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# Synthesis of Reduced Graphene Oxide Using Reducing Lime Juice (*Citrus aurantifolia*) and Its Application as Malachite Green Adsorbent in Aquatic Environments

Said Ali Akbar[1*], Hasby[2]

[1] Department of Aquaculture, FKP, Universitas Syiah Kuala, Banda Aceh, Aceh, Indonesia
[2] Department of Chemistry Education, FKIP, Universitas Samudra, Langsa, Aceh, Indonesia.

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Corresponding Author:
Said Ali Akbar
saidaliakbar@usk.ac.id

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**Abstract:** The cytotoxicity of fishes resulting from the presence of various industrial dyes in industrial effluents is a significant global concern. This study aims to synthesize Reduced Graphene Oxide (RGO) using a natural reducing agent from lime juice. The synthesis process uses a modified Hummer method. The Langmuir and Freundlich Isothermal equations are used to study the adsorption character of MG using RGO. The adsorption kinetics was studied using first and second order equations. The adsorption capacity was measured with concentration 2, 10, 50, and 100 mg $L^{-1}$ of MG respectively. The results obtained are for the adsorption isotherms following Langmuir and Freundlich. ΔG° shows a negative value which means the adsorption process is spontaneous. The adsorption kinetics follows the first order equation. The adsorption capacity obtained was $Q_m$ = 223.21 mg $g^{-1}$ and a contact time of 20 minutes which made it a strong adsorbent for removing MG from water samples. The removal efficiency of MG by prepared adsorbents from real environmental water sample on the Ulee Lheue beach, Alue Naga beach, and the Krueng Aceh river has been implemented. The results obtained confirmed good work in the applicability of adsorbents for environmental.

**Keywords:** Citrus aurantifolia; Lime Juice; Malachite Green; Reduced Graphene Oxide

## Introduction

The contamination water by textile dyes is a global problem. By way of gill respiration, these dyes come into intimate contact with fish. Fish are susceptible to a wide variety of toxic substances (Pipoyan et al., 2020). The presence of dyes in the water sample prevents light from entering the water for plants, and therefore, with the disruption of the photosynthesis process, the dissolved oxygen concentration required for living organisms is drastically reduced (Amiri-Hosseini & Hashempour, 2021). A comparative toxicological study of textile dye wastewater (untreated and treated) on freshwater fish, *Gambusia affinis* showed reduced mortality and a marked cytotoxic effect on red blood cells and also found a decrease in the number and percent of deformation (poikilocytosis) and their variation. size. Another study on *Mastacembelus armatus* which is an edible proteinaceous freshwater fish exposed to textile waste caused changes in ion regulation of tissues such as liver, kidney and muscle with decreased concentrations of sodium and chloride ions and increased concentrations of potassium, calcium ion and magnesium. Then, a study was also conducted on the impact of textile dyeing industry waste on several hematological parameters of freshwater fish Oreochromis mossambicus. The main histological changes observed in the liver are hyperemia, necrosis and degeneration (Teymori et al., 2020).

Malachite green (MG) is a dye with the cationic category, which has wide applications in textiles. In addition, because MG has a low price, through its antibacterial properties it is widely used as a fungicide in the aquaculture industry as well as the treatment of Saprolegnia in fish. Long-term exposure can cause skin

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sensitization or even skin cancer (Pathy et al., 2022). Therefore, taking into account the health risks and hazards posed by these chemicals, an innovation is needed that can be used to remove the presence of these dyes from water samples. Among the various methods developed to overcome this problem, adsorption is one of the easiest techniques to use because of its advantages, namely, the availability of a wide variety of adsorbents, high efficiency, and low cost.

Some adsorbents such as modified graphene oxide (Ghahramani et al., 2018), magnetically reduced graphene oxide (Liu et al., 2015), activated carbon (Ahmad et al., 2021), carbon nanotubes (Luo & Li, 2022), and Natural zeolites (Radoor et al., 2021) are widely used as MG adsorbents. Graphene oxide (GO) is the best adsorbent candidate due to its high surface area, abundant availability, good electrical conductivity, low cost, and the ability to modify its surface via the available functional groups. GO is synthesized through the oxidation of graphite, in this process exfoliation of graphite oxide occurs (Chen et al., 2020). Reduced Graphene Oxide (RGO) can be synthesized using GO. RGO has better material properties than GO (Rout et al., 2021). The transformation of GO into RGO can be carried out through a chemical reduction process (Söğüt et al., 2020).

