–EDX are shown in Figure 5. Based on Figure 5, the EDX spectra confirmed the presence of silver (Ag) as the predominant element in the synthesized nanoparticles, together with minor amounts of carbon (C), oxygen (O), and chlorine (Cl), which were likely associated with phytochemical compounds from the plant extract and residual components of the synthesis process. The SEM–EDX analysis of AgNPs for replicates 1, 2, and 3 showed that the average Ag content was 69.9825 wt%. This relatively

high silver content indicates the successful formation of AgNPs with high purity [21]. Furthermore, the consistency of the elemental composition among the three replicates demonstrates the good reproducibility of the green synthesis method using gandaria stem bark extract. The presence of carbon and oxygen also supports the role of plant-derived biomolecules in reducing Ag^+ ions and stabilizing the synthesized nanoparticles, thereby preventing excessive particle growth and contributing to nanoparticle stability [21].



Element	(keV)	Mass%	Sigma	Atom%	Compound	Mass%	Cation	K
C K	0.277	7.82	0.06	21.16	-	-	-	6.6086
O K	0.523	25.10	0.27	50.39	-	-	-	14.2618
Cl K	2.621	24.39	0.29	22.44	-	-	-	4.4159
Ag L	2.983	60.38	0.31	20.04	-	-	-	69.4159
Total		100.00		100.00				

Element	(keV)	Mass%	Sigma	Atom%	Compound	Mass%	Cation	K
C K	0.277	7.62	0.06	21.60	-	-	-	6.3576
O K	0.523	25.70	0.27	51.18	-	-	-	14.7891
Cl K	2.621	25.09	0.29	22.28	-	-	-	4.3955
Ag L	2.983	61.18	0.34	20.86	-	-	-	72.3514
Total		100.00		100.00				

Element	(keV)	Mass%	Sigma	Atom%	Compound	Mass%	Cation	K
C K	0.277	7.82	0.06	21.78	-	-	-	7.2912
O K	0.523	24.96	0.27	50.56	-	-	-	20.4761
Cl K	2.621	25.39	0.31	22.41	-	-	-	4.0325
Ag L	2.983	61.63	0.50	19.42	-	-	-	68.1803
Total		100.00		100.00				

Figure 5. EDX spectra and elemental composition of AgNPs synthesized using *Bouea macrophylla* stem bark extract at extract concentrations of (a) 1%, (b) 2%, and (c) 4% (w/v).

The particle size distribution of the synthesized AgNPs was further evaluated by measuring 50 randomly selected particles from the SEM image at 10,000× magnification using ImageJ software, and the results are presented in Figure 6. As shown in Figure 6, the particle diameters ranged from approximately 30 to 210 nm, with the majority of particles distributed between 80 and 140 nm. The particle size distribution followed a log-normal distribution, with the fitted curve showing a maximum frequency at approximately 108 nm, indicating that most nanoparticles were concentrated around this size. The log-normal fitting also demonstrated a satisfactory correlation (R^2 = 0.859), suggesting that the measured particle sizes were reasonably represented by the fitted model. Although a small proportion of larger particles was observed, likely due to particle agglomeration during synthesis or sample preparation, the overall distribution indicated relatively uniform nanoparticle formation. These findings are consistent with the SEM observations (Figure 4), confirming that the green synthesis using *Bouea macrophylla* stem bark extract successfully produced AgNPs with a relatively homogeneous nanoscale size distribution. Particle size based on Image G analysis from SEM results was obtained at a magnification of 10.000. The nanoparticle diameter distribution shows that most particles fall within the range of 100–200 nm. Overall, the distribution of silver particles ranges from 20 to 220 nm, which is still classified as the nanoparticle size range [18].

