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Adsorption of Malachite Green Using Rice Husk-Based Adsorbents

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Adsorption, alginate, chitosan, Malachite Green, rice husk

Abstract

This study investigates the adsorption of Malachite Green using chitosan alginate rice husk beads and rice husk powder. The methodology involved adsorbent preparation, adsorption study, kinetic and isotherm models evaluation and adsorbent characterization. The selected malachite green adsorption conditions were 50% removal at 1.35 g for chitosan alginate rice husk beads and 0.02 g for rice husk powder, 60 minutes contact time and 5 mg/L initial concentration. Both adsorbents were fitted to the pseudo-second-order kinetic model rather than the pseudo-first-order kinetic model, indicating chemisorption is the rate-limiting step. Chitosan alginate rice husk beads were better fitted to the heterogeneous Freundlich isotherm model, while rice husk powder matched better with the homogeneous monolayer Langmuir isotherm model. Chitosan alginate beads showed lower adsorption than rice husk powder due to blocked functional groups. The FTIR characterization showed functional groups -OH, -NH, -C-H and C=C present on both adsorbents. For chitosan alginate beads, the shift, disappearance and appearance of new peaks after Malachite Green adsorption indicated bonding breakdown and reaction occurred with Malachite Green, thus new bonding was formed. SEM micrographs illustrated surface changed from smooth to rough and porous were filled by particles after Malachite Green adsorption. In this study, rice husk-based adsorbent has the potential to be used for removing Malachite Green from aqueous solutions, especially in pilot study or industrial applications.

1. Introduction

Malachite Green, a triphenylmethane dye, is a multi-purpose chemical that has been used as a fungicide in aquaculture, dye in the textile industry, food colouring agent and food additive in the food industry [1], [2]. Wastewater from the industry that contains Malachite Green affects human, animal, and aquatic life as it is poisonous and has harmful effects on the immune and reproductive systems. Other than that, Malachite Green also negatively impacts on the environment [2], [3]. Therefore, an effective method for removing malachite green is needed.

Among the available treatment methods, adsorption is a passive mass transfer mechanism where gases or solutes bind to solid or liquid surfaces. It is the most efficient method to remove dye from wastewater [4]. Since activated carbon is expensive, wastewater treatment using low-cost by-products from the agricultural, domestic, and industrial sectors has been known as a viable option. The low-cost by-products such as *Limonia acidissima* (Wood Apple) shell [5], coffee husk [6], eggshells [7] pomegranate peel [8], rice husk [9] and groundnut shell [10] have been tested for the removal of dye from wastewater while contributing to waste minimization, recovery and reuse [11]. Therefore, most researchers focus their studies on low-cost sustainable carbon sources, as they are cost-effective, easy to operate and minimize waste production.

There is research on low-cost and sustainable adsorbents for the adsorption of Malachite Green such as rice husk [12], chitosan [13]-[15], and alginate [16]. However, there is a lack of study on the use of rice husk together with chitosan and alginate for Malachite Green removal and comparison studies with rice husk powder. The surface characteristics of adsorbents play a vital role in the removal of dyes in wastewater [6].

The study aims to evaluate the Malachite Green adsorption using rice husk-based adsorbents. The objectives of this study are to investigate the optimum parameters for Malachite Green adsorption using rice husk-based adsorbents, to evaluate the adsorption performance through kinetic and isotherm studies and to characterize the adsorbents before and after Malachite Green adsorption.

2. Methodology

2.1 Materials

2.1.1 Adsorbent Preparation

Rice husk was collected from a paddy farm in Kapit, Sarawak, Malaysia. The rice husk was ground into smaller sizes using a rice mill machine and sieved into 125 µm in order to prepare the rice husk powder. Next, it was washed using ultrapure water three times for the removal of contaminants dust and soluble impurities. Then, the rice husk was dried in the oven at 60 °C until a constant weight was obtained. For the chitosan alginate rice husk beads preparation, 0.6 g of alginate (Sigma-Aldrich) was dissolved in 30 mL of ultrapure water at 100 °C with magnetic stirring for five minutes. 0.6 g of chitosan powder (Sigma-Aldrich) was then added to the solution and dissolved in 1 mL of 1% acetic acid. Next, 0.6 g of rice husk powder was added and stirred homogeneously. By using a syringe, 10 mL of this solution was dripped into 50 mL of 2% calcium chloride solution to form smooth chitosan alginate rice husk beads. Lastly, the beads were rinsed three times using ultrapure water and dried at 60 °C until a constant weight.

2.1.2 Malachite Green Solutions Preparation

A stock solution of Malachite Green dye (Chemiz) was prepared by adding 1 g of dye to 500 mL of ultrapure water inside a volumetric flask, with a 2000 mg/L concentration. The desired concentrations of Malachite Green dye were prepared by dilution of the stock solution with a suitable volume of ultrapure water. Next, the calibration curves were constructed using absorbance values versus concentrations of the Malachite Green. Then, this concentration was measured at 617 nm [17] using a UV-VIS Spectrophotometer (Thermal Scientific Genesys 30 Visible Spectrophotometer).