RGO is the best innovation of carbon-derived materials which can be adsorbents to remove heavy metals and dyes in wastewater (Khan et al., 2014). This is supported by the high surface area of RGO reaching 2620 $m^2/g$ and easily obtained through natural graphite raw materials, so that it can be made on a large scale (Razaq et al., 2022; Tingshun et al., 2015). Currently the chemical reduction method is the easiest and best step in making RGO in large quantities and for commercial applications. Among several reducing agents that have been used have been reported such as Zn metal, Fe metal, Sn metal, $NaBH_4$, and $LiAlH_4$. All of these reducing agents are used to reduce GO to RGO (Söğüt et al., 2020). However, all of these reagents are classified as toxic chemicals, so the waste products from synthesis will become environmental pollution if not managed properly (Lopez et al., 2017). Therefore, another reducing agent that is environmentally friendly is needed for the RGO manufacturing process. Several green reducing agents that are easy, inexpensive, and environmentally friendly have been reported, such as *Psidium guajava*, *Eucalyptus*, Vitamin C, and Thiourea dioxide, (Silva et al. 2018).

Lime is a plant with fruit that is small and tastes sour. Fruit production is very fast and very fertile when planted in the lowlands. Lime (*Citrus aurantifolia*) contains a citric acid-like chemical compound. This compound serves as a natural preservative, a cleaning agent, and a sour flavour enhancer in foods and beverages. Various fruits and vegetables contain citric acid, but only limes contain concentrations of up to 8% dry weight (Lin et al., 2019). Therefore, lime is a potential reducing agent for the synthesis of RGO. This innovation can reduce the cost of RGO synthesis, which is costly and environmentally hazardous.

## Method

This research is an experimental research conducted at the Chemistry Education Laboratory at Syiah Kuala University. This research was conducted to determine the adsorption characteristics of RGO which was synthesized using a natural reducing agent from lime juice. The research flow chart is presented in Figure 1.

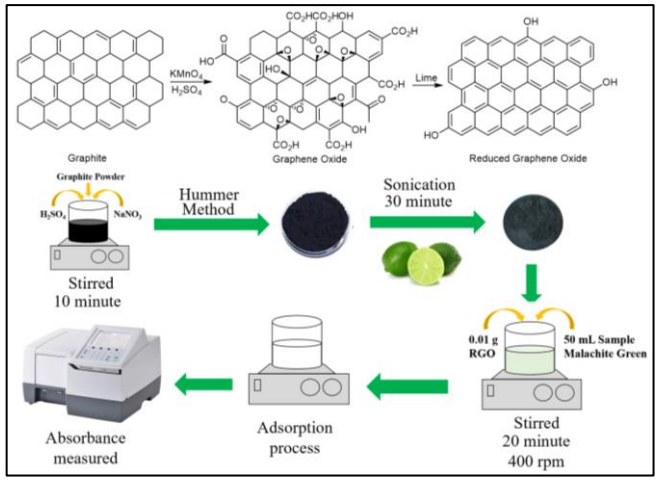


**Figure 1.** Schematic flow chart of this research

*Tools and materials*

Graphite powder, 97% $H_2SO_4$, 37% HCl, $NaNO_3$, $KMnO_4$, 37% $H_2O_2$, Deionized water, lime, and Malachite Green (in the form of chloride salt).

*Synthesis of Graphene Oxide*

A modified Hummer method is utilised to prepare GO (Agharkar et al., 2014). 1 g of graphite powder and 23 mL of $H_2SO_4$ solution were added to a 500 mL glass, which was then stirred in an ice bath. After 10 minutes of stirring, 0.5 g $NaNO_3$ was then added. Then, 3 g of $KMnO_4$ was added slowly while stirring the solution for 48 minutes. Lastly, 3 mL of an $H_2O_2$ solution containing 30% was added to the mixture. The obtained GO was filtered and purified via acetone rinsing. In addition, the obtained GO was oven-dried at 60°C for 60 minutes.

