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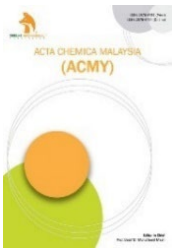

RESEARCH ARTICLE

ENHANCED PHOTOCATALYTIC DECOLORIZATION OF ORANGE II AND RHODAMINE B DYES IN AQUEOUS SOLUTION WITH G-C_3N_4 OBTAINED FROM UREA AND 1,3,5-TRIHYDROXYBENZENE

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ABSTRACT

Graphitic carbon nitride (g-C_3N_4) photocatalysts were synthesized via thermal polymerization of urea in the presence of 1,3,5-trihydroxybenzene (THB) as a molecular modifier. The incorporation of 10 mg THB resulted in enhanced visible-light-driven photocatalytic performance for dye decolorization in aqueous solution. Under 450 nm irradiation, the modified g-C_3N_4 (T10) achieved over 90% degradation of Orange II within 60 min, with a pseudo-first-order rate constant approximately five times higher than that of pristine g-C_3N_4. Kinetic analysis based on the Langmuir–Hinshelwood model confirmed improved charge transfer efficiency and reduced electron–hole recombination. In contrast, Rhodamine B degradation exhibited substrate-dependent behavior, indicating that surface modification influences adsorption characteristics and interfacial reaction pathways. The enhanced performance toward Orange II is attributed to extended π-conjugation, improved visible-light absorption, and surface hydroxyl functionalities introduced by THB, which promote dye adsorption and reactive oxygen species formation. These findings demonstrate that molecular-level modification of g-C_3N_4 using aromatic hydroxyl compounds provides an effective strategy to tailor photocatalytic selectivity and improve visible-light-driven wastewater treatment performance.

KEYWORDS

Visible light; Photocatalytic decolarization; g-C_3N_4; Dye solution; 1,3,5-trihydroxybenzene

1. INTRODUCTION

Water pollution caused by synthetic dyes such as Orange II and Rhodamine B has become a serious environmental concern due to their high chemical stability, toxicity, and resistance to conventional wastewater treatment methods. These dyes are widely used in textile, paper, leather, and plastic industries, and even at low concentrations, they can significantly disrupt aquatic ecosystems and pose risks to human health. Their complex aromatic molecular structures render them chemically and photochemically stable, limiting their biodegradation and leading to persistent contamination of natural water bodies. Consequently, the development of efficient and sustainable technologies for dye degradation has become an urgent research priority (Rania et al., 2022).

Graphitic carbon nitride (g-C_3N_4), a metal-free polymeric semiconductor composed of tri-s-triazine (heptazine) units, has attracted considerable attention over the past decade owing to its suitable band gap (~2.7 eV), excellent thermal and chemical stability, and low-cost synthesis (Deluga et al., 2004; Weifeng et al., 2020). Unlike conventional photocatalysts such as TiO_2 and ZnO, which are primarily activated under ultraviolet light, g-C_3N_4 can be effectively activated by visible light, making it particularly attractive for solar-driven photocatalytic applications. Despite these advantages, the photocatalytic efficiency of pristine g-C_3N_4 remains limited by intrinsic drawbacks, including a narrow light absorption range, rapid recombination of photogenerated electron–hole pairs, low specific surface area, and an insufficient number of surface-active sites. These limitations collectively hinder charge transfer efficiency and overall catalytic performance (Weifeng et al., 2020).

In this context, the incorporation of small organic molecules containing aromatic rings and hydroxyl groups has emerged as a promising modification strategy. Aromatic compounds such as 1,3,5-trihydroxybenzene (phloroglucinol) possess rich π-conjugated systems and multiple hydroxyl functionalities. When introduced during the thermal polymerization of urea, these molecules can interact with g-C_3N_4 precursors through π–π stacking interactions or act as carbon dopants, thereby extending the conjugated network and shifting the light absorption edge toward longer wavelengths (Min et al., 2022). In addition, surface hydroxyl groups can enhance hydrophilicity, promote dye adsorption via hydrogen bonding, and facilitate the formation of surface-bound •OH radicals during photocatalysis. These dual effects—electronic structure modification and surface activation—are expected to synergistically improve visible-light-driven photocatalytic performance (Maeda et al., 2018).

Therefore, this study aims to synthesize modified g-C_3N_4 materials through the thermal polymerization of urea in the presence of controlled amounts of 1,3,5-trihydroxybenzene. The photocatalytic performance of the resulting materials was evaluated through the visible-light-induced decolorization of Orange II and Rhodamine B, with particular attention to

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the influence of molecular modification on photocatalytic efficiency.