2.2 Methods

2.2.1 Adsorption Study

The removal of Malachite Green by the chitosan alginate rice husk beads was studied by using a batch sorption experiment conducted in a 50 mL centrifuge tube. 50 ml of 5 mg/L Malachite Green was added to each 0.5 g to 2.0 g of the chitosan alginate rice husk beads, respectively. Using the multi-rotator (Programmable Rotator Multi RS-60), the solution was agitated at 125 rpm under room temperature for 60 minutes. Next, the Malachite Green solution was separated and analysed at 617 nm using a UV-VIS Spectrophotometer (Thermal Scientific Genesys 30 Visible Spectrophotometer). The recorded data was converted from absorbance to concentration using the calibration curve. Lastly, the adsorption performance for removal percentage and uptake were calculated using Eq. (1) and Eq. (2):

The percentage of Malachite Green removal, $R(\%)$,

$$R(\%)=\frac{C_{initial}-C_{final}}{C_{initial}}\times 100\% \tag{1}$$

The uptake of Malachite Green, q_e (mg/g),

$$q_e(mg/g)=\frac{(C_{initial}-C_{final})v}{M} \tag{2}$$

where $C_{initial}$ is the Malachite Green concentration before adsorption (mg/L), C final is the Malachite Green concentration after adsorption (mg/L), V is the volume of Malachite Green solution (L), and M is the mass of adsorbent (g).

The experiment was repeated by using rice husk powder. The adsorbent amount used ranged from 0.005 g to 0.1 g. Next, the experiment was repeated using both adsorbents to investigate the optimisation parameters of contact time at 0.5 to 120 minutes and initial Malachite Green concentration at 1 to 50 mg/L.

2.2.2 Kinetic and Isotherm Models Evaluation

The Malachite Green adsorption optimisation contact time results were examined for the kinetic analysis. The pseudo-first-order kinetic and pseudo-second-order kinetic models as shown in Eq. (3) and Eq. (4) were used:

$$\log(q_e-q_t)=\log q_e-\frac{tk_1}{2.303} \tag{3}$$

$$\frac{t}{q_t}=\frac{1}{2k_2q_e^{\,2}}+\frac{t}{q_e} \tag{4}$$

where t is time (min), q_e and q_t are the Malachite Green adsorption uptake at equilibrium and at time (mg/g) and K_1 and K_2 are the consonants for pseudo-first order and pseudo-second order kinetic models.

The results of the adsorption studies for initial Malachite Green concentration were fitted to linearized Langmuir and Freundlich isotherm model equations for isotherm analysis using Eq. (5) and Eq. (6).

$$\frac{C_e}{q_e}=\frac{C_e}{q_{\max}}+\frac{1}{bq_{\max}} \tag{5}$$

$$Inq_e=Ink_F+\frac{1}{nInC_e} \tag{6}$$

where q_e is the equilibrium of Malachite Green adsorption uptake (mg/g), q_{max} is the maximum Malachite Green adsorption uptake (mg/g), C_e is the Malachite Green concentration at equilibrium (mg/L), and b is the constant for Langmuir, while K_F and n are the constants for Freundlich.

2.2.3 Characterization Study

For Fourier-transform infrared spectroscopy (FTIR), chitosan alginate rice husk beads and rice husk powder before and after Malachite Green adsorption was characterised using FTIR (Bruker Alpha I) (ATR-FTIR). The spectra on chitosan alginate rice husk beads and rice husk powder surface were obtained by averaging 16 scans in the range of 650 to 4000 cm^{-1} at a resolution of 4 cm^{-1}.

For SEM sample preparation, samples were coated with gold using a sputter coater. The scanning electron microscopy (SEM) images of the chitosan alginate rice husk beads and rice husk powder surfaces, before and after adsorption of Malachite Green were obtained by using a SEM (Hitachi tabletop microscope TM3030 Plus) at a voltage of 20 kV and a magnification of 1000x and 2000x.

3. Results and Discussion

3.1 Adsorption Study

A series of adsorption studies on the effects of adsorbent amount, contact time and initial concentration of Malachite Green was presented and discussed in the following sub-sections.

3.1.1 Adsorbent Amount

Fig. 1 shows the effect of the adsorbent amount on the removal and uptake of Malachite Green using chitosan alginate rice husk beads and rice husk powder. For chitosan alginate rice husk beads, the removal progressively increased from 23% to 54% when the adsorbent amount was increased from 0.5 g to 2.0 g (Fig. 1(a)). For rice husk powder, the removal showed a rapid increase from 11% to 62% when the amount of the adsorbent was increased from 0.005 g to 0.025 g (Fig. 1(b)). Then, a gradual rise towards the saturation phase could be observed from 62% to 96%, when the amount of the adsorbent was increased from 0.025 g to 0.1 g. An increase in the availability of adsorption sites was the reason for the rapid increase trend [18]. Meanwhile, the removal for the gradual increase was due to Malachite Green moved further in macropores for available adsorption sites when the majority of the adsorption sites on the surface were occupied. A similar removal trend for Malachite Green adsorption was observed using industrial waste coffee husk [6] and groundnut shell waste-based powdered activated carbon [10]. The results obtained were consistent with the optimum and removal trends although using different materials.