*Synthesis of Reduced Graphene Oxide (RGO)*

The lime juice is diluted with 200 mL of distilled water in a container. The solution's concentration is then adjusted as desired. The solution was then sonicated for

30 minutes while 0.1 g GO was added. Observe this process until a uniformly dispersed brown solution forms in the solution. Then the solution was centrifuged for 30 minutes at 4000 rpm, then the two solutions were put into a microwave oven with a power of 800 W for 1 minute. The mixture is filtered, then rinsed with acetone. Furthermore, the filtered RGO was dried at 60°C for 60 minutes using an oven.

*Characterization of Synthesized Products*

X-ray Diffraction (XRD) testing was carried out to determine the crystal character of Graphite, GO, and RGO. The diffraction pattern was obtained using a Rigaku-Denki diffractometer with $K_\alpha$ : 0.154 nm Cu-sealed tube. Then, the character of the functional groups found in Graphite, GO, and RGO were studied with Infrared (IR) spectra obtained with Bruker Alpha FTIR, with a wave number range of 550 - 4000 $cm^{-1}$. Lastly is the solubility test on each Graphite, GO, and RGO, solubility is tested using water and ethanol solvents.

*Adsorption Procedure*

A total of 50 mL of sample solution was prepared with various concentrations of MG, namely 2, 10, 50, and 100 mg $L^{-1}$. Into each sample was added 0.01 g of RGO. The mixture was stirred for 20 minutes at 400 rpm. This process was carried out at room temperature at room temperature (25 ± 2 °C). The absorbance of the remaining MG in solution was measured to obtain a standard calibration curve at 620 nm with UV-vis spectroscopy (Shimadzu UV-3150 spectroscopy) (Rehman et al., 2021). Determination of the adsorption capacity of RGO according to Equation 1:

$$q_e = \frac{(C_o - C_e)V}{W} \quad (1)$$

where $C_o$ is the initial concentration of MG, W is the amount of RGO (g), $q_e$ is the adsorption capacity (mg $g^{-1}$), V is the volume of sample solution (L), and $C_e$ is the equilibrium concentration of MG (mg $L^{-1}$).

*Real Water Sample*

The actual sample was used to study the efficacy of the adsorbent for MG removal. Original water samples were collected from the waters on the Ulee Lheue beach, Alue Naga beach, and the Krueng Aceh river. A total of 0.01 g of RGO was mixed in a 50 mL plastic vial with 50 mL of water sample at maximum conditions, the amount adsorbed was calculated by comparing the concentration of MG before and after adsorption. The mixture was stirred for 20 minutes at 400 rpm.

## Result and Discussion

*Characterization of Synthesized RGO*

*X-Ray Diffraction (XRD)*

Figure 2 shows the X-ray diffraction patterns of graphite, GO and RGO. At 2θ = 26.4° a graphite diffraction peak was found with a d-spacing of 0.34 nm. Nevertheless, the graphite diffraction pattern reveals a greater d-spacing distance than graphite, specifically at 2θ = 10° with a d-spacing distance of 0.88 nm.. It can be concluded that the d-spacing distance of GO is far due to the insertion of functional groups containing oxygen (Kyzas et al., 2018). Then another characteristic of GO is the emergence of turbostratic interference at 2θ = 43°. In contrast to RGO, GO displays a diffraction peak of 2θ = 23.8° with a d-spacing of 0.37 nm, indicating that the decrease in d-spacing is the result of GO's reduced functional groups. (Al-Gaashani et al., 2019).

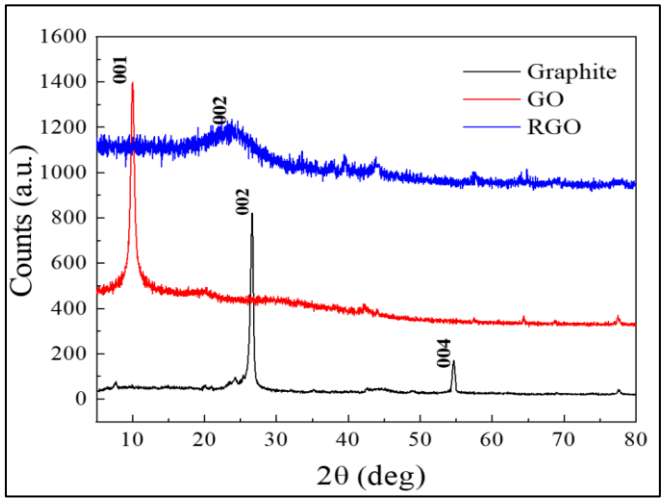


**Figure 2.** X-ray diffraction patterns on graphite, GO, and RGO samples

Figure 2 depicts RGO as having a more amorphous appearance than graphite and GO. RGO lacks the sharp peaks observed in graphite and GO. Therefore, RGO has a low level of crystallinity (Liu et al., 2016).

