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CATALYTIC ACTIVITY OF GREEN-SYNTHESIZED AgNPS IN THE REMOVAL OF DYE POLLUTANTS FROM WASTEWATER

Ana Kesić[1]*, Nikola Srećković [2], Vladimir Mihailović[2], Katarina Marković[1], Mirjana Grujović[1]

Abstract: Dye-containing wastewater represents a serious environmental problem due to the toxicity and persistence of synthetic dyes. In this study, the catalytic activity of silver nanoparticles (AgNPs) obtained by green synthesis using *Agrimonia eupatoria* L. plant extract as a reducing agent was investigated. The catalytic performance of AgNPs was evaluated through the reduction of Congo red in the presence of $NaBH_4$, monitored by UV–Vis spectrophotometry. The results demonstrated efficient catalytic degradation of the dye, with a calculated pseudo-first-order rate constant of $k' = 0.0299\ min^{-1}$. The biogenic AgNPs exhibited high catalytic efficiency, indicating their strong potential for application in wastewater treatment.

Keywords: Silver Nanoparticles, Green Synthesis, *Agrimonia eupatoria L*, Catalytic potential, Congo red.

Introduction

Dye-containing wastewater has become an increasingly pressing environmental issue due to its harmful effects on living organisms and the abiotic environment. Many synthetic dyes are highly carcinogenic, chemically stable, and resistant to biodegradation, allowing them to persist in aquatic systems for extended periods (Kayani et al., 2025). Azo dyes, such as Congo red, are particularly concerning due to their widespread industrial use and their potential toxic, mutagenic, and carcinogenic effects.

Conventional wastewater treatment methods, including adsorption, coagulation–flocculation, and biological processes, often show limited

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efficiency, incur high operational costs, and carry the risk of secondary pollution. As a result, catalytic degradation of organic pollutants using nanomaterials has emerged as a promising and effective alternative for dye removal (Chaturvedi et al., 2012). Numerous studies have demonstrated that nanoparticles obtained via green synthesis possess significant potential to catalyze the degradation of various organic pollutants, including synthetic dyes (Mihailović et al., 2023).

Silver nanoparticles (AgNPs) have attracted considerable attention due to their large specific surface area, excellent catalytic properties, and efficient electron-transfer capability. Compared to traditional physical and chemical synthesis methods, green synthesis provides a sustainable and environmentally friendly approach, using plant extracts as reducing and stabilizing agents while avoiding toxic chemicals. Plant-derived biomolecules, particularly polyphenols and flavonoids, play a crucial role in forming stable and catalytically active AgNPs (Srećković et al., 2021).

In the present study, silver nanoparticles were synthesized using *Agrimonia eupatoria* L. extract as a reducing agent, representing a sustainable and eco-friendly approach. The catalytic activity of the biogenic AgNPs was evaluated through the reduction of Congo red in the presence of sodium borohydride. his study further highlights the potential application of AgNPs for effective and sustainable dye removal from wastewater.

Materials and methods

Aqueous plant extracts were prepared using the previously described maceration method (Muruzović et al., 2023). The dried and ground plant material was extracted in distilled water at room temperature, with solvent replacement every 24 h. The obtained filtrates were concentrated under reduced pressure at 40 °C, and the dry extracts were stored at 4 °C until further use. Silver nanoparticles (AgNPs) were synthesized using $AgNO_3$ as a precursor, while the aqueous plant extracts acted as reducing and stabilizing agents. The synthesis conditions were optimized by varying $AgNO_3$ concentration, pH, temperature, and reaction time, in accordance with previously reported studies (Marković et al., 2022; Kesić et al., 2023). The formation of AgNPs was monitored by the visual color change of the solution and UV–Vis spectrophotometry. After synthesis, the nanoparticles were isolated by centrifugation, washed with demineralized water, dried at 40 °C, and stored at

4 °C, and UV–Vis spectra were recorded in the wavelength range of 200–800 nm.