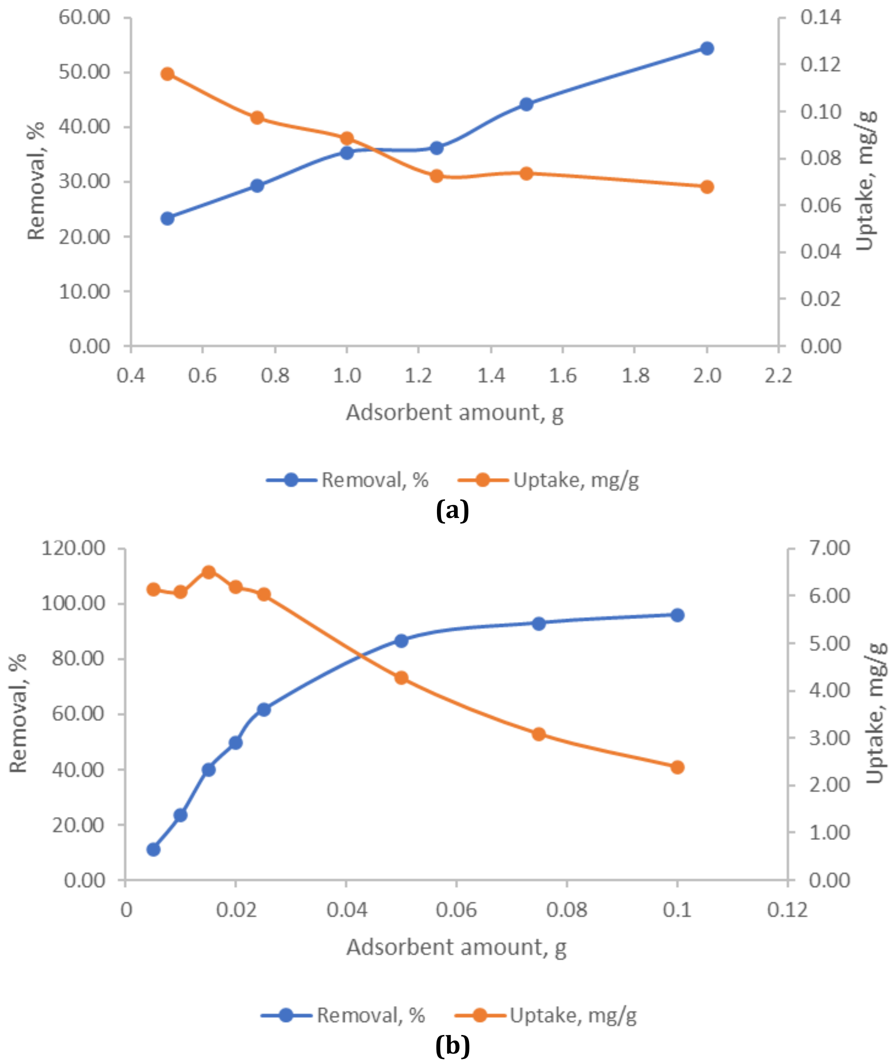


Fig. 1 *Effect of adsorbent dosage on the removal and uptake of Malachite Green by (a) Chitosan alginate rice husk beads; and (b) Rice husk powder (60 minutes, 5 mg/L Malachite Green)*

The uptake of Malachite Green using chitosan alginate rice husk beads showed a gradually decreasing trend from 0.12 mg/g to 0.11 mg/g for the adsorbent amount of 0.5 g to 2.0 g. (Fig. 1(a)). Similarly, a general gradual decrease was also observed from 6.51 mg/g to 2.39 mg/g when the amount of rice husk powder adsorbent was increased (Fig. 1(b)). When a large amount of adsorbent was added, it clumped together, reducing the number of active binding sites and surface area, thus decreasing the uptake of Malachite Green [18]. A similar uptake trend was reported for the adsorption of Malachite Green as a cationic dye onto functionalized multi-walled carbon nanotubes [19].

The chosen amount of adsorbent was 1.35 g for chitosan alginate rice husk beads. Meanwhile, for rice husk powder, the chosen amount was 0.02 g. For chitosan alginate rice husk beads and rice husk powder, the adsorbent amount for removing Malachite Green at 50% was chosen to avoid waste of chemicals, adsorbents, time, and materials. This amount of absorbent was also used as a reference point for further research [20].

3.1.2 Contact Time

Fig. 2 illustrates the effects of contact time on the removal and uptake of Malachite Green using chitosan alginate rice husk beads and rice husk powder. Both of adsorbents showed a similar trend in removal and uptake with three phases – an initial rapid phase, a gradual phase and finally saturation phase. A rapid increase from 0.5 to 15 minutes recorded Malachite Green removal percentage from 7% to 28% and uptake from 0.01 mg/g to 0.05 mg/g for chitosan alginate rice husk beads. There was a gradual increase from 28% to 60% removal and uptake from 0.05 mg/g to 0.11 mg/g as the duration of the experiment increased from 15 to 90 minutes. Finally, the saturation phase was achieved at 90 to 120 minutes with 61% removal and 0.11 mg/g uptake of Malachite Green.

