Study Kinetic of Photocatalytic Degradation of Methylene Blue Dye Under UV and Daylight Using g-C3N4

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**Abstract**   
Methylene blue (MB) is a widely applied synthetic dye in industries such as textiles, paper, and pharmaceuticals; however, its toxic nature, chemical stability, and resistance to biodegradation make it a significant environmental pollutant. In this study, graphitic carbon nitride g-C3N4 was employed for the removal of MB through a combination of adsorption and photocatalytic degradation under both UV and daylight conditions. The adsorption process was best described by a pseudo-second-order kinetic model, indicating that the mechanism is largely influenced by surface interactions and the presence of active sites on g-C3N4. Meanwhile, the photocatalytic degradation followed a first-order kinetic model, with a stronger correlation observed under daylight exposure. MB removal efficiencies reached 56.20% under daylight and 22.46% under UV light, demonstrating the efficiency performance of g-C3N4 in the visible light range. This enhanced activity is attributed to the suitable bandgap of g-C3N4 (~2.98 eV), which enables efficient absorption of visible light and facilitates the generation of reactive species. Overall, the findings support the use of g-C3N4 as an effective photocatalyst for the treatment of dye-contaminated wastewater through a dual mechanism involving both adsorption and photocatalytic degradation.

*Keyword: g-C3N4, Methylene Blue, Daylight,**Photocatalytic*

# INTRODUCTION

The production of synthetic dyes is rising because of their demand, particularly in textile and clothing industries. Every year, thousands of tons of these chemicals are produced worldwide, with the annual industrial output of dye compounds being approximately 700.000 tons[1]. Methylene blue (MB) is synthetic dye widely used as a colorant for paper, wool, silk, and cotton. It is a widely recognized organic dye that dissolves stability in water at room temperature and MB is known to be highly toxic at elevated concentrations [2]. Additionally, most organic dye is difficult to biodegrade due to their high resistance to environmental conditions, making their removal from wastewater a critical and challenging task. To mitigate the risk posed by organic dye, several treatment methods have been developed, including adsorption, photocatalysis, advanced oxidation processes, ion exchange, and microbiological decomposition.

Photocatalysis is widely recognized as an effective and environmentally friendly method for degrading organic pollutants such as Methylene Blue (MB) due to its ability to harness light energy to drive chemical reactions without the need for additional reagents. In photocatalytic reactions, a semiconductor material can be excited by light to produce electron–hole pairs that initiate redox reactions on the catalyst surface. These charge carriers can react with water and oxygen to produce reactive oxygen species (ROS), including hydroxyl radicals (•OH) and superoxide anions (O₂•⁻), which are extremely effective in decomposing complicated dye molecules like MB into non-toxic end products like CO2 and H2O [3]. Compared to conventional methods like adsorption or chemical oxidation, photocatalysis offers the advantages of reusability of the catalyst, low operational cost, and the ability to utilize solar energy as a renewable light source [4]. Therefore, photocatalysis is considered a promising and sustainable approach for the treatment of dye-contaminated wastewater. The photocatalytic degradation under daylight has several significant advantages. One of the key benefits is its energy efficiency since it makes use of plentiful and renewable solar energy, lowering its dependence on artificial UV light sources that consume electricity. This makes the process more sustainable and economic, particularly for large-scale or outdoor applications like wastewater treatment. Additionally, numerous advanced photocatalysts have been designed to respond to visible light, enabling efficient pollutant degradation under natural light conditions.

Several studies have been conducted on various types of semiconductor materials, such as TiO₂ and ZnO, which are commonly used in the photocatalytic degradation of dyes. However, these materials are primarily active under UV light and exhibit limited activity under visible light due to their relatively wide bandgap energies of approximately 3.3 eV for TiO₂ [5],[6] and 3.2 eV for ZnO [7],[8]. Graphitic carbon nitride (g-C3N4) has emerged as a metal-free semiconductor with remarkable potential for photocatalytic applications, particularly under visible light irradiation. Its moderate band gap energy of approximately 2.7 eV enables the absorption of a broad range of the visible spectrum, making it suitable for daylight-driven photocatalysis [9]. While several studies suggest that g-C3N4 performs more efficiently under daylight than under ultraviolet (UV) irradiation. Therefore, it is important to evaluate the photocatalytic degradation kinetics under both UV and daylight irradiation to obtain a clearer understanding of the reaction rate and efficiency under each condition. This approach allows for a more accurate comparison of g-C3N4 activity and helps clarify its true potential as a visible-light-driven photocatalyst.

This study aims to demonstrate the superior photocatalytic efficiency of g-C3N4 under daylight irradiation compared to UV light. The study involves a comprehensive kinetic evaluation of both the adsorption and photocatalytic degradation stages of methylene blue. By analyzing and comparing the reaction kinetics under different lighting conditions, the study aims to provide clear evidence that daylight more effectively activates g-C3N4. The findings are expected to offer strong evidence of g-C3N4 enhanced performance under visible light, contributing to the development of more efficient and sustainable photocatalytic systems for environmental applications.