*Infrared Spectrum*

The results of the IR measurements on the three samples are presented in Figure 3. The vibrational peak at 3412 $cm^{-1}$ is the vibrational mode of the hydroxyl group. Then at 1723 $cm^{-1}$ is the vibrational character of carbonyl (C=O). Then the vibrations at 1178 and 1073 $cm^{-1}$ are vibrational modes of the epoxide group with C—O bonds (Lopez et al., 2017). The high intensity of the main vibrational peaks in GO indicates that many groups containing O atoms appear during the graphite oxide process. After the reduction of GO to RGO, the vibration peak at 1620 $cm^{-1}$ increased in RGO. This peak

is the character of C=C, this bond is formed from the process of reducing GO to RGO (Liu et al., 2019).

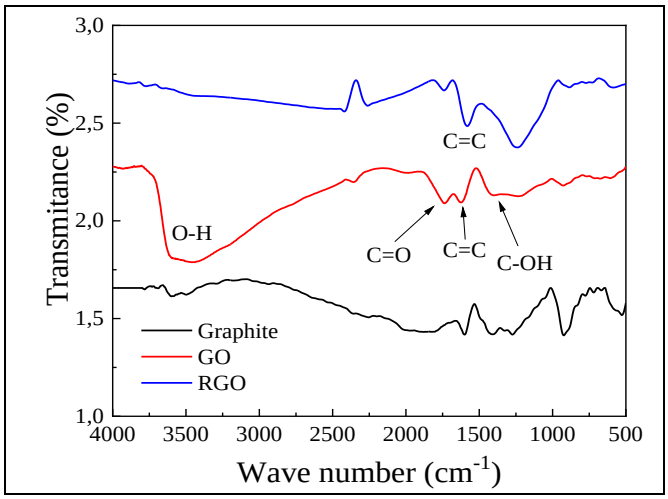


**Figure 3.** FTIR spectra of graphite, GO, and RGO samples

*Solubility*

The results of the solubility test are presented in Figure 4. The test results show that GO is more soluble in water than in ethanol. This is consistent with the results in the IR spectrum that GO has many functional groups containing O atoms. However, RGO is slightly soluble in ethanol as a result of some of the functional groups in GO that have been reduced. This causes RGO to lack groups containing O atoms. In addition, graphite has a low solubility in ethanol compared to water due to the presence of numerous polar functional groups (Kyzas et al., 2018).

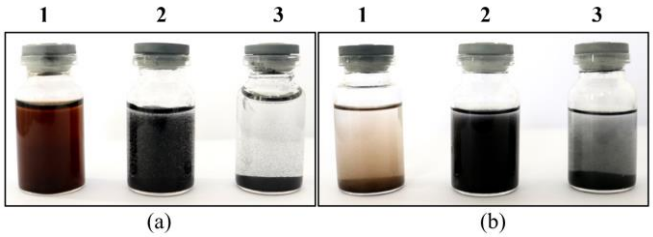


**Figure 4.** Dissolving samples sequentially, namely (1) GO, (2) RGO and (3) graphite in a) water and b) ethanol

*Effect of contact time and initial concentration of Malachite Green*

Figure 5 depicts the effect of contact time on the MG adsorption capacity. Based on this observation, the maximum MG adsorption capacity was reached in 20 minutes and remained constant thereafter. In addition, an increase in MG adsorption capacity indicates that RGO is a rapid adsorbent for MG removal from water samples; in this case, an increase in MG concentration can explain why the diffusion process of MG from solution to the RGO surface increases.

