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RESEARCH ARTICLE

PHOTOCATALYTIC REMOVAL OF TEXTILE DYE POLLUTANTS FROM WATER USING C-TiO_2 NANOPARTICLES

Nusrat Zahan[a], Shahriar Atik Fahim[a,b], Humayra Gazi[a], Md. Shakhawoat Hossain[c], Md. Ashraful Islam Molla[a*]

[a]Department of Applied Chemistry and Chemical Engineering, Faculty of Engineering and Technology, University of Dhaka, Dhaka 1000, Bangladesh
[b] Department of Chemistry, American International University-Bangladesh, Dhaka 1229, Bangladesh
[c] Department of Arts and Sciences, Ahsanullah University of Science and Technology, Dhaka 1208, Bangladesh
****Corresponding Author Email: ashraful.acce@du.ac.bd***

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ABSTRACT

Water pollution caused by textile dyes is a critical environmental issue, significantly impacting aquatic ecosystems and human health. Photocatalysis has emerged as a promising solution to this problem, using light to activate catalysts that degrade organic pollutants. Titanium dioxide (TiO_2), in particular, is a widely used photocatalyst for its strong oxidative properties under UV light. This study explores the degradation of Rhodamine B (RhB), a common and persistent textile dye, using carbon-doped TiO_2 (C-TiO_2) photocatalysts under UV-C light. The impact of various operating parameters, such as pH, initial RhB concentration, photocatalyst amount, and irradiation time, has been investigated. The results showed that at a pH of 9, a dye concentration of 10 mg/L, a photocatalyst amount of 25 mg, and an irradiation time of 90 min, the 6% C-TiO_2 achieved the highest photocatalytic RhB removal of 74% under UV-C light.

KEYWORDS

Photocatalysis, C-TiO_2, Rhodamine B, Nanoparticles, UV-C irradiation

1. INTRODUCTION

In recent times, there has been a growing concern about environmental issues. The rapid growth of industries results in the release of large quantities of pollutants into the environment. Among them the textile dyeing industry significantly contributes to water pollution (Saeed et al., 2022). Releasing untreated effluents poses a substantial risk to both aquatic ecosystems and human well-being. It's essential to implement effective treatments for dye-containing wastewater to mitigate its adverse effects. Photocatalysis has become increasingly popular in recent years as a method for decomposing and photodegrading organic compounds. Titanium dioxide (TiO_2) has attracted considerable interest as a photocatalyst for purifying organic pollutants in water, due to its plentiful availability, affordability, superior electrical conductivity, exceptional resistance to photo-corrosion, stability in aquatic environments, minimal environmental harm, and potent photocatalytic prowess (Fahim et al., 2023).

Significant research has focused on enhancing TiO_2 photocatalysts due to their potential for removing various organic compounds in both water and air phases. However, TiO_2 has a relatively large band gap (3.2 eV) which means activation of TiO_2 photocatalyst requires quite high photon energy (Shathy et al., 2022). Besides the fast recombination of charge carriers (electrons and holes) within TiO_2 can considerably reduce its effectiveness as a photocatalyst. Given these challenges, several approaches have been proposed to narrow the bandwidth gap of TiO_2 by adding dopants to it. Currently, researchers employ various methods to add dopants to TiO_2, including noble metal doping, transition metal doping, non-metal doping, and so on (Li et al., 2015; Chen et al., 2015; Thirupathi et al., 2018). Notably, doping with non-metals has garnered more interest than metal doping for enhancing the photoelectronic properties of TiO_2 and shifting its absorption edge towards longer wavelengths (Hua et al., 2020).

Contemporary research indicates that introducing carbon into TiO_2 significantly boosts its ability to catalyze reactions using light (Yang et al., 2015). The incorporation of carbon atoms into TiO_2 leads to an electron coupling interaction between the carbon and the TiO_2 (Hua et al., 2020). This results in a reduction of the bandgap of TiO_2. Research has shown that carbon-doped TiO_2 is more effective in breaking down organic contaminants. The study highlighted that carbon doping not only expanded the surface area of TiO_2 but also its capacity to absorb visible light, which in turn increased the rate at which it could photo catalytically degrade the dye methylene blue, outperforming its undoped counterpart (Zhang et al., 2020).