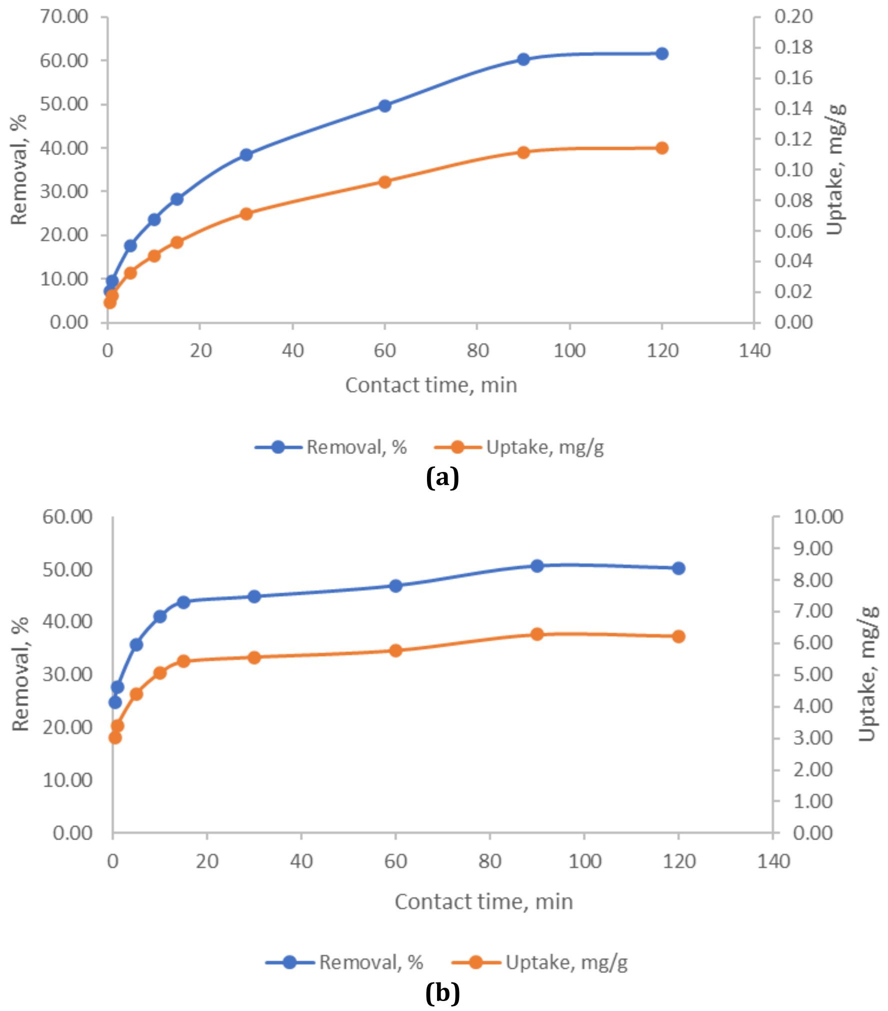


Fig. 2 *Effect of contact time on removal and uptake of Malachite Green by (a) Chitosan alginate rice husk beads; and (b) Rice husk powder (Adsorbent amounts (a) 1.35 g, and (b) 0.02 g, 5 mg/L Malachite Green)*

For rice husk powder, a rapid increase in removal of 25% to 44% and uptake of 3.03 mg/g to 5.42 mg/g was recorded from 0.5 to 15 minutes. The uptake of malachite green gradually increased from 5.42 mg/g to 6.28 mg/g, and the removal percentage was from 38% to 50%. The saturation phase was achieved at 50% removal and 6.25 mg/g uptake for contact time from 90 to 120 minutes. Due to the abundance of empty sites on the surface of the adsorbent, the removal and uptake of Malachite Green increased quickly during the initial phase [21]. At a later phase, the removal and uptake of Malachite Green gradually increased as the empty sites on the surface of the adsorbent gradually decreased over time [21], [22]. The saturation phase was reached as the active binding sites had been fully occupied by the Malachite Green and were no longer able to adsorb the dye. Similar results for removal were reported for the adsorption of Malachite Green using rice husk-activated carbon [23] and functionalized multi-walled carbon nanotubes [19].

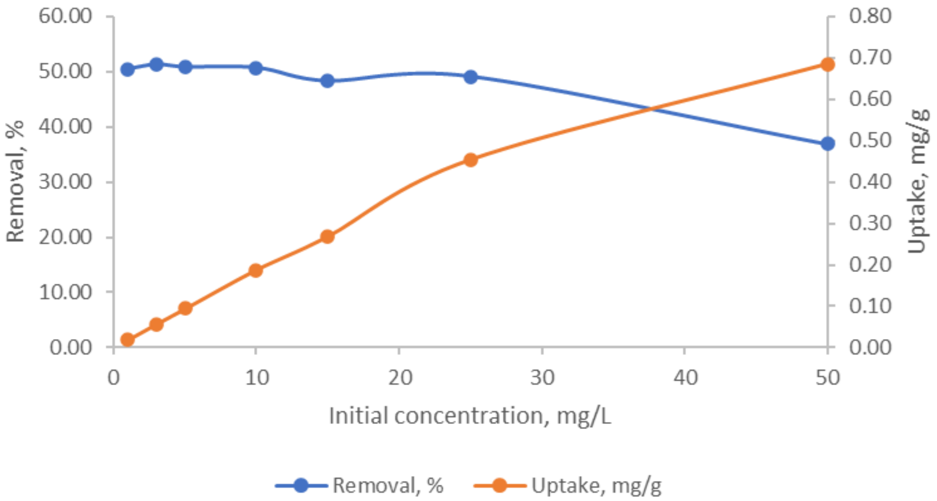
The contact time of 60 minutes was selected and consistent with the adsorbent amounts chosen for chitosan alginate rice husk beads and rice husk powder as it indicated the 50% removal. In addition, it saves a lot of time and energy, and the result before saturation is important for kinetic studies.

