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Dosage-Controlled Photocatalysis of ZnO/TiO_2/Biochar for Efficient Removal of Tartrazine under UV–Visible Irradiation

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Abstrak

Tartrazin merupakan zat warna sintetis yang banyak digunakan dalam industri pangan, farmasi, dan minuman serta memiliki struktur kimia yang relatif stabil sehingga sulit dihilangkan melalui proses pengolahan limbah konvensional. Fotokatalisis menggunakan material semikonduktor menjadi salah satu pendekatan yang potensial untuk menguraikan zat warna melalui pembentukan spesies oksidatif reaktif. Kombinasi TiO_2, ZnO, dan biochar dikembangkan untuk meningkatkan kinerja fotokatalitik melalui sinergi antara aktivitas semikonduktor dan kemampuan adsorpsi biochar. Proses fotodegradasi dilakukan menggunakan fotokatalis TiO_2/ZnO/Biochar dengan variasi dosis 0; 0,1; dan 0,5 g dalam 500 mL larutan model selama 120 menit menggunakan penyinaran UV-Vis. Konsentrasi tartrazin dianalisis menggunakan spektrofotometer UV-Vis berdasarkan perubahan absorbansi selama proses degradasi. Dosis 0,1 g memberikan kinerja fotokatalitik terbaik dengan efisiensi degradasi sebesar 74% lebih tinggi dibandingkan penyinaran UV A-B sebesar 48% dan adsorpsi dalam kondisi gelap sebesar 19% setelah 120 menit. Peningkatan dosis katalis tidak meningkatkan efisiensi secara proporsional, yang diduga berkaitan dengan meningkatnya penghamburan cahaya dan aglomerasi partikel. Hasil ini menunjukkan bahwa TiO_2/ZnO/Biochar berpotensi digunakan sebagai fotokatalis untuk pengolahan limbah yang mengandung zat warna tartrazin.

Kata kunci: *TiO_2/ZnO/Biochar, photocatalysis,catalyst dosage, tartrazine.*

Abstract

Tartrazine is a synthetic dye widely used in the food, pharmaceutical, and beverage industries. Its relatively stable chemical structure makes it difficult to remove through conventional wastewater treatment processes. Photocatalysis using semiconductor materials is a promising approach for dye degradation through the generation of reactive oxidative species. A TiO_2/ZnO/Biochar photocatalyst was developed to enhance photocatalytic performance through the synergistic effects of semiconductor activity and biochar adsorption capacity. Photodegradation was conducted using TiO_2/ZnO/Biochar at catalyst dosages of 0, 0.1, and 0.5 g in 500 mL of model solution for 120 minutes under UV-Vis irradiation. Tartrazine concentration was analyzed using a UV-Vis spectrophotometer based on changes in absorbance during the degradation process. A catalyst dosage of 0.1 g showed the highest photocatalytic performance, achieving 74% degradation efficiency after 120 minutes, compared with 48% under UV A-B irradiation and 19% under dark adsorption conditions. Increasing the catalyst dosage did not proportionally improve degradation efficiency, presumably due to increased light scattering and particle agglomeration. These findings demonstrate the potential of TiO_2/ZnO/Biochar as a photocatalyst for treating wastewater containing tartrazine dye.

Keywords: *ZnO/TiO_2/Biochar; photocatalysis; catalyst dosage; tartrazine*

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INTRODUCTION

Water pollution caused by synthetic dyes has become a serious environmental issue due to their persistence, toxicity, and resistance to conventional biodegradation processes. Tartrazine is an azo dye widely used in the food, beverage, and pharmaceutical industries because of its high color stability and low production cost. Its persistence in aquatic environments can pose potential risks to ecosystems and human health, particularly when degradation is incomplete and potentially harmful intermediate compounds are formed (Tuoi et al., 2024).

In wastewater generated from food industries, pharmaceutical manufacturing, and domestic activities, synthetic dyes such as tartrazine may be present and require effective treatment before being discharged into the environment. The chemical stability of tartrazine makes its removal challenging using conventional biological treatment processes. Therefore, the development of effective technologies capable of degrading tartrazine into simpler and less harmful compounds is necessary.

Conventional wastewater treatment technologies, including coagulation, filtration, and adsorption, generally transfer contaminants from one phase to another and may be less effective for the complete degradation of persistent organic pollutants. Advanced oxidation processes, particularly heterogeneous photocatalysis, have attracted considerable attention because they can degrade organic contaminants through the generation of highly reactive species and potentially convert them into simpler compounds such as carbon dioxide and water without producing hazardous secondary residues (Pant et al., 2020).

