

Biosynthesis of gold and silver nanoparticle using water hyacinth (*Eichhornia crassipes*) extract for photocatalytic degradation of organophosphate and organochlorine pesticide

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Abstract

The study aimed to assess the efficiency of synthesized gold (Au) and silver (Ag) nanoparticles in the degradation of organochlorine and organophosphate pesticides through photocatalysis. The synthesis of gold and silver nanoparticles was achieved using *Eichhornia crassipes* (water hyacinth extract). Photocatalytic degradation tests were conducted on organochlorine and organophosphate pesticides using gold and silver nanoparticles, with the absorbance of the samples measured by a UV spectrophotometer. The photocatalytic degradation rates of organochlorine and organophosphate were determined, with varied concentrations of the synthesized nanoparticles. The results showed high degradation rates at lower concentrations (10–20 ppm), with degradations of 51.789%, 47.954%, 47.983%, 44.088%, 41.565%, and 36.749% for 25/75, 50/50, and 75/25 Au nanoparticle ratios, respectively.

The results also revealed that higher degradation rates were observed at longer reaction times (70–80 minutes), with percentage degradations of 44.344% and 49.987%, 41.754% and 45.937%, 36.773% and 40.458% for 25/75, 50/50, and 75/25 Au nanoparticle ratios, respectively. Lower degradation efficiencies were observed at shorter reaction times (10–20 minutes), with percentage degradations of 15.356% and 19.982%, 13.746% and 17.082%, and 10.976% and 15.167% for 25/75, 50/50, and 75/25 ratios, respectively.

Additionally, the results showed high degradation rates at lower concentrations (10–20 ppm) for Ag nanoparticles, with percentage degradations ranging from 40.814% to 44.822% across AgNP ratios (25/75, 50/50, 75/25), indicating efficient degradation at lower concentrations. Conversely, at higher concentrations (60–80 ppm), the degradation efficiency was notably lower, with percentage degradations ranging from 7.004% to 13.539% across different AgNP ratios.

In conclusion, Au nanoparticles exhibited higher photocatalytic efficiency than Ag nanoparticles, particularly in degrading organophosphate (Sniper) pesticides. It is recommended that these synthesized nanoparticles be considered as environmentally friendly and cost-effective options for pesticide degradation.

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1. Introduction

Nanoparticles are microscopic particles with a diameter of less than about 100 nanometers (nm). In aerosol science, the term is often reserved for particles less than 50 nm in diameter, while the term "ultrafine particles" is used for particles less than 100 nm in diameter (Woo *et al.* [20]).

Nanobiotechnology is an important emerging field of nanotechnology that has developed dramatically for a variety of applications. The biosynthesis of different nanoparticles has seen enormous growth in popularity since the beginning of the twenty-first century (Woo *et al.* [20]).

Biosynthesis (or green synthesis) is more eco-friendly and cost-effective due to its chemical stability, excellent biocompatibility, easy fabrication process, convenient surface bio-conjugation with molecular probes, excellent optical properties related to surface plasmon resonance, and low toxicity (López-Millán *et al.* [13]).

The synthesis of silver and gold nanoparticles has gained significant attention due to their unique properties and potential applications in various fields. Silver and gold nanoparticles, in particular, have demonstrated excellent magnetic properties and biocompatibility. Recently, there has been a growing interest in developing environmentally friendly and cost-effective synthesis methods for magnetite nanoparticles. One such approach involves the utilization of natural extracts as reducing and stabilizing agents. Based on different chemically synthesized, silver and gold nanoparticles obtained by the biogenic routes are free from toxic contamination with by-products. On the other hand, the biological synthesis of silver and gold nanoparticles has advantages such as the simple, single-step, cost-effective, environmentally friendly, and biocompatible nature of synthesized gold nanoparticles (Cheng *et al.* [7]).

Gold nanoparticles (AuNPs) are particles of gold that range in size from 1 to 100 nm. AuNPs have gained a lot of interest because of their exceptional electronic optical (Carrillo-Cazares *et al.* [6]), mechanical, chemical, and physical capabilities AuNPs have unique qualities of employing bigger surface area to volume ratio, diverse sizes and shapes, and therefore they demonstrate enormous potential in various domains of biotechnology and biomedicine, such as protein and DNA degradation (Amendola *et al.* [4]).