# Methodology

## Materials

Urea (99.9%, Sigma-Aldrich) was used as precursors of g-C­3N4. Deionized (DI) water as solvent. Methylene blue (MB) synthetic was used as dye for photocatalytic activity.

## Synthesis of g-C3N4

The synthesis of g-C3N4 was carried out by polycondensation, utilizing urea as the precursor, adapting a previously reported method[10]. The process began by placing 10 g of urea in a crucible. This crucible was transferred to a muffle furnace and subjected to heat treatment at 525 °C for 3 hours. Following calcination, the sample was allowed to cool to room temperature before being collected and mechanically ground into powder. The steps for this synthesis are summarized in **Figure 1**.



Figure 1. Schematic synthesis of g-C3N4

## Photocatalytic Test

To evaluate the photocatalytic activity, a standardized procedure was applied for the g-C₃N₄ samples. First, 0.25 g of the designated catalyst was suspended in 500 mL of 10 ppm methylene blue (MB) solution. The suspension was kept in the dark for 25 minutes to establish adsorption–desorption equilibrium. After this step, the mixture was exposed to daylight irradiation and UV irradiation provided by 12 W and 20 W, respectively. The photocatalytic reaction was initiated under these conditions, and samples were collected after 60 minutes. Each sample collected was centrifuged for 10 minutes at 300 rpm to separate the catalyst. The resulting filtrate was analyzed with a UV-Vis spectrophotometer at 664 nm to determine the final MB concentration and calculate the removal percentage using Equation 1.

|  |  |
| --- | --- |
|  | (1) |

with C0 and Ct was initial concentration (ppm) and concentration at variation time (ppm), respectively.

## Kinetic Studies

The kinetic studies of methylene blue (MB) removal using g-C₃N₄ were carried out to evaluate both the adsorption and photocatalytic degradation processes. For the adsorption process (in dark conditions), the kinetic data were fitted using pseudo-first-order, pseudo-second-order models and diffusion intra-particle , as described in the following equations (1), (2), and (3):

|  |  |
| --- | --- |
|  | (1) |
|  | (2) |
|  | (3) |

In these equations, qt (mg/g) is the amount of MB adsorbed at time (minute), qe is the adsorption capacity at equilibrium, C is boundary layer thickness (mg/g) and k1ads, k2ads and kid are the rate constants of the pseudo-first-order, pseudo-second-order models, and diffusion intra-particle, respectively.

For the photocatalytic degradation process under UV and daylight irradiation, the kinetic behavior was evaluated using first order and second-order kinetic models [11],[12], given by equations (4) and (5), respectively:

|  |  |
| --- | --- |
|  | (4) |
|  | (5) |

in the above equation, C0 is initial MB concentration at t = 0 min (ppm), Ct is final MB concentration at time (ppm) and k1 and k2 are the first order and second order rate constant, respectively [13].

## Characterization

The characterization used to support the results of degradation photocatalytic is Fourier Transform Infrared Spectroscopy (FTIR), X-Ray Diffraction (XRD), and optical characterization. Fourier Transform Infrared Spectroscopy (FTIR) is employed to identify the functional groups present in the composite catalyst. The samples are positioned on a holder and exposed to infrared light with a wavelength range of 400-4000 cm-1. X-ray diffraction (XRD) was employed to determine change in the material structure following the calcination process. This analysis utilized CuKα radiation. The X-ray scanning range was set from 5-50° to identify alterations in the crystal structure and intermolecular distance between polymer intersegmental chain influenced by the pyrolysis heating rate. The optical properties were investigated using UV-Vis spectroscopy to determine the band gap energy of the g-C3N4. The band gap energies of the samples were estimated from Tauc plots derived from UV-Vis Diffuse Reflectance Spectroscopy (DRS) data.

# Result and Discussion

## Characterization

**Figure 2** illustrates the FTIR spectrum of synthesized g-C₃N₄ (Figure 2) shows a number of sharp absorption bands that are indicative of the successful development of its typical structure. The broadband in the range of 3000–3400 cm⁻¹ is attributed to stretching vibrations of N–H groups and remaining –OH groups, usually present as a result of insufficient condensation of precursor molecules of urea or adsorbed surface moisture. A sharp absorption peak located at 1628 cm⁻¹ is attributed to the stretching vibration of C=N bonds, and it is an indication of the presence of conjugated heteroaromatic regions. Absorption bands located at 1538, 1403, and 1233 cm⁻¹ are assigned to stretching vibrations of aromatic C–N and C–N–C bonds in the heptazine (s-triazine) backbones. It is noteworthy that the sharp and strong peak located at 808 cm⁻¹ is indicative of the typical breathing mode of the s-triazine units, acting as a fingerprint of g-C₃N₄ and presenting clear evidence for its successful polymerization. The existence of these functional groups emphasizes g-C₃N₄'s graphitic-layered-like framework, in which the conjugated π-electron system is retained inside the heptazine network. Specifically, the rich nitrogen functionalities (C–N, C=N, and terminally bonded –NH₂ groups) are recognized as active adsorption sites of cationic dyes like methylene blue via electrostatic attraction and π–π accumulation. Such a structural property accounts for the robust correlation between the detected functional groups via FTIR and pseudo-second-order adsorption kinetics manifested in the research. In addition, the nitrogen-rich sites are crucial in trapping photo-excited charge carriers and hence contributing significantly to the formation of reactive oxygen species in the case of photocatalysis. Accordingly, FTIR analysis not only verifies the chemical nature of g-C₃N₄ but also sheds light on its dual application as an adsorbent and photocatalyst in wastewater remediation. FTIR spectrum of the prepared g-C3N4, providing compelling evidence of its chemical bonding environment. A prominent absorption peak observed at 1628 cm-1 is attributed to the C=N stretching modes. The presence and integrity of the core aromatic C-N structure are confirmed by multiple absorption bands at 1233, 1403, and 1538 cm-1. Critically, the sharp and intense band at 808 cm-1 serves as a fingerprint for g-C3N4, representing the out-of-plane bending vibration (or breathing mode) of the s-triazine (or heptazine) heterocyclic units. These results are confirmed by the literature and are in excellent agreement with the reference spectrum for g-C3N4 reported by Warda et al. (2025)[14].

