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Green Synthesis of Silver Nanoparticles Using Penicillium Species: pH-Dependent Formation and Catalytic Performance

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Abstract

Background: The utilization of Penicillium species for the environmentally friendly synthesis of silver nanoparticles has demonstrated effectiveness, as supported by experimental evidence. This method offers a sustainable alternative for nanoparticle production.

Purpose: The goal of this study was to look into the influence of pH on the optical characteristics, structural attributes, and catalytic performance of silver nanoparticles synthesized using Penicillium species. Understanding the pH dependency provides insights into controlling and optimizing nanoparticle properties.

Research Methodology: Silver nanoparticle synthesis was confirmed through visual inspection and UV-visible spectroscopy, displaying an absorption band for surface plasmon resonance between 408 and 415 nm. Fourier-transform infrared spectroscopy identified biomolecules responsible for nanoparticle stabilization and reduction. X-ray diffraction analysis confirmed the tiny-particles' cubic crystalline structure. The size distribution and shape of the nanoparticles were clarified by transmission and scanning electron microscopy. Photoluminescence spectroscopy examined luminescence properties, while catalytic activity was assessed through methylene blue reduction.

Findings: Higher pH values led to a blue shift and surface plasmon resonance curve that is sharper, influencing the intensity and location of the absorption peak. The nanoparticles exhibited a cubic crystalline structure with a crystallite size of approximately 16.69 nm. They appeared spherical, uniformly distributed, and ranged from 9 to 19 nm in size. Luminescence characteristics were observed, with emission peaks at 530 nm upon excitation at 400 nm. The nanoparticles demonstrated efficient catalytic activity, evidenced by rapid methylene blue reduction when sodium borohydride is present.

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1. Introduction

The dimensions and form of metallic tiny particles influence their electrical, optical, and chemical characteristics [1-4]. The oscillation of electrons on metallic nanoparticle surfaces is caused by the interaction of incoming light with free electrons. Plasmons are a cloud of pulsating electrons that form on the surface of these nanoparticles. The main force behind nanoscale engineering and the focus of deep investigation are plasmons. When the oscillation frequency of free electrons and the frequency of incident light collide, resonance is produced. We refer to this phenomena as surface plasmon resonance. This SPR is in charge of the colours that the metallic tiny-particles display[5]. The metallic tiny-particles' surface plasmon resonance is dependent on their dimensions, form, and the liquid medium they are suspended in. Nanoparticles are tolerated by a shift in the LSPR wavelength when they are used to detect biological and chemical substances [6]. The spectrum's visible region contains the SPR absorption band for Nobel metal tiny-particles like gold and silver. Tiny-particles of gold and silver showed size-dependent colour change [7].

It has been observed that the optical [8,9], catalytic [10, 11] , and characteristics of silver nanoparticles that are antibacterial [12] are depending on their size and structure. Because plasmon is size-dependent, it may be able to grow electrical or photonic apparatuses founded onplasmon stimulation and identification. The reaction solution's pH level affects how dark the brown colour of the silver tiny-particles is [7].

The Manufacturing of silver tiny particles by standard methods involving chemical and physical processes is the subject of several publications. However, synthesis of nanoparticles using these technologies required the application ofhazardous chemicals that have a detrimental impact on the surroundings. Therefore, the application of organic stabilising and reducing chemicals in green synthesis techniques is favoured for producing silver nanoparticles. Extracellular creation of nanoparticles is made possible by the huge number of extracellular enzymes found in fungi [13]. A number of fungus have been shown to be capable of producing silver nanoparticles, including Aspergillus flavus [14], Aspergillus niger [15], Trichoderma asperelum [16], and Penicillium species [16–18].

Due to their catalytic performance, green synthesised silver nanoparticles have been reported to be useful for environmental maintenance. Size-dependent catalytic performance is demonstrated by silver nanoparticles synthesised with Kashayam Guggulutiktham in the reduction of MB [19]. Catalytic activity of silver nanoparticles is based on their shape, synthesised by a straightforward solvothermal technique, was observed for the oxidation of styrene [20].