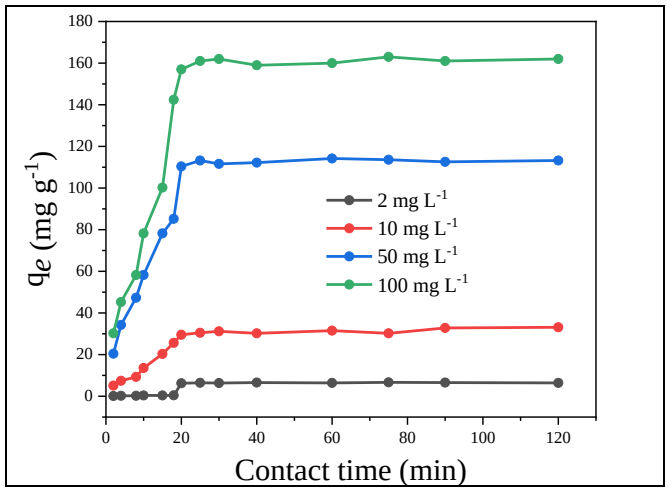


**Figure 5.** The influence of contact time and initial MG concentration on the adsorption capacity of RGO

*Adsorption Isotherm*

This study employed two isothermal models, Langmuir and Freundlich, to comprehend the adsorption behaviour of MG on RGO.

*Langmuir and Freundlich*

From this equation by plotting the curve between $C_e/q_e$ vs Ce, which of the slopes and intersections of the resulting plots (Table 1), can be used to determine the value of $K_{ads}$ (Langmuir constant, mg $L^{-1}$) and Qmax (maximum adsorption capacity theoretical, mg $g^{-1}$). The calculation obtained by $Q_{max}$ is 223.21 mg $g^{-1}$, therefore RGO can be used as an MG adsorbent in water samples efficiently and well.

$$\frac{C_e}{q_e} = \frac{C_e}{Q_{max}} + \frac{K_{ads}}{Q_{max}} \quad (2)$$

**Table 1.** Isothermal parameters for the MG adsorption on RGO

| Model | Parameter | Value |
|---|---|---|
| Langmuir | $Q_m$ (mg $g^{-1}$) | 223.21 |
| | $K_{ads}$ | 88.03 |
| | $R^2$ | 0.963 |
| Freundlich | $1/n$ | 0.72 |
| | $K_f$ | 8.81 |
| | $R^2$ | 0.984 |

The equation 3 is a linear form of the Freundlich equation:

$$\log q_e = \log K_f + \frac{1}{n} log C_e \quad (3)$$

From this equation, by plotting the curve between $\log q_e$ and $\log C_e$, which of the slopes and intersections of the resulting plots (Table 1), the values of $K_f$ and $1/n$ will

be obtained. $K_f$ is the Freundlich constant while 1/n is the favorability index. The results obtained are presented in table 2. From the two isothermal equation models, an $R^2$ of 0.963 is obtained for Langmuir and 0.984 for Freundlich. Therefore the adsorption of MG on RGO is suitable using the two isothermal models (Figure 6).

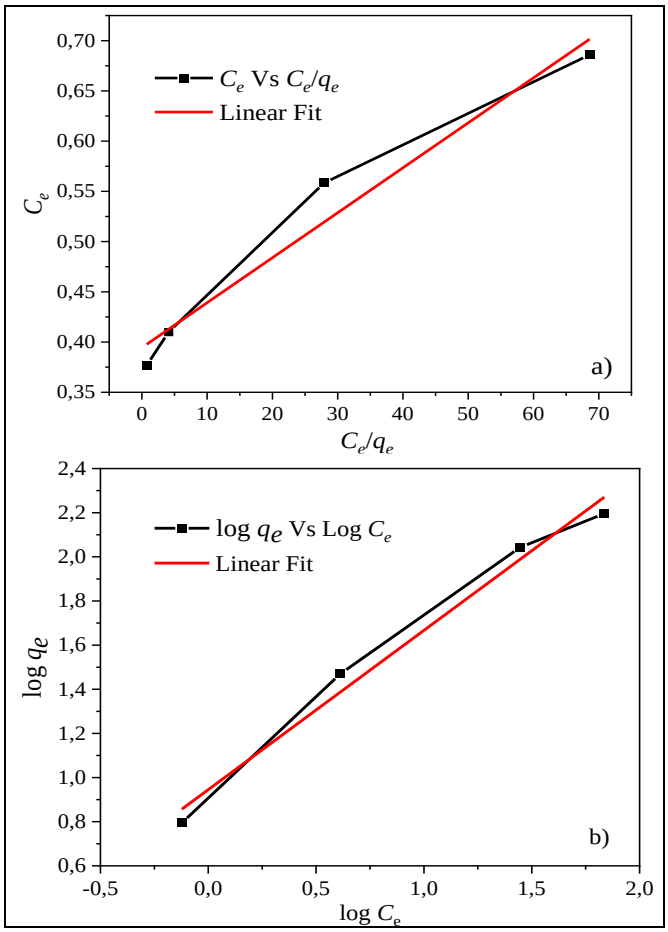


**Figure 6.** Fitting curve of a) Langmuir Isothermal and b) Freundlich Isothermal

*Kinetic model for adsorption of MG with RGO*

*First order equations*

The first order kinetic model is shown in equation 4 (Rout et al., 2021):