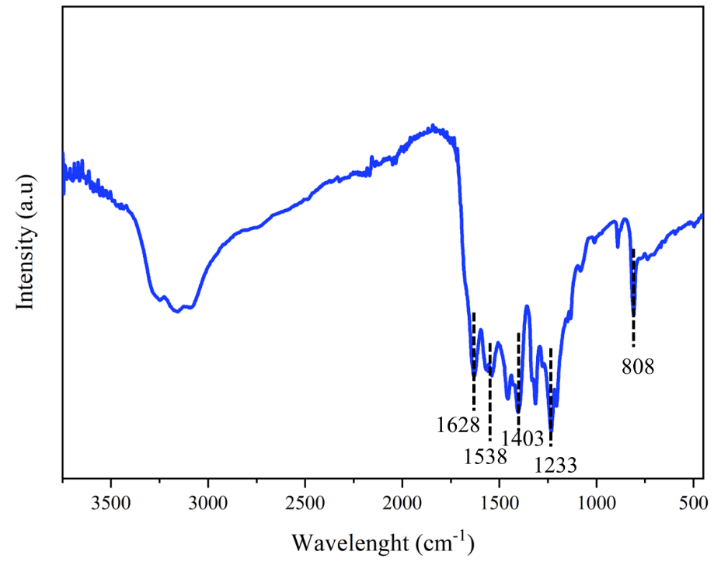


Figure 2. FTIR of g-C3N4

**Figure 3a** presents the XRD results for the g-C₃N₄ material. The analysis shows two main diffraction peaks. A high-intensity peak detected at 27.85° is assigned to the (002) plane, which is indicative of the interlayer stacking within the graphitic structure[15],[16]. Additionally, a peak observed at 12.85° is indicated as the (100) plane and is associated with the in-plane periodicity arising from the nitride pore structure[17]. These diffraction features, as further illustrated in **Figure 3b**, confirm the typical of g-C₃N₄ characterized by its layered morphology and extended conjugated structure.