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The current study describes how Penicillium species are used to synthesise silver nanoparticles quickly and outside of cells. It was looked at how the pH levels of the reaction liquids influenced the way the silver nanoparticlesoptical characteristic SPR.

2. Literature Review

Talank et al. (2022) examined the physicochemical and biological characteristics of silver nanoparticles (AgNPs) produced via green synthesis. The characterization techniques used were FT-IR, DLS, XRD, SEM, and TEM. AgNPs showed a crystalline structure and spherical shape. MIC was comparable to tetracycline at 8 μg mL−1 against Staphylococcus aureus. AgNPs shown strong antimicrobial and biofilm-inhibiting properties. They also showed notable antioxidant and anticoagulant qualities. The potential of biogenic AgNPs for a range of biomedical applications is highlighted by these findings.[21]

In 2023, Kumari et al. offered a critical analysis of environmentally friendly methods for size and sculpting metal nanoparticles (MNPs) for use in environmental applications. They emphasised the importance of MNPs and the drawbacks of traditional synthesis techniques. The review's main objective was to investigate several green synthesis approaches, such as ecologically friendly and biological methods. There was a lot of discussion about environmental applications including heavy metal removal and pollution degradation. Green-synthesized MNPs were evaluated against their conventionally synthesised counterparts in terms of performance. We also looked at the regulatory obstacles to scaling up green synthesis methods. Overall, the review highlighted how green synthesis techniques shape MNPs for long-term environmental solutions in an environmentally responsible manner.[22]

Radulescu et al. (2023)gave a thorough analysis of the fundamentals and biological uses of green metal and metal oxide nanoparticle production. They emphasised the growing curiosity about nanotechnology and the enhanced development of metallic tiny particles in tissue engineering, with a focus on the ability to manage scaffold characteristics and their antibacterial qualities. The review focused on the negative environmental effects of conventional synthesis techniques and the benefits of green synthesis, such as reduced waste and the use of cleaner solvents. It was explored how biomolecules could be used in green synthesis to increase antibacterial action and decrease toxicity. The study's objectives were to discuss green-synthesized nanoparticle production methods, chosen solvents, parameters, and the most recent biomedical uses.[23]

Michael et al. (2022)examined biologically mediated approaches for synthesising nanoparticles, with a particular emphasis on processes mediated by algae and fungi. To serve as a guide for upcoming study, they thoroughly examined the synthesis techniques, applications, composition, and characterization of nanoparticles in five distinct categories. It has been observed that conventional approaches to the process of creating nanoparticles havedrawbacks, comprising the toxicity of precursor materials, the need to regulate high temperatures, and expensive synthesis costs. Utilising bioreduction by biomolecules or

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enzymes found in fungus and algae, greener synthesis has been emphasised as a low-energy, low-cost, and low-temperature method of producing different metallic nanoparticles utilised in industries and healthcare.[24]

In 2021, Satapathy et al.talked about the harmful effects of industrial pollution, especially as it relates to the textile sector, which releases about 50,000 tonnes of synthetic dyes yearly. As an environmentally acceptable way to reduce water pollution, they suggested use nanoparticles of metal and metal oxide that have been green synthesised for photocatalytic destruction. The writers highlighted the benefits of the green synthesis technique, emphasising how environmentally friendly and sustainable it is. They investigated the various uses of nanoparticles, such as antibacterial activity, bioimaging, catalysis, and sensing. They also looked into the function of natural extracts in the manufacturing of the possibility of Investigating the degradation of dyes through photocatalysis using nanoparticle which opens up new avenues for the environmentally safe treatment of wastewater.[25]

3. Experimental Methods

Two analytical-grade chemicals were used: AgNO3 and 0.1 N NaOH. NCIM 1313 Penicillum species a type of fungal, was procured in Pune is home to the National Chemical Laboratory Culture Collection Centre.