2. Experimental

2.1 Preparation of g-C_3N_4

The g-C_3N_4 powders were synthesized by a poly-condensation method. Typically, 10 g of urea,5 mL water and 1,3,5-trihydroxybenzene 10 mg was transferred into a 30 mL ceramic crucible with a lid. Then, the crucible was covered by an aluminum foil, placed in a muffle furnace, heated to 500 °C at a heating increment rate of 2 °C min^{-1}, and held for 2 h at 500 °C. After natural cooling to room temperature, the sample was manually ground in an agate mortar, and the product was collected as a yellow powder.

2.2 Photocatalytic performance test

The photocatalytic decolorization experiments were conducted in a Pyrex glass reactor with an inner volume of 123 mL. In a typical procedure, 100 mL of an aqueous 5 ppm Orange II or Rhodamine B (RhB) solution and 30 mg of the g–C_3N_4(T10) were placed into the reactor and magnetically stirred both before and during light irradiation. Prior to illumination, the suspension containing solutions was kept in the dark for 30 min to establish adsorption–desorption equilibrium. Subsequently, the reaction mixture was irradiated for 60 min under visible light (λ = 450 nm). During the photocatalytic decolorization process, 2 mL aliquots of the suspension were withdrawn at predetermined time intervals and centrifuged at 10,000 rpm for 5 min to separate the photocatalyst. The absorbance of the resulting supernatant was then measured using a UV–Vis spectrophotometer (UV-2450, Shimadzu, Kyoto, Japan) at 485 nm for Orange II and 552 nm for RhB to determine the residual dye concentration.

3. Results

3.1 Visible-Light-Induced Decolorization

Figure 1 shows the visible-light-induced photocatalytic decolorization of (a) Orange II and (b) Rhodamine B over pristine g-C_3N_4 and THB-modified g-C_3N_4 (T10). Both dyes exhibited gradual concentration decreases under 450 nm irradiation, confirming that the synthesized materials are active in the visible-light region. For Orange II, the T10 sample demonstrated markedly enhanced photocatalytic activity compared with pristine g-C_3N_4. More than 90% of Orange II was decolorized within 60 min using g-C_3N_4 (T10), whereas significantly lower removal efficiency was observed for the unmodified catalyst under identical conditions. The faster degradation rate suggests that THB incorporation effectively promotes visible-light absorption and facilitates interfacial charge transfer processes. In contrast, Rhodamine B degradation showed a different trend. Although both catalysts were capable of decomposing RhB under visible light, the enhancement effect of THB modification was less pronounced. This indicates that photocatalytic activity is not solely determined by intrinsic electronic properties but is also influenced by dye-specific adsorption behavior and surface interactions. The improved performance observed for Orange II can be attributed to the extended π-conjugation and surface hydroxyl groups introduced by THB during thermal polymerization. These modifications likely enhance light-harvesting capability and suppress electron–hole recombination, while simultaneously improving adsorption of anionic dye molecules. The results demonstrate that molecular modification of g-C_3N_4 can effectively tailor photocatalytic efficiency depending on substrate characteristics.

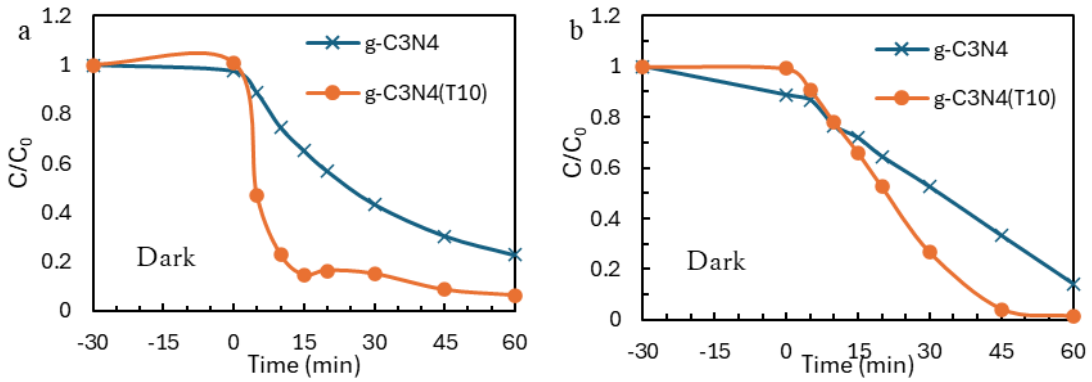

Figure 1: Photocatalytic decolorization of (a) Orange II and (b) Rhodamine B under visible-light irradiation