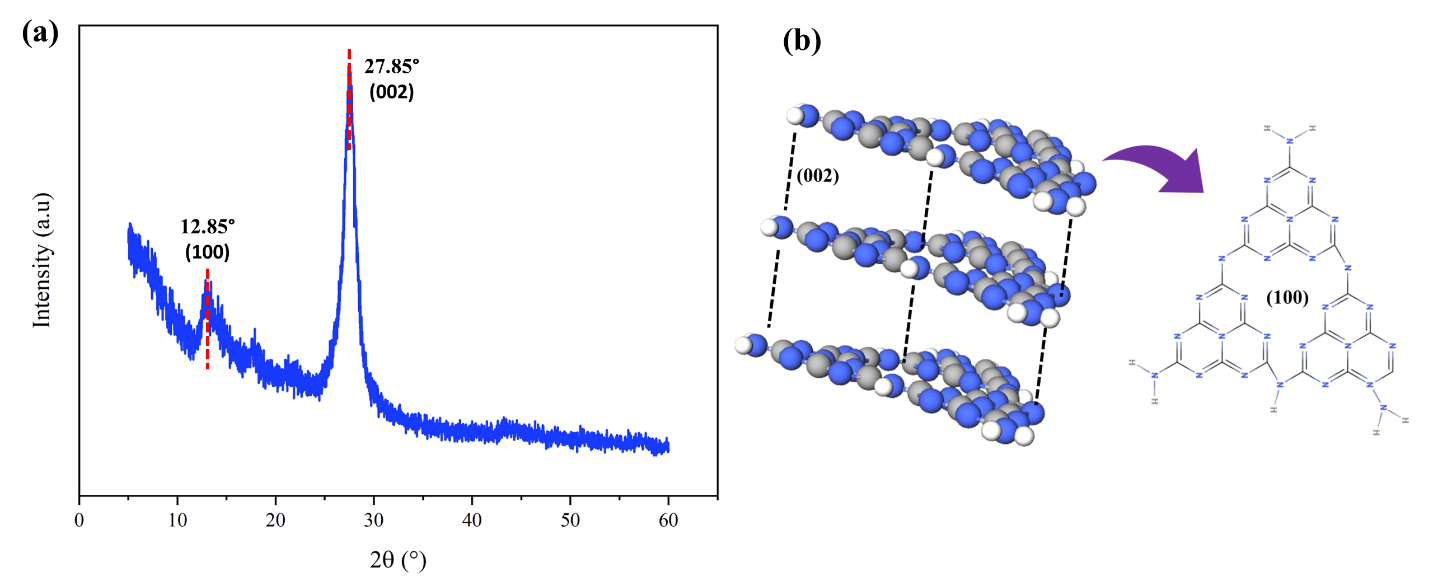


Figure 3. XRD (a) and structure (b) of g-C₃N₄

The SEM micrograph of g-C3N4 (**Figure 4**) reveals that the material exhibits an aggregated morphology with a dominant flake-like appearance. These flake-like structures indicate the layered nature of g-C3N4, which is consistent with its graphitic structure composed of stacked conjugated sheets. The observed morphology also demonstrates irregular shapes and varying sizes of the flakes, suggesting partial exfoliation and aggregation during the synthesis process [18]. Such features are commonly reported for g-C3N4 prepared via thermal polymerization methods, where the condensation of precursor molecules leads to the formation of nanosheets that subsequently restack [19]. This phenomenon is further attributed to the thermal expansion of the air trapped between the layers, which causes the cleavage of hydrogen bonds and facilitates the separation of g-C3N4 sheets.

A close-up of a grey object

AI-generated content may be incorrect.

Figure 4. SEM of g-C3N4

The nitrogen adsorption–desorption isotherm of g-C3N4 exhibits a typical type IV profile, which is characteristic of mesoporous materials. The presence of a hysteresis loop further confirms the existence of mesopores formed by the stacking of g-C3N4 layers and the interparticle voids[20]. The specific surface area, calculated by the Brunauer–Emmett–Teller (BET) method, was found to be 53.08 m2g-1. Enhancement in surface area provides a greater number of accessible active sites and facilitates mass transfer during photocatalytic processes. This mesoporous structure is favorable for the diffusion of reactant molecules, thereby contributing to improved photocatalytic activity.

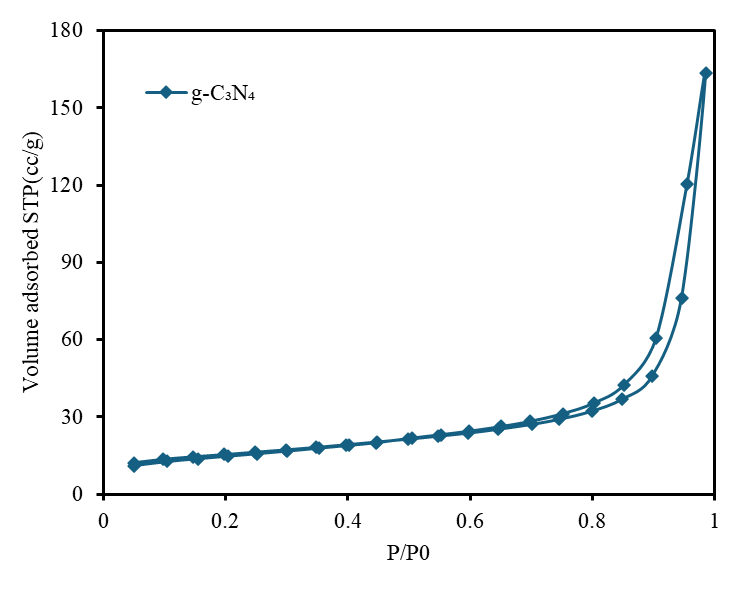


Figure 5. BET of g-C3N4

The Tauc plot in **Figure 6** is utilized to assess the optical properties of g-C3N4, specifically its bandgap energy. As seen from the plot, the bandgap of g-C3N4 is calculated as 2.98 eV, which falls in the near-visible region. This appropriate bandgap makes g-C3N4 capable of absorbing visible light effectively and hence is greatly promising for photocatalytic applications. Specifically, g-C3N4 has exhibited great potential in the photodegradation of MB dye under visible light irradiation, due to its ability to generate reactive species that break down MB molecules. Therefore, its visible-light-responsive nature makes g-C3N4 a strong candidate for use in dye wastewater treatment through photocatalytic processes.