Titanium dioxide (TiO_2) and zinc oxide (ZnO) are among the most widely investigated semiconductor photocatalysts owing to their excellent chemical stability, relatively low toxicity, and strong oxidative capabilities (Suryandari et al., 2021). Nevertheless, both materials have limitations associated with relatively wide band gap energies and rapid electron–hole recombination, which can reduce their photocatalytic performance (Pant et al., 2020). The incorporation of biochar has been proposed as an effective strategy to improve photocatalytic activity because biochar possesses a porous structure, high surface area, and conductive properties that can facilitate electron transport and suppress charge recombination (Zhou et al., 2022).

Previous studies have demonstrated the promising performance of semiconductor–biochar composites for dye degradation. Paulu and Bart (2021) reported that TiO_2/Biochar achieved approximately 94% degradation efficiency toward Rhodamine B after 120 min of irradiation, whereas pristine TiO_2 only reached 62%. Heltina et al. (2025) found that ZnO/Biochar exhibited degradation efficiencies ranging from 88–92% for methylene blue under visible-light irradiation. El et al. (2023) synthesized TiO_2/Biochar composites for methyl orange degradation, demonstrating the potential of biochar-based semiconductor composites for the treatment of dye-containing wastewater. These findings indicate that the combination of semiconductor photocatalysts with biochar can improve the removal of persistent organic dyes.

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However, photocatalytic efficiency is strongly influenced by the amount of catalyst used. An insufficient catalyst dosage may provide limited active sites for pollutant adsorption and degradation, whereas excessive catalyst dosage can increase light scattering and particle agglomeration, reducing the effective penetration of irradiation into the reaction system. Therefore, optimization of catalyst dosage is an important factor in achieving efficient photocatalytic degradation.

The novelty of the present study lies in the application of a ternary ZnO/TiO_2/Biochar photocatalyst for tartrazine degradation under UV–visible irradiation, with particular emphasis on catalyst dosage optimization without the addition of external oxidizing agents. The combination of ZnO, TiO_2, and biochar is expected to provide synergistic effects through improved pollutant adsorption, enhanced charge separation, and more effective utilization of irradiation.

Therefore, this study aims to evaluate the effectiveness of dosage-controlled ZnO/TiO_2/Biochar photocatalysts in degrading tartrazine under UV–visible irradiation and to determine the optimum catalyst dosage for achieving maximum degradation efficiency.

RESEARCH METHOD

Tartrazine (≥99%, Sigma-Aldrich, USA) was used as the model pollutant. Titanium dioxide (TiO_2, anatase phase, Merck, Germany), zinc oxide (ZnO, Merck, Germany), biochar derived from corn cob, hydrochloric acid (HCl, 37%, Merck, Germany), sodium hydroxide (NaOH, Merck, Germany), and distilled water were used throughout the experiments.

Instruments:

The instruments used in this study included an analytical balance (Ohaus Pioneer PA214, USA), a UV–Vis spectrophotometer (Shimadzu UV-1800, Japan), a pH meter (Hanna Instruments HI2211, Romania), a magnetic stirrer-hotplate (IKA C-MAG HS7, Germany), a UV–Visible lamp (250 W Mercury Lamp, Philips, Netherlands), a UV A-B lamp (20 W, Philips, Netherlands), volumetric glassware (Pyrex®, USA), and Whatman No. 1 filter paper (Cytiva, UK).

Preparation of Tartrazine–Tetracycline Mixed Solution

A stock solution of 100 mg L^{-1} was prepared by dissolving 50 mg of tartrazine in distilled water and diluting the solution to 500 mL in a volumetric flask. Subsequently, standard solutions with concentrations of 2, 4, 6, 8, and 10 mg L^{-1} were prepared by serial dilution and used to construct the tartrazine calibration curve.The synthetic tartrazine solution used in the photocatalytic experiments was prepared by appropriate dilution of the stock solution with distilled water to obtain an initial concentration of 10 mg L^{-1}. The solution was homogenized using magnetic stirring before the photocatalytic experiments.

Photocatalytic Experiments

Photocatalytic degradation experiments were conducted in a batch reactor containing 500 mL of tartrazine solution. The ZnO/TiO_2/Biochar photocatalyst was added at dosages of 0, 0.10, and 0.50 g. The experiments were conducted under UV–Visible irradiation for 120 min to evaluate the effect of catalyst dosage and determine the optimum dosage.

Samples were collected at irradiation times of 0, 30, 60, 90, and 120 min. Each sample was filtered using Whatman No. 1 filter paper to separate the photocatalyst particles before UV–Vis analysis.

