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# Adsorption of Brilliant Green Dye in Aqueous Medium Using Magnetic Adsorbents Prepared from Rice Husk Ash

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**Abstract.** Large scale production and extensive application of synthetic dyes in the textile industry have caused considerable environmental pollution. The treatment of colored effluents using efficient technologies, are sought after due to this concern. This research evaluates the effectiveness of magnetic adsorbent (MRHA) prepared from rice husk ash (RHA) by a chemical processing method to remove brilliant green (BG) dye from an aqueous medium. The resulting MRHA adsorbents were tested at different initial concentration of 5–400mg/L, adsorbents amount of 0.5–2.3g, shaking rate of 50–300rpm, contact time of 15–120 min, pH of 3–11, and temperature of 27–60 C. A maximum removal of 96.65% was obtained at initial dye concentration of 200mg/L, adsorbents amount of 2g, shaking rate of 150rpm, contact time of 60min, and temperature of 50 C. The fresh and spent MRHA adsorbents were analyzed through scanning electron microscopy (SEM) and particle size distribution. The particle analysis of the fresh and spent adsorbents indicated bimodal pore size of 90 and 900 $\mu$ m, respectively. The adsorption behavior of the adsorbent followed those of the Langmuir, Freundlich, and Temkin isotherm models. However, Temkin isotherm model displayed the best fit with the coefficient of determination,  $R^2$  of 0.811, suggesting a strong interaction, equivalent to that of chemical absorption between BG dye molecules and the surface of MRHA adsorbent for effective removal of BG dye from the aqueous medium. The magnetic character of the adsorbents allowed the spent sample to be isolated successfully and conveniently by using an external magnetic field.

**Key words:** Dye; textile; environment; biodegradable; magnetic adsorbent

## INTRODUCTION

Environmental pollution related to water and wastewater has been one of the most serious issues affecting the world today. Aquatic environment is constantly polluted by human activities such as the release of effluent or polluted water

containing oil and heavy metals [1-2] and dyes from textile industries [3-5]. Among the dyes used in the textile industries, brilliant green (BG) is one of the most common types being applied. BG is also heavily applied in paper printing industries, photosensitizer in the making of photogalvanic cells, and in saloon bars to dye hair [6-8]. In paper printing industries, about 0.8–1.0kg of BG is required for every ton of the colored paper produced [9]. The release of the spent BG from these industries into the water stream without treatment however, has been reported to affect the aquatic life and human beings. BG dye is known to have genotoxic and carcinogenic properties, which can cause a significant health risk to living beings [10-12]. Therefore, its removal from contaminated waste water is imperative.

Novel techniques in dye removal from the polluted water have been reported rather extensively. The adsorption technique by activated carbon (AC) however, has gained widest acceptance in research because of its effectiveness, inexpensiveness, practicality and simplicity [13-16]. The current trend in research lately is on lowering the cost of adsorbent preparation further, in addition to its renewability [17-20]. As a result, low-cost adsorbents prepared from industrial solid and biomaterial wastes such as chitosan [21] and date palm [22] are gaining momentum in research. Researches on the exploitation of the other agricultural solid waste materials such as rice husk ash (RHA), palm oil fuel ash (PFA) and coal fly ash (CFA) [23-29] are also on the rise.

A common trait that these adsorbents share is on the spent adsorbent's reusability. Most of the time, the spent adsorbent became waste at the end of the cycle and it had to be disposed of, together with the sequestered dye. This in turn, creates another issue of waste disposal. In order to overcome this problem, the adsorbent is synthesized with magnetic materials such as iron oxide, so that it can be separated from the treated water by way of magnetic separation [30-42]. By magnetic separation, the adsorbent is easily removed from the solution using an external magnetic field.

In this work, we prepared magnetic adsorbents derived from RHA for removal of BG dye. This adsorbent was synthesized by a chemical processing method, which is simpler in approach than the other conventional techniques discussed in the literatures [30-42]. The effectiveness of the prepared adsorbent from this method for BG removal, its adsorption behaviour, recyclability and its characteristics from the analysis of scanning electron microscopy (SEM) and particle size distribution shall be discussed at greater length in the paper.