3.1.3 Initial Malachite Green Concentration

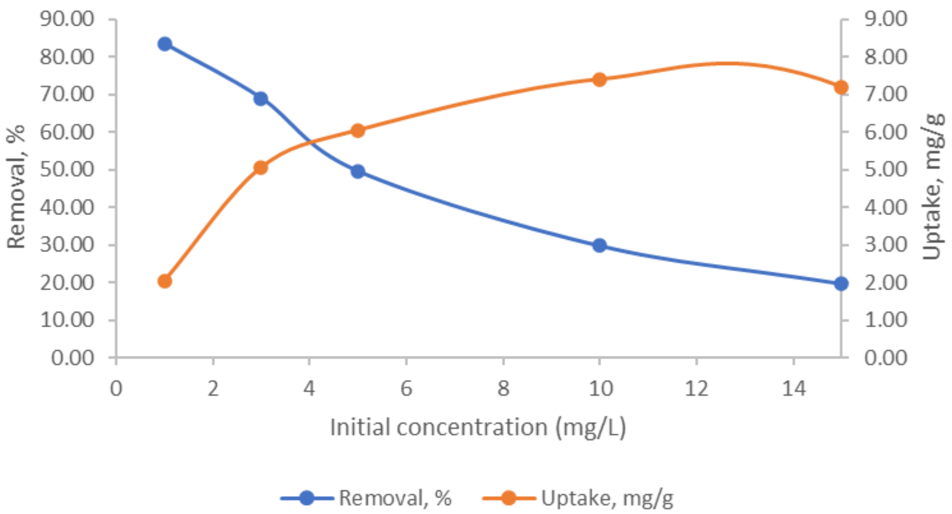
Fig. 3 presents the effect of initial concentration on the removal and uptake of Malachite Green using chitosan alginate rice husk beads and rice husk powder. The removal of Malachite Green by chitosan alginate rice husk beads and rice husk powder were generally in a decreasing trend. When the concentration of Malachite Green was increased from 5 mg/L to 50 mg/L, the removal gradually decreased from 50% to 37% and from 84% to 20% for chitosan alginate rice husk beads and rice husk powder, respectively. Due to the increased ratio of Malachite Green to adsorbent, the Malachite Green removal decreased.

The uptake of Malachite Green by chitosan alginate rice husk beads rapidly increased from 0.02 mg/g to 0.68 mg/g as the concentration of Malachite Green was increased from 5 mg/L to 50 mg/L. at the similar condition, the uptake of rice husk powder rapidly increased from 2.04 mg/g to 7.40 mg/g. Malachite Green was taken up easier because of its strong driving force in high concentrations, which sped up the adsorption process [6], [24].

A similar removal and uptake trend of Malachite Green was observed using industrial waste coffee husk [6]. Other than that, Chowdhury & Das [7] also reported a similar removal trend for Malachite Green using eggshells.



(a)



(b)

Fig. 3 *Effect of initial concentration on removal and uptake of Malachite Green by (a) Chitosan alginate rice husk beads and (b) Rice husk powder (Adsorbent amount (a) 1.35 g, and (b) 0.02 g, 60 minutes)*

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3.2 Evaluation Study

The results for pseudo-first-order and pseudo-second-order kinetic models in the kinetic study as well as Langmuir and Freundlich isotherm models used in the isotherm study were evaluated.

3.2.1 Kinetic Study

Table 1 shows the comparison of the kinetic study of Malachite Green using various adsorbents. For regression coefficient analysis, the pseudo-first-order kinetic model using chitosan alginate rice husk beads ($r^2 = 0.9944$) fits better than the pseudo-second-order kinetic model ($r^2 = 0.9782$). For rice husk powder, the regression coefficient analysis of the linearized pseudo-second-order kinetic model ($r^2 = 0.9986$) showed an excellent fit than the pseudo-first-order kinetic model ($r^2 = 0.7100$). However, the calculation uptake (q_t) for both chitosan alginate rice husk beads and rice husk powder for experimental values at 0.11 mg/g and 6.28 mg/g corresponded to the pseudo-second-order kinetic model at 0.12 mg/g and 6.31 mg/g, compared to the pseudo-first-order kinetic model at 1.03 mg/g and 2.40 mg/g. The findings supported the fact that both materials were better fitted to the pseudo-second-order kinetic model than the pseudo-first-order kinetic model. The pseudo-second-order kinetic model indicated that chemisorption is the rate-limiting phase when compared to physisorption for the pseudo-first-order kinetic model. Table 1 also summarizes consistent findings where the adsorption of Malachite Green is well corresponded to the pseudo-second-order kinetic model than the pseudo-first-order kinetic model.