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The catalyst dosage providing the highest degradation efficiency was subsequently used for comparative experiments under UV A-B irradiation and dark adsorption conditions. The dark adsorption experiment was conducted without irradiation to distinguish the contribution of adsorption from the photocatalytic degradation process.

UV–Visible Spectrophotometric Analysis

The maximum absorption wavelength (λmax) of tartrazine was determined using a UV–Vis spectrophotometer within the wavelength range of 300–500 nm. Calibration curves were established using tartrazine standard solutions with concentrations ranging from 2 to 10 mg L^{-1}. The residual concentration of tartrazine after treatment was determined from its absorbance using the corresponding calibration equation (Fayazi, 2024).

Degradation Efficiency and Kinetic Analysis

Photocatalytic degradation efficiency was calculated according to Equation (1):Degradation (%) $= ((C_0 - C_t)/C_0) \times 100$ Where C_0 is the initial concentration (mg L^{-1}) and C_t is the concentration at irradiation time t (mg L^{-1}). The degradation kinetics were evaluated using a pseudo-first-order model: $\ln(C_0/C_t) = kt$ Where k is the apparent rate constant (min^{-1}), and t is the irradiation time (min). Sampling Technique and Data Analysis

Sampling was conducted using a purposive sampling approach by collecting aliquots from predetermined catalyst dosages and irradiation times according to the experimental design. All experiments were performed in triplicate, and the obtained data were expressed as mean ± standard deviation. Graphical analysis and kinetic modeling were used to determine optimum photocatalytic conditions and evaluate the performance of ZnO/TiO_2/Biochar toward degradation of tartrazine mixtures (Sharma et al., 2023).

RESULTS AND DISCUSSION

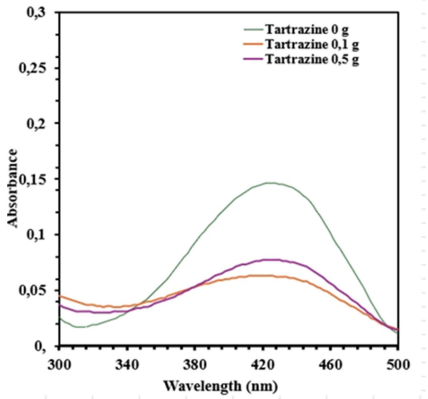


Figure 1. UV–Vis absorption spectra of tartrazine at different ZnO/TiO_2/Biochar catalyst dosages.

Figure 1 presents the UV–Vis absorption spectra of tartrazine at different ZnO/TiO_2/Biochar catalyst dosages (0, 0.1, and 0.5 g) over the wavelength range of 300–500 nm. All samples exhibit a broad absorption band in the visible region, with the maximum absorption occurring at approximately 425 nm. This result is consistent with previous reports indicating that tartrazine has a characteristic

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maximum absorption wavelength around 425–428 nm in aqueous solution (Green, 1990; Jain et al., 2003; Safi et al., 2014).

The absorption intensity differs among the catalyst dosages. The sample without catalyst (0 g) exhibits the highest absorbance, whereas the samples containing 0.1 and 0.5 g of ZnO/TiO_2/Biochar show lower absorbance. The absorption maximum around 425 nm is associated with the chromophoric system of tartrazine and is commonly used as the analytical wavelength for monitoring tartrazine concentration during degradation experiments (Jain et al., 2003; Rojas et al., 2016).

The decrease in absorbance intensity after treatment indicates a reduction in the amount of tartrazine remaining in the solution. However, the degradation efficiency should be determined quantitatively from the change in concentration using the initial and residual concentrations rather than from the absorbance spectrum alone. In this study, the subsequent degradation analysis showed that the 0.1 g catalyst dosage provided the highest degradation efficiency.

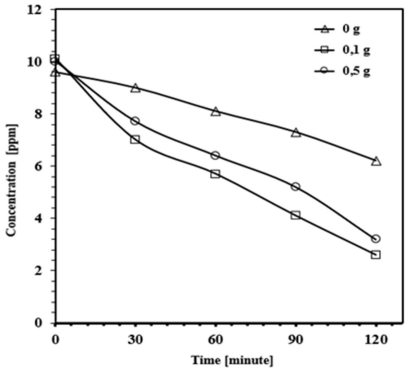


Figure 2. Effect of ZnO/TiO_2/Biochar catalyst dosage on the degradation of tartrazine over irradiation time

Figure 2 illustrates the changes in tartrazine concentration during 120 min of UV–Visible irradiation at different ZnO/TiO_2/Biochar catalyst dosages (0, 0.1, and 0.5 g). The initial tartrazine concentration was approximately 10 mg L^{-1} for all treatments. Overall, the tartrazine concentration decreased progressively with increasing irradiation time, indicating that the treatment effectively reduced the concentration of tartrazine in the solution.