In a similar vein, a group of researcher observed that TiO_2 doped with carbon was more efficient in photo catalytically degrading phenol under both UV and visible light (Zhang et al., 2020). The ability of carbon-doped TiO_2 to break down complex organic compounds into less harmful substances was highlighted in a recent work thereby reducing environmental impact (Shathy et al., 2022). This study stands out from prior research by methodically examining the influence of different carbon doping concentrations in TiO_2. Herein, the effects of several parameters, including pH, initial RhB concentration, photocatalyst amount, and irradiation time, were investigated to achieve the highest possible elimination of RhB from wastewater under UV-C light.

2. EXPERIMENTAL METHODS

The synthesis method and characterization of C-TiO_2 nanoparticles were discussed in the previously published paper (Zahan et al., 2024). The

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photocatalytic performance of C-TiO_2 nanoparticles was investigated by degrading RhB under a UV-C light (λ = 254 nm, 25 W, HNS G13, Osram, Russia) at ambient temperature. Typically, 50 mL of RhB dye (10 mg/L) solutions and 25 mg of C-TiO_2 were added to a 100 mL beaker, as displayed in Figure 1. A magnetic stirrer was used in the dark to equilibrate the suspension for 30 min. The suspension in the glass beaker was then exposed to a UV-C lamp at various intervals of time. Approximately 3 mL of RhB solution were collected and filtered using a 0.45 µm Advantec membrane filter. The concentration of the RhB was evaluated using a UV-visible spectrophotometer. Following the Beer-Lambert rule, the relative RhB concentration (C/C_0) was measured at a relative absorbance (A/A_0) of λ_{max} = 552 nm, where A_0 and A were the absorbances of aqueous RhB dye at an initial time (t_0) of photocatalysis and at any time t, correspondingly.

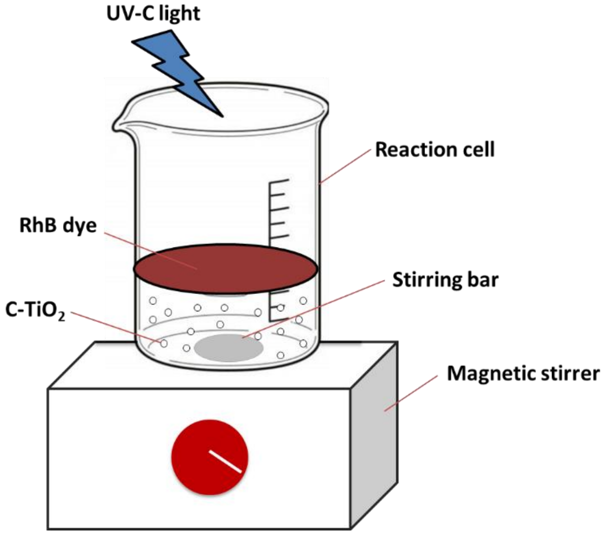

Figure 1: Schematic diagram for the photocatalytic removal of RhB dye.

3. RESULTS AND DISCUSSION

3.1 Photocatalytic removal of RhB dye

The self-degradation of RhB dye was investigated as well both in darkness and under UV-C light to study the efficiency of the C-TiO_2 photocatalysts. The absorption spectrum of RhB dye exhibits a prominent peak at a wavelength of 552 nm. This specific wavelength serves as a reference line which is stated in the Figure 2(a). In the absence of light (dark condition), the adoption of RhB dye was minimal, accounting for less than 10%. However, upon introducing C-TiO_2 photocatalysts and UV-C light under various experimental conditions, the degradation efficiency significantly improved. Specifically, after 90 min of UV-C irradiation, the RhB removal efficiencies were found to be 56%, 46%, 74%, and 52% for TiO_2, 4% C-TiO_2, 6% C-TiO_2, and 8% C-TiO_2 respectively in Figure 2(b). This is because when C-TiO_2 is illuminated by light below 380 nm, it definitively excites valence band electrons across the band gap into the conduction band, unequivocally leaving holes behind in the valence band (Parra et al., 2000).

The holes in TiO_2 react with water molecules or hydroxide ions (OH^-), producing hydroxyl radicals (•OH). Organic pollutants that are adsorbed on the catalyst's surface will then be oxidized by •OH. On the other hand, the improvement of photocatalytic performance is occurred due to the incorporation of C, which further reduces the forbidden bandwidth of the co-doped catalyst (Wang et al., 2021). A decrease in band gap energy could have led to the improved photocatalytic activity of C-TiO_2 as it requires less energy to activate a photocatalytic reaction. Another reason may be that C-TiO_2 is known to demonstrate exceptional quantum efficiency (Irfan et al., 2022). This high level of quantum efficiency implies that a greater number of electron-hole pairs are engaged in the photocatalytic reaction, which in turn contributes to an enhanced degradation efficiency (Irfan et al., 2022).

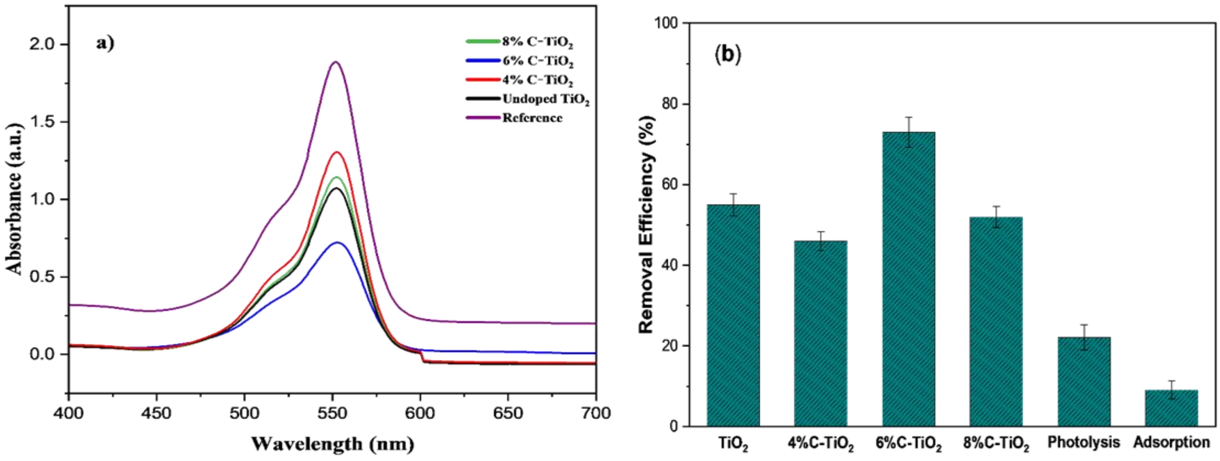

Figure 2:1 (a) UV–visible results showing the decay in the intensity of RhB solution (b) Photocatalytic removal, adsorption under dark, and photolysis for RhB (photocatalyst: 25 mg; initial RhB concentration: 10 mg/L; pH: 9; UV-C irradiation: 90 min).

3.2 Effect of pH

The pH level of the dye solution is a key factor affecting photodegradation performance. It notably influences the electrostatic interactions between the catalyst, dye molecules, and reactive oxygen species during the degradation process (Saeed et al., 2022). For comparison, the normal pH of the RhB solution was measured to be approximately 6 in Figure 3. The pH of the solution was varied at different time interval while maintaining another parameters constant. At 90 min of irradiation time, the photocatalytic performance was consistently better in every case. It was found that the photocatalytic removal efficiency after 90 min of irradiation time was 39% at pH 5 and reached its peak efficiency at pH 11 which was 75%, as shown in Figure 3. At pH 9, the catalytic efficiency was found to be 74%. This behavior observed in the degradation of RhB can be attributed to the adsorption characteristics of RhB on the catalyst's surface (Islam et al., 2020). The photocatalytic process predominantly takes place on the surface of the photocatalyst, rather than in the bulk solution. The adsorption of substrates onto the catalyst's surface is a crucial step for their subsequent photocatalytic degradation.

The iso-electric point of the catalyst plays a significant role in influencing the adsorption of organic substrates and intermediates during a

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photoreaction (Islam et al., 2020). Under acidic conditions, the catalyst's surface acquires a positive charge, which discourages the adsorption of positively charged particles like RhB molecules due to electrostatic repulsion. However, when the pH value exceeds the iso-electric point of the catalyst, the surface charge becomes negative. This change in surface charge attracts RhB molecules, leading to enhanced catalytic activity. After optimizing the pH condition, despite the marginal difference in degradation efficiency between pH 9 and pH 11, it is important to consider the environmental implications. Considering that pH 9 already achieves considerable degradation efficiency and is more environmentally friendly, pH 9 was chosen as the optimized condition for the degradation process.