Table 1 *Comparison of Malachite Green adsorption for pseudo-first-order and pseudo-second-order kinetic models using various adsorbents*

Sample	Pseudo-first-order			Pseudo-second-order			Experimental value	Reference
	r^2	k_1	*qt* (mg/g)	r^2	k_2	*qt* (mg/g)	*qt, exp* (mg/g)	
Industrial waste coffee husk	0.9446	0.0136	55.12	0.9942	0.0005	84.03	81.50	[6]
Chitosan beads 1-butyl-3-methylimidazolium acetate A	0.9740	0.4470	62.66	1.00	0.1010	7.50	7.20	[25]
Chitosan beads 1-butyl-3-methylimidazolium B	0.8810	0.1470	4.33	1.00	0.0550	9.70	9.20	[25]
Rice Husks	0.7800	0.0140	0.05	0.9860	4.3000	0.63	0.62	[12]
Chitosan alginate rice husk beads	0.9944	2.3917	1.03	0.9782	0.5680	0.12	0.11	This study
Rice husk powder	0.7100	0.0458	2.40	0.9986	0.0722	6.31	6.28	This study

3.2.2 Isotherm Study

Table 2 compares the Malachite Green adsorption for linearized Langmuir and Freundlich isotherm models of different adsorbents. For the regression coefficient (r^2), the Freundlich isotherm model for chitosan alginate rice husk beads 0.9864 presented a better fit compared to the Langmuir isotherm model with 0.9102. However, the Langmuir isotherm model for rice husk powder ($r^2 = 0.9866$) was better corresponded than the Freundlich isotherm model ($r^2 = 0.9671$). This revealed that the chitosan alginate rice husk beads have a heterogeneous surface with multiple functional groups as adsorption sites. Meanwhile, the chitosan alginate rice husk beads exhibited monolayer homogeneity at the surface of the adsorbent. Generally, adsorbents in Table 2 were better fitted to the Langmuir isotherm model due to the surface characteristics of the adsorbents.

The estimated maximum uptake (q_{max}) for the Langmuir isotherm models were 1.56 mg/g for chitosan alginate rice husk beads, and 8.22 mg/g for rice husk powder. Based on the q_{max}, it showed that rice husk powder was five times higher than chitosan alginate rice husk beads. This indicated that the rice husk powder has the characteristics of a good adsorbent having a large specific likely surface area and available adsorption sites. However, for chitosan alginate rice husk beads, it did not show a better result because the functional groups on the surface of chitosan alginate rice husk were blocked by the formation of the matrix, thus reducing the adsorption performance. The performance of chitosan alginate rice husk beads and rice husk powder were comparable with previous studies as shown in Table 2.

The high *b* value of 15.463 L/mg in the Langmuir isotherm model in Table 2 implied that chitosan alginate rice husk beads showed high selectivity towards Malachite Green. Meanwhile, the low *b* value of 0.0087 L/mg for rice husk powder revealed that it has low selectivity towards Malachite Green. The low values of k_F and *n* for the Freundlich isotherm model for both adsorbents in this study indicated slow adsorption of Malachite Green (Table 2).

Table 2 *Comparison of Malachite Green adsorption for Langmuir and Freundlich isotherm models of adsorbents*

Sample	Langmuir			Freundlich			Reference
	q_{max} (mg/g)	*b* (L/mg)	r^2	k_F	*n*	r^2	
Industrial waste coffee husk	78.00	4.0968	0.9988	59.7585	2.9630	0.9039	[6]
Chitosan beads 1-butyl-3-methylimidazolium acetate A	8.07	-12.3990	0.9980	8.8700	-9.9010	0.9810	[25]
Chitosan beads 1-butyl-3-methylimidazolium B	0.24	-0.9670	0.9910	8.3800	-0.7020	0.9210	[25]
Chitosan alginate rice husk beads	1.56	15.4632	0.912	0.0401	1.1188	0.9864	This study
Rice husk powder	8.22	0.0087	0.9866	4.4326	2.5478	0.9671	This study

3.3 Characterization Study

The results for the characterization of chitosan alginate rice husk beads and rice husk powder, before and after adsorption using FTIR and SEM/EDX were presented and discussed in the following sections.

3.3.1 Fourier-Transform Infrared Spectroscopy (FTIR)

Fig. 4 and Tables 3 present the changes in the wavelength of the FTIR spectra before and after adsorption for chitosan alginate rice husk beads and rice husk powder. For chitosan alginate rice husk beads, the wideband shifted from 3264 to 3236 cm^{-1} corresponded to the -OH and -NH of hydroxyl groups. The peak at 2321 to 2322 cm^{-1} was due to -CH bond and the shifted wavelength from 1590 to 1604 cm^{-1} was attributed to the amides 1 C=O and C=C. Shifted peaks from 1417 to 1415 cm^{-1} and 1012 to 1020 cm^{-1} were assigned to the alcohol C-O of cellulose. After Malachite Green adsorption, it was found that the disappearance of peak at 2110 cm^{-1} and the appearance of peaks at 1523, 1317 and 773 cm^{-1}. This revealed that -C≡C from alkynes was broken down and converted to C-C, -C-H and high possibility to bind with -OH. This condition supported that heterogeneous surface with various functional groups for chitosan rice husk beads.

For rice husk powder as shown in Table 4 and Fig. 4, the broad peak shifted from 3283 to 3284 cm^{-1} was due to the presence of -OH and -NH groups. The shifted peak from 1640 to 1641 cm^{-1} and 1525 to 1516 cm^{-1} represented -NH and C=C from the aromatic group, respectively. A new peak was formed at 1364 cm^{-1} was attributed to -C-H. This also proposed that Malachite Green had been adsorbed onto rice husk powder and new bonds had been established between Malachite Green and the rice husk powder.

