



## Bio-mediated Synthesis of Silver Nanoparticles Utilizing Aqueous Leaf Extract of *Lantana camera* and Their Application as Nanophotocatalysts and Antimicrobial agents

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### ABSTRACT

This present study aimed toward the bio-mediated synthesis of silver nanoparticles (AgNPs) by utilizing aqueous leaf extract of *Lantana camera* with silver nitrate ( $\text{AgNO}_3$ ) as the precursor. The color transformation from light yellow to dark brown of the reaction media initially verified the synthesis of silver nanoparticles which was further confirmed by observing the characteristics Surface Plasmon Resonance (SPR) band peak of silver nanoparticles at 458 nm. The presence of any functional groups in silver nanoparticles was determined using Fourier Transformed Infrared (FT-IR) spectroscopy with the stretch of bonds of functional groups. Surface morphology and dispersity of nanoparticles were characterized with a Field-Emission Scanning Electron Microscope (FE-SEM) and the spherical polydispersed nanoparticles were observed with the particle sizes between 15 to 35 nm. The crystallinity and purity of AgNPs were revealed by X-ray Diffraction (XRD) analysis and Energy Dispersive Spectroscopy (EDS), respectively. Photocatalytic activity of AgNPs was observed against Methylene blue (MB) dye under solar light irradiation. The AgNPs efficiently degraded the dye by nearly 95% in 120 min of exposure time. The degradation of dye with time was fitted to pseudo-first-order and pseudo-second-order kinetic models where the data best fit the pseudo-first-order kinetic model. Synthesized AgNPs showed moderate anti-microbial activity against four *Gram-positive* and *Gram-negative* bacterial strains while they showed perfect activity against two anti-fungal strains.

**Keywords:** Green synthesis, Silver nanoparticles, Methylene blue, Dye degradation.

### INTRODUCTION

Nanotechnology pledges scientific progress in the field of modern research of fabrication, manipulating, and individualizing nanoparticles<sup>1</sup>. In the fields of biomedical, electronics, food and health care, drug-gene delivery, the importance

of nanotechnology is increasing day by day<sup>1,2</sup>. A nanoparticle or discerning particle is usually described as a particle with a diameter of between 1 and 100 nm but is sometimes used for large particles up to 500 nm or for fiber and tubes less than 100 nm in only two dimensions<sup>3</sup>. These particles have special properties in terms of their very tiny size



and a very large and high surface-to-volume ratio which are completely different from their bulk nature of counterparts thus their properties are varied with their morphology. Numerous metals are found in nature but all of them are silver, gold, platinum, and palladium synthesized in nanostructure formed. Staggering physicochemical, electrical, optical, and magnetic peculiarities are shown by noble metal nanoparticles. Due to their extraordinary properties, they are extensively used in various fields. Among noble metal nanoparticles, silver is synthesized widely because of its identical features<sup>4</sup>. The salient characteristics have made them proper to use in pharmaceuticals, agriculture, water detoxification and filtration, air filtration, and dye degradation in the textile industry, catalysis, and chemical industries<sup>5</sup>. Nanoparticles have some emerging properties such as large surface area, faster rates to equilibrium, less resistance to diffusion, and high adsorption. Because of these properties nanoparticles have gained great importance in the last few decades. Researchers explored the application of silver nanoparticles as a dye-removal agent. Having some supercilious physical, chemical, and biological properties silver nanoparticles are very versatile for implementation in various industries. Silver nanoparticles also possess antibacterial and anti-fungal besides these mentioned characteristics and facilitate their application in wastewater treatment<sup>6,7</sup>. There are some conventional synthesis processes like physical and chemical methods but these methods have some major drawbacks. Using the physical method to synthesize nanoparticles, a large number of particles can be produced e.g., controlled yield, and product can be gained in powder form with fine particles. However, it has some disadvantages such as being very time-consuming and the large space needed<sup>8</sup>.

