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EDITORS

Pn. Rosliza Ali
Pn. Nunshaimah Salleh
Pn. Norsakina Zurina Zulkifli
Pn. Adibatul Husna Fadzil
Pn. Yanti Yaacob
Pn. Lili Widarti Zainuddin

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Preface

The Scientific Project Colloquium offers a platform for publishing Diploma Science final year projects (FYP). The objective is to effectively distribute research findings throughout all scientific disciplines. The primary objective of including final year projects into the course curriculum is to encourage students to put their theoretical knowledge into practical applications.

We would like to express our gratitude to our primary establishment, the Faculty of Applied Sciences and Universiti Teknologi MARA, Perak Branch, for their invaluable assistance.

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Editors

Rosliza Ali
Nunshaimah Salleh
Norsakina Zurina Zulkifli
Adibatul Husna Fadzil
Yanti Yaacob
Lili Widarti Zainuddin

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SORPTION ENERGIES ESTIMATION USING DUBININ-RADUSHKEVICH AND TEMKIN ADSORPTION ISOTHERMS FOR g-C_3N_4/Fe_3O_4 COMPOSITE

[1,2*]Nurul Izza Taib, [1,2,3]Rozaina Saleh, [1]Aunie Aqelah Fisol, [1]Isma Iwana Ismail, [1]Nur Najieha Ismail

[1]Faculty of Applied Sciences, Universiti Teknologi MARA Perak Branch, Tapah Campus, 35400 Tapah Road, Perak, Malaysia
[2]Advanced Materials for Environmental Research Initiative Group, Universiti Teknologi MARA Perak Branch, Tapah Campus, 35400 Tapah Road, Perak, Malaysia
[3]Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

*izza257@uitm.edu.my

Abstract: This study investigates the efficacy of utilizing a composite of g-C_3N_4/Fe_3O_4 as an adsorbent for the removal of methylene blue (MB) from aqueous solutions. The sorption characteristics were analyzed using the Dubinin-Radushkevich and Temkin isotherm models. The findings indicate that the sorption of MB onto the composite is effectively described by the Dubinin-Radushkevich isotherm, exhibiting a high correlation coefficient (R^2 = 0.98). The calculated mean adsorption energy (β) was determined to be 0.0015 mol²/J², and the mean free energy (E) was found to be 0.586 kJ/mol, suggesting that the adsorption mechanism is predominantly physisorption. Furthermore, the maximum adsorption capacity (q_D) was estimated to be 40.14 mg/g.

Keywords: *g-C_3N_4, iron oxide nanoparticles, Dubinin-Radushkevich, Temkin, sorption energies*

INTRODUCTION

The discharge of dyes from various industrial activities has raised significant environmental concerns due to their persistence in the environment and potential to enter the food chain, posing threats to public health even at low concentrations. The textile industry significantly contributes to dye wastewater, representing approximately two-thirds of the total dye waste generated on a global scale (Mittal et al., 2009; Ryu et al., 2023). Each year, approximately 300,000 tons of textile dyes are discharged into wastewater, leading to adverse effects on aquatic ecosystems and human health due to their toxic properties (Ogugbue & Sawidis, 2011; Zong et al., 2024).

Methylene blue (MB), a widely used cationic dye, has been associated with various serious health issues such as increased heart rate, vomiting, shock, cyanosis, and jaundice, which are critical indicators of toxicity (Cai et al., 2020; Ilias et al., 2024). Moreover, methylene blue toxicity has implications beyond immediate health effects; chronic exposure can lead to tissue necrosis and hemolytic anemia (Xiang et al., 2019). The persistence of the dye in the environment and its ability to bioaccumulate in aquatic systems complicate the health risks associated with methylene blue. Effective removal strategies from wastewater are necessary to mitigate these dangers.