Peaks shifted after adsorption for chitosan alginate rice husk beads indicated the reaction occurred between functional groups and the Malachite Green. In addition, the disappearance of peaks corresponded to the new peaks where such circumstances provided evidence on the relevant functional group was broken down and a new functional group formed due to its involvement in the Malachite Green adsorption process [26].

3.3.2 Scanning Electron Microscopy (SEM)

Fig. 5 shows the SEM micrographs images for chitosan alginate rice husk beads and rice husk powder, before and after adsorption of Malachite Green. Before adsorption, the slightly smooth and porous surface morphology of chitosan alginate rice husk beads and rice husk powder were clearly observed in the SEM image. However, the surface became rough and pores were filled after the Malachite Green adsorption This indicated that the adsorption of Malachite Green has changed the surface morphology of the rice husk powder as it is deposited on the surface and blocked the formation on the surface. Such circumstance confirmed that the Malachite Green was successfully adsorbed on the chitosan alginate rice husk beads. These observations are consistent with those of Ngah et al. [14] and Murthy et al. [30].

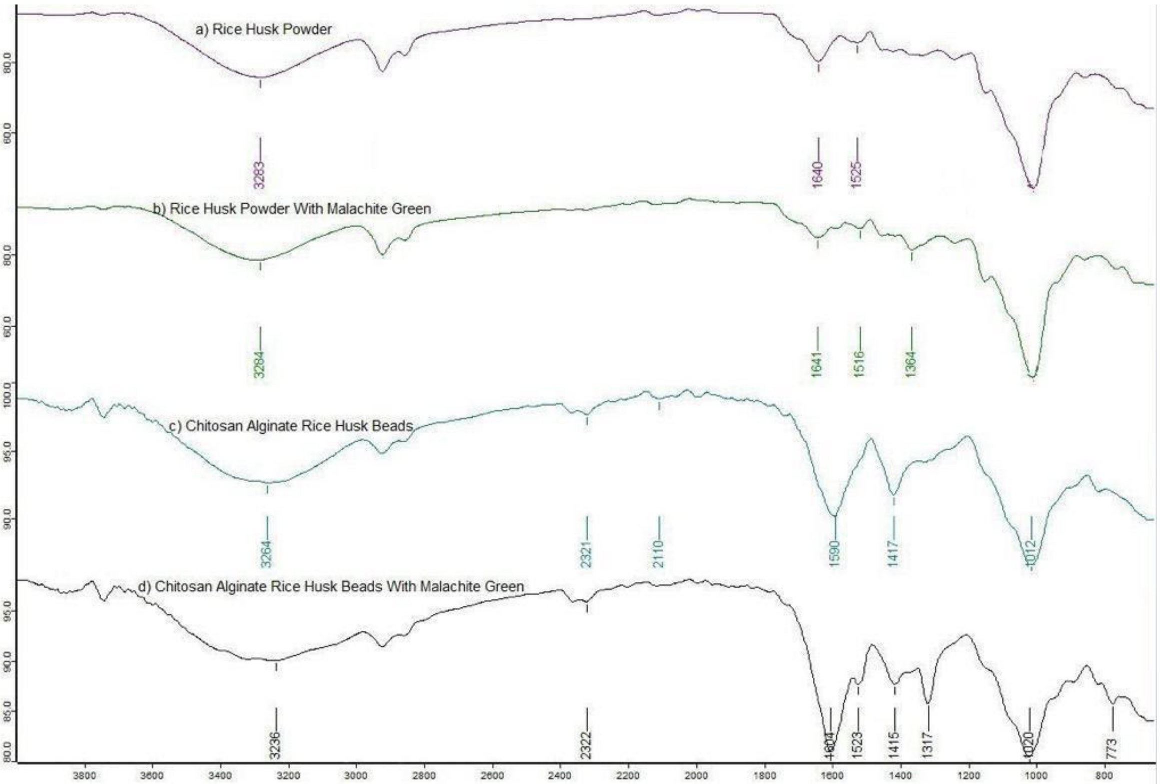

Fig. 4 *FTIR spectra of the (a) Rice husk powder; (b) Rice husk powder with Malachite Green; (c) Chitosan alginate rice husk beads; (d) Chitosan alginate rice husk beads with Malachite Green within the 650-4000 cm⁻¹*

Table 3 *Peak assignments for chitosan alginate rice husk beads before and after adsorption*

Wavelength (cm^{-1})			
Before adsorption	After adsorption	Assignment	Reference
3264	3236	-OH, -NH	[27]
2321	2322	-C-H	[28]
2110	-	-C≡C	[12]
1590	1604	C=O, C=C	[5], [26]
-	1523	C=C	[29]
1417	1415	C-O	[29]
-	1317	C-C	[5]
1012	1020	C-O	[29]
-	773	-OH, -C-H	[27]

Table 4 *Peak assignments for rice husk powder before and after adsorption*

Wavelength (cm^{-1})			
Before adsorption	After adsorption	Assignment	Reference
3283	3284	-OH, -NH	[26]
1640	1641	-NH	[14]
1525	1516	C=C	[5], [29]
-	1364	-C-H	[12]