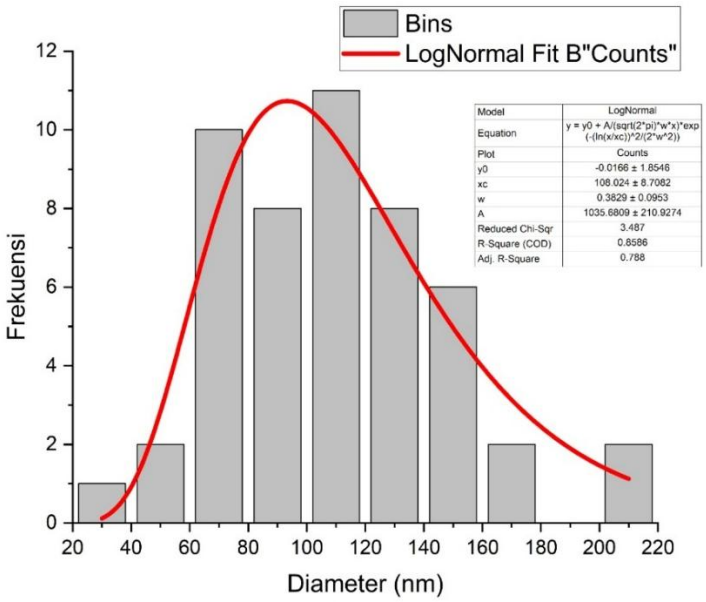


Figure 6. Silver particle size measured from 50 points using Image G at a magnification of 10.000.

3.6. Aplication test of 4% AgNPS on facial creams.

The application test was conducted using two commercially available whitening creams purchased from Keranggot Market, Cilegon. The AgNPs used in this study were synthesized using a 4% (w/v) gandaria stem bark extract. The analysis included a negative control consisting of whitening cream mixed with distilled water and a positive control consisting of a 0.01 M $HgCl_2$ solution mixed with AgNPs. The test samples were prepared by weighing 0.5 g of each whitening cream (A and B), dissolving the samples in 2 mL of distilled water, and subsequently adding 2 mL of the AgNP colloidal solution. The results showed that both whitening cream samples exhibited a rapid and pronounced color change compared with the negative control. The intensity of the color change was also greater than that observed for the positive control, indicating the possible presence of a relatively high concentration of mercury in both cream samples. These findings demonstrate that the synthesized AgNPs can serve as a rapid preliminary qualitative colorimetric sensor for detecting mercury in facial whitening creams. However, further quantitative analysis using instrumental techniques is recommended to confirm the mercury concentration in the tested samples.

4. Conclusions

Silver nanoparticles (AgNPs) were successfully synthesized through a green synthesis approach using *Bouea macrophylla* (gandaria) stem bark extract as a natural reducing and stabilizing agent. Among the tested extract concentrations, the 4% (w/v) extract produced AgNPs with the most favorable characteristics, exhibiting the highest selectivity toward Hg^{2+} ions based on the colorimetric response. Morphological characterization by SEM revealed relatively uniform nanoparticles with some degree of agglomeration, while particle size analysis confirmed a predominantly nanoscale distribution. SEM–EDS analysis further verified the successful synthesis of AgNPs, with silver identified as the predominant element, indicating high nanoparticle purity. The synthesized AgNPs demonstrated excellent selectivity toward mercury compared with Pb^{2+} and Ca^{2+}, producing a rapid and distinct color change upon interaction with Hg^{2+} ions. Furthermore, the application test on commercially available facial

whitening creams demonstrated that the AgNPs synthesized using the 4% extract concentration could effectively serve as a rapid preliminary qualitative colorimetric sensor for mercury detection. These findings highlight the potential of gandaria stem bark as a sustainable and eco-friendly precursor for AgNP synthesis and demonstrate the applicability of the synthesized nanoparticles as a simple, low-cost, and environmentally friendly sensing material for the preliminary screening of mercury contamination in cosmetic products. Further studies should focus on improving sensitivity, selectivity, and quantitative detection performance for practical analytical applications.

Author Contribution

Bramantyo contributed to the conceptualization, data collection, data analysis, writing of the original draft, and funding acquisition. Juwarin Pancawati contributed to the methodology. Yus Rama Denny supervised the research. All authors have read and agreed to the published version of the manuscript.

Competing Interest

All authors should disclose any financial, personal, or professional relationships that might influence or appear to influence their research.

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