## METHODOLOGY

### Adsorbent preparation

The rice husk ash (RHA) was supplied directly by Kilang Beras & Minyak Sin Guan Hup Sdn. Bhd. Nibong Tebal, Penang. It was in the form of black coarse dry ash with the broken rice husk-like structure upon received. Prior to use, raw RHA was sieved to a fine particle size of less than 75 $\mu$ m. Then the ash was washed with deionized water to remove impurities and dried in the oven with 110°C overnight. Brilliant green (BG) dye, FeCl<sub>3</sub> and FeSO<sub>4</sub> were purchased from Sigma-Aldrich, Malaysia. All chemical reagents used in this experiment were of analytical grade. Double distilled water was used throughout this experiment.

The magnetic RHA adsorbent (MRHA) was prepared according to the chemical processing method described previously [43], but with an improvement. About 50 g of RHA was suspended in 500mL of distilled water. A ferric chloride solution was freshly prepared by adding 18g of FeCl<sub>3</sub> into 150mL of distilled water. Also, a ferrous sulfate solution was prepared by adding 20g of FeSO<sub>4</sub> into 150mL of distilled water. Both solutions were combined and vigorously stirred at 60–70 C. The suspension formed was then added into RHA solution at room temperature and slowly stirred for 30min. After mixing, 10M of NaOH solution was added drop wise into the suspension until the pH raised to 10–11. During NaOH addition, the suspension became dark brown at pH around 6 and then black at pH around 10. After mixing for 60min, the suspension was aged at room temperature for 24h and then repeatedly washed with distilled water followed by ethanol. The MRHA adsorbent was vacuum filtered and dried in the oven overnight at 50 C and stored in a desiccator before used in batch BG adsorption studies.

The surface morphologies of the selected (prepared and spent) MRHA adsorbents were analyzed using scanning electron microscopy (SEM, Leo Supra 35VP microscope). While the particle size distribution of the selected MRHA adsorbents were investigated using Mastersizer 2000.

### BG adsorption experiment

Batch BG adsorption studies were performed in a Certomat S-II horizontal shaker (Sartorius AG) to observe the effect of important parameters, i.e. initial BG concentration (5–400mg/L), adsorbents amount (0.5–2.3g), shaking rate (50–

300rpm), contact time (15–120min), pH (3–11), and temperature (27–60 C) on the adsorption of BG in the form of BG removal efficiency. All experiments were conducted in 250ml conical flasks covered by aluminium foil with 100mL of the prepared dye solution without any pH adjustment, except during pH study. At appropriate time intervals during each batch experimental study, the MRHA adsorbents were separated from the suspension using a strong magnetic separator shown in **FIGURE 1(c)**. After the separation, the residual dye samples were withdrawn, and the dye concentration was analyzed using DR 2800 Spectrophotometer (Hach Co.). The equilibrium adsorption capacity,  $q_e$  (mg/g) and the removal efficiency (RE, %) of BG by MRHA adsorbents were calculated using the following equations, respectively:

$$q_e = \frac{(C_0 - C_e) V}{W} \quad (1)$$

$$RE = \frac{C_0 - C_e}{C_0} \times 100 \quad (2)$$

where  $C_0$  and  $C_e$  (mg/L) are BG concentrations at initial and equilibrium, respectively.  $W$  (g) is the mass of MRHA adsorbent used and  $V$  (L) is the volume of the prepared dye in aqueous medium. All the experiments were conducted in triplicate to increase the precision of the results, and only the average value was reported throughout this study.

### Adsorption isotherm studies

To study the adsorption equilibrium relationship between BG dye concentration and MRHA adsorbent, the adsorption isotherm of BG dye on MRHA adsorbent were measured. Three of the most established isotherm models, such as Langmuir, Freundlich, and Temkin isotherm were used to test the experimental data. The linear form of Langmuir [44], Freundlich [45], and Temkin [46] isotherms are given in equation (3), (4) and (5), respectively.

$$\frac{C_e}{q_e} = \frac{1}{Q_0 b} + \frac{C_e}{Q_0} \quad (3)$$

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \quad (4)$$

$$q_e = B_1 \ln K_t + B_1 \ln C_e \quad (5)$$

where, the constant  $Q_0$  is the maximum monolayer adsorption capacity (mg/g) and  $b$  is Langmuir constant (L/mg),  $K_F$  and  $n$  are Freundlich constants, and  $K_t$  and  $B_1$  are Temkin isotherms which are related to the maximum binding energy and heat of adsorption, respectively. The applicability of the isotherm model was assessed by the coefficient of determination ( $R^2$ ). Apart from that, the essential characteristics of the Langmuir isotherm can be expressed in term of dimensionless constant separation factor  $R_L$  [47] defined by equation (6),

$$R_L = \frac{1}{1 + C_0 b} \quad (6)$$

The value of  $R_L$  indicates the type of the isotherm to be either irreversible ( $R_L=0$ ), favorable ( $0 < R_L < 1$ ), linear ( $R_L=1$ ), or unfavorable ( $R_L > 1$ ).