The removal of heavy metals from wastewater is critical, and various conventional technologies such as chemical coagulation, precipitation, flocculation, membrane separation, and ion exchange have been employed. However, these methods often come with limitations, including high operational costs and generating secondary waste (Aragaw & Bogale, 2021; Hung et al., 2020). The adsorption method has attracted many researchers due to its simplicity, high efficiency, and cost-effectiveness, which is widely applied to most water treatment plants (Renu et al., 2017). The composite material g-C_3N_4/Fe_3O_4 has emerged as an effective adsorbent for the removal of dyes from aqueous solutions due to its unique structural and chemical properties. This composite has a high surface area and porosity, which facilitates the adsorption process. The interconnected porous networks in g-C_3N_4 composites enable rapid mass transfer of dye molecules, increasing its overall adsorption capacity (Pan et al., 2022). The incorporation of iron oxide (Fe_3O_4) in the composite further enhances its magnetic properties, facilitating the facile separation of the adsorbent from the solution after the adsorption process. This feature significantly enhances process efficiency and cost-effectiveness (Azali et al., 2023). The main objective of the study was to investigate the feasibility of using g-C_3N_4/Fe_3O_4 as an adsorbent for the removal of MB at different initial concentrations and to assess the sorption energies using Dubinin-Radushkevich and Temkin isotherms.

METHODOLOGY

The g-C_3N_4 was synthesized using the thermal polymerization method. A crucible with 10.0 g of urea was heated in a furnace to 550 ºC at a rate of 2 ºC/min for 3 hours. After cooling at room temperature, the yellow product was ground into a fine powder and stored for further use. The g-C_3N_4/Fe_3O_4 composite was synthesized using a mass ratio 2:1 of g-C_3N_4 to Fe_3O_4. In brief, 0.8 g-C_3N_4 was dispersed in a mixture of 60 ml of ethanol and 120 ml of water in a round bottom flask. The yellow suspension was sonicated for 30 minutes to achieve a homogeneous solution. Subsequently, 0.9340 g of $FeCl_3.6H_2O$ and 0.3436 g of $FeCl_2.4H_2O$ were dissolved with 100 ml of distilled water in a beaker. Then, the solution was added to a round bottom flask, which formed a brown suspension. Then, the suspension was stirred for 30 minutes at 80 ºC. Following this, 10 ml of 30% NH_3 was introduced drop by drop to the mixture under a vigorous stirring. After 60 minutes of stirring, a dark brown g-C_3N_4/Fe_3O_4 composite suspension was obtained. After cooling at room temperature, the mixture was washed twice with water and ethanol before being magnetically separated. The dark brown precipitate was obtained and dried in an oven for 12 hours at 70 ºC. Adsorption isotherms were performed at room temperature using 40 mg of g-C_3N_4/Fe_3O_4 with initial concentrations ranging from 0 mg/L to 300 mg/L of 50 mL of MB solutions. The mixture was agitated in an orbital shaker at 100 rpm for 1 hour at 298 K. The reaction mixture was filtered using filter paper to separate the supernatant, and the concentration of methylene blue in the filtrate was measured with a UV-visible spectrophotometer (UV/Vis Spectrometer Lambda 25 Perkin Elmer). The percentage removal (%) was calculated using Equation (1):

$$\text{Removal (\%)} = \frac{(C_o - C_e)}{C_o}\ x\ 100 \quad (1)$$

where C_0(mg/L) is the initial MB concentration, and C_e(mg/L) is the final MB concentration.

FINDINGS

To assess the sorption energies associated with the interaction between g-C_3N_4/Fe_3O_4 and MB, the adsorption equilibrium data were analyzed by fitting the Dubinin-Radushkevich and Temkin isotherm models. The Dubinin–Radushkevich (D–R) isotherm has been utilized to differentiate between the physical and chemical adsorption of metal ions. It was postulated based on the mean free energy *E* per molecule of the adsorbent. The corresponding equation is provided as follows (Husein et al., 2019):

$$ln\ ln\ q_e = ln\ ln\ q_D - \beta\left[RT\ ln\ ln\ \left(1 + \frac{1}{C_e}\right)\right]^2 \quad (2)$$