3.1 Preparation of Fungal Filtrate

Potato starch broth was employed as a sterile liquid medium to cultivate the fungus species Penicillium [26]. Following a 7-day incubation period at 30°C in static conditions, the mycelial mat was spotted on the media. Utilizing Whatman No. 1 filter paper, isolate the mycelia. To get rid of all traces of the media component, deionized water was thoroughly rinsed over the mycelia that had been harvested on the paper. One hundred millilitres of deionized water were used to suspend ten fresh grammes of mycelia, which were then incubated for seventy-two hours at 30 degrees Celsius with 110 revolutions per minute of shaking. After the incubation period, the fungal mycelia were divided utilising Whatmann No. 1 filter paper, and silver nanoparticles were made from the filtrate[27].

3.2 Production of Silver Nanoparticles

An equal volume of AgNO3 solution and fungal filtrate at a final 1 mM concentrations were contained in each of the five flasks. The pH values of the flasks were changed to range from 7 to 11, and they were all incubated for 72 hours at 30 °C with trembling at 110 rpm. 0.1 N NaOH solution was applied to change the pH of the reaction solution.

3.3 Characterization

By monitoring colour changes, the creation of silver nanoparticles may be seen visually. The criterion used to investigate how pH influences how silver nanoparticles are made is variation in colour intensity. The method employed to verify the creation of nanoparticles made from silver and investigate how the reaction's solution's pH influences nanoparticle synthesis is

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called UV-visible spectroscopy. Changes in the absorption band and colour intensity in UV-visible spectra were utilised as a gauge to identify how pH affected the silver nanoparticles' optical properties (SPR).

Using FTIR spectroscopy, the fungal filtrate was scanned between 500 and 4000 cm-1. FTIR analysis was used to assess the existence of viable biomolecules that were in charge of stabilising the produced nanoparticles and reducing the silver ions.

To ascertain the crystal structure of the materials, which were in powder form, diffraction using X-ray analysis was performed. 2θ was used to measure the diffracted intensities using CuKα radiation in the 20–80° range. Electron microscopy with scanning (SEM S-4800) was utilized to look over the size and shape of thin films made from a colloidal mixture containing silver tiny-particles that were generated on glass slides. The silver nanoparticles were captured utilizing a Philips CM 200 transmission electron microscope, which allowed for the acquisition of TEM images. The produced silver nanoparticles' photoluminescence spectra were captured at room temperature using a Spectrophotometer Perkin Elmer LS 55. At a rate of 100 nm/minute, the emission and excitation slit widths were both maintained at 5 nm. The produced silver nanoparticles' catalytic effectiveness was assessed for NaBH4's decrease of MB.

4. Results and Discussion

4.1 Visual Observation

The AgNO3 solution (not shown) and the fungal filtrate (Fig. 1a) do not change colour after 72 hours of incubation; on the other hand, the fungal filtrate treated with AgNO3 turns brown, which is the initial indication that silver nanoparticles are developing (Fig. 1b). Figure 2 shows five flasks showing how the intensity of colour changes as the reaction medium's pH rises. As the pH value increased, so did the color's intensity. It implies that the silver ion reduction rate rises with increasing pH value, leading to a faster formation of nanoparticles [28]. The produced silver nanoparticles' surface plasmon resonance was what gave them their brown hue [29].

Fig. 1 (a) Filtrate of Penicillium fungus and (b) synthesized AgNPs

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Fig. 2 Changes in color intensity in relation to pH

4.2 UV-Visible Spectroscopy

The method utilised to investigate the Plasmon resonance at the surfacedisplayed by a nanoparticle of silver generated at various pH values is UV-visible absorption spectroscopy. The visible and UV spectrums of the silver nanoparticles prepared at different pH levels and the fungal filtrate are displayed in Fig. 3. The absorbance of fungal filtrate is attributed to the amide band at 237 nm and the residues of tyrosine and tryptophan in the protein molecule at 277 nm [30]. There isn't an absorbance peak in the visible area for the fungal filtrate.