The assessment of the catalytic activity of AgNPs was determined by the reduction of aqueous Congo red (CR) solutions in the presence of $NaBH_4$. In this reaction, 1 mL of AgNPs aqueous solution (0.1 mg mL^{-1}) was mixed with 5 mL of 10 μM CR solution and 1.5 mL of 1 mM $NaBH_4$ solution. The reaction took place in the dark. The speed of the CR catalytic degradation process in different time intervals was monitored with a UV-Vis spectrophotometer at 25°C in the wavelength range of 300 to 700 nm (λ_{max}= 500 nm).

The reaction rate constant (k) for the catalytic degradation of CR was calculated using the pseudo-first-order kinetic equation:

$$ln\frac{A_0}{A_t} = k \cdot t$$

where $\boldsymbol{A_0}$ is the absorbance at 0 min, $\boldsymbol{A_t}$ is the absorbance at different time points, $\boldsymbol{k}$ is the reaction rate constant, and $\boldsymbol{t}$ is the reaction time. This kinetic equation assumes that the concentration of $NaBH_4$ remains in large excess and that the reaction follows pseudo-first-order kinetics. The obtained rate constant allows comparison of the catalytic efficiency with previously reported results of AgNPs synthesized via green methods.

Results and discussion

The ability of AgNPs to behave as catalysts in the degradation of dye is reflected in their ability to transfer an electron from donor molecules ($NaBH_4$) to acceptor molecules (azo bond in CR). The absorbance of CR decreased steadily over time at 338 and 500 nm, and finally, the solution became colorless, and the absorption maxima in the UV-Vis spectrum disappeared.

The catalytic degradation of Congo Red (CR) was investigated in the presence of $NaBH_4$, both in the absence and presence of green-synthesized AgNPs, and the reaction progress was monitored by UV–Vis spectrophotometry (Figures 1–2).

As shown in **Figure 1**, the UV–Vis spectra of CR recorded in the presence of $NaBH_4$ without AgNPs exhibit no significant changes in absorbance intensity over time. The characteristic absorption band of CR at approximately 500 nm remains almost unchanged, indicating that $NaBH_4$ alone does not have a significant effect on CR degradation under the applied experimental conditions. This observation confirms that direct electron transfer from $NaBH_4$ to CR molecules is kinetically unfavorable in the absence of a catalyst.

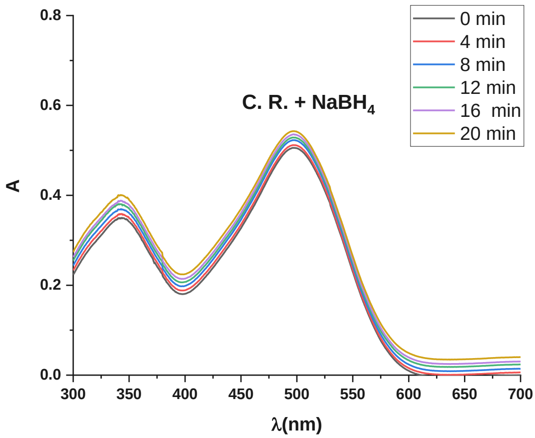

Figure 1. UV–Vis spectra of the catalytic degradation of Congo Red in the presence of NaBH4 without AgNPs.

In contrast, a pronounced catalytic effect is observed upon the addition of AgNPs (**Figure 2**). A gradual decrease in absorbance intensity at ~500 nm with increasing reaction time clearly demonstrates the efficient degradation of CR in the presence of AgNPs and $NaBH_4$. The disappearance of the characteristic absorption band is associated with the cleavage of the azo bond responsible for the dye′s color. In this system, AgNPs act as electron relay centers, facilitating electron transfer from BH_4^- ions to CR molecules adsorbed on the nanoparticle surface, thereby significantly accelerating the reduction process.