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (4)$$

$$log\left(\frac{q_e}{q_e - q_t}\right) = \frac{k_1}{2{,}303} \quad (5)$$

$$log(q_e - q_t) = log\ q_e - \frac{k_1}{2{,}303}t \quad (6)$$

The linear form of equation 4 is shown in equations 5 and 6, where $k_1$ is the value of the rate constant with units in $min^{-1}$. The values of $k_1$ and qe can be determined from the slopes and intersections of the lines via the log $(q_e - q_t)$ vs $t$ plots for MG (Figure 7). Table 2 shows the results of $R^2$, $k_1$ and qe obtained for the MG adsorption process on RGO. Based on the $R^2$ value obtained, the first order kinetic model is suitable to be applied to MG adsorption. This can be seen in the experimental qe values that are almost close to $q_e$ (*cal*) in Table 2. Therefore the kinetic model for MG adsorption follows the first order.

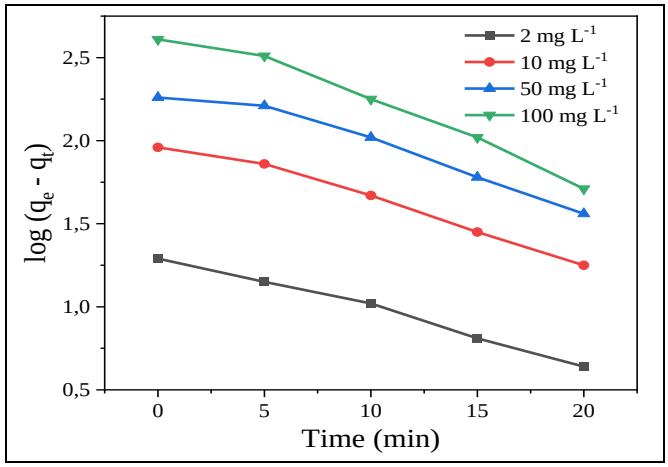


**Figure 7.** The adsorption of MG onto RGO follow first-order kinetics

**Table 2.** Parameters of MG adsorption on RGO with first order kinetic model

| Concentration | First order kinetic model | | | |
|---|---|---|---|---|
| (mg $L^{-1}$) | $q_e$ (exp) | $q_e$ (cal) | $k_1$ | $R^2$ |
| 2 | 19.49 | 21.41 | 0.0328 | 0.992 |
| 10 | 91.20 | 100.92 | 0.0366 | 0.985 |
| 50 | 181.97 | 214.70 | 0.0366 | 0.981 |
| 100 | 407.38 | 476.43 | 0.0498 | 0.976 |

*Second order equations*

The first order kinetic model is shown in equation 7 (Rout et al., 2021):

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (7)$$

The linear form of equation 7 is shown in equations 8 :

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}t \quad (8)$$

h is the initial absorption rate in mg $g^{-1}$ $min^{-1}$ as $k_2qe^2$, then $k_2$ is the rate constant in g $mg^{-1}$ $min^{-1}$. the values of $q_e$ and $h$ can be obtained from the slopes and intersections of the plots between $t/q_t$ vs t (Figure 8)