On the other hand, the chemical synthesis method involves toxic and hazardous materials, which is not an environment-friendly method. And also prohibited for medical uses as an application. Because nanoparticles synthesized via this route bind chemicals on their surface<sup>2</sup>. An eco-friendly synthesis method is necessary to overcome these conventional methods<sup>9,10</sup>. Without any of the toxic and hazardous chemicals or any kind of exterior agents environmentally friendly processes are being developed by the researcher recently as a safe way of approach. As a way of green approach various

microorganism-mediated syntheses are done by bacteria, fungi, and plants because of their reducing and capping ability to transform metal compounds into reduced nanoparticles<sup>11</sup>. Also, cellulosic materials, algae, and yeast are used to synthesize various nanoparticles as a way to approach green synthesis<sup>12</sup>. Plants parts such as leaves, roots, stems, fruit, flower, and rhizome are used for the synthesis of nanoparticles<sup>13,14</sup>. Above all, the extract of various parts of the plant is used for the synthesis procedure, because plant extract contains various phytochemicals that act as a capping agent<sup>15</sup>. Plant extract consists of various metabolites e.g. alkaloids, terpenoids, amino acids, alcoholic compounds, flavones, flavonoids, proteins, polysaccharides, and phenolic compounds are very much responsible for reducing Ag ions into nanoparticles. However, the mechanism is very unknown of the synthesis of nanoparticles with the parts of plant<sup>16</sup>. Some of the phytochemicals in plants act as capping agents and stabilizers. *Lantana camara* (common lantana) is a species of flowering plant that belongs to the family Verbenaceae and is a genus of about 150 species of herbs<sup>17</sup>. *Lantana* is a native species to tropical and subtropical America by origin. And introduced as a decorative and verge plant to other countries in the world. It spread into various areas such as crop fields, railway tracks, to distributed areas as roadsides and hillsides<sup>18</sup>. Thus, the *Lantana camera* contains essential phytochemicals to synthesize nanoparticles using its leaf extract.

In this study, we attempted to establish a robust and reliable green synthesis method for AgNPs by *Lantana camera* aqueous leaf extract, which is an eco-friendly, cost effective, and free from any kind of toxic materials. Synthesized silver nanoparticles were subjected to investigate photocatalytic activity against dye and also microbial activities were evaluated against bacterial strains and fungal strains.

## EXPERIMENTAL

### Materials

Silver nitrate ( $\text{AgNO}_3$ ) (Merck KGaA, Germany), Deionized water (18 mili-Q), Methylene blue (PT. SMART LAB, INDONESIA), Dimethyl sulfoxide (DMSO) (Germany), Mueller Hinton Agar (MHA) media (HIMDIA, India) and Potato Dextrose Agar (PDA). Leaves of *Lantana camera*.

### Synthesis of Silver Nanoparticles

Fresh leaves of *Lantana camera* collected from various places of Chattogram hill tracks such as rode side, crops filed side etc. Fresh leaves were splashed many times with deionized water to remove any kind of dirt and contamination. Leaves were dried under shaded conditions avoiding sun drying because sun light may affect the metabolites present in the leaves. Then dried leaves were ground to make a fine powder. To make 10% (w/v) aqueous leaf extract, 1 g of leaf powder was put into 10 mL of di-ionized water and stirred for 60 minutes at 50°C. Then the prepared extract was cooled down to room temperature and filtered with Whatman No-1 filter paper and kept at 4°C for further procedure.

A 5 mM aqueous solution of AgNO<sub>3</sub> (precursor) was prepared in dark conditions with di-ionized water. Silver nitrate solution and aqueous leaf extract were slowly mixed considering a ratio of 9:1 while stirring at 60 and 70°C in dark conditions for 2 h until the color of the reaction media showed a significant change. And then it was kept for 24 h at room temperature to acquire complete reduction and nucleation of silver nanoparticles. During the reaction, Ag<sup>+</sup> was reduced to Ag<sup>0</sup> by metabolites present in the aqueous leaf extract which resulted in the formation of silver nanoparticles (AgNPs).