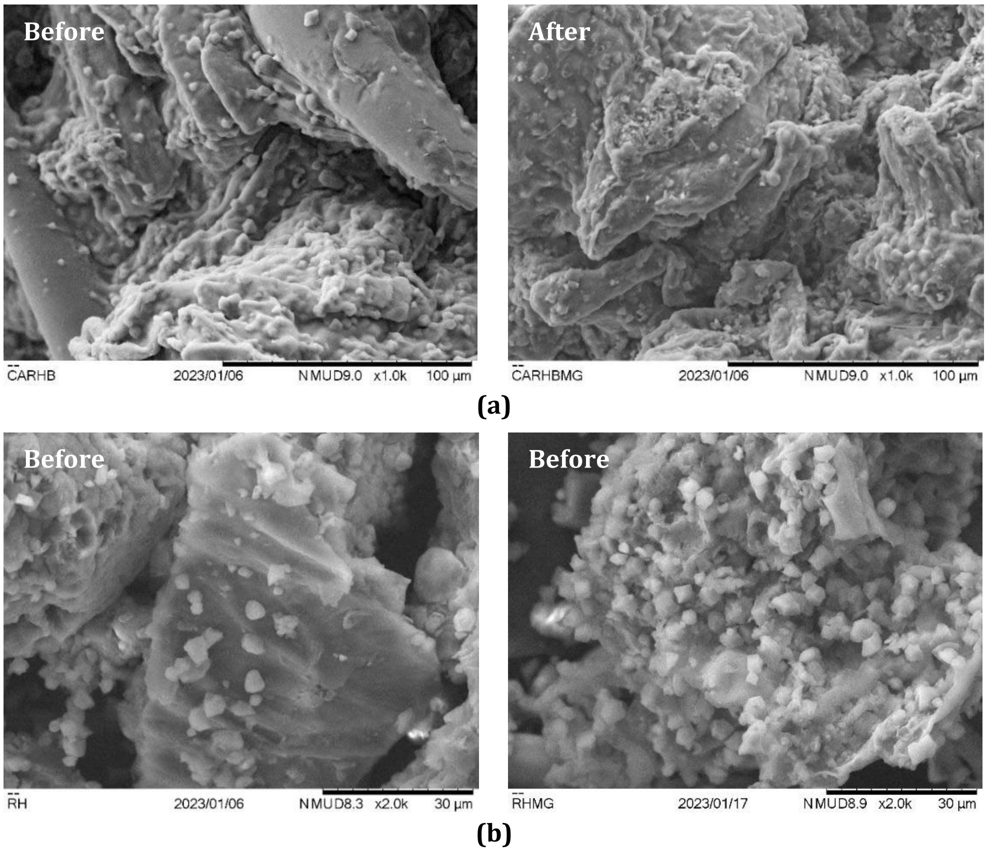

Fig. 5 *SEM micrographs before and after Malachite Green adsorption by (a) Chitosan alginate rice husk beads (1000x); and (b) Rice husk powder (2000x)*

4. Conclusion

In this study, the adsorption of Malachite Green using rice husk-based adsorbents was investigated. A 50% removal efficiency of Malachite Green was determined for chitosan alginate rice husk beads and rice husk powder at 1.35 g and 0.02 g, respectively. The selected adsorption conditions for chitosan alginate rice husk beads and rice husk powder were found to be 60 minutes and a 5 mg/L Malachite Green concentration. The Malachite Green adsorption kinetic for chitosan alginate rice husk beads and rice husk powder were better fitted with the pseudo-second-order kinetic model compared to pseudo-first-order kinetic model. This indicated that chemisorption is the rate-limiting phase in the adsorption process. For adsorption isotherm, chitosan alginate rice husk beads had a better fit to the Freundlich isotherm model, meanwhile, rice husk powder was better fitted to the Langmuir isotherm model. Such conditions revealed a heterogeneous multilayer surface for chitosan alginate rice husk beads and a homogeneous monolayer surface for rice husk powder. FTIR spectra analysis showed chitosan alginate rice husk beads exhibited extra functional groups of -C≡C, C-O, C-C, C=O, C-O besides -OH, -NH, -C-H and C=C in both rice husk-based adsorbents. SEM analysis showed that the surface morphology of adsorbents from rough and porous became rough and filled with particles after the adsorption of Malachite Green. In conclusion, rice husk powder has a better adsorption performance of Malachite Green compared to chitosan alginate rice husk beads as the matrix beds have blocked and prevented the functional groups from binding with Malachite Green. Rice husk-based adsorbents are potential for the removal of Malachite Green from aqueous solutions.

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Conflict of Interest

Authors declare that there is no conflict of interest regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design:*** *Chia Chay Tay, Rui Hong Wu;* ***data collection:*** *Angelie Uni Mapong Anak Manok, Farhanny Ismail;* ***analysis and interpretation of results:*** *Dhia Zur'Ain Noor Salizam, Nor Atikah Husna Ahmad Nasir, Rui Hong Wu;* ***draft manuscript preparation:*** *Angelie Uni Mapong Anak Manok, Chia Chay Tay, Eric D. Van Hullebusch. All authors reviewed the results and approved the final version of the manuscript.*

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