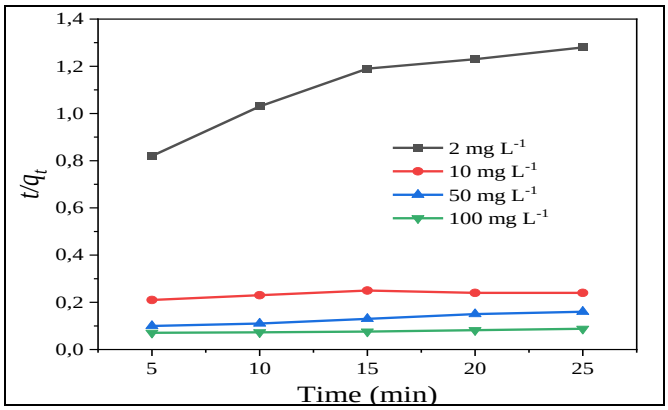


**Figure 8.** The adsorption of MG onto RGO follow second-order kinetics

**Table 3.** Parameters of MG adsorption on RGO with second order kinetic model

| Concentration (mg L-1) | | Second order kinetic model | | | |
|---|---|---|---|---|---|
| | $q_e$ (exp) | $q_e$ (cal) | $k_2$ | $h$ | $R^2$ |
| 2 | 19.49 | 22.54 | 3.3 x 10-3 | 1.29 | 0.894 |
| 10 | 91.20 | 104.88 | 5.6 x 10-4 | 4.69 | 0.532 |
| 50 | 181.97 | 549.02 | 3.65 x 10-5 | 1.21 | 0.984 |
| 100 | 407.38 | 427 | 9.25 x 10-5 | 15.36 | 0.953 |

The results of $R^2$ in the second order kinetic equation, obtained a value range of 0.532–0.95 (Table 3). In addition, the $q_e$ value (calculation) does not approach the experimental $q_e$ value (Table 3). Consequently, the adsorption kinetics of MG on RGO cannot be predicted using a second-order kinetics model (it is unsuitable).

*Adsorption thermodynamics*

To determine the effect of temperature on MG adsorption, the thermodynamic parameters standard Gibbs free energy change (ΔG°), standard enthalpy change (ΔH°), and standard entropy change (ΔS°) were calculated using Eq. 9–11. (Söğüt et al., 2020) :

$$Ln\ K_d = \frac{\Delta S^o}{R} - \frac{\Delta H^o}{RT} \quad (9)$$

$$K_d = \frac{C_o - C_e}{C_e} x \frac{V}{m} \quad (10)$$

$$\Delta G = \Delta H - T\Delta S \quad (11)$$

where $V$, $m$, $R$, $K_d$, $C_o$, $C_e$, dan $T$ are volume of sample solution in L, amount of adsorbent in g, universal gas constant (8.314 J mol$^{-1}$ K$^{-1}$), distribution coefficients, initial concentration, equilibrium MG, and temperature in K. Calculating ΔH° and ΔS° by plotting ln $K_d$ against $1/T$ and using the slope and intersection of the plots, respectively (Table 4). The outcomes demonstrate that ΔG° is negative, proving that MG adsorption to RGO occurs voluntarily. Further evidence that the adsorption process is more effective at higher temperatures comes from the fact that the value of ΔG° increases as temperature rises. Additionally, a positive value of ΔH° indicates that the process of MG adhering to RGO is endothermic.

**Table 4.** Thermodynamic parameters for MG adsorption on RGO

| $C_o$ (mg L-1) | ΔHo (kJ mol-1) | ΔSo (J mol-1 K-1) | ΔGo, 298,15 K (kJ mol-1) | ΔGo, 308,15 K (kJ mol-1) | ΔGo, 318,15 K (kJ mol-1) |
|---|---|---|---|---|---|
| 10 | 37.8021 | 142.9093 | -4.80633 | -6.23542 | -7.66451 |
| 50 | 37.46455 | 137.0895 | -3.4087 | -4.7796 | -6.15049 |

*Comparison with Other Adsorbents*

Table 5 displays the efficacy of the proposed RGO in removing MG. Although the adsorption capacity of the RGO synthesised in this study was lower than that of other adsorbents, the RGO synthesised in this study has the advantage that the new reducing agent used is natural, thereby reducing production costs. With an adsorption capacity of $Q_m$ = 223.221 mg g$^{-1}$ and a contact time of 20 minutes, it works well to remove MG from water samples..