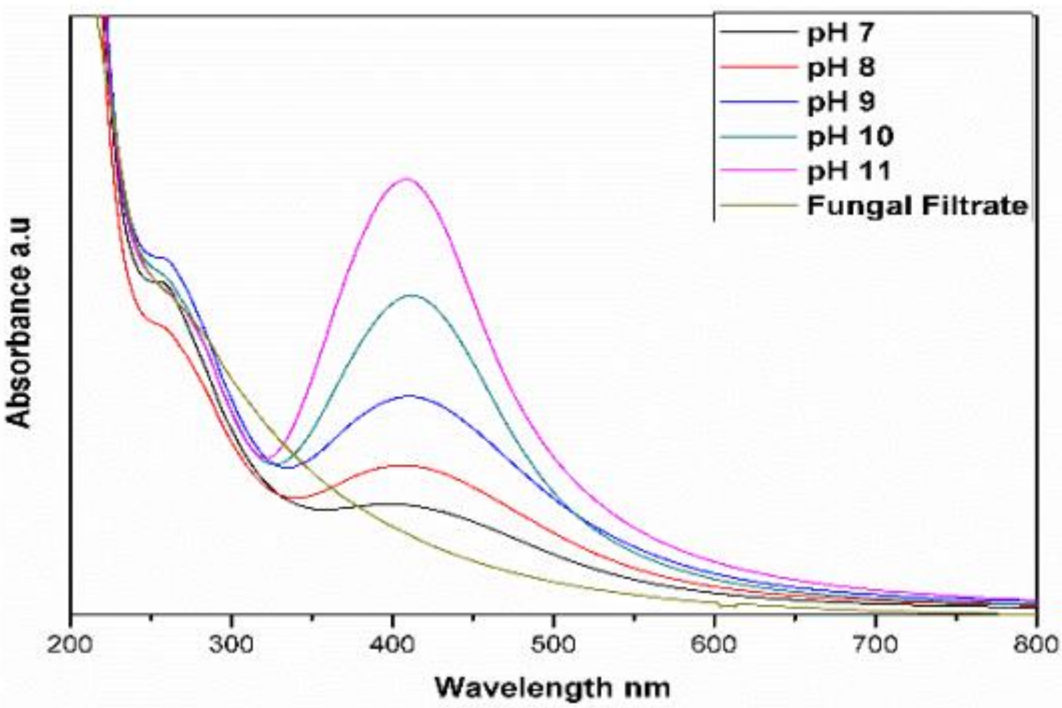

Fig. 3 At various pH levels, the fungal filtrate and the filtrate treated with AgNO3 had their UV-visible absorption spectra recorded.

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The fungus-filtered liquid, after applying AgNO3 solution, shows absorbance peaks in the range of 408 to 415 nm. This indicates the presence of silver tiny-particles. When free electrons on the surface of metal nanoparticles oscillated coherently, it created the SPR absorption band [31]. Figure 3 shows that the SPR curve sharpens and shifts towards the blue end of the spectrum as the pH value rises. A possible explanation for the shift in surface plasmon resonance to higher energy could be a reduction in nanoparticle size [8] significant quantity of functional groupings is accessible to attach silver ions in the alkaline form of the reaction media, leading to the production of a significant number of tiny sized AgNPs [32].

The absorption spectra clearly show that when the pH was raised from 7 to 11, Blue shifted its wavelength of maximum absorbance (λmax) from 415 to 408 nm.According to the metal nanoparticle quantum theory, there is a connection between the wavelength of maximum absorbance (λmax) and the conduction band's energy is quite fascinating [33].