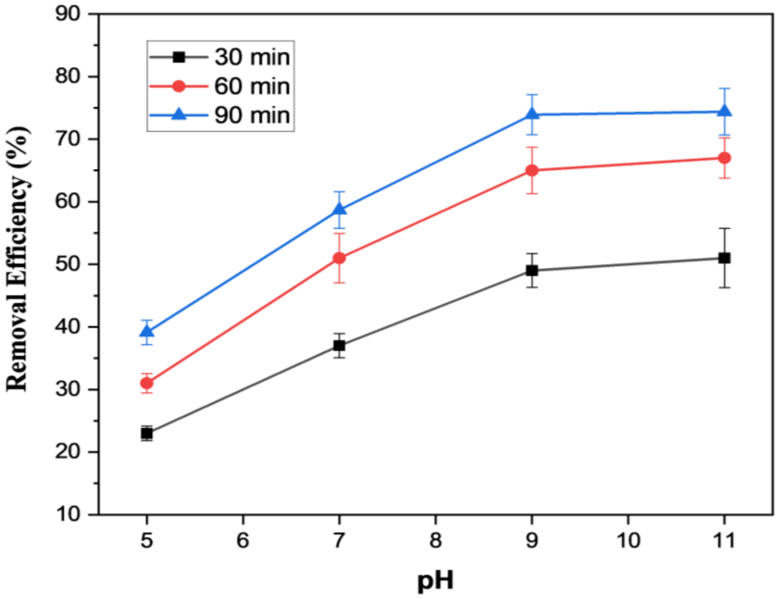

Figure 3:2 Effect of solution pH on photocatalytic removal of RhB with 6% C-TiO$_2$ (photocatalyst: 25 mg; initial RhB concentration: 10 mg/L; pH: 5–11; UV-C irradiation: 30, 60 and 90 min).

3.3 Effect of initial RhB concentration

The photocatalytic removal depends on the dye concentration (Hanafi and Sapawe 2020), as the degradation depends on the active site of the catalyst molecule, which varies with the change in concentration. The degradation efficiency of RhB dye was studied at various time intervals and different initial dye concentrations. It was observed that as the initial concentration of the dye solution increased, the degradation efficiency decreased in Figure 4. At 90 min of irradiation, the photocatalytic performance consistently showed improvement in all instances. Specifically, at a concentration of 5 mg/L, the degradation efficiency was measured 81%, while at 20 mg/L, it decreased to 32%. At 10 mg/L the degradation efficiency was found to be 74%. This can be attributed to the fact that a higher dye concentration leads to more dye molecules being adsorbed onto the catalyst surface, thereby blocking the active sites on the catalyst.

Therefore, at high dye concentrations, the generation of reactive radicals on the surface of the catalyst is reduced since the active sites are covered by dye molecules which in turn affect the catalytic activity of the photocatalyst. Moreover, a rise in substrate concentration can lead to the production of intermediates that may adsorb onto the catalyst surface (Hanafi and Sapawe, 2020). The increased presence of these intermediates can cause a slow dispersion across the catalyst surface, affecting the active sites and ultimately leading to a decrease in removal efficiency. In contrast, at lower concentrations, the number of catalytic sites is not a limiting factor, and the degradation rate is directly proportional to the substrate concentration (Hanafi and Sapawe, 2020). Despite this decrease in efficiency with increasing concentration, an intermediate concentration of 10 mg/L was selected as the optimized condition. While there is a decrease in efficiency from 5 mg/L to 10 mg/L, the 74% degradation efficiency at 10 mg/L remains sufficiently high for practical application and environmental remediation purposes. Furthermore, selecting 10 mg/L offers practical advantages such as scalability and feasibility in real-world scenarios.

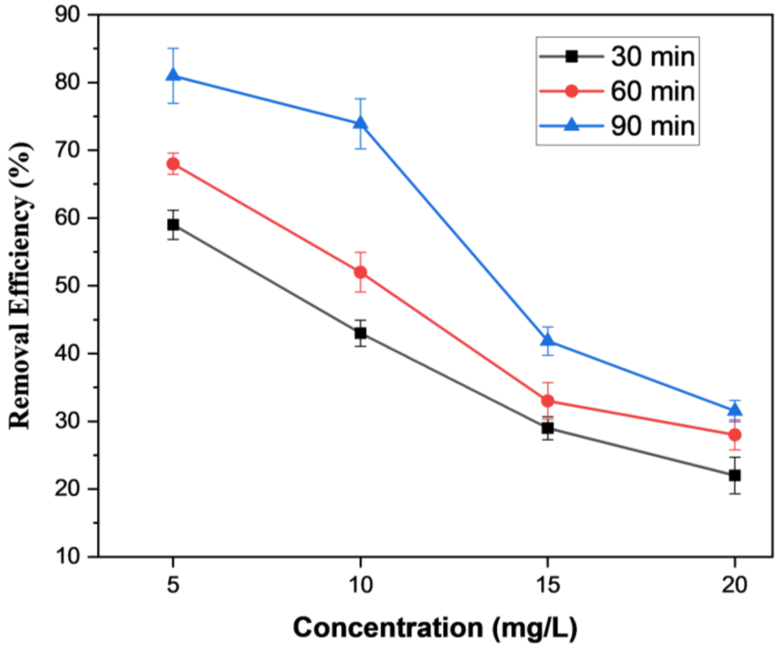

Figure 4:3 Effect of initial concentration on the photocatalytic removal of RhB with 6% C-TiO$_2$ (photocatalyst: 25 mg; initial RhB concentration: 5–20 mg/L; pH: 9; UV-C irradiation: 30, 60 and 90 min).

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3.4 Effect of photocatalyst amount

The influence of catalyst quantity was investigated by varying the amount of catalyst introduced into the RhB dye solution and the irradiation time, while maintaining other conditions constant. The investigation revealed a discernible trend. As the catalyst dose increased from 15 mg to 30 mg, there was a notable variation in degradation efficiency (Figure 5) in every time interval. Specifically, at doses of 15 mg, 20 mg, 25 mg, and 30 mg, the degradation efficiencies at 90 min of irradiation time were measured at 49%, 58%, 74%, and 68%, respectively which are higher than the degradation observed at 30 min and 60 min of irradiation. Notably, the highest degradation efficiency of 74% was achieved at a catalyst dose of 25 mg. However, at 30 mg, a slight decrease in efficiency to 68% was observed. The enrichment in the photocatalytic activity with the increase in catalyst dose was attained mainly because of the increase in the surface area of the catalyst. Increasing the dose of the catalyst results in a larger number of active sites for the reaction (Molla et al., 2019). In other words, the improvement in photocatalytic activity resulted due to the generation of more photogenerated electron-hole pairs. Finally, the photogenerated electron-hole carriers further the formation of reactive species for photocatalytic degradation. Conversely, with the addition of more catalyst doses, the removal efficiency is observed to be decreased. An overly high dose of the catalyst might result in an excess absorption of light by the catalyst particles and this could potentially diminish the light accessible for the photocatalytic reaction, consequently leading to a decrease in the efficiency of dye removal (Buu et al., 2023; Mamun et al., 2017). Again, there exists a saturation point beyond which an increase in the catalyst dose no longer enhances the removal efficiency. This is attributed to the fact that all dye molecules have already established interactions with the catalyst, consequently, any surplus catalyst does not contribute to additional photodegradation (Shathy et al., 2022). Based on these findings, a catalyst dose of 25 mg emerges as the most favorable condition, achieving a substantial degradation efficiency of 74%.

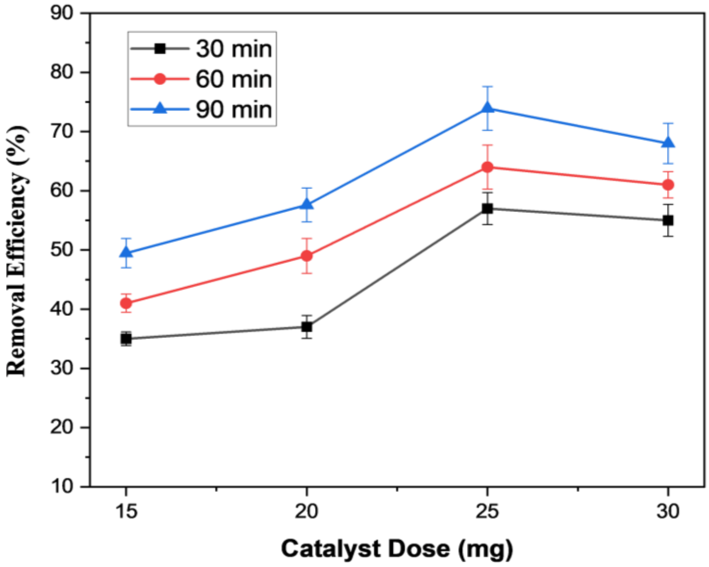

Figure 5: Effect of photocatalyst amount on the photocatalytic removal of RhB with 6% C-TiO_2 (photocatalyst: 10–25 mg; initial RhB concentration: 10 mg/L; pH: 9; UV-C irradiation: 30, 60 and 90 min).

3.5 Effect of irradiation time

The duration of irradiation time significantly impacts the degradation process. As the irradiation time varies, the dye molecules have differing amounts of time to bind to the catalyst's active sites. This directly influences the extent of dye adsorption, thereby affecting the degradation rate. The influence was observed by varying the time of irradiation, while maintaining other conditions constant. The degradation rate of RhB was recorded for undoped TiO_2, 4% C-TiO_2, 6% C-TiO_2, 8% C-TiO_2 with irradiation times of 30, 60, and 90 min, respectively (Figure 6). The figure clearly demonstrates that in each instance, 6% C-TiO_2 exhibited superior photocatalytic efficiency, achieving 34%, 52%, and 74% after 30, 60, and 90 min of irradiation, respectively. Based on these results, the optimized irradiation time was determined to be 90 min, yielding the highest degradation rate of 74%. This prolonged engagement facilitates the adsorption of a larger number of dye molecules, which subsequently undergo photodegradation (Islam et al. 2020). Moreover, the catalytic efficiency increases over time as the formation of •OH and $•O_2^-$ increases with irradiation time (Fahim et al., 2023).

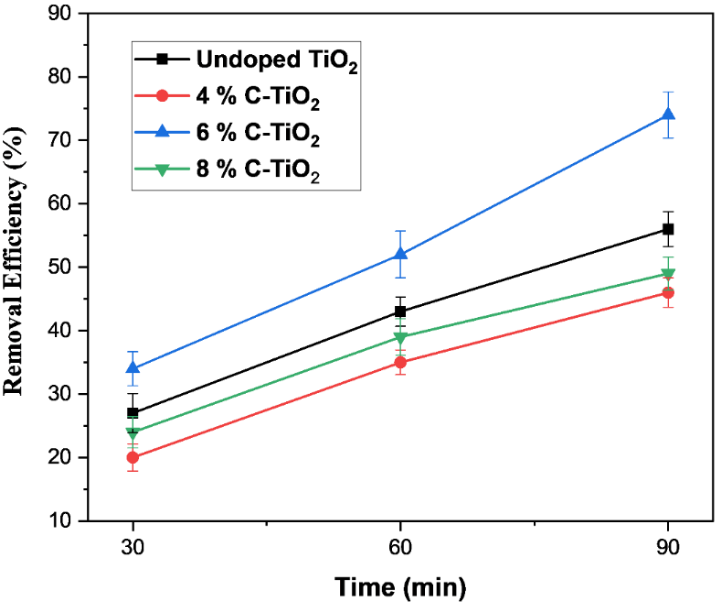

Figure 6: Effect of irradiation time on the photocatalytic removal of RhB with Undoped TiO_2, 4% C-TiO_2, 6% C-TiO_2, 8% C-TiO_2 (photocatalyst: 25 mg; initial RhB concentration: 10 mg/L; pH: 9; UV-C irradiation: 30–90 min).

4. CONCLUSION

The study convincingly showcased the remarkable enhancement in photocatalytic removal of RhB dye using C-TiO_2 under UV-C light irradiation. Within 90 min of UV-C light irradiation, with an initial RhB concentration of 10 mg/L and 25 mg of 6% C-TiO_2, the photocatalytic removal efficiency of RhB was found to be 74% at a pH of 9. Compared to undoped TiO_2, 6% C-TiO_2 nanoparticles demonstrate superior photocatalytic performance in RhB dye degradation. Therefore, the C-TiO_2 photocatalyst offers itself as a viable substitute for the elimination of hazardous textile dyes.

CREDIT AUTHORSHIP CONTRIBUTION STATEMENT

Nusrat Zahan: Investigation, Methodology, Formal analysis, Writing – original draft, Writing – review & editing. Shahriar Atik Fahim: Formal analysis, Writing – review & editing. Humayra Gazia: Formal analysis, Writing – review & editing. Md. Shakhawoat Hossain: Formal analysis, Visualization. Md. Ashraful Islam Molla: Conceptualization, Project administration, Supervision, Funding acquisition, Methodology, Formal analysis, Visualization, Writing – review & editing.

DECLARATION OF COMPETING INTEREST

The authors declare no competing financial interests.

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