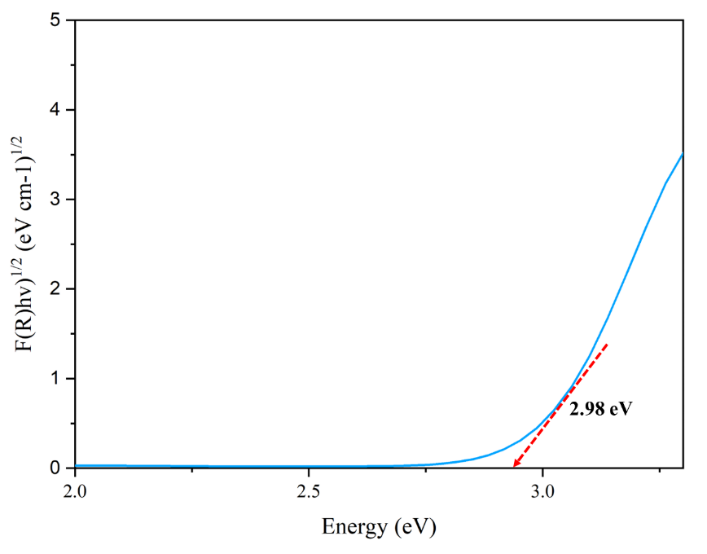


Figure 6. Tauc plot of g-C₃N₄

## Photocatalytic Performance

In **Figure 7a**, the removal efficiency (%) of MB is plotted against time. A noticeable enhancement in photocatalytic performance is observed under daylight conditions compared to UV. During the dark phase, no photocatalytic reaction occurs due to the absence of light energy; thus, any observed decrease in MB concentration is solely attributed to the adsorption of dye molecules onto the surface of the g-C₃N₄ catalyst. This initial adsorption is essential because it facilitates better interaction between the catalyst and dye once light irradiation begins, allowing more efficient photodegradation [21]. Moreover, the similar adsorption trends observed for both UV and daylight samples confirm that the subsequent differences in degradation performance are primarily because of light rather than variations in initial dye binding.

Under daylight, g-C3N4 achieved approximately 56.20% removal of MB within 60 minutes, whereas under UV light, the degradation efficiency remained below 22.46% over the same time frame. This indicates that g-C3N4 is responsive to visible light, which is consistent with its known band gap (2.98 eV), making it an effective photocatalyst under visible-light irradiation. The steep increase in removal percentage after light exposure (t = 0) under daylight further confirms the significant photocatalytic activity of g-C3N4. This result clearly demonstrates that g-C3N4 exhibits enhanced photocatalytic activity under daylight compared to UV light. This is because the bandgap value of g-C3N4 is 2.98 eV, making it active under daylight than under UV light. The absorption spectrum analysis in **Figure 7b** further supports this observation. The UV–Vis spectra show a significant reduction in the characteristic MB absorption peak at around 664 nm after photocatalytic treatment, with the greatest reduction occurring under daylight exposure. The initial MB solution exhibited strong absorbance, while the sample treated under UV showed moderate peak suppression. The daylight-treated sample showed the most pronounced decrease in peak intensity, confirming more extensive degradation of the dye. The inset images in **Figure 7b** visually support the spectral data, showing a noticeable color fading in the MB solution, particularly under daylight exposure. This visual evidence, combined with the spectral and kinetic data, confirms that g-C3N4 is efficient under daylight.

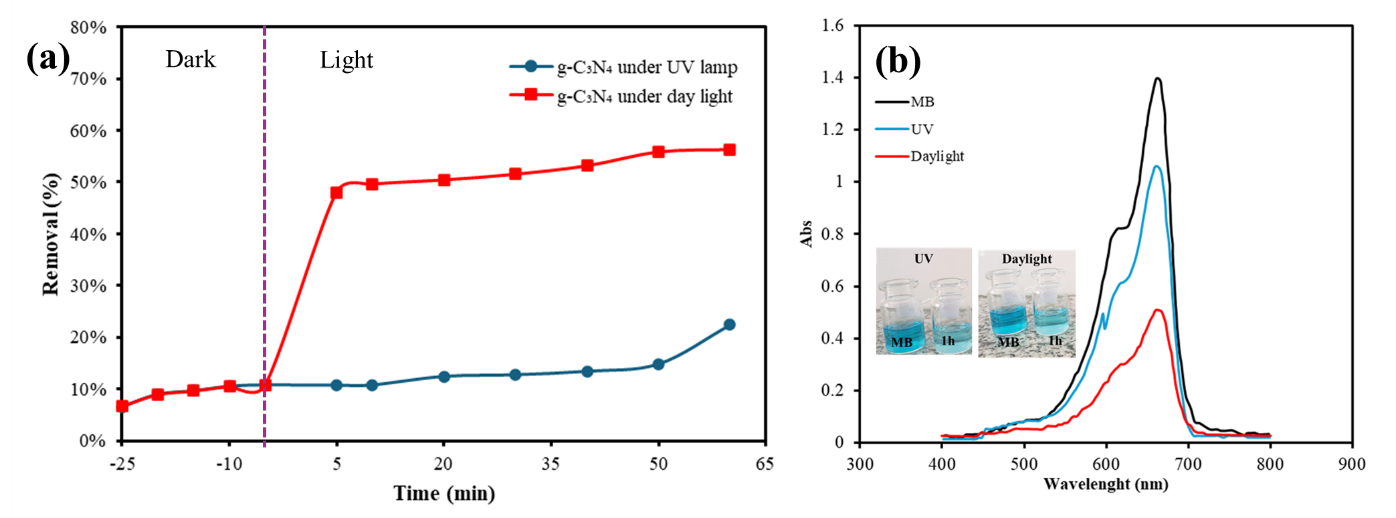


Figure 7. percentage removal of MB (a) and spectra UV-Vis of MB (b)

## Kinetic Evaluation

The adsorption kinetics of methylene blue (MB) onto g-C3N4 were evaluated using pseudo-first order, pseudo-second order and diffusion intra-particle kinetic models, as depicted in **Figure 8a**, **Figure 8b**, and **Figure 8c** respectively. The pseudo-first-order model, represented by the linear plot of ln(Qe−Qt) versus time, yielded a correlation coefficient (R²) of 0.949, indicating a moderate fit. In contrast, the pseudo-second-order kinetic model, shown by the linear plot of t/Qt versus time, exhibited an excellent linear correlation with an R² value of 0.998. This significantly higher correlation coefficient suggests that the adsorption of MB onto g-C₃N₄ follows the pseudo-second-order kinetic model more accurately. The values of the correlation coefficients (R²) and kinetic rate constants (k1 and k2) for each model are summarized in **Table 1**.

The conformity to the pseudo-second-order model indicates that the adsorption process is highly dependent on the availability and utilization of active sites on the g-C3N4 surface. The primary active sites for methylene blue adsorption on g-C3N4 are the nitrogen-based functional groups (NH2) and the carbon sites in heptazine rings, which facilitate π–π stacking and electrostatic interactions **(Figure 8d)** [22]. These active sites play a crucial role in capturing dye molecules prior to photocatalytic degradation. Efficient adsorption on these sites ensures that the dye molecules are positioned close to the surface, allowing more effective interaction with photogenerated reactive species such as electrons, holes, or radicals under light irradiation. Therefore, the observed pseudo-second-order kinetics support the notion that g-C3N4 provides abundant and reactive surface sites, which significantly enhance the adsorption step and subsequently improve the overall photocatalytic efficiency.