The sample without photocatalyst (0 g) showed the lowest decrease in concentration, from approximately 9.6 to 6.2 mg L^{-1} after 120 min. In contrast, the addition of ZnO/TiO_2/Biochar enhanced tartrazine removal. At a dosage of 0.1 g, the concentration decreased from approximately 10.1 to 2.6 mg L^{-1}, representing the greatest reduction among the tested dosages. The 0.5 g dosage also reduced the concentration substantially, reaching approximately 3.2 mg L^{-1} at 120 min.

The superior performance obtained at 0.1 g indicates that increasing the photocatalyst dosage does not necessarily result in a proportional increase in degradation efficiency. An appropriate catalyst dosage provides sufficient active sites for pollutant adsorption and photocatalytic reactions, whereas excessive catalyst loading can increase light scattering, solution turbidity, and particle aggregation, thereby limiting effective light penetration and the availability of active sites (Khan et al., 2024; Zhang et al., 2023)

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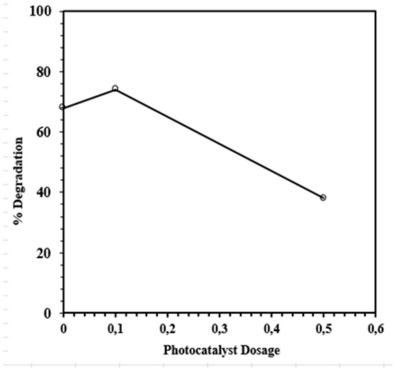


Figure 3. Effect of ZnO/TiO₂/Biochar catalyst dosage on tartrazine degradation efficiency

Figure 3 shows the effect of ZnO/TiO₂/Biochar catalyst dosage on the degradation efficiency of tartrazine after 120 min of UV–Visible irradiation. The catalyst dosages investigated were 0, 0.1, and 0.5 g. The degradation efficiency increased from 38% at 0 g to 74% at 0.1 g, indicating that the presence of ZnO/TiO₂/Biochar substantially enhanced tartrazine removal. This improvement can be attributed to the availability of additional active sites on the photocatalyst surface, which facilitates the interaction between tartrazine molecules and the photocatalyst and promotes photocatalytic reactions. Similar behavior has been reported for tartrazine degradation using TiO_2, where increasing catalyst loading initially enhanced degradation due to the increased availability of active sites (Gupta et al., 2011).

However, increasing the catalyst dosage from 0.1 to 0.5 g resulted in a decrease in degradation efficiency from 74% to 68%. This indicates that increasing the photocatalyst amount beyond the optimum level did not improve the degradation performance. Excessive catalyst loading can increase suspension turbidity and light scattering, thereby reducing the penetration of irradiation into the reaction medium. In addition, increased particle concentration can promote particle–particle interactions and agglomeration, which may reduce the effective surface area and accessibility of active sites (Ahmed et al., 2015; Tolosana-Moranchel et al., 2021).

The highest degradation efficiency was therefore obtained at a catalyst dosage of 0.1 g, corresponding to 74% degradation. This result demonstrates that the photocatalyst dosage is an important operational parameter in the ZnO/TiO₂/Biochar system, and 0.1 g was selected as the optimum dosage for the subsequent irradiation-condition comparison. The occurrence of an optimum catalyst loading followed by a decrease in degradation at higher dosages has also been reported in previous photocatalytic dye degradation studies (Gupta et al., 2011; Tan et al., 2011).

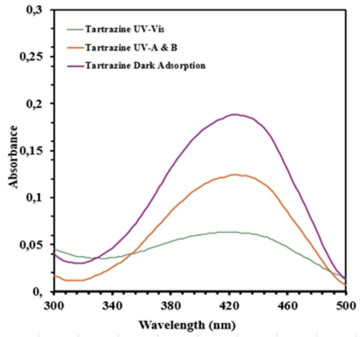


Figure 4. UV–Vis absorption spectra of tartrazine under different treatment conditions

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Figure 4 presents the UV–Vis absorption spectra of tartrazine under three different treatment conditions: UV–Visible irradiation, UV-A&B irradiation, and dark adsorption, using the optimum ZnO/TiO_2/Biochar catalyst dosage of 0.1 g. The spectra were recorded over the wavelength range of 300–500 nm. All three treatment conditions exhibit a characteristic absorption band in the visible region, with the maximum absorption occurring at approximately 425 nm. This wavelength is consistent with previously reported λmax values for tartrazine in aqueous solution, which are generally around 425–429 nm (Gupta et al., 2011; Huanhuan et al., 2020).