Water hyacinth (*Eichhornia crassipes*) is a floating, invasive aquatic plant that is highly toxic and contains significant amounts of phenolic compounds with antioxidant properties. Multiple studies have shown that water hyacinth extract has the potential to create metal nanoparticles through its reducing and capping properties. Despite numerous attempts to eradicate this species, *Eichhornia* persists in spreading across many inland water systems, posing a substantial threat to aquatic life. Organophosphate and organochlorine pesticides also pose significant environmental and human health risks, emphasizing the need for an effective treatment method using biosynthesized gold and silver nanoparticles. The accumulation of these chemicals in water sources can pollute water supplies and disrupt aquatic ecosystems, with contamination having serious consequences for biodiversity and the overall health of ecosystems.

A pesticide is a substance used to control, prevent, kill, or reduce the population of pests. Synthetic pesticides known as organochlorine (OC) insecticides are extensively used globally. These chlorinated hydrocarbon derivatives have a wide range of applications in the chemical industry and agriculture. Short-term exposure to organochlorine pesticides can cause convulsions, headaches, dizziness, nausea, vomiting, tremors, confusion, muscle weakness, slurred speech, salivation, and sweating (Dabholkar *et al.* [9]).

Prolonged exposure to organochlorine insecticides can have detrimental effects on the liver, kidneys, central nervous system, thyroid, and bladder. Numerous chemicals have been associated with higher rates of liver or kidney cancer in animals. Some studies indicate that organochlorine insecticides may also contribute to

the development of cancer in humans. Organochlorine insecticides have half-lives ranging from months to decades, and in some cases, even up to a century (Dabholkar *et al.* [9]).

Organophosphate and organochlorine pesticides are widely recognized for their harmful impacts on nontarget organisms, their tendency to accumulate in the food chain, and their ability to contaminate soil and water sources. Consequently, there is a growing need for innovative and sustainable approaches to address the issue of pesticide pollution and its detrimental effects. One promising approach is photocatalytic degradation, which employs catalysts to decompose pesticides when exposed to light energy. In recent years, scientists have examined the use of natural resources, specifically plant-based materials, as catalysts for environmental remediation. The water hyacinth plant (*Eichhornia crassipes*), often considered an invasive aquatic species, has gained attention as a potential option due to its unique characteristics and abilities (Dabholkar *et al.* [9]).

2. Materials and Methods

The materials used included a rotary evaporator, rotary shaker, UV-visible spectrophotometer, and other necessary laboratory apparatus and equipment. All materials were of high quality and met standard requirements, with solutions and reagents purchased from Sigma-Aldrich.

2.1. Methods

2.1.1. Sampling point

Water hyacinth (*Eichhornia crassipes*) was collected along Hayingada–Mubi Road in the Girei Local Government Area of Adamawa State, at coordinates 9°17'16" N latitude and 12°26'48" E longitude.

2.1.2. Sample collection

Plant samples were collected at HayenGada, Mubi Road, in the Girei Local Government Area, Adamawa State, Nigeria, using a simple random sampling approach. The samples were gathered by hand using gloves and placed in polythene bags before being transferred to the laboratory for further processing. The plant samples were identified using an application called *PictureThis* and validated by a botanist, Mal Musa Ibrahim, from the Botany Division of the Department of Biological Sciences at MAU Yola, Nigeria.

2.1.3. Preparation of extract

A 10 g powdered sample was weighed with an analytical balance and transported to a 250 ml beaker containing 100 ml of deionized water, where it was heated for 20 minutes. After cooling, the mixture was then filtered through Whatman number 1 filter paper using a suction pump. The resulting extract was kept in a 50°C oven for subsequent examination.

2.1.4. Biosynthesis of the *Eichhornia crassipes* silver nanoparticles (EC-AgNPs)

As a silver ion reduction agent, 25 mL of leaf extract was mixed with 75 mL of 1 mM silver nitrate $(AgNO_3)$ solution. The reaction's effect was investigated at various temperatures (250, 500, and 750 degrees celsius). The reduction of silver nitrate over time was also assessed at various time intervals (25, 50, and 75

minutes) at each temperature. The creation of silver nanoparticles was established when the mixture turned yellowish-brown in color.