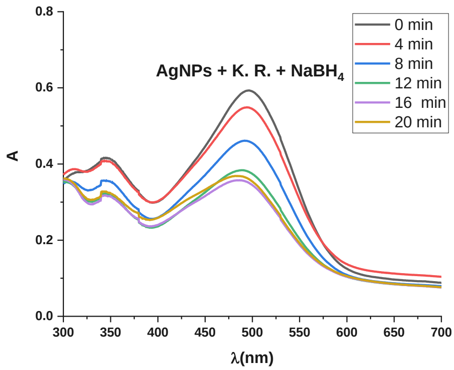

Figure 2. Catalytic degradation of Congo Red in the presence of AgNPs (**C**)

To quantitatively evaluate the reaction kinetics, the degradation data were analyzed using a pseudo-first-order kinetic model, assuming an excess concentration of $NaBH_4$. The linear relationship between $\ln(A_0/A_t)$ and reaction time (**Figure 3**) confirms that the catalytic degradation of CR follows pseudo-first-order kinetics. The apparent rate constant was calculated to be **k = 0.0299 min⁻¹**, indicating a relatively fast degradation rate and effective catalytic performance of the synthesized nanoparticles.

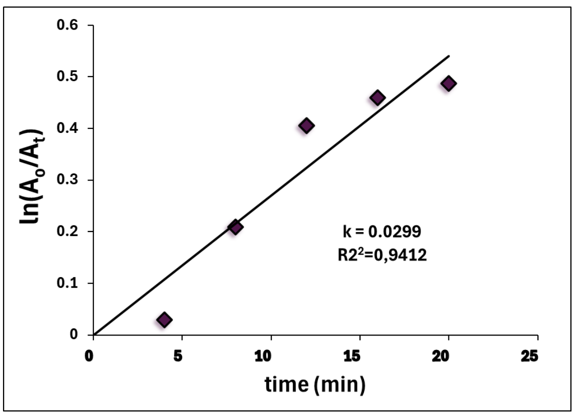

Figure 3. Pseudo-first-order kinetic plot ($\ln(A_0/A_t)$ versus time) for the catalytic degradation of Congo Red in the presence of AgNPs and $NaBH_4$.

Overall, the obtained results demonstrate that silver nanoparticles synthesized via a green approach using *Agrimonia eupatoria* L. plant extract significantly enhance the catalytic degradation of Congo Red in aqueous media, highlighting their strong potential for environmentally friendly wastewater treatment applications.

The obtained rate constant (k = 0.0299 min^{-1}) demonstrates similar catalytic efficiency compared to previously reported results for AgNPs synthesized via green methods (Mihailović et al., 2023). These results confirm the reliability of green synthesis for producing effective catalysts for the degradation of azo dye.

Conclusion

In general, the biogenic AgNPs exhibited excellent catalytic activity, achieving high degradation efficiency of Congo Red in the presence of $NaBH_4$. The UV–Vis spectrophotometric analysis confirmed that $NaBH_4$ alone had no significant effect on dye degradation, while the introduction of AgNPs led to a pronounced and rapid decrease in the characteristic absorption band of Congo Red. The degradation process followed pseudo-first-order kinetics, with an apparent rate constant of k = 0.0299 min^{-1}, indicating efficient catalytic performance. Therefore, the green-synthesized silver nanoparticles can be successfully applied as effective and environmentally friendly catalysts for wastewater treatment applications. The obtained results are consistent with previously published studies demonstrating the high catalytic potential of green-synthesized AgNP.

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4th INTERNATIONAL SYMPOSIUM ON BIOTECHNOLOGY
(31st SYMPOSIUM ON BIOTECHNOLOGY
WITH INTERNATIONAL PARTICIPATION)

University of Kragujevac, Faculty of Agronomy in Čačak

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CERTIFICATE OF ATTENDANCE

Ana Kesić, Nikola Srećković, Vladimir Mihajlović, Katarina Marković, Mirjana Grujović

CATALYTIC ACTIVITY OF GREEN-SYNTHESIZED AGNPS IN THE REMOVAL OF DYE POLLUTANTS FROM WASTEWATER

Chair of Organizing Committee
Prof. dr Pavle Mašković, Ph.D.
Pavle Mašković

УНИВЕРЗИТЕТ У КРАГУЈЕВЦУ АГРОНОМСКИ ФАКУЛТЕТ ОПШТА СЛУЖБА ЧАЧАК

Chair of Scientific Committee
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Vladimir Kurćubić