The absorbance intensity differs considerably among the treatment conditions. The dark adsorption sample exhibits the highest absorbance, followed by UV-A&B irradiation, whereas the UV–Visible irradiation treatment shows the lowest absorbance at approximately 425 nm. The lower absorbance observed after UV–Visible treatment indicates a greater reduction in the concentration of tartrazine compared with the other treatment conditions. UV irradiation can activate semiconductor photocatalysts, promoting the generation of reactive oxygen species that contribute to the degradation of organic dye molecules (Gupta et al., 2011).

The relatively high absorbance observed under dark adsorption indicates that adsorption alone contributes to tartrazine removal but is less effective than the combined adsorption and photocatalytic process. Similar experimental approaches have used dark conditions as an adsorption control to distinguish the contribution of adsorption from photocatalytic degradation (Bensalah et al., 2024).

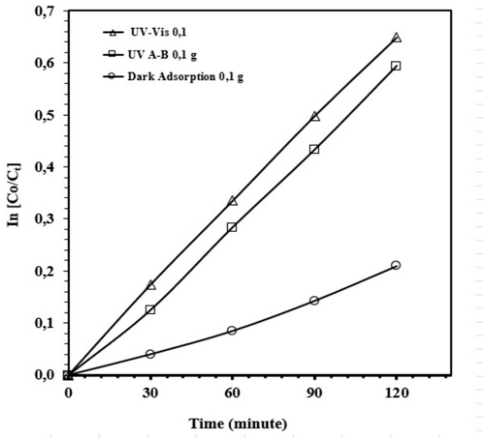


Figure 5. Pseudo-first-order kinetic plots for tartrazine degradation under different irradiation conditions

Figure 5 presents the pseudo-first-order kinetic plots of tartrazine degradation using 0.1 g ZnO/TiO_2/Biochar under three different treatment conditions: UV–Visible irradiation, UV A-B irradiation, and dark adsorption. The relationship between In (Co/Ct) and irradiation time shows an approximately linear increase for all three conditions, indicating that the degradation process can be described by a pseudo-first-order kinetic model. This kinetic model is commonly applied to heterogeneous photocatalytic degradation of tartrazine and other organic dyes (Gupta et al., 2011; Zeghioud et al., 2024).

The UV–Visible treatment exhibited the steepest slope, followed by UV A-B irradiation, whereas dark adsorption showed the lowest slope. Since the slope of the InCo/Ct) versus time plot corresponds to the apparent rate constant (k), the results indicate that the UV–Visible condition had the highest degradation rate, while dark adsorption had the lowest reaction rate. Similar kinetic behavior has been reported

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for tartrazine photocatalytic degradation, where a linear In(Co/Ct) versus irradiation-time relationship was used to demonstrate pseudo-first-order kinetics (Gupta et al., 2011; Husein et al., 2026).

At 120 min, the (Co/Ct) values reached approximately 0.65 for UV–Visible irradiation, 0.59 for UV-A&B irradiation, and 0.21 for dark adsorption. The higher value obtained under UV–Visible irradiation reflects a greater reduction in tartrazine concentration and is consistent with the degradation efficiencies obtained previously, namely 74% for UV–Visible, 48% for UV A-B, and 19% for dark adsorption.

The relatively low slope under dark adsorption indicates that adsorption contributes to tartrazine removal but is less effective than the irradiation-assisted photocatalytic process. This distinction is important because dark adsorption provides an indication of the contribution of surface adsorption, whereas irradiation activates the semiconductor photocatalyst and promotes the formation of reactive species responsible for pollutant degradation (Husein et al., 2026; Ahmed et al., 2023).

CONCLUSION

The ZnO/TiO_2/Biochar ternary photocatalyst demonstrated effective performance in degrading tartrazine under UV–Visible irradiation. Catalyst dosage influenced the photocatalytic activity, as indicated by the decrease in tartrazine concentration during the degradation process. Among the investigated dosages, 0.1 g exhibited the highest degradation efficiency, reaching 74% after 120 min of irradiation, compared with 38% at 0 g and 68% at 0.5 g. Increasing the catalyst dosage to 0.5 g did not further improve the degradation efficiency, which may be attributed to increased light scattering and particle agglomeration that can limit light penetration within the reaction system. Comparison of irradiation conditions showed that UV–Visible irradiation achieved the highest degradation efficiency (74%), followed by UV A-B irradiation (48%) and dark adsorption (19%). These findings indicate that 0.1 g ZnO/TiO_2/Biochar under UV–Visible irradiation represents the optimum condition among the investigated experimental conditions for tartrazine degradation.

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