2.1.5. Biosynthesis of the Eichhornia crassipes gold nanoparticles (EC-AuNPs)

A 25 ml leaf extract was added to 75 ml of a 1 mM HAuCl₄· $3H_2O$ solution and introduced into a 250 ml beaker while stirring for 60 seconds. Seventy-five ml of the prepared 1 mM HAuCl₄· $3H_2O$ solution was introduced into a 250 ml beaker with constant stirring using a magnetic stirrer at different times, concentrations, and temperatures. The centrifuging process was repeated three times. Thereafter, the mixture was washed. The purified suspension was dried in a hot air oven to obtain a dried powder, which is the gold nanoparticle. Finally, the dried gold nanoparticles were analyzed.

3. Result and Discussion

60 50 % Degradation Rate 40 25/75 AuNPs 30 50/50 AuNPs 75/25 AuNPs 20 10 0 0 100 20 80 Concentration mg/dm³

3.1. Effect of concentration on the photodegradation of organophosphate

Figure 1. Comparison diagram of photocatalytic organophosphate degradation with different AuNPs concentration.

The data indicate a clear inverse relationship between the initial concentration of organophosphate and its percentage degradation. As the concentration increases, the degradation efficiency decreases. High experimental degradation rates were observed at lower concentrations (10-20 ppm) with degradation percentages of 51.789% and 47.954%, 47.983% and 44.088%, and 41.565% and 36.749% for 25/75, 50/50, and 75/25 AuNPs, respectively. Lower experimental degradation efficiency was observed at higher concentrations (60-80 ppm), with degradation percentages of 21.876% and 17.541%, 18.844% and 15.834%, and 15.483% and 11.903% for 25/75, 50/50, and 75/25, respectively

At lower concentrations, the photocatalyst has more available active sites relative to the number of organophosphate molecules, leading to higher degradation efficiency. As the concentration increases, the active sites become saturated, limiting the photocatalyst's ability to effectively degrade all the organophosphate molecules. A higher concentration of organophosphates can lead to a reduction in the penetration of light through the solution, which is essential for activating the photocatalyst. Reduced light penetration results in lower photocatalytic activity thus lower degradation percentages. With a higher concentration of

organophosphate, there is a greater chance of recombination of the electron-hole pairs generated by photocatalyst, reducing the number of active species like (hydroxyl radicals) available for degradation.



Figure 2. Comparison diagram of photocatalytic degradation of time with different AuNPs concentration.

The data illustrate a positive correlation between the reaction time and the percentage degradation of organophosphate. As the reaction time increases, the degradation efficiency also increases. This trend is common in photocatalytic processes.

Higher % degradation rates were observed at longer reaction times (70-80 minutes), with % degradations of 44.344 and 49.987%, 41.754 and 45.937%, and 36.773% and 40.458% for 25/75, 50/50, and 75/25 AuNPs, respectively. In contrast, lower experimental degradation efficiency was observed at shorter reaction times (10-20 minutes), with % degradations of 15.356 and 19.982%, 13.746 and 17.082%, and 10.976 and 15.167% for 25/75, 50/50, and 75/25, respectively. Longer exposure time allows more interaction between the photocatalyst and the organophosphate molecules. This extended interaction results in more organophosphate molecules being degraded over time. The trend suggests that the reaction is ongoing and has not yet reached a plateau within the 80-minute time frame, implying that further degradation could occur with additional time.

Over time, the accumulation of reactive species such as hydroxyl radicals and superoxide anions increases. The reactive species are responsible for breaking down organophosphate molecules. The longer the photocatalytic process runs, the more reactive species are generated, enhancing the degradation process. With increased time, there is a higher probability of electron-hole pairs participating in degradation reactions before recombination can occur. Effective separation of electron-hole pairs over extended periods can lead to higher degradation rates.



3.2. Effect of concentration on the photodegradation of organochlorine

Figure 3. Comparison diagram of photocatalytic degradation of time with different AuNPs concentration.

The data indicate a clear inverse relationship between the initial concentration of organophosphate and their percentage degradation. As the concentration increases, degradation efficiency decreases. High experimental degradation rates were observed at lower concentrations (10-20 ppm), with % degradations of 45.407 and 41.794%, 41.267 and 38.654%, and 38.667 and 35.401% for 25/75, 50/50, and 75/25 AuNPs, respectively. In contrast, lower experimental degradation efficiencies were observed at higher concentrations (60-80 ppm), with % degradations of 20.753 and 14.911%, 17.543 and 11.577%, and 15.422 and 9.107% for 25/75, 50/50, and 75/25, respectively.