The photon energy equation and the UV-visible absorption spectra of Einstein can be used to directly determine Ecb (eV), the conduction band energy, as follows:

$$Ecb = \frac{hc}{\lambda max}$$

With the given variables, we have h as the Planck constant, c as the speed of light, and λmax as the maximum absorption wavelength. Considering that the energy level of the band of conduction of silver nanoparticles produced at pH 11 is found to be 3.02 eV,

4.3 FTIR Analysis

Fig. 4 displays the Penicillium filtrate's FTIR spectrum. The bands that showed up at 657, 1070, 1249, 1357, 1643, 2077, and 3363 cm-1 show that many substances and groups are involved in the production of nanoparticles of silver.

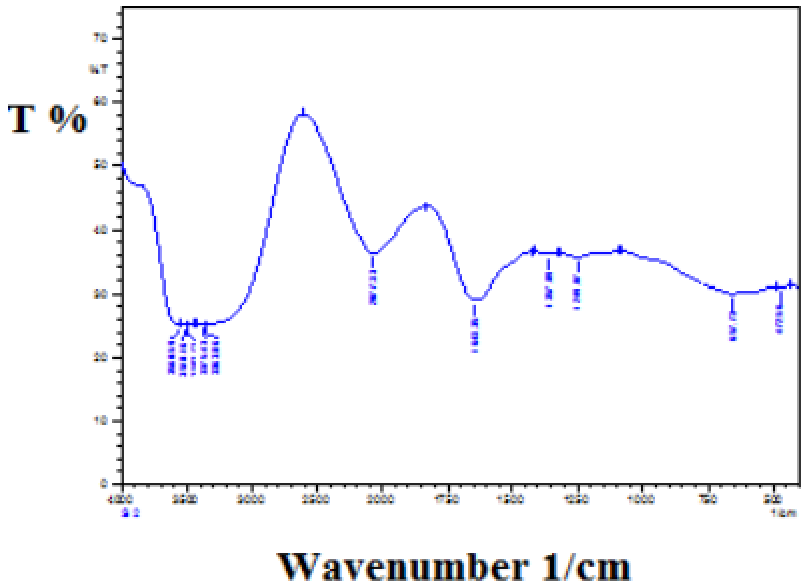

Fig. 4 FTIR spectra of species of Penicillium

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4.4 X-Ray Diffraction Analysis

Figure 5 displays the pattern of XRD for the produced nanoparticles of silver, with 37.9°, 43.5°, 63.8°, and 77.5° 2θ values marking the intensity maxima. The peaks correspond to the planes (111), (200), (220), and (311), in that order. According to XRD examination, the produced silver nanoparticles had a fcc structure and were crystalline in form. The standard JCPDS No. 43-0649 was matched with it. Additionally, The XRD graphs' high-intensity peak (1 1 1) can be broadened utilizing the FWHM peak and the Scherer equation, Table 1 demonstrates how the samples' crystallite size was calculated to be 16.69 nm.

$$D(nm) = \frac{k\lambda}{\beta\cos\theta}$$

where is the diffraction line's entire width, expressed in radians, at half its greatest intensity; is the size of the crystalline (nm); is the Bragg angle; that is Cu K = 0.154 nm's X-ray wavelength.For sphere particles, the factor of shape, k, is equal to 0.94.