### Characterization

Formation of AgNPs preliminary ensured by observing the physical appearance as color change of reaction media from pale yellow to dark brown. Then, Surface Plasmon Resonance (SPR) band peak from an UV-Visible spectrophotometer (T80+ by PG instrument) was taken for confirmation. FT-IR spectroscopy was employed to investigate the presence of any functional groups. Surface morphology and shape of the AgNPs were evaluated by FE-SEM (JSM-7600F) analysis. Crystalline nature and size were evaluated by XRD (PANalytical) analysis and EDX (JSM-7600F) analysis employed to measure the composition and purity of AgNPs.

### Photocatalytic Activity

Synthesized AgNPs were subjected to investigate the photocatalytic activity against

organic methylene blue dye. This study was investigated under solar light irradiation. The photocatalytic activity was evaluated against various concentrations of methylene blue dye solutions with different amounts of AgNPs catalysts. At the preliminary stage of this photocatalytic reaction, the mixture of methylene blue dye and silver nanoparticles was stirred in the dark until it reached equilibrium and then continued the reaction under sunlight. The absorbance of methylene blue dye was measured during the reaction at different time intervals at 664 nm using UV-Visible spectrophotometer by taking de-ionized as control. The degradation percentage of dye was calculated with the following Equation (1)

$$\text{Degradation (\%)} = \frac{C_0 - C_t}{C_0} \quad (1)$$

Where C<sub>0</sub> = the initial concentration and  
C<sub>t</sub> = the concentration after time t.

### Microbial Activity Assay

Synthesized AgNPs using leaf extract of *Lantana camera* were subjected to four bacterial strains (two *Gram-positive* and two *Gram-negative*) and two fungal strains to evaluate the microbial activity. Microbial activity assay was examined through Agar disc diffusion method<sup>19</sup> where Mueller Hinton Agar (MHA) media (HIMIDA, India) and Potato Dextrose Agar (PDA) were the basal media for bacterial and fungal strains, respectively. Ceftriaxone and Amphotericin B. were used as standard and Dimethyl sulfoxide (DMSO) was used as control for this study.

## RESULTS AND DISCUSSIONS

### Silver Nanoparticles synthesis and Characterizations

In the initial step of synthesizing silver nanoparticles (AgNPs), the reaction media turned from a pale yellow to a deep brown, signifying the full reduction of Ag<sup>+</sup> (ions) to Ag<sup>0</sup> (atoms) and the formation of AgNPs shown in Fig 1. Absorbance measurements taken at various points in time throughout the reaction allowed us to validate the bio-synthesis of AgNPs by revealing a peak at 458 nm in the Surface Plasmon Resonance (SPR) band (Figure 2).



Fig. 1. Physical appearance (color change) of reaction media during synthesis of nanoparticle using *Lantana camera* leaf extract

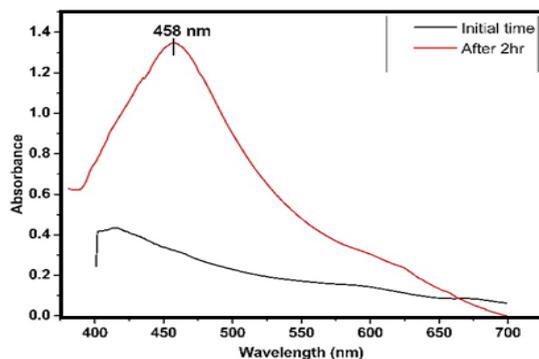


Fig. 2. UV-Visible spectrum of synthesized AgNPs using *Lantana camera* (created using Origin Pro 8.5)