## RESULTS AND DISCUSSION

### Characterization of MRHA adsorbents

**FIGURE 1** shows the surface of the selected fresh and spent MRHA samples through the lens of SEM at 500× magnification. The MRHA sample exhibited a porous surface structure with fractured and elongated cell walls around

the pores. The pore structure of the spent MRHA sample became less porous than before, indicating that they were covered by the BG dye molecules.

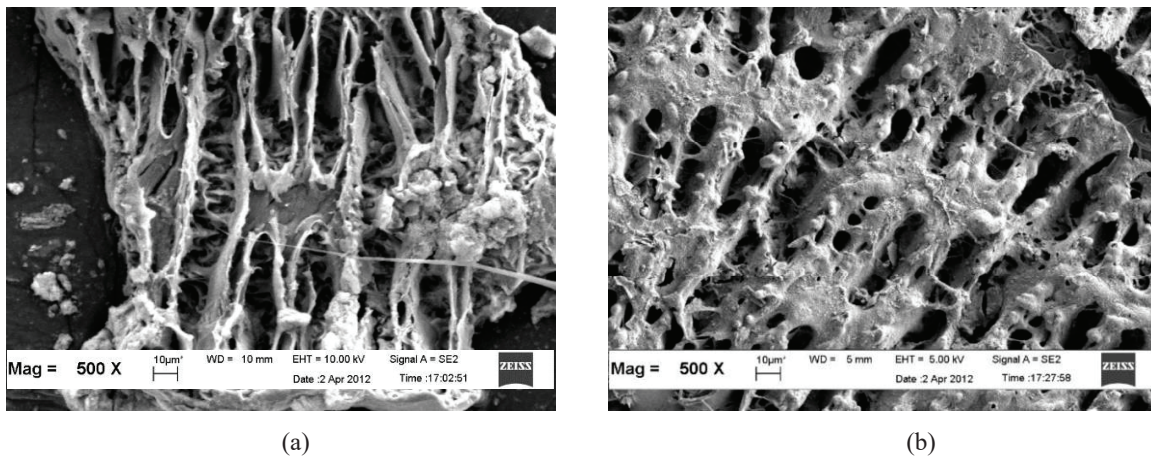


FIGURE 1. Image of scanning electron micrograph (SEM) of (a) prepared and (b) spent MRHA adsorbents

The particle size and distribution of the selected fresh and spent MRHA samples are shown in FIGURE 2. Both samples exhibited a bimodal-type particle size distribution, with two different ranges resulting from the breaking up of particles or variable growth mechanisms during the preparation method. The higher pore volume shown by the dotted line as observed for the spent MRHA sample could be due to the additional volume contributed by the BG dye molecules that were adsorbed and remained on the MRHA surface. This is consistent with the SEM images depicted in the previous figure.

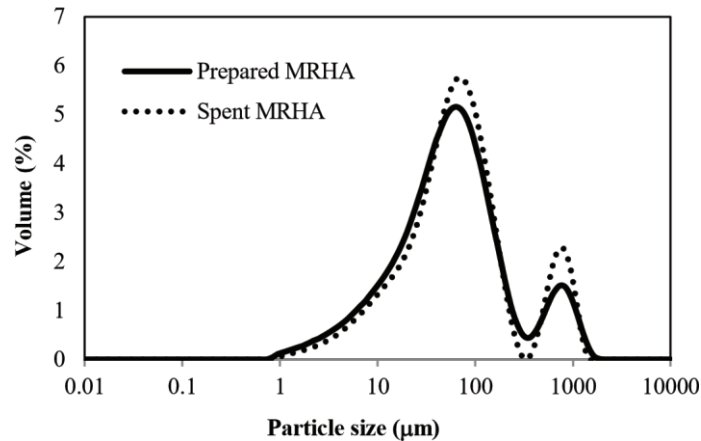
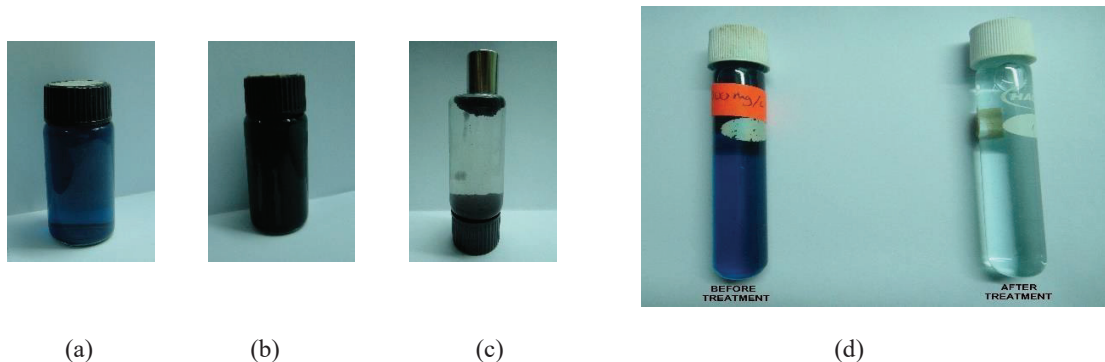