Table 1 XRD analysis of synthesized Ag nanoparticles

2θ	θ	cosθ	FWHM in radians	Size nm
37.9	19	0.946	0.0087	16.69

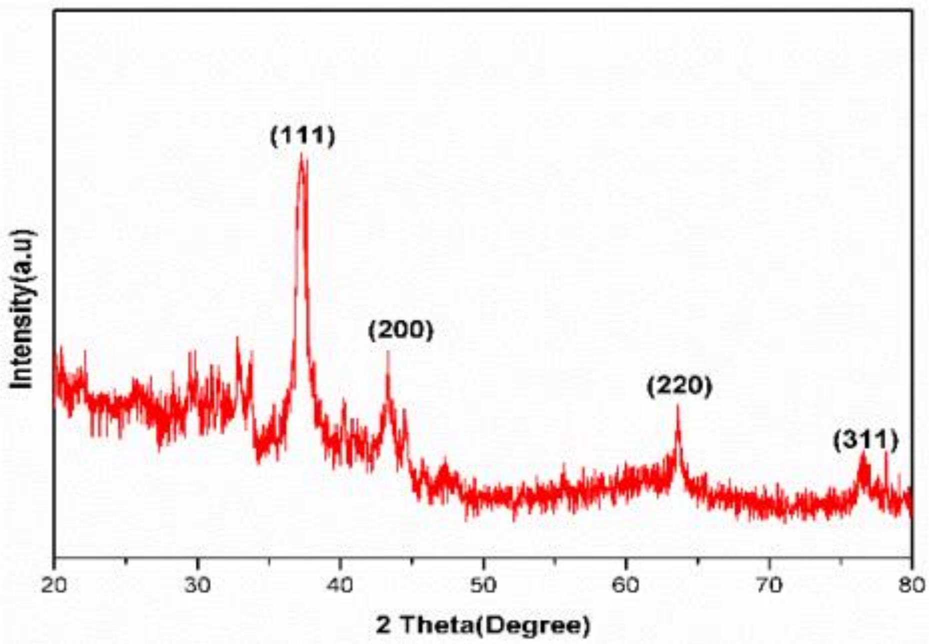

Fig. 5 Pattern of AgNPs in XRD

4.5 Scanning Electron Microscopy

Figure 6 displays a scanning electron micrograph of the produced nanoparticles of silver. It amply demonstrates the size range of the nanoparticles was 11–15 nm, were spherically shaped, and were evenly distributed.

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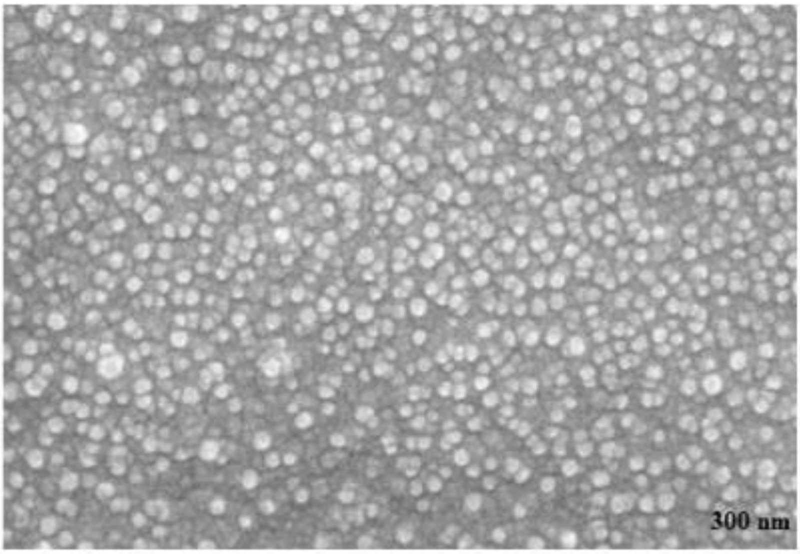

Fig. 6 The produced AgNPs are visible in the scanning electron micrograph

4.6 Transmission Electron Microscopy

The produced nanoparticles of silver TEMpictures are shown in Fig. 7(a), indicated their spherical shape. The tiny-particles were between 9 and 19 nm in size. The generated nanoparticles appear to be crystalline and have a face-centered cubic geometry based on the electron diffraction pattern of the specified area depicted in Figure 7(b).