FTIR spectroscopy was employed to elucidate the responsible functional groups for the reduction of Ag from  $\text{Ag}^+$  those present in the leaf of plant *Lantana camera*. In (Fig. 3) this analysis, it shows four absorption peaks at wavenumbers  $1615\text{ cm}^{-1}$ ,  $1510\text{ cm}^{-1}$ ,  $1209\text{ cm}^{-1}$ ,  $958\text{ cm}^{-1}$  corresponding to C=O stretching vibration of carbonyl groups,  $-\text{NO}_2$  aliphatic nitro group, C-O stretching of the carboxylic group and unsaturated C=C bond, respectively which indicate the presence of these functional bands, a shift in absorption bands of the phytochemical compounds from aqueous extract of *Lantana camera* as a stabilizing agent.

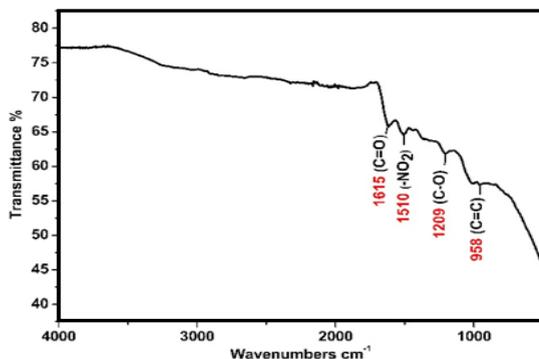


Fig. 3. FTIR spectrum of synthesized AgNPs using *Lantana camera* (created using Origin Pro 8.5)

At different magnifications, SEM analysis

was conducted to evaluate the surface nature and morphology of plant-mediated produced AgNPs. It displays some nanoparticle morphological characteristics, such as size, shape, and environment and is shown in the (Fig. 4). The spherical nanoparticles in the size range between 15 to 35 nm were observed.

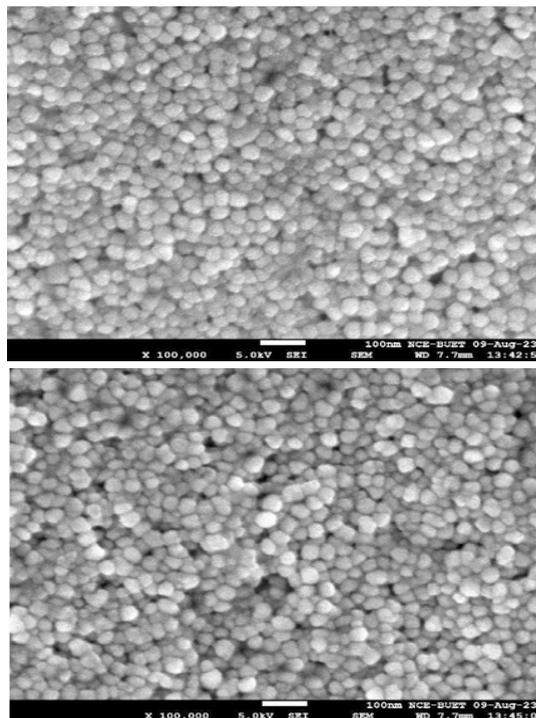


Fig. 4. FE-SEM image of synthesized AgNPs using *Lantana camera*

Using another spectroscopic analysis, XRD investigation peak pattern of the synthesized AgNPs by the leaf extract of *Lantana camera* is shown in (Fig. 5). The angle of diffraction data was taken for a range  $30 \leq 2\theta \leq 80$  degrees with a step of 0.01313 degrees. There are four peaks in the diffractogram at  $38.15991^\circ$ ,  $44.10789^\circ$ ,  $64.54581^\circ$ , and  $77.47817^\circ$ , these are the main characteristic peaks for AgNPs. The four diverse diffraction peaks at  $38.16^\circ$ ,  $44.10^\circ$ ,  $64.56^\circ$  and  $77.48^\circ$  of the  $2\theta$  values can be allotted to the planes of 111, 200, 220 and 300 respectively<sup>20</sup>. By using Debye-Scherrer formula crystalline size and average crystallite size have been estimated from the diffractogram synthesized of AgNPs. Each one of the four peaks was fitted with a Gaussian function to find out the FWHM for the calculation of crystallite size. The FWHM of the fitted Gaussian non-linear curve is taken as FWHM of the peaks in the diffractogram. These values are estimated through the software