At lower concentrations, the photocatalyst has more available active sites relative to the number of organophosphate molecules, leading to higher degradation efficiency. As concentration increases, the active sites become saturated, limiting the photocatalyst's ability to effectively degrade all the organophosphate molecules. A higher concentration can also reduce light penetration through the solution, which is essential for activating the photocatalyst. Reduced light penetration results in lower photocatalytic activity and, thus, lower degradation percentages. Additionally, at higher concentrations, there is a greater chance of recombination of the electron-hole pairs generated by the photocatalyst, reducing the number of active species, such as hydroxyl radicals, available for degradation.



3.3. Effect of time on the organochlorine photodegradation

Figure 4. Comparison diagram of photocatalytic degradation of time with different AuNPs concentration.

The data illustrate a positive correlation between reaction time and the percentage degradation of organochlorine. As the reaction time increases, degradation efficiency also increases, a trend common in photocatalytic processes. Higher % degradation rates were observed at longer reaction times (70-80 minutes), with % degradations of 37.832 and 44.744%, 35.832 and 41.034%, and 33.423 and 38.248% for 25/75, 50/50, and 75/25 AuNPs, respectively. In contrast, lower experimental degradation efficiencies were observed at shorter reaction times (10-20 minutes), with % degradations of 10.311 and 14.697%, 8.617 and 12.243%, and 7.107 and 10.193% for 25/75, 50/50, and 75/25, respectively.

Longer exposure times allow more interaction between the photocatalyst and organochlorine molecules, leading to more degradation over time. The trend suggests that the reaction is ongoing and has not yet reached a plateau within the 80-minute time frame, implying that further degradation could occur with additional time. Over time, the accumulation of reactive species, such as hydroxyl radicals and superoxide anions, increases. These reactive species are responsible for breaking down organochlorine molecules. The longer the photocatalytic process runs, the more reactive species are generated, enhancing degradation efficiency. With increased time, there is a higher probability of electron-hole pairs participating in degradation reactions before recombination occurs. Effective separation of electron-hole pairs over extended periods can lead to higher degradation rates.



3.4. Effect of concentration on the photodegradation of organophosphate

Figure 5. Comparison diagram of photocatalytic organophosphate degradation with different AgNPs concentration.

The experiment involved a range of organophosphate concentrations from 10 to 80 mg/dm³. The results, illustrated in Figure 5, clearly show that as the organophosphate concentration increased, catalytic degradation decreased. Analyzing the experimental data revealed a concentration-dependent trend in percentage degradation. It was observed that with an increase in the initial concentration of the pollutant, the percentage degradation rate decreased, indicating an inverse relationship between concentration and percentage degradation. This trend is consistent with photocatalytic degradation rates, as discussed by Zhang *et al.* [21]. The authors highlighted that higher initial concentrations of pollutants often lead to reduced efficiency due to maximum saturation effects and increased competition for active sites on the catalyst surface.

The experimental results displayed high degradation rates at lower concentrations (10-20 ppm), with percentage degradations ranging from 40.814% to 44.822% for different AgNP ratios (25/75, 50/50, 75/25), indicating efficient degradation at lower concentrations. Conversely, at higher concentrations (60-80 ppm), the experimental degradation efficiency was notably lower, with percentage degradations ranging from 7.004% to 13.539% for different AgNP ratios. This further reinforces the concentration-dependent trend observed in the experiment.

Moreover, it was observed that a higher concentration of organophosphates can lead to reduced light penetration through the solution, which is essential for activating the photocatalyst. Reduced light penetration results in lower photocatalytic activity and, consequently, lower degradation percentages. Additionally, a higher concentration of organophosphate increases the chances of recombination of the electron-hole pairs generated by the photocatalyst, reducing the number of active species, such as hydroxyl radicals, available for degradation.



3.5. Effect of concentration on the photodegradation of organochlorine

Figure 6. Comparison diagram of photocatalytic organochlorine degradation with different AgNPs concentration.