FIGURE 2. Particle size distribution of prepared and spent MRHA adsorbents

FIGURE 3 shows the adsorption process behavior of MRHA adsorbent on the BG dye solution. FIGURE 3(c) shows that the spent MRHA adsorbent was effectively separated from the solution after the BG adsorption. MRHA sample was observed to cling towards the permanent magnet placed externally on top of the inverted bottom test tube. FIGURE 3(d) shows that BG dye was successfully removed from the solution, leaving only clear solution after the adsorption treatment.

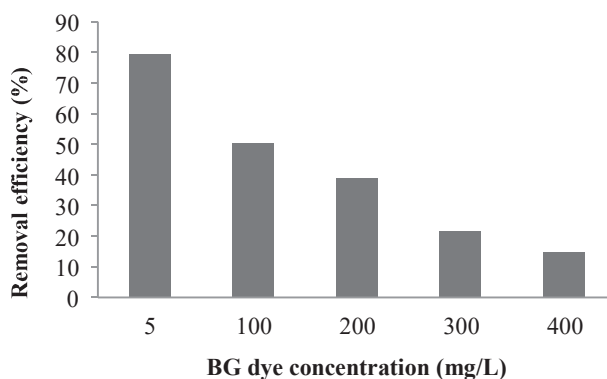




**FIGURE 3.** Photographs of (a) prepared BG dye, (b) MRHA+BG dye, (c) demonstration of magnetic separation of MRHA adsorbent, and (d) comparison of BG dye solution before and after adsorption by MRHA adsorbent.

### Effect of initial BG concentration

**FIGURE 4** shows the effect of initial dye concentration on BG dye adsorption by MRHA adsorbent. The BG dye removal efficiency was observed to decrease from 79.41% to 14.81% with the increasing initial dye concentration from 5 mg/L to 400mg/L at constant adsorbent dosage of 0.5g, contact time of 1h, shaking rate of 150rpm, and room temperature. The reason for the decreasing removal efficiency can be explained by the decreasing mass gradient between the dye solution and adsorbent as the initial BG concentration was increased. At low initial dye concentration, BG dye adsorption took place at the higher energy site. As the initial dye concentration was increased, the higher energy sites became saturated and the adsorption had to be initiated at the lower energy sites, thus resulting in the decrease of the adsorption efficiency [48].



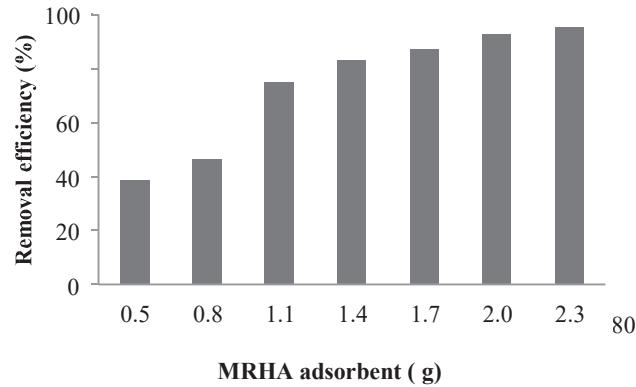
**FIGURE 4.** Effect of initial concentration on the adsorption of BG dye by MRHA adsorbents (Experimental conditions: adsorbent dosage of 0.5g, contact time of 1h, shaking rate of 150rpm, and room temperature)

A similar trend was also observed in the literature that reported the use of a corn derived activated carbon [49], citrus limetta peel [50], red clay [51], rice husk [52], and ZnO nanoparticles [53] as an adsorbent. Nevertheless, other studies revealed contradictory results. For an example, the removal efficiency of white rice husk ash at a fixed adsorbent dosage (0.5g per 100mL) reportedly increased as BG dye concentration was increased from 3mg/L to 100mg/L [54]. In another work [52], it was reported that the zwitterionic adsorbent-based composite successfully removed BG dye at higher than 90% efficiency as the initial concentration was increased from 50ppm to 200ppm.