Origin Pro 8.5. From the data of the four peaks "D" has been calculated. The mean crystalline size of the synthesized silver nanoparticles is found to be approximately 10 nm. Crystallites size and average crystallites size investigated and calculated from XRD data using Scherrer Equation (2) and presented in Table 1.

$$D = k\lambda/\beta \cos\theta \quad (2)$$

Here, crystal size,  $D$  = (nm)  
 (Scherrer-constant),  $K = 0.9$   
 (FWHM),  $\beta$  = Full Width at Half Maximum in radians  
 (wave length of X-ray sources),  $\lambda = 0.15406$  nm  
 Peak position,  $\theta$  = (radians)

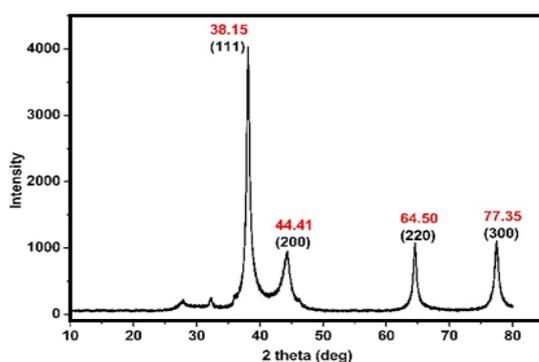


Fig. 5. X-ray Diffraction peak pattern of synthesized silver nanoparticles (created using Origin Pro 8.5)

Table 1: Table of Crystalline size calculation from XRD data

Peak No	2 $\theta$ on x-axis	FWHM (deg)	Crystalline size 'D' (nm)	Average crystalline size (nm)
1	38.15991	0.86833	9.68	9.95
2	44.10789	2.13106	4.02	
3	64.54581	0.67211	13.98	
4	77.47817	0.83819	12.15	

Elemental analysis of silver nanoparticle was performed using EDX spectroscopy which demonstrated in the (Fig. 6). Synthesized AgNPs was confirmed by the intense high signal seen in the characteristics silver (Ag) region of the EDX spectra<sup>21</sup>. Metallic synthesized silver nanocrystals exhibit an optical absorption peak at about 3 keV due to the existence of the SPR band. The purity of silver nanoparticles can be described by this analysis hence synthesized silver nanoparticles exhibit high purity. The EDX study provides details on how the aqueous leaf extract of *Lantana camara* forms pure AgNPs.

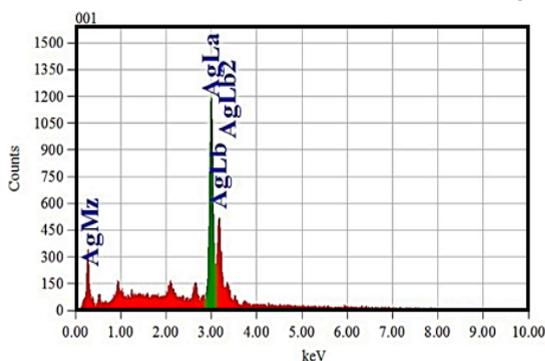
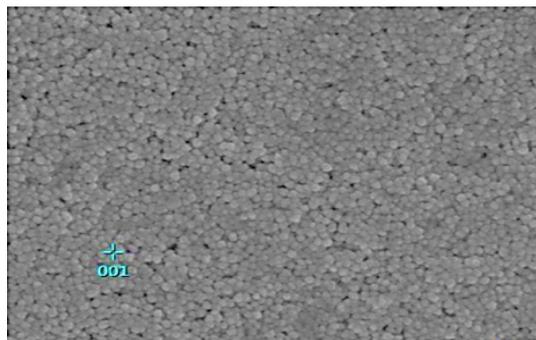