**Table 5.** Comparison of the adsorption capacity ($Qm$) of MG with various adsorbents

| Adsorbent | Reductor | $Q_m$ (mg/g) | Reference |
|---|---|---|---|
| rGO–PANI | $NaBH_4$ | 666.7 mg g$^{-1}$ | Ghahramani et al., 2018 |
| α-$Fe_2O_3$–rGO | $FeCl_3 \cdot 6H_2O$ | 438.8 mg g$^{-1}$ | Liu et al., 2015 |
| GO-S-$La_2O_2(CO_3)$ | - | 555.5 mg g$^{-1}$ | Eftekhari et al., 2022 |
| GO/aminated lignin aerogels | - | 113.5 mg g$^{-1}$ | Chen et al., 2020 |
| ZIF-67@GO | - | 134.79 mg g$^{-1}$ | Rehman et al., 2021 |
| RGO/Zeolit imidazolat | Iron sulfate ($FeSO_4$) | 3000 mg g$^{-1}$ | Lin et al., 2016 |
| RGO | $NaBH_4$ | 1111.11 mg g$^{-1}$ | Söğüt et al., 2020 |
| RGO | Hidrazin monohidrate | 279.85 mg g$^{-1}$ | Rout et al., 2021 |
| RGO | Lime Juice | 223.21 mg g$^{-1}$ | This work |

*Malachite Green removal performance of RGO from a real water sample*

To investigate the applicability of the proposed dyes adsorbent, MG removal by prepared adsorbents from real environmental water samples was investigated. The water used in this measurement was taken from the sea and rivers, while the water parameters used are in accordance with Table 6.

**Table 6.** Parameters physics and chemistry of real water sample in Banda Aceh city

| Parameter | Sample Water Point | | |
|---|---|---|---|
| | Ulee Lheue Beach | Alue Naga Beach | Krueng Aceh River |
| Salinity (ppt) | 23,4 | 21.6 | 0.2 |
| $NH_3$ (mg.$L^{-1}$) | 0.06 | 0.08 | 0.15 |
| $PO_4^{3-}$ (mg.$L^{-1}$) | 0.134 | 0.262 | 0.108 |
| $NO_3^-$ (mg.$L^{-1}$) | 0.308 | 0.261 | 0.026 |
| pH | 7.81 | 7.96 | 8.4 |
| Temperature | 30.2 °C | 30.4 °C | 29 °C |

Figure 8 demonstrates that RGO green synthesis is highly effective at removing the majority of MG from actual water samples. The removal efficacy decreased to approximately 82.5% at Ulee Lheue beach, 84.5% at Alue Naga beach, and 92.4% in the Krueng Aceh river. This confirms the environmental applicability of adsorbents.

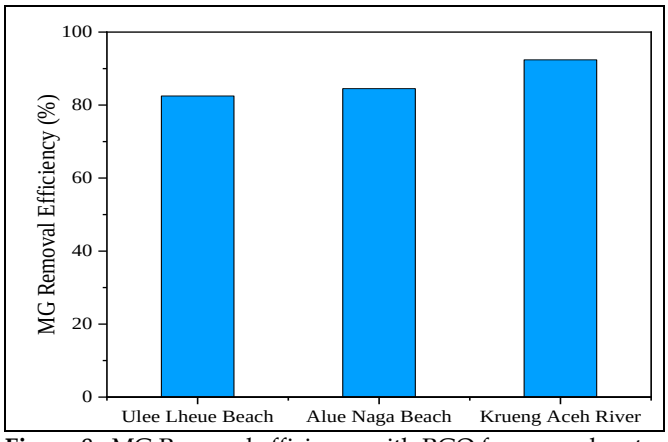


**Figure 8.** MG Removal efficiency with RGO from a real water sample.

## Conclusion

RGO was used in this study as a novel and effective adsorbent to remove MG from water samples. The Hummers method was used to synthesise GO, which was then reduced using a natural reducing agent derived from lime juice. Then ultrasonication is carried out to become RGO. The characterization of the synthesized product was used for XRD, IR Spectrum, and Solubility. From the characterization results it was found that the GO reduction product had correctly formed RGO. Both the Langmuir and the Freundlich adsorption isotherms are consistent with the MG adsorption data, as shown by the studied data. The first and second order equations were used to study the kinetics of RGO adsorption. After looking into the MG adsorption process, it was found to be in agreement with the first order kinetic equations. Thermodynamic investigations have shown that the adsorption of MG onto RGO is endothermic, and that the process is spontaneous if $\Delta G°$ is negative.. With an adsorption capacity of $Q_m$ = 223.221 mg $g^{-1}$ and a contact time of 20 minutes, this adsorbent is highly effective at removing MG from water samples. With the aid of adsorbents from actual environmental water samples, the removal of MG has been prepared. The outcome demonstrated the efficacy of adsorbents in their use as environmental adsorbents.

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