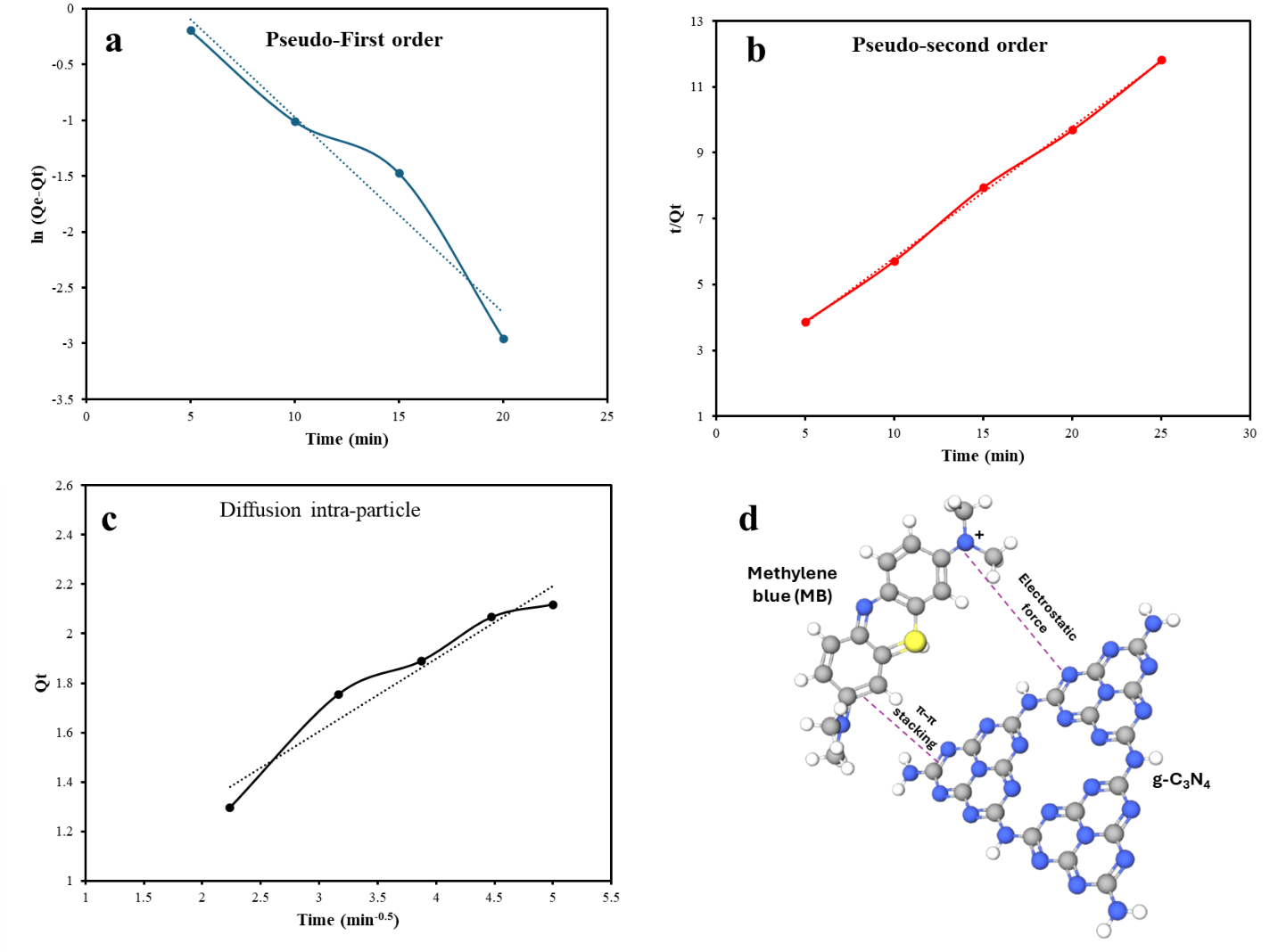


Figure 8. Pseudo-first order (a) pseudo-second order graphic (b) diffusion intra-particle (c) interaction of g-C3N4 with MB (d)

The intra-particle diffusion model was used to analyze the adsorption mechanism of methylene blue (MB) onto g-C3N4. The plot of Qt versus t0.5 shows a good linearity with an R² value of 0.9434. The relatively high correlation coefficient indicates that intra-particle diffusion contributes to the adsorption process. The adsorption of MB onto g-C3N4 following the intra-particle diffusion model suggests that the process involves the transport of MB molecules from the bulk solution into the internal pores or surface structure of g-C3N4 [23]. MB molecules, which are cationic and planar, can interact with the nitrogen-rich sites of g-C3N4 via electrostatic attraction and π–π interactions [22]. These interactions, combined with the gradual diffusion into g-C3N4 porous matrix, support the applicability of the intra-particle diffusion model in describing the adsorption behavior of MB on g-C3N4.

Table 1. Properties of kinetic adsorption process

|  |  |  |
| --- | --- | --- |
| **Kinetic Model** | **R2** | **k (rate constant)** |
| Pseudo-first order | 0.949 | 0.175 (min-1) |
| Pseudo-second order | 0.998 | 8.4 (g/mg.min) |
| Diffusion Intra-particle | 0.943 | 0.293 (mg/g. min0.5) |

**Figure 10** shows the kinetic modelling results for methylene blue degradation under UV and daylight illumination, evaluated using both first order and second-order reaction models. In **Figure 10a**, the first-order model demonstrates a stronger correlation with the experimental data, as indicated by higher R² values of 0.9725 for daylight and 0.7275 for UV, compared to the second-order model in **Figure 10b**, which yields R² values of 0.969 for daylight and 0.7117 for UV. The strong linearity observed in the first-order plots indicates that the degradation process is better described by first-order kinetics, especially under daylight. In a first-order reaction, the degradation rate is directly dependent on the concentration of methylene blue; thus, the reaction proceeds faster at higher concentrations and slows as the dye is consumed [24],[25]. This pattern suggests that dye concentration is the main limiting factor, while the photocatalyst is present in excess. This behavior typically reflects that the dye concentration limits the reaction rate, while the photocatalyst is present in sufficient quantities. Additionally, the rate constant under daylight (k = 0.0032 min⁻¹) is higher than that under UV (k = 0.0020 min⁻¹), confirming better photocatalytic performance under visible light. **Table 2** summarizes the kinetic parameters, including rate constants and R² values for both models. The degradation results also align with previous adsorption studies that followed pseudo-second-order kinetics, indicating that surface interactions and active site occupation are key factors in both the adsorption and photocatalytic processes.