Fig. 6. EDX analysis of synthesized AgNPs using *Lantana camara*

### Photocatalytic Activity of AgNPs

Photocatalytic activity of synthesized AgNPs were inspected against cationic methylene blue dye under sunlight. The temperature and pH were in optimum condition at 25°C and 7 respectively during the procedure. This photocatalytic activity was examined against concentrations of methylene blue dye of 5 mgL<sup>-1</sup>. The amount of synthesized AgNPs catalyst used for the degradation of methylene blue in each investigation was 125 mg, 250 mg, and 500 mg per liter of methylene blue solution. At first AgNPs added to methylene blue aqueous solution and continued the reaction in dark until it reaches equilibrium then the reaction media exposed to sunlight hence the degradation of methylene blue continued. During the experiment absorbance of methylene blue dye was observed at 665 nm at different time intervals e.g. 0 min, 30 min, 60 min, 90 min, and 120 minute. For the 125 mg, 250 mg, and 500 mg of catalyst against 5 mgL<sup>-1</sup> of methylene blue solution, the percentage of degradation was 95.05%, 95.77%, and 95.55% respectively (Fig. 7). The degradation of the dye is measured by observing the absorbance at 665 nm, which is gradually decreased as the

reaction continues until it reached equilibrium after 120 minutes. The same procedure was employed also for 10 mgL<sup>-1</sup> of methylene blue dye solution. For 10 mgL<sup>-1</sup> solution the degradation efficiency

is around 50% with catalyst amounts 125 mg, 250 mg, and 500 mg. Hence the photocatalytic activity of AgNPs exhibited better results against the 5 mgL<sup>-1</sup> solution of methylene blue dye.