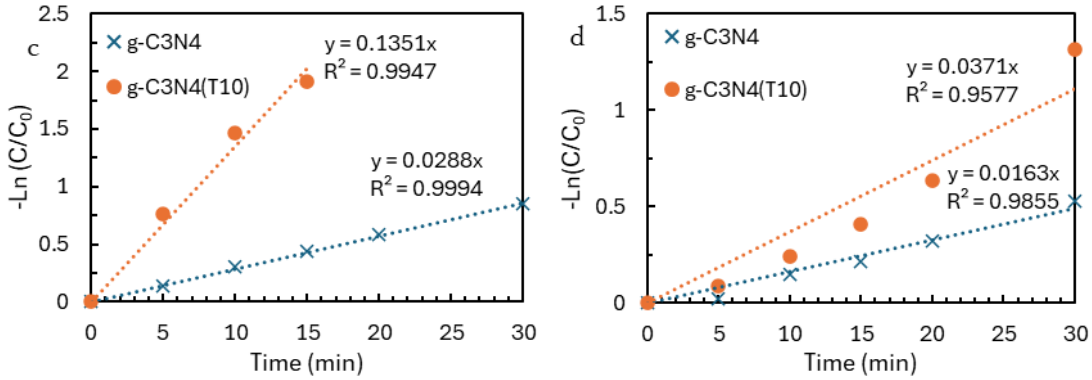

Figure 2: Corresponding kinetic plots based on the Langmuir–Hinshelwood model for Orange II (c) and Rhodamine B (d).

3.2 Reaction rate

Langmuir-Hinshelwood model is often used in the reaction rate theory of photocatalysts (Ouedraogo et al, 2018). The Langmuir-Hinshelwood formula shown here, is a generalization of Langmuir's equation of energy-absorbing isotherm applied to surface reactions:

$$r = -\frac{dC}{dt} = \frac{kKC}{1+KC} \quad (1)$$

where k is the reaction rate constant (mg (L $min)^{-1}$), K is the adsorption coefficient of the reactant (L mg^{-1}), and C is the concentration of the reactant (mg L^{-1}).

When the concentration of the reactant C is minimal, it can be approximated by the following formula (Deluga, G. A. et al. 2004):

$$-\frac{dC}{dt} = kKC \quad (2)$$

$$-Ln\left(\frac{C}{C_0}\right) = k_{app}t \quad (3)$$

where k_{app} is apparent first-order rate constant.

Figures 2 (c) and (d) present the kinetic analysis of Orange II and Rhodamine B decolorization, respectively, based on the Langmuir–Hinshelwood (L–H) model. The linear fits of -Ln(C/C_0) versus irradiation time indicate that both reactions follow pseudo-first-order kinetics.

Table 1: Photocatalytic decolorization parameters for Orange II.			
Photocatalyst	**Pseudo first order rate constant k (min^{-1})**	**Correlation coefficient**	**Substrate half-live $t_{1/2}$ (min)**
g-C_3N_4	0.029	0.995	24
g-C_3N_4 (T10)	0.14	0.999	5.0

Table 2: Photocatalytic decolorization parameters for Rhodamine B.			
Photocatalyst	**Pseudo first order rate constant k (min^{-1})**	**Correlation coefficient**	**Substrate half-live $t_{1/2}$ (min)**
g-C_3N_4	0.037	0.986	19
g-C_3N_4 (T10)	0.016	0.958	43

Table 1 summarizes the kinetic parameters for the photocatalytic decolorization of Orange II under visible-light irradiation. The linear correlation between $-\ln(C/C_0)$ and irradiation time confirms that the degradation process follows pseudo-first-order kinetics according to the Langmuir–Hinshelwood model. The apparent rate constant (k_{app}) of pristine g-C_3N_4 was calculated to be 0.029 min^{-1} with a half-life ($t_{1/2}$) of 24 min. In contrast, the modified g-C_3N_4 (T10) exhibited a significantly higher rate constant of 0.14 min^{-1} and a shortened half-life of only 5.0 min. This represents nearly a fivefold enhancement in photocatalytic activity upon incorporation of 1,3,5-trihydroxybenzene. The high correlation coefficients ($R^2 \geq 0.995$) indicate excellent agreement with the kinetic model, demonstrating stable and reproducible reaction behavior. The substantial acceleration of Orange II degradation suggests that THB incorporation effectively improves charge separation efficiency and promotes interfacial electron transfer processes. The enhancement can be attributed to several synergistic factors (Guo et al., 2024; Ohnishi et al. 2024; Omori et. al., 2025; Watanabe et al., 2025a, b): 1) extended π-conjugation induced by THB, which broadens visible-light absorption. 2) improved surface hydrophilicity due to hydroxyl functionalities. 3) enhanced adsorption of anionic Orange II molecules through hydrogen bonding and electrostatic interactions. 4) reduced electron–hole recombination, facilitating reactive oxygen species generation. These results clearly demonstrate that molecular-level modification during thermal polymerization is an effective strategy for improving photocatalytic performance.