The Temkin isotherm postulates that the heat of adsorption for all molecules within the adsorbent layer decreases linearly with sorption coverage as a result of the indirect interaction between the absorbent and the absorbate ((Dubinin, 1947). The isotherm can be represented in its linear form as follows:

$$q_e = B\ ln\ ln\ A_T + B\ ln\ ln\ C_e \quad (3)$$

where $B = \frac{RT}{b_T}$, q_e = equilibrium Cu(II) concentration (mg/g), A_T = Temkin equilibrium binding constant represents the maximum bonding energy (L/mg), *B* = Temkin constant associated with the heat of sorption (J/mol), b_T = constant of Temkin isotherm, q_D = maximum adsorption capacity (mg/g), β = constant of Dubinin-Radushkevich associated with the mean adsorption energy (mol^2/J^2), *R* = universal gas constant (8.314 J/mol K), *T* = temperature at 298.15 K. The mean free energy of adsorption *E* (kJ/mol) was calculated using:

$$E = \frac{1}{\sqrt{2\beta}} \quad (4)$$

In reference to the D-R plot in Figure 1, the parameter values were calculated as follows: q_D = 40.14 mg/g, $\beta =$ 0.0015 mol^2/J^2 with the R^2 = 0.98. The calculated energy value based on this model (0.586 kJ/mol) was found to be significantly lower than 8 kJ/mol. This isotherm postulates the multilayer adsorption of MB onto g-C_3N_4/Fe_3O_4.

This process typically involves weak van der Waals forces between adsorbates and adsorbents, indicative of physical adsorption.

In accordance with the Temkin plot (Figure 2), the parameter following values were estimated: A_T = 0.03437 L/mg, B = 4.78 J/mol and $b_T = 518.2$ with R^2 = 0.60. The Temkin parameter, A_T is 0.03437 L/mg, suggesting a low affinity of the MB for the g-C_3N_4/Fe_3O_4, indicating weak bonding between g-C_3N_4/Fe_3O_4 and MB. The value of B constant was less than 8 kJ/mol, which confirms that the interaction between g-C_3N_4/Fe_3O_4 and MB is weak, indicative of physical adsorption rather than chemical adsorption.

The adsorption energy is positive for MB adsorption from the aqueous solution, which indicates that the adsorption is exothermic. Meanwhile, b_T value is < 80, further suggesting that the adsorption process is primarily physical in nature and that the physical adsorption process is involved (Temkin & Pyzhev, 1940). The finding suggests that the Dubinin-Radushkevich model accurately describes the sorption energies of the interaction between g-C_3N_4/Fe_3O_4 based on its high regression coefficient value, R^2.

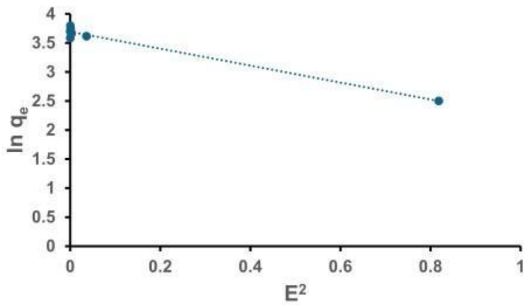

Figure 1. Dubinin-Radushkevich plot for the adsorption of MB onto g-C_3N_4/Fe_3O_4 composite

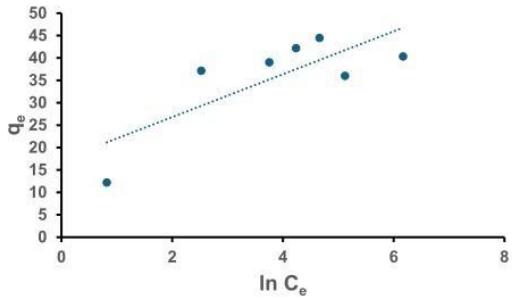

Figure 2. Temkin plot for the adsorption of MB onto g-C_3N_4/Fe_3O_4 composite

CONCLUSIONS

The sorption energy data for MB adsorption on g-C_3N_4/Fe_3O_4 can be effectively represented by the Dubinin-Radushkevich model, which exhibited strong correlation coefficients for the studied concentrations. Sorption energy value (β), mean free energy (E), and heat of sorption (B) were estimated as 0.0015 mol^2/J^2, 0.586 kJ/mol, and 4.78 J/mol, respectively, which reveals the physisorption process dominates chemisorption and ion exchange.

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