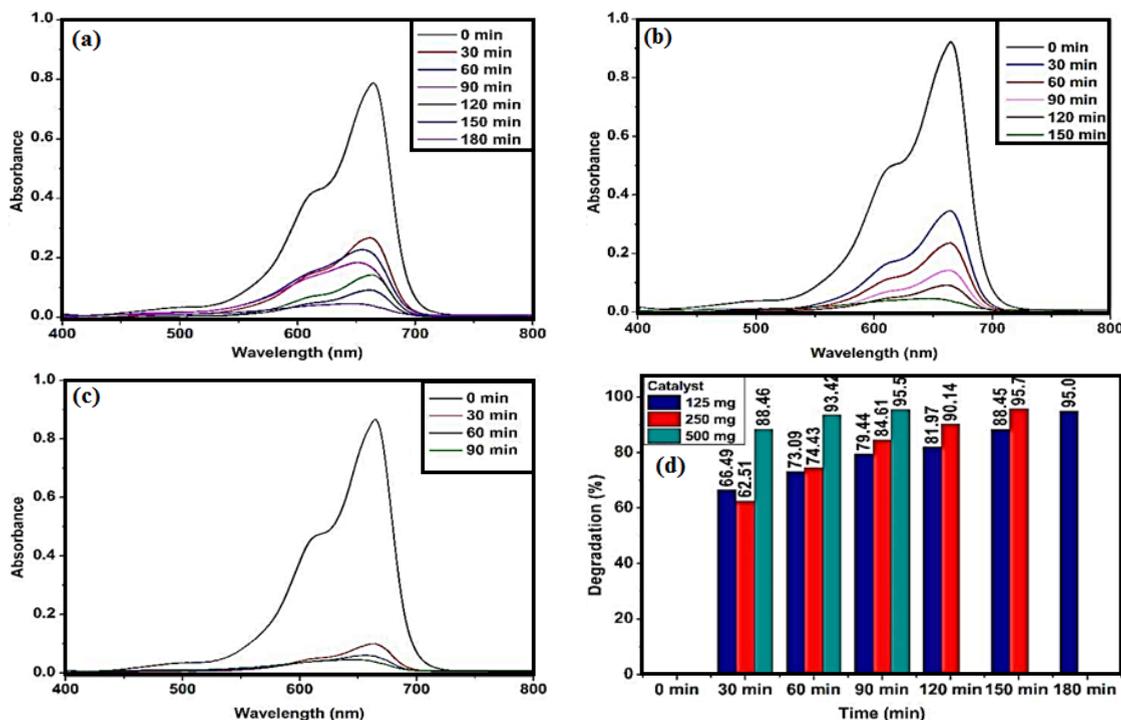


Fig. 7. Absorption spectra (UV-Visible) of dye degradation (a) 125 mg, (b) 250 mg and (c) 500 mg of synthesized AgNPs for 5 mgL<sup>-1</sup> concentration, (d) degradation of MB dye with different amount of silver nanoparticles catalyst (created using Origin Pro 8.5)

### Reaction kinetics study

Degradation rate of methylene blue by synthesized AgNPs was investigated through kinetics study (Fig. 8)<sup>22</sup>. To evaluate the kinetics of degradation of methylene blue dye by synthesized AgNPs, the following experiments were carried out for a concentration of 10 mgL<sup>-1</sup> of methylene blue at room temperature 25°C, the amount of catalyst being applied for the procedure was 250 mg per liter of methylene blue solution. The experiment was carried out for 0 min to 120 min when the reaction reached the equilibrium. The kinetics of degradation of methylene blue was investigated with a pseudo-first-order (Equations 3 and 4, Fig. 8b) and pseudo-second-order model (Equations 5 and 6, Figure 8c)<sup>23,24</sup>.

Pseudo-first-order Eq.

$$q_t = q_e (1 - e^{-kt}) \quad (3)$$

linear form of pseudo first-order Eq.

$$\ln(q_e - q_t) = \ln q_e^{-kt} \quad (4)$$

Pseudo-second-order Eq.

$$q_t = kq_e^2 t / (1 + kq_e t) \quad (5)$$

linear form of pseudo second-order Eq.

$$t/q_t = 1/kq_e^2 + t/q_e \quad (6)$$

Where  $q_e$  = concentration of methylene blue at equilibrium and  $q_t$  = concentration of methylene blue at time (t), respectively, also the co-efficient of each model is listed in the Table 2

The linearity in the pseudo first-order plots fitted better than that of pseudo second-order plots shown in the Fig. 8, as confirmed by the higher R<sup>2</sup> values. This pseudo first-order kinetic model exhibits the chemical reaction nature of the experiment and its rate of removal of methylene blue dye.



## CONCLUSION

The current work reports the successful biogenic synthesis of silver nanoparticles utilizing an aqueous leaf extract of the wild plant *Lantana camera*. A sustainable, economical, and non-hazardous chemical-free green synthesis technique has been devised for the synthesis of silver nanoparticles. The photocatalytic and microbiological capabilities of these nanoparticles have been extensively assessed. Synthesized silver nanoparticles were seen to be of reduced dimensions. The inclusion of metabolites presents in the aqueous leaf extract of the *Lantana camera* served as capping and stabilizing agents during the production of silver nanoparticles, therefore enhancing their versatility in several applications. Under sunlight, the photocatalytic activity of silver nanoparticles was studied, and it was shown that the produced silver

nanoparticles were capable of degrading methylene blue dye to a great degree. The reaction kinetics including the degradation of methylene blue were also investigated. Reaction kinetics of degradation of methylene blue were also investigated which showed best fitting with pseudo first order kinetic model with best correlation value  $R^2$  0.99237.

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## Conflict of Interest

The authors declare that they do not possess any known conflicting financial interest.

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