CATALYTIC ACTIVITY OF GREEN-SYNTHESIZED AgNPS IN THE REMOVAL OF DYE POLLUTANTS FROM WASTEWATER

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Introduction:
Dye-containing wastewater represents a serious environmental problem due to its harmful effects on living organisms and the abiotic environment. Azo dyes, such as **Congo red**, are of particular concern because of their widespread industrial use and potential toxic, mutagenic, and carcinogenic effects. **Green-synthesized nanoparticles** have shown significant potential for the catalytic degradation of organic pollutants, including synthetic dyes. Silver nanoparticles (**AgNPs**) have attracted considerable attention due to their large specific surface area, excellent catalytic properties, and efficient electron-transfer capability, while green synthesis using plant extracts as reducing and stabilizing agents represents a sustainable and environmentally friendly approach. Plant biomolecules, especially polyphenols and flavonoids, play a key role in the formation of stable and catalytically active AgNPs. In this study, AgNPs were synthesized using *Agrimonia eupatoria L.* extract, and their catalytic activity was evaluated through the reduction of Congo red in the presence of sodium borohydride, highlighting their potential for efficient and sustainable dye removal from wastewater.

Keywords:
- Silver Nanoparticles,
- Green Synthesis,
- *Agrimonia eupatoria L,*
- Catalytic potential,
- Congo red