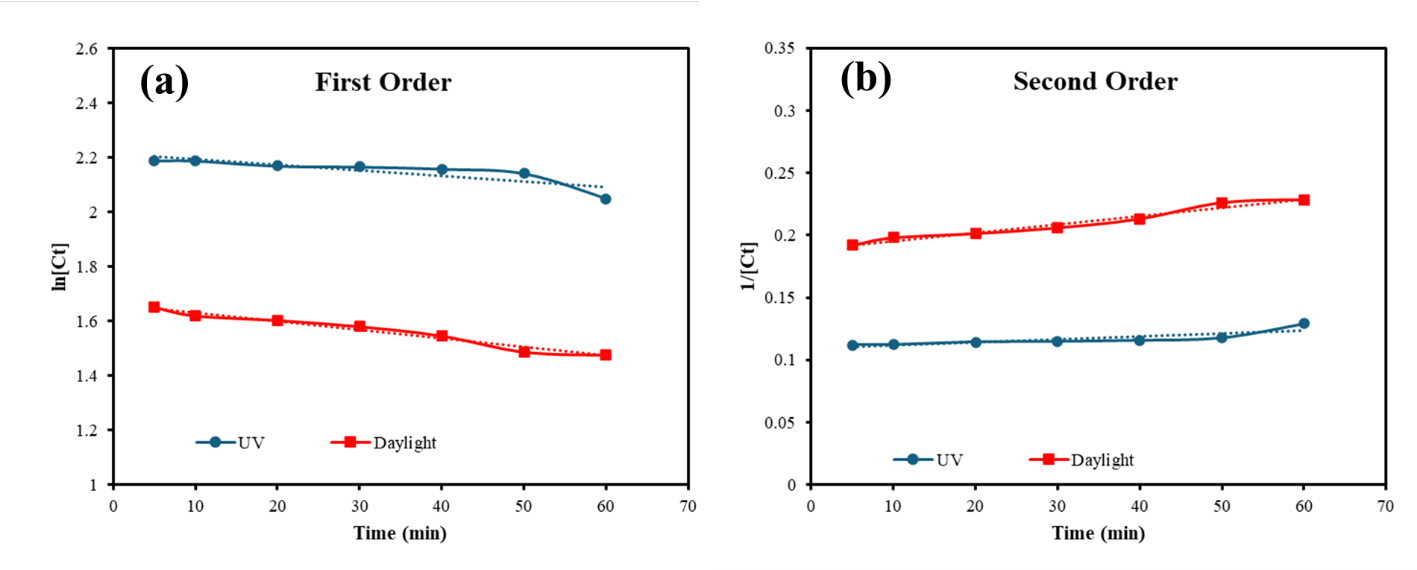


Figure 9. first order (a) and second order (b) of degradation photocatalytic kinetic

The combination of pseudo-second-order kinetics in the adsorption stage and first-order kinetics in the photocatalytic degradation stage offers significant advantages for an efficient dye removal process. The pseudo-second-order model indicates that adsorption is primarily controlled by the availability of active sites and involves chemisorption, suggesting a strong interaction between dye molecules and the surface of g-C3N4. This facilitates the effective accumulation of dye molecules near the photocatalyst's reactive sites, enhancing the readiness for subsequent degradation. In the photocatalytic stage, the first-order kinetic model implies that the degradation rate is directly proportional to the concentration of the dye, which is favorable at the lower concentrations typically remaining after the adsorption step. This kinetic behavior supports a continuous and effective breakdown of pollutants without saturation effects. Together, this sequential mechanism maximizes the utilization of surface-active sites and light-driven degradation efficiency, making the system highly suitable for practical applications in environmental remediation.

Table 2. Properties of photocatalytic process

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  | **First order** | | **Second order** | |
|  | **K1 (min-1)** | **R2** | **K2 (g/mg.min)** | **R2** |
| UV | 2 x 10-3 | 0.728 | 2 x 10-4 | 0.712 |
| Daylight | 3.2 x 10-3 | 0.973 | 7 x 10-4 | 0.969 |

# Photocatalys mechanism

**Figure 11** illustrates a predicted photocatalyst under visible light irradiation. Based on the Tauc plot, the band gap of g-C3N4 is 2.98 eV. When g-C3N4 absorbs photons with energy equal to or greater than its bandgap (~2.7 eV), electrons are excited from the valence band to the conduction band, leaving behind holes in the valence band. These photogenerated charge carriers migrate to the catalyst surface, where electrons reduce dissolved oxygen to form reactive superoxide radicals (•O2-). At the same time, the valence band holes can either oxidize MB directly or react with water to produce hydroxyl radicals (•OH). The combined activity of these reactive oxygen species, along with the strong oxidative capacity of the holes, drives the sequential degradation of MB via N-demethylation, breakdown of aromatic structures, and ultimate mineralization into CO2, H2O, and inorganic ions [26].

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Figure 10. g-C3N4 photocatalyst mechanism

# CONCLUSION

The removal of methylene blue using g-C3N4 achieved efficiencies of 56.20% under daylight and 22.46% under UV light, indicating a significantly enhanced performance under visible light. This observation is strongly supported by the kinetic analysis, which shows a higher rate constant and stronger correlation with the first-order kinetic model under daylight conditions. Effective photocatalytic activity in daylight can be attributed to the bandgap of g-C3N4 (2.98 eV), which allows efficient absorption of visible photos for dye degradation. The adsorption process followed a pseudo-second-order kinetic model, indicating that the uptake of MB was primarily controlled by surface interactions and the availability of active sites on the g-C3N4 surface. This was followed by a photocatalytic degradation step, which conformed to a first-order kinetic model, suggesting that the degradation rate depended on the concentration of MB and was more efficient when the dye concentration was higher. The consistency between the adsorption and photocatalytic stages highlights that surface interactions and active site utilization are key factors influencing the overall efficiency of MB removal. These findings confirm that g-C3N4 is an effective and visible-light-responsive material for dye remediation through photocatalytic mechanisms.

# Acknowledgments

The author gratefully acknowledges the supported funding from the Ministry of Higher Education, Sciences and Technology of Republic Indonesia through Penelitian Fundamental Reguler under contract no: 1316/PKS/ITS/2025 and the financial support from the Institut Teknologi Sepuluh Nopember for this work, under project scheme of the Publication Writing and IPR Incentive Program (PPHKI) 2025.

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