### Effect of MRHA adsorbent amount

The effect of the MRHA dosage (0.5g to 2.3g) on BG removal efficiency was studied by keeping initial BG dye concentration at 200mg/L, contact time at 1h, shaking rate at 150rpm, and at room temperature. The results from the

experiment are shown in **FIGURE 5**. The removal efficiency of BG dye was observed to increase with an increase in the MRHA dosage from 0.5g to 1.7g. Although the maximum BG dye removal of 95.42% could be obtained at the highest MRHA dosage of 2.3g, the removal efficiency of MRHA did not improve significantly at higher than 1.7g. The removal efficiency increased up to the optimum dosage, which in this case was 1.7g, beyond which the removal efficiency did not change with the increase in the adsorbent dosage. This could be due to the greater difficulty of the BG dye molecules to access the higher energy sites and easier access to lower energy sites as the adsorbent amount was increased in the solution medium [1-2, 4-5, 55-56].

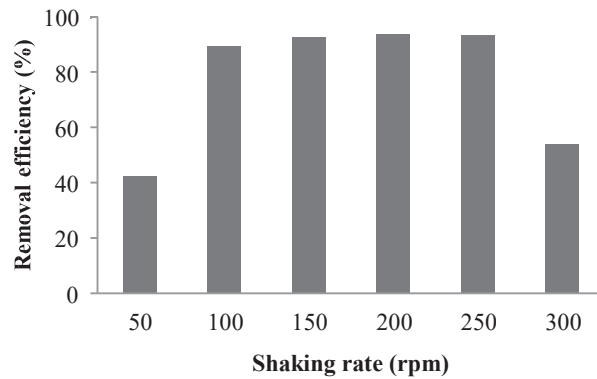


**FIGURE 5.** Effect of adsorbent amount on the adsorption of BG dye by MRHA adsorbents (Experimental conditions: initial BG dye concentration of 200mg/L, contact time of 1h, shaking rate of 150rpm, and room temperature)

### Effect of shaking rate

Shaking rate is one of the main factors influencing the adsorption process of BG dye because it is related to the diffusion or mobility of the BG dye molecules towards the surface of the MRHA adsorbents [26]. In this work, the effect of shaking rate on the BG removal efficiency was studied by varying it from 50 to 300rpm while keeping the other parameters constant. The result shown in **FIGURE 6** shows that increasing the shaking rate from 50 to 150rpm caused the BG dye removal efficiency to increase from 42.35% to 92.74%. The increase was as expected since increasing shaking rate reduced the film boundary layer surrounding the MRHA particles, thus increasing the external film transfer coefficient and agility of the adsorbate. The increasing trend was also observed when kaolin was used to remove BG dye at increasing rate from 610 to 1000rpm [56].

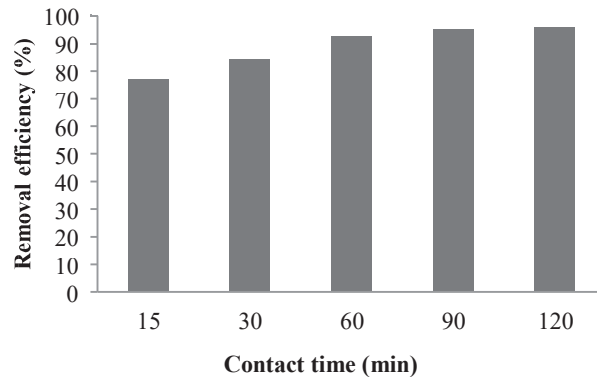
However, increasing the shaking rate of up to 250 rpm in this work did not increase the BG dye removal efficiency at all. In fact, increasing the shaking rate further from 250 to 300 rpm caused the BG dye removal efficiency to start dipping to 53.97%. This happened as a result of higher boundary layer resistance that led to the decrease of the BG diffusion. As the rate was increased beyond a certain threshold (in this case 300rpm), the adsorbed BG molecules started to be stripped off from the adsorbent sites. The kinetic energy that BG molecules gained from the high shaking rate was also greater than the adsorption energy, causing the former molecules to rather mingle in the swirling solution than being adsorