Silver Nanoparticles: Unveiling Catalytic Warriors Against Organic Dyes: A Current Review

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Abstract- **Dye wastewater discharged following inadequate treatment poses a significant threat to the environment and biodiversity, leading to a substantial increase in pollution levels. The presence of dyes in the effluent presents a challenge for conventional water treatment techniques due to their high resistance to removal. As a result, there is a pressing demand for more sophisticated and cost-efficient methodologies. A variety of metallic nanoparticles have demonstrated the capability to eliminate diverse contaminants from wastewater streams. Among these nanoparticles, silver nanoparticles (AgNPs) have garnered considerable attention for their adjustable optical characteristics attributed to the plasmonic effect. The biogenic route to nanoparticle synthesis offers a promising alternative to traditional physiochemical synthesis methods. Biological approaches, using plants, may be particularly suitable for large-scale production, thereby offering a sustainable avenue for utilization. Moreover, the rapid advancement of eco-friendly metallic nanoparticle synthesis is driven by its broad applications across various scientific disciplines. Biogenic Ag-NPs have been highlighted for their photocatalytic effectiveness, reusability, and environmentally friendly synthesis process, showcasing their efficacy in water purification applications. The catalytic potential of green-synthesized Ag-NPs has been evidenced in the removal of dyes and color detoxification processes.**

Keywords: **Dye, wastewater, catalytic degradation, silver nanoparticles.**

Introduction

Industrial and population growth in the recent past has caused a significant rise in the pollution of water resources in developing countries. Most of these industries release untreated waste in surrounding water reserves polluting them with various types of contaminants including widely utilized, organic dyes and nitro compounds. [1] Textiles, paper, pharmaceuticals, leather, plastic, and food industries are leading contributors to wastewater contamination with toxic organic dyes.[2][3] Dyes are commonly classified based on their solubility into anionic, non-ionic, and cationic types. Anionic dyes pose significant challenges due to their vivid hues and water solubility, making them difficult to remove using traditional wastewater treatments. On the other hand, non-ionic dyes, like disperse dyes, remain unchanged in aqueous systems. However, cationic disperse dyes tend to bioaccumulate and undergo reduction of their azo and nitro groups in soil sediments, resulting in the formation of toxic aromatic amines.[4] Azo dyes stand out as exemplary models for studying dye pollution because of their extensive production, high consumption rates constituting 60–70% of total dye usage, and significant discharge into aquatic environments.[5]. Dyes exhibit photochemical stability under normal environmental temperatures, and they are non-biodegradable.[6] Even at concentrations as low as 1 mg L^{-1} in effluents, dyes can display toxicity, contributing to environmental degradation and various diseases in living organisms. They pose toxigenic, carcinogenic, and mutagenic risks to both humans and the environment. [7] This risk is particularly pronounced with azo dyes, where carcinogenicity can arise from both the dye compound itself and its metabolized by-products.[8] This persistent dye accumulation has become a major concern as it singlehandedly affects several tropical levels of the ecosystem starting with primarily changing the aesthetics and subsequently disturbing aquatic flora and fauna by preventing penetration of sunlight into water.[4,9] Various methods classified into physical, and chemical methods like ozonation, adsorption, coagulation, electrochemical oxidation, and filtration have been tested for removal and have been employed over the years. However, they are not very convenient due to their high costs, formation of toxic by-products, stringent reaction conditions, and limited effectiveness. [10,11]

Nitroaromatic compounds play a crucial role in synthetic organic chemistry, serving as vital building blocks for various pharmaceuticals, agricultural chemicals, and dyes manufacturing industries.[12] However, nitrogen-containing organic compounds are significant sources of pollution in surface waters, said to have carcinogenic and mutagenic effects, leading to various adverse health effects ranging from inflammatory reactions affecting the skin and eyes to neurotoxicity. [13, 14,15,16]

Nitro compounds and azo dyes have been found to be reduced in aquatic environments, giving rise to toxins.[17] The development of mild, efficient, and economical reduction methods is an area of active research. Currently, several semiconductor metallic nanoparticles, such as zinc oxide and titanium dioxide, are efficiently employed for the degradation of these organic dyes. [18,19] Photocatalytic degradation of organic dyes using metal nanoparticles represents a significant approach for converting toxic molecules into environmentally friendly by products. This method is extensively utilized for the remediation of persistent pollutants due to its excellent catalytic activity and eco-friendly attributes.[20] However, their exclusive absorption of UV light, rather than visible light, renders them less practical for real-world applications. [21] Silver nanoparticles (Ag NPs), on the other hand, possess a remarkable ability to absorb both visible and UV light through the surface plasmon resonance (SPR) effect and Interbrand transitions, respectively. Physicochemical properties such as small size, shape, larger surface area, surface chemistry, and particle morphology distinguish silver nanoparticles from bulk silver metal. Additionally, AgNPs are renowned for their antimicrobial, optical, electrical, and catalytic properties, holding immense promise for effectively combating harmful dyes via photocatalytic processes. [22,23,24]. Silver nanoparticles have demonstrated effectiveness in efficiently reducing and removing dyes and various nitro compounds through both photocatalytic and catalytic means. With their unique physicochemical properties and diverse applications, silver nanoparticles hold immense promise for effectively combating harmful dyes and nitro compounds in aquatic environments.

Synthesis of silver nanoparticles

Since the beginning of nanotechnology, various methods of synthesis of silver nanoparticles have been developed and are still being explored to optimize their properties and maximize their efficiency for various applications. The fundamental synthesis routes in which silver nanoparticles have been synthesized include the conventional physical and chemical methods as well as biological methods of synthesis. [25,26] However, over the years researchers have shown a growing interest in the biological synthesis of silver nanoparticles due to its green and sustainable nature. physical methods are widely used but have very few advantages and are often expensive, less productive, and require harsh conditions that are not always environmentally friendly. In physical methods, the particle size of nanoparticles can be finely controlled, allowing for precise manipulation of their dimensions. Furthermore, the purity of the resulting silver nanoparticles is remarkably high.[27]

Chemical methods of synthesis are numerous and most commonly used. They involve metal precursors, reducing and stabilizing agents to produce nanoparticles with the desired size, shape, and stability. Chemical synthesis produces diverse shapes and sizes of silver nanoparticles and is mostly cost and time-efficient. However, the reducing agent and solvents used during synthesis can be toxic and impact the use of resultant silver nanoparticles in various biomedical applications like drug delivery, biosensing, and imaging.[28]

Hence a green approach for the synthesis of silver nanoparticles which is safe, sustainable, and economical is highly desirable. Biological route is considered a great alternative to the synthesis of silver nanoparticles of a nontoxic nature. [29,30,31] Biological synthesis uses stabilizing and reducing agents derived from natural sources such as plants, microorganisms, algae, fungi, and even some animal waste products. Moreover, biological methods yield a substantial quantity of nanoparticles characterized by well-defined shapes and uniform sizes. However, metal nanoparticles tend to aggregate easily due to factors such as high surface energy, small size, and van der Waals forces. This aggregation diminishes their catalytic activity and reusability, underscoring the necessity for approaches to enhance nanoparticle stabilization and prevent aggregation. Plant-derived capping agents offer stability and inhibit aggregation, making them specifically suitable for catalysis. [32,33]

Plant-Mediated Silver Nanoparticle Synthesis

Plants are abundant in biomolecules such as proteins, vitamins, enzymes, phenolic compounds, and secondary metabolites that possess stabilizing and reducing properties essential for nanoparticle synthesis.[34]] Using plant extracts for green synthesis of silver nanoparticles aligns with the principle of green chemistry, while also producing stable nanoparticles in well-defined shapes, desired sizes, and high purity.[35] Fruits, leaves, stems, seeds, and even flowers of various plant species have been used for the synthesis of silver nanoparticles. This reaction process resembles with chemical synthesis as the plant extract acts as both a reducing and stabilizing agent while eliminating the need for toxic chemicals. These plants, namely *Cornus officinalis, Atrocarpus altilis, Azadirachta indica, Coffea arabica, Melia azedarach* have been harnessed for the synthesis of silver nanoparticles. [36-40]

The antioxidant activity of the biomolecules presents in *Syzygium cumini* fruit extract played a crucial role in the reduction of silver ions into silver nuclei. The strong antioxidant properties of the phytoconstituents facilitate the successful reduction of silver ions (Ag^{+}) into zero-valent silver ions (Ag^{0}). These flavonoids act as reducing agents, initiating the formation of Ag^{0} nuclei from Ag^{+} ions further leading to the formation of stable silver nanoparticles.[41]

In another study, it was observed that the size and shape of silver nanoparticles synthesized using *Crocus haussknechtii bois* extract are significantly influenced by temperature and pH optimization. It was determined that by optimizing the

temperature to 75°C and pH to 7, spherical nanoparticles ranging from 10-25 nm could be obtained within 120 minutes.[42]

The pH of the solution impacts the surface charge of the nanoparticles, affecting their stability and interactions with other molecules. It also influences the reduction rate of silver ions and the nucleation process, ultimately shaping the size and morphology of the nanoparticles. Likewise, the temperature of the reaction solution plays a crucial role in the kinetics of the reaction, nucleation rate, and nanoparticle growth. Higher temperatures promote faster reduction rates and nucleation, leading to smaller nanoparticles, while lower temperatures favor the formation of larger nanoparticles. [43,44]

In addition to pH and temperature, factors such as leaf extract concentration, the volume of extract, stirring rate, and reaction time also play pivotal roles in both the synthesis and performance of silver nanoparticles using phytosynthetic methods. The precise control of these parameters is essential for fine-tuning the synthesis process and crafting silver nanoparticles with tailored characteristics to meet specific requirements.[45]

Kinetic Profiling: Analysing photodegradation Dynamics and Mechanisms

Photodegradation of the organic dye with silver nanoparticles undergoes a complex mechanism mediated by the interaction of light energy with the nanoparticles and the dye molecules. Under the light, especially of the visible range, AgNPs absorb the light energy, and by undergoing the SPR mechanism they excite the electrons which attain higher energy levels leaving holes on their surface that conduct electron-hole pairs e^{-}-h^{+}. [46,48] Organic dye molecules in the solution are also adsorbed through several bonding interactions. The excited electrons on the nanoparticles transfer to the adsorbed dye molecules which reduce the molecule by electron transfer from the dye molecule. The process separates the dye molecules into smaller and less harmful entities. [47] The holes in the AgNPs generated in the reaction combine with the molecule to form an atom. Conversely, the holes created on the silver nanoparticles react with molecular oxygen or water to form hydroxyl radicals and superoxide radicals. The ROS which is extremely susceptible causes the oxidation of dye molecules through disintegration into smaller institutions or are wholly mineralized into carbon dioxide and water.[48] Consequently, the silver nanoparticles may undergo surface reconstruction or oxidation, and regeneration can restore them for other catalytic processes.[49]

In conclusion, the photocatalytic degradation mechanism of organic dyes by AgNPs is based on the synergistic cooperation of light energy, silver nanoparticles, dye molecules, and hydroxyl radicals to effectively eliminate dye pollutants from aqueous systems. The effectiveness of the photocatalytic dye degradation process depends on several parameters, including the concentrations of AgNPs and dye, the pH of the solution, reaction duration, and the intensity and source of light. [46-50] Research focused on exploring the photocatalytic capabilities of diverse bio-fabricated AgNPs for degrading dyes, alongside investigating the impact of different parameters on photocatalysis is discussed below:

Radish Microgreens have been utilized in the synthesis of AgNPs and were employed as a photocatalyst to facilitate the degradation process of hazardous crystal violet and pyronin Y dyes, demonstrating a notable degradation efficiency of 96.90% and 94.46%, respectively.[51]

A separate study focused on bio-reduced silver nanoparticles that were synthesized utilizing *Trigonella foenum-graecum* leaf extract, showcasing photocatalytic degradation capabilities of approximately 88% and 86% for reactive blue 19 and reactive yellow 186 dyes under sunlight irradiation. The assessment of COD analysis of the solution was carried out to evaluate the mineralization of organic dye species, indicating the transformation of organic compounds into simpler and less harmful substances like CO_2 and water within the degraded medium.[52]

The photocatalytic activity of green-synthesized *P. Speciosa* AgNps of 35 nm was found to be significant, achieving an 84% degradation rate in 180 min in the presence of Methylene blue(MB) and AgNPs under sunlight irradiation, surpassing the efficiency of solutions containing MB alone or with NaOH. Upon conducting a pseudo-first-order kinetics analysis, a degradation rate constant of -K = 0.0098 was determined, with the transition of the MB solution from deep blue to colourless indicating effective degradation.[53]

The decolorization potential of silver nanoparticles that were synthesized using *Aloe barbadensis* miller leaves extract was evaluated for various dyes including eosin, crystal violet, carbol fuchsin, MB, and saffranine, showcasing maximum degradation percentages of 70.76%, 95.54%, 94.63%, 93.53%, and 90.12%, respectively, under UV illumination. [54]

The influence of different parameters on the photocatalysis process was also highlighted, particularly in the comparison involving herbal plant *Kalanchoe pinnata* leaves mediated AgNPs, demonstrating the photocatalytic degradation of Rhodamine B (RhB) dye under UV-light and dark conditions. The research findings indicated that the dye exhibited faster degradation under UV irradiation, with approximately 83% degradation occurring in 45 minutes, characterized by a rate constant (k) of 0.042 min^{-1}, and 87% degradation in the dark within 150 minutes, featuring a k value of 0.013 min^{-1}. Furthermore, the study explored the impact of various parameters such as pH, temperature,

reactant concentrations, and reaction time, confirming that alkaline pH and high temperature were more conducive to the synthesis of AgNPs.[55]

Another investigation delved into the photocatalytic effectiveness of AgNPs derived from *C. pangorei* against RhB dye under ultraviolet exposure at specific intervals, revealing that these synthesized nanoparticles could decompose Rh-B dye by up to 86% within a 2-hour irradiation period.[56]

Additionally, a comparative analysis was conducted during the photodegradation process of *Alpinia officinarum*-synthesized AgNPs on malachite green (MG) and MB dyes under UV (365 nm) and visible light (400 W) exposure, indicating significant degradation activity by AgNPs, with MG dye degradation reaching approximately 91% and MB dye degradation at 38% under UV light within a 2-hour irradiation period. However, under visible light, MG dye degradation was observed in approximately 38% of samples, while MB dye degradation reached 80% after 2 hours of irradiation.[57]

A comprehensive research study was also conducted on the removal of dye, considering factors such as contact time, UV light exposure time, nanoparticle dosage, temperature, and pH by *Albizia procera* AgNPs. Numerous conclusions were drawn from this extensive study.[58] Furthermore, the investigation into a variety of bio-fabricated AgNPs for dye degradation highlights their significant promise in environmental remediation efforts.

CATALYTIC PROWESS OF SILVER NANOPARTICLES

Nitroaromatic compounds are a threat to almost every living thing. nitrophenols are worse as they are the most common pollutants from manufacturing units of various industries and they are nonbiodegradable as well as carcinogenic.[59]

AgNPs demonstrate efficacy as catalysts in the degradation of synthetic dyes via $NaBH_4$-based processes. Notably, $NaBH_4$ alone in dye solution fails to catalyze the reduction process due to a thermodynamic favorability mismatch, despite its inherent feasibility. [60] This discrepancy arises from substantial differences in redox potential between the donor and acceptor species. The introduction of the silver nanocatalyst effectively mitigates this limitation by providing an alternative pathway characterized by a lower activation energy for the reduction reaction. Consequently, this augmentation renders the reduction process not only thermodynamically favorable but also kinetically feasible. Silver nanoparticles, in case of dyes act as electron mediators, enhancing the electron transfer process between the BH_4-ion and the dye. This dual role involves facilitating the transfer of electrons from the BH_4-ion to the dye while concurrently becoming saturated with BH_4-ions, which subsequently transfer electrons to the dye. Through this mechanism, silver nanoparticles play a pivotal role in expediting electron transfer from the reducing agent BH_4- to the dye. Consequently, this process leads to the disintegration of the chromophore structure of the dye into smaller molecules. Functioning as a redox catalyst, silver nanoparticles effectively diminish the kinetic barrier between the nucleophilic $NaBH_4$ and the electrophilic dye, thus facilitating the reaction. [61,62]

Peltophorum pterocarpum leaves extract stabilized AgNPs were utilized for the chemical degradation of methylene blue in the presence of AGNPS and $NaBH_4$. The investigation focused on optimizing pH and temperature parameters to achieve the highest degradation efficiency, particularly at a neutral pH and 70°C temperature, with a rapid reaction speed of 2.5 minutes recorded. The catalyst exhibited consistent catalytic performance, maintaining stability without any observable changes in morphology even after 5 cycles, as evidenced by the HRTEM micrograph of the retrieved NPs.[63]

Biogenic AgNPs derived from *Imperata cylindrica* extract demonstrated the enhancement of methylene blue degradation, reaching up to 92.06% within a span of 14 minutes. [64]

Furthermore, the catalytic performance of nanoparticles synthesized from *Crocus haussknechtii boiss* extract was evaluated for the degradation of Congo Red dye, showing that 16.67 µM of the dye could be degraded in just 220 seconds, with a first-order kinetic model constant (Kapp) of 0.011 s^{-1} for 5.39 µg of synthesized AgNPs. The degradation rate achieved stands as one of the highest values reported for the catalytic reduction of Congo Red dye.[65]

Conclusion

Nanobiotechnology presents numerous compelling advantages and provides a cost-effective approach to safeguarding environmental bioremediation. The utilization of green synthesized nanoparticles can serve as an efficient method for engaging in antimicrobial and photocatalytic activities concurrently. The utilization of various natural compounds found in plant extracts is recognized for their ability to function as both reducing and stabilizing agents during the production of silver nanoparticles. The stability of plant-mediated silver nanoparticles is attributed to the existence of inherent capping agents, which play a role in averting particle aggregation. Moreover, silver nanoparticles created through green synthesis exhibit promising potential in applications, particularly as photocatalysts, although further exploration is necessary to transition their use from laboratory settings to practical water remediation scenarios. Evidently, the potential environmental and health implications stemming from the presence of silver nanoparticles may present challenges in their broad implementation. Therefore, a comprehensive investigation into the accumulation and mode of

action of Ag NPs within the human body is imperative. Hence, the assessment of silver nanoparticle toxicity and the associated risks linked to their use in treating dye effluents is essential.

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