The concentration of organochlorine in the experiment ranged from 10 to 80 mg/dm³. The results, shown in Figure 6, indicate that as organochlorine concentration increases, catalytic degradation decreases. The experimental data reveal a clear trend where percentage degradation depends on concentration: as the initial pollutant concentration rises, the percentage degradation rate decreases. This inverse relationship between concentration and percentage degradation aligns with known photocatalytic degradation patterns. Higher initial concentrations of pollutants often reduce efficiency due to saturation effects and increased competition for active sites on the catalyst surface (Zhang *et al.* [21]).

Higher experimental degradation rates were observed at lower concentrations (10-20 ppm), with percentage degradations of 39.265% and 34.557%, 34.992% and 30.007%, and 30.437% and 26.741% for 25/75, 50/50, and 75/25 AgNPs, respectively. Conversely, lower experimental degradation efficiency was observed at higher concentrations (60-80 ppm), with percentage degradations of 15.044% and 11.338%, 9.018% and 7.541%, and 7.409% and 6.111% for 25/75, 50/50, and 75/25, respectively.

Higher organochlorine concentrations can reduce light penetration through the solution, which is crucial for photocatalyst activation. This reduced light penetration lowers photocatalytic activity, resulting in lower degradation percentages. Additionally, at higher organochlorine concentrations, the likelihood of recombination of electron-hole pairs generated by the photocatalyst increases, reducing the number of active species, such as hydroxyl radicals, available for degradation.



3.6. Effect of time on the photodegradation of organophosphate

Figure 7. Comparison diagram of photocatalytic degradation of time with different AuNPs concentration.

Time is a critical factor when assessing material degradability. The percentage degradation increases as the duration of the photocatalytic test increases, indicating a positive correlation between reaction time and organophosphate degradation (Wang *et al.* [19]). The degradation rate remains relatively gradual over time. The highest degradation rate was observed at 39.513% after 80 minutes with 25/75 AgNPs, while the lowest was recorded at 7.448% after 10 minutes with 75/25 AgNPs. This suggests that the photocatalytic process is effective over prolonged exposure periods.

In the initial time intervals (10 to 30 minutes), 25/75 AgNPs, 50/50 AgNPs, and 75/25 AgNPs demonstrated moderate degradation levels. As the reaction progresses, the accumulation of reactive species such as hydroxyl radicals and superoxide anions increases. These reactive species play a crucial role in breaking down organophosphate molecules, enhancing the degradation process with extended reaction times. Moreover, longer durations allow for a higher probability of electron-hole pairs participating in degradation reactions before recombination occurs. Effective separation of electron-hole pairs over extended periods can lead to increased degradation rates.



3.7. Effect of time on the photodegradation of organochlorine

Figure 8. Comparison diagram of photocatalytic degradation of time with different AgNPs concentration.

Time is a crucial factor in assessing a material's degradation capability. The observed percentage degradation values indicate an upward trend as time progresses, suggesting a positive correlation between the duration of photocatalytic activity and organochlorine degradation. The degradation rate remains relatively gradual, with the highest percentage degradation recorded at 34.703% for 25/75 AgNPs after 80 minutes, and the lowest at 6.281% for 75/25 AgNPs after 10 minutes. This pattern indicates that the photocatalytic process maintains effectiveness over extended periods. During the initial intervals (10-30 minutes), moderate degradation percentages were observed for 25/75 AgNPs, 50/50 AgNPs, and 75/25 AgNPs.

Over time, the accumulation of reactive species, such as hydroxyl radicals and superoxide anions, increases significantly. These reactive species are key agents in breaking down organophosphate molecules, intensifying the degradation process as time increases. With prolonged exposure, there is a greater likelihood of electron-hole pairs participating in degradation reactions before recombination occurs. Effective separation of these electron-hole pairs over longer durations results in higher degradation rates.

4. Conclusions

The study introduced an innovative approach for synthesizing silver (AgNPs) and gold (AuNPs) nanoparticles using water hyacinth, an invasive aquatic plant. These nanoparticles were explored for their potential in environmentally friendly and cost-effective pesticide degradation. The research focused on the photocatalytic degradation of organophosphate and organochlorine pesticides using various concentrations of AgNPs and AuNPs. The results indicated that AuNPs exhibited superior photocatalytic efficiency compared to AgNPs, especially in the degradation of organophosphate (Sniper) pesticides. In contrast, AgNPs showed relatively lower degradation activity. Overall, both AgNPs and AuNPs were effective in the photocatalytic degradation of organophosphate pesticides, with AuNPs demonstrating greater efficacy.

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