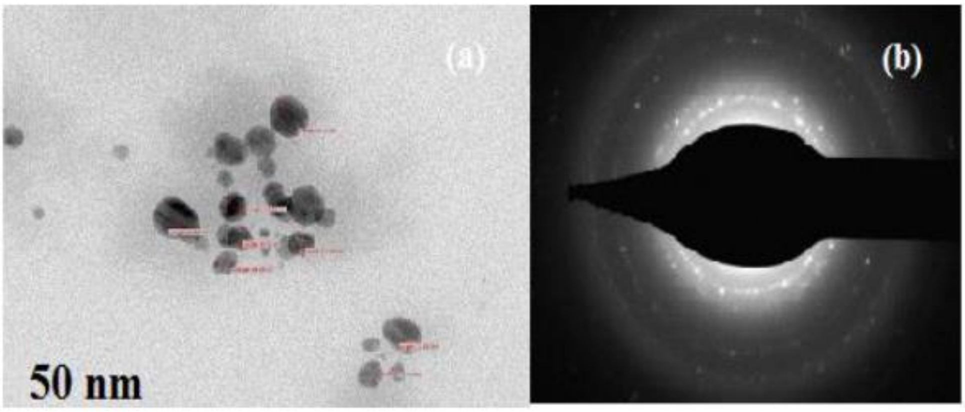

Fig. 7 (a) TEM image and (b) SAED pattern of AgNPs

4.7 Photoluminescence Spectra

Figure 8 shows the produced colloidal solution of silver nanoparticles' PL spectra. In the visible region, the emission peak was generated at 530 nm upon stimulation at 400 nm. The tiny-particles emit light because of the stimulation of electrons moving from one energy level to another, leading to the energy being released. The generation of luminosity is a consequence of the energy losses experienced by electrons. The silver tiny-particles' optical

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characteristics are determined by the interband and intraband transitions between electronic states 34.

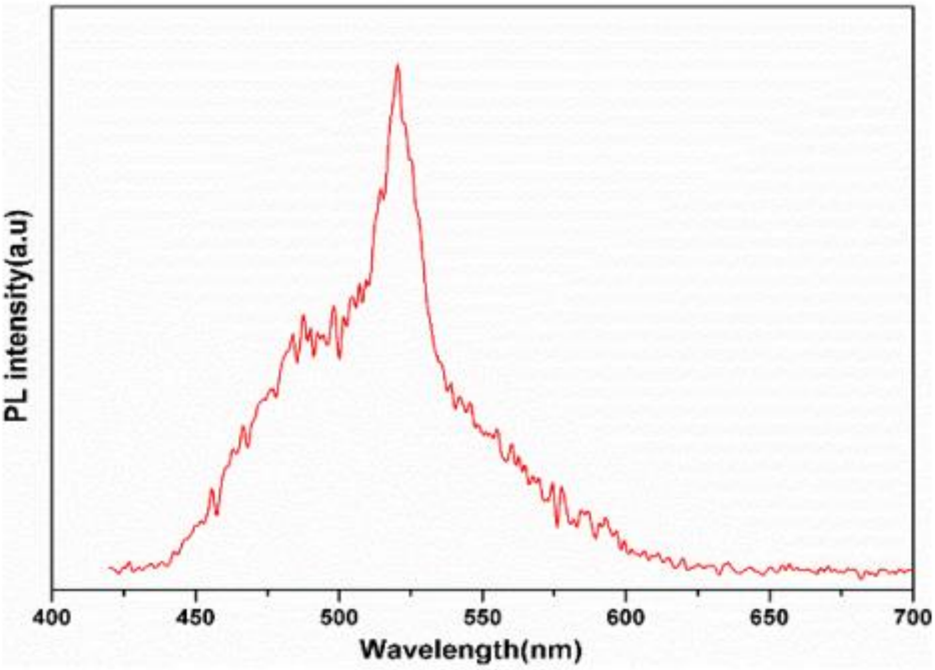

Fig. 8 PL emission spectra of AgNPs

4.8 Catalytic Activity

The reduction of MB by NaBH4 was investigated to assess the level of catalysis of produced nanoparticles of silver [35, 36]. Two tubes were used to conduct the reaction, each holding two millilitres of blue M.B. The two tubes were filled with 0.5 mL of NaBH4. A 0.3 ml colloidal solution was added to tube 2's solution. Both visually and with the aid of a UV-visible spectrometer, the reaction was observed. The reaction begins right away, and For several hours, the solution in tube 1 stays blue, the fluid in tube 2 becomes colourless after just two minutes. Figure 9(a).