Table 2 presents the kinetic parameters for Rhodamine B (RhB) degradation. Similar to Orange II, the photocatalytic process follows pseudo-first-order kinetics, as evidenced by the linear relationship between $-\mathrm{Ln}(C/C_0)$ and irradiation time. However, unlike the behavior observed for Orange II, the kinetic trend for RhB differs. The pristine g-C_3N_4 exhibited a rate constant of 0.037 min^{-1} with a half-life of 19 min, whereas the modified g-C_3N_4 (T10) showed a lower apparent rate constant of 0.016 min^{-1} and an extended half-life of 43 min. This contrasting behavior indicates that photocatalytic performance is strongly substrate-dependent. The difference may arise from variations in molecular structure, adsorption orientation, and electrostatic interactions between the dye molecules and catalyst surface. Rhodamine B is a cationic xanthene dye with a bulky molecular framework, whereas Orange II is an anionic azo dye. The incorporation of THB likely modifies the surface functional groups and electronic distribution of g-C_3N_4. While this modification enhances adsorption and degradation of anionic species, it may reduce favorable adsorption of cationic RhB due to altered surface charge distribution or steric hindrance effects.

Furthermore, photocatalytic degradation involves multiple steps, including light absorption, charge separation, radical formation, dye adsorption, and interfacial electron transfer. Even if electronic properties are improved, inefficient adsorption can limit overall reaction rates. Therefore, the results emphasize that surface chemistry plays a decisive role in determining substrate selectivity.

To clarify the photocatalytic pathway, the following mechanism can be proposed: Under visible-light irradiation (λ = 450 nm), electrons are excited from the valence band (VB) to the conduction band (CB) of g-C_3N_4, leaving holes in the VB:

$$g\text{-}C_3N_4 + h\nu \rightarrow e^-(CB) + h^+(VB) \quad (4)$$

The photogenerated electrons react with dissolved oxygen to produce superoxide radicals ($\bullet O_2^-$), while holes either directly oxidize dye molecules or generate hydroxyl radicals (•OH). The THB-modified structure enhances π-electron delocalization, which facilitates charge carrier migration and suppresses recombination (Khatun et al., 2024; Suhag et al., 2024a, b; Tateishi et al., 2024; Uzzaman et al., 2024). The different degradation behaviors observed for Orange II and RhB indicate that adsorption affinity and molecular orientation significantly influence radical attack efficiency.

4. Conclusion

In this study, THB-modified g-C_3N_4 photocatalysts were successfully synthesized via thermal polymerization of urea in the presence of 1,3,5-trihydroxybenzene as a molecular modifier. The incorporation of 10 mg THB significantly enhanced visible-light-driven photocatalytic performance toward Orange II degradation, achieving over 90% decolorization within 60 min and exhibiting an apparent rate constant approximately five times higher than that of pristine g-C_3N_4. The kinetic behavior followed pseudo-first-order reaction kinetics based on the Langmuir–Hinshelwood model, confirming efficient charge transfer and improved reaction dynamics.

In contrast, Rhodamine B degradation displayed substrate-dependent behavior, indicating that surface modification influences adsorption characteristics and interfacial reaction pathways. The distinct degradation trends highlight that photocatalytic efficiency is governed not only by electronic structure enhancement but also by dye–catalyst interactions and molecular orientation at the catalyst surface.

The improved performance toward Orange II can be attributed to extended π-conjugation, enhanced visible-light absorption, surface hydroxyl functionalities, and suppressed electron–hole recombination induced by THB incorporation. These findings demonstrate that molecular-level modification of g-C_3N_4 provides an effective and tunable strategy for tailoring photocatalytic selectivity under visible-light irradiation.

Overall, this work offers valuable insight into substrate-dependent photocatalytic mechanisms and presents a simple yet efficient approach for designing advanced g-C_3N_4-based materials for sustainable dye wastewater treatment applications.

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