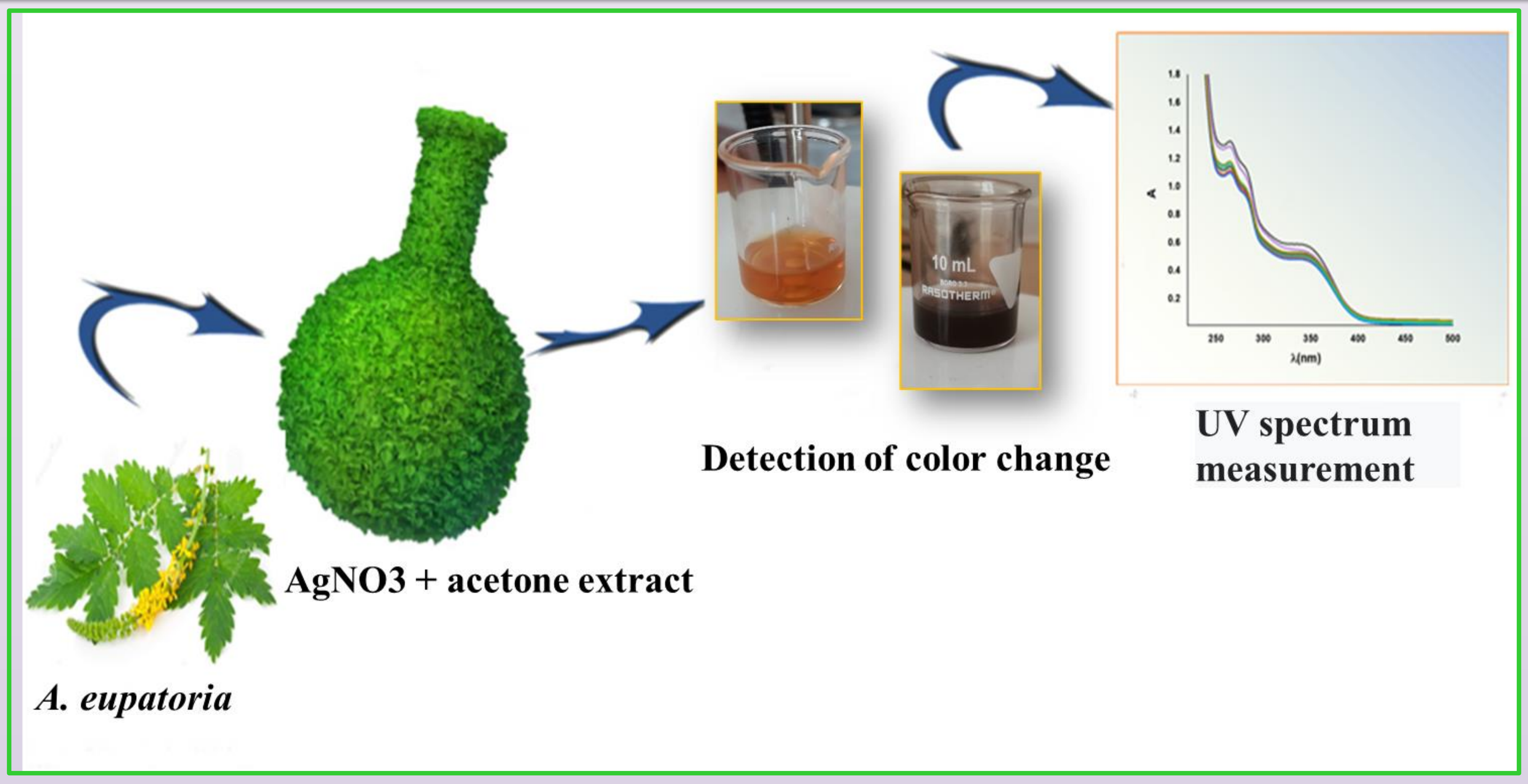

***Figure 1**. Schematic illustration of green synthesis of AgNPs using plant extract Agrimonia eupatoria L[1]*

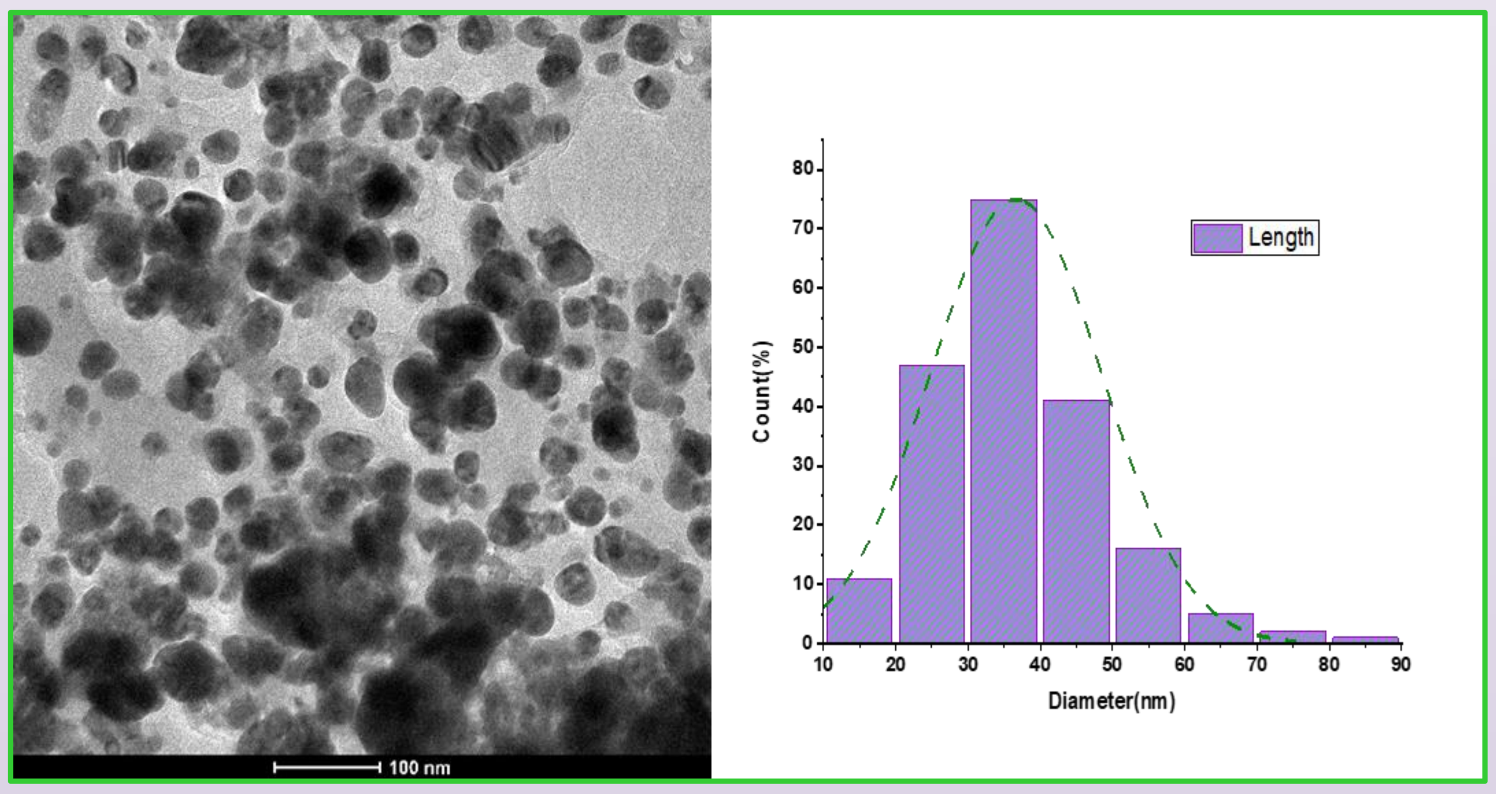

***Figure 2**. TEM image and size distribution of AgNPs synthesized using A. eupatoria aqueous extrac[1,2]*

Experimental:
Aqueous plant extracts were prepared by maceration of dried and ground plant material in distilled water at room temperature, with solvent replacement every 24 h. The extracts were concentrated under reduced pressure and stored at 4 ° C until use. **Silver nanoparticles** (AgNPs) were synthesized using $AgNO_3$ as a precursor, while the plant extracts served as reducing and stabilizing agents. The synthesis conditions ($AgNO_3$ concentration, pH, temperature, and reaction time) were optimized, and the formation of AgNPs was monitored by visual color change and UV–Vis spectrophotometry (200–800 nm). The nanoparticles were isolated by centrifugation, washed with demineralized water, dried at 40 ° C, and stored at 4 ° C. **The catalytic activity of AgNPs** was evaluated by the reduction of Congo red (CR) in the presence of $NaBH_4$. The degradation process was monitored by UV–Vis spectrophotometry at 25 ° C (300–700 nm, λmax = 500 nm). The reaction rate constant was calculated using a *pseudo*-first-order kinetic model.