MB's highest absorbance is measured at 663.5 nm, as shown in Fig. 9(b); however, the peak disappears in a matter of minutes (two) upon the addition of the AgNPs solution in tube 2. This suggests that silver nanoparticles catalysed the reduction of MB. There was one absorbance peak at 400 nm, which could have been caused by silver nanoparticles.

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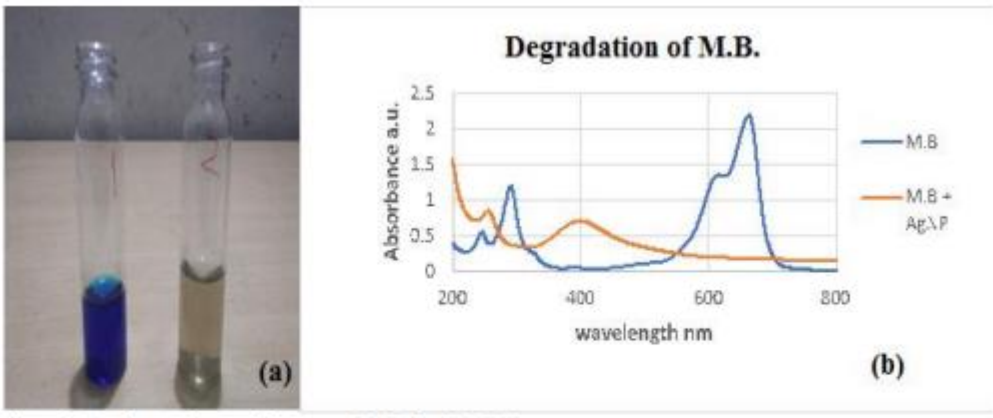

Fig. 9 Catalytic degradation of MB by NaBH4

5. Conclusion

The manufacture of nanoparticles of silver utilising Penicillium species as a green technique has been effectively proven, according to the provided experimental outcomes and conversations. The effects of pH on the synthesised nanoparticles' optical qualities, structural traits, and catalytic activity were examined in this investigation.

The creation of minuscule silver particles was verified by visual inspection and Spectroscopy in the UV and visible range; surface plasmon resonance absorption band was evident between 408 and 415 nm. It was discovered that the reaction medium's pH affected the SPR peak's intensity and location, with higher pH values producing a blue shift and sharper SPR curve.

FTIR research showed that there was various biomolecules in the fungal filtrate in charge of stabilizing and reducing ions of silver during nanoparticle synthesis. The silver nanoparticles' crystalline structure—a cubic construction centred on the face with a crystallite size of roughly 16.69 nm—was validated by X-ray diffraction research.

The spherical shape of the generated nanoparticles was shown by the pictures obtained from scanning electron microscopy (SEM) and transmission electron microscopy (TEM), evenly dispersed, and varied in size between 9 and 19 nm.

Photoluminescence spectroscopy indicated luminescence characteristics of the nanoparticles of silver, with peaks in emission detected at 530 nm upon 400 nm excitation.

The methylene blue decrease (MB) by NaBH4 served as evidence of the synthesised The nanoparticles of silver' catalytic activity. The quick transition of blue to colourless when silver nanoparticles are present indicated their catalytic efficiency in the reduction reaction.

In conclusion, the study highlights the potential of Penicillium species-mediated green synthesis for the efficient production of silver nanoparticles containing tunable optical properties and catalytic activity. These nanoparticles hold promise for several uses in industries like catalysis, sensing, and biomedical engineering. Further research could focus on exploring the specific mechanisms underlying the pH-dependent synthesis and optimizing the nanoparticle properties for targeted applications.

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