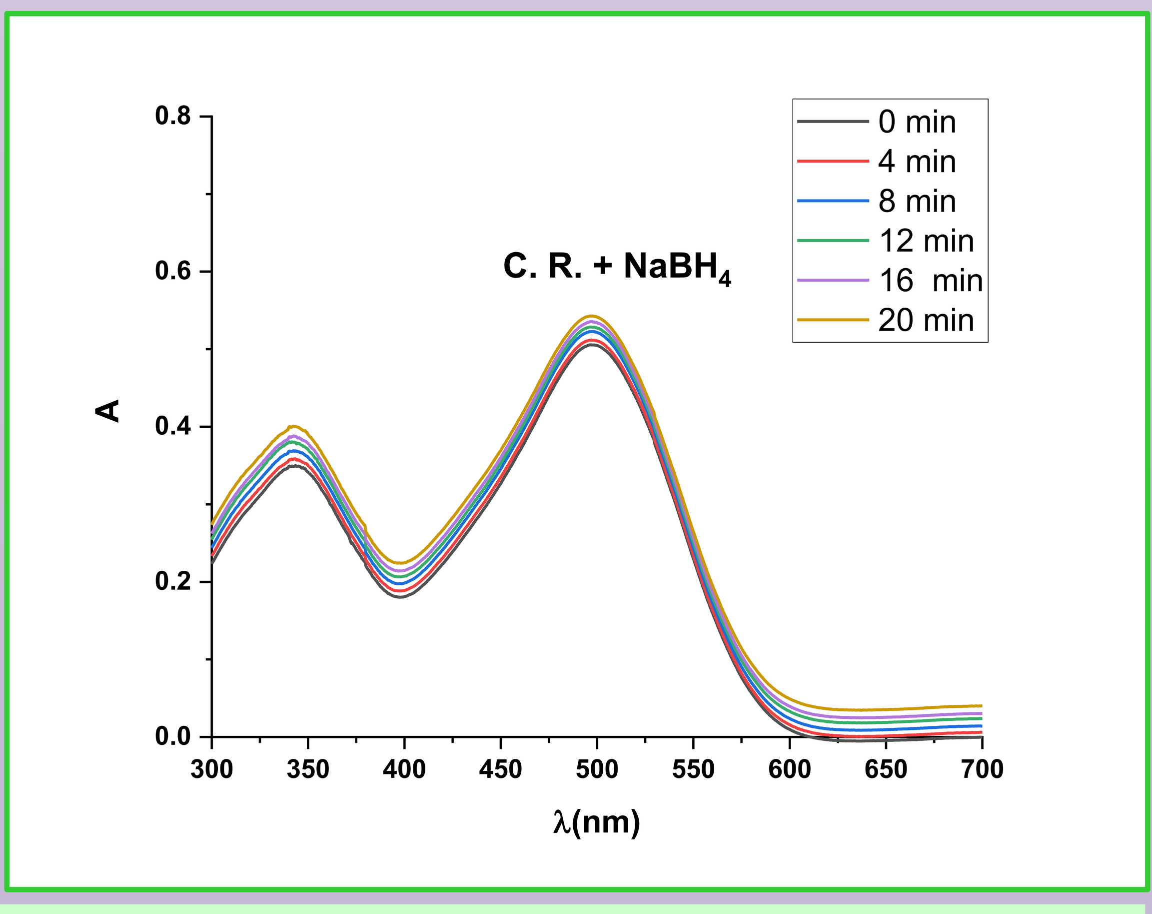

***Figure 3**. UV–Vis spectra of the catalytic degradation of Congo Red in the presence of NaBH4 without AgNPs.*

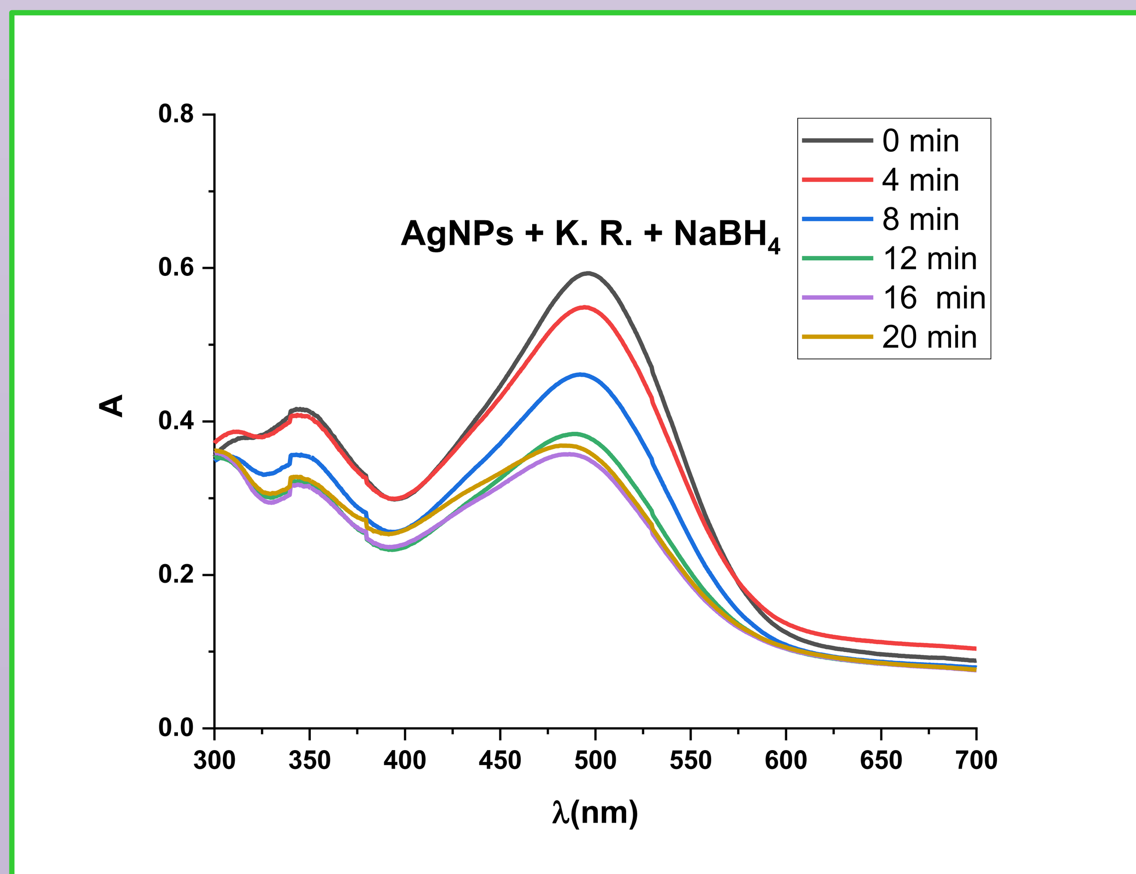

***Figure 4**. Catalytic degradation of Congo Red in the presence of AgNPs*

The reaction rate constant (k) for the catalytic degradation of CR was calculated using the **pseudo-first-order** kinetic equation:

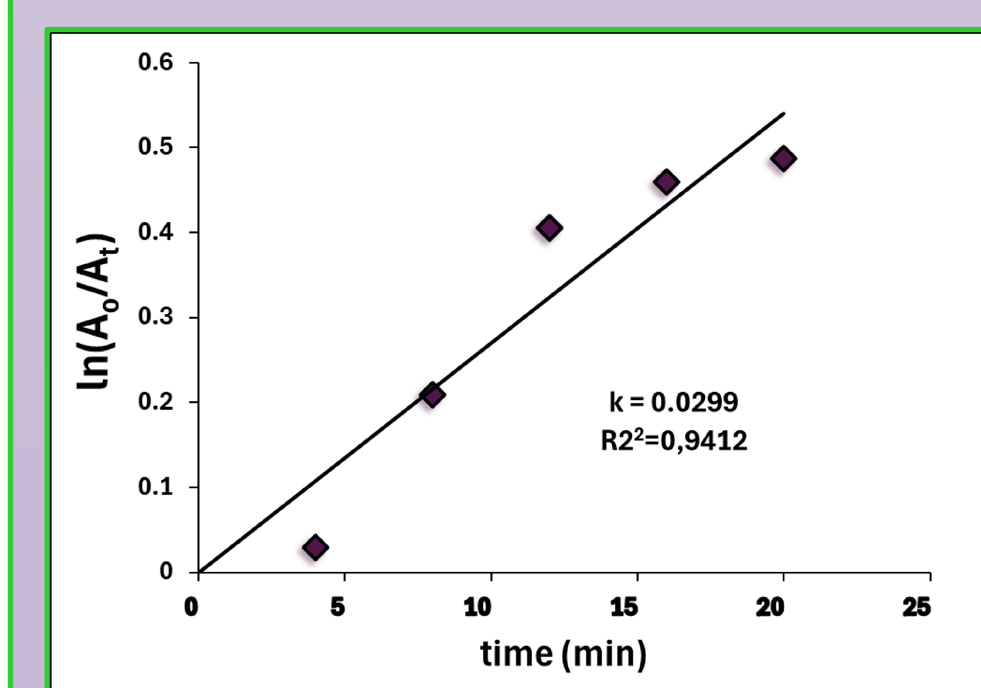

***Figure5**. Pseudo-first-order kinetic plot of Congo red degradation.*

$$ln\frac{A_0}{A_t} = k \cdot t$$

Conclusions:
- Biogenic AgNPs showed **excellent catalytic activity** toward Congo red degradation in the presence of $NaBH_4$.
- UV–Vis analysis confirmed negligible dye degradation with $NaBH_4$ alone, while AgNPs caused a rapid decrease in the characteristic absorption band.
- The degradation process followed **pseudo-first-order** kinetics (**k = 0.0299 min^{-1}**).
- Green-synthesized AgNPs demonstrated **high catalytic efficiency** and potential for environmentally friendly wastewater treatment.
- The results **are consistent** with previously reported studies on green-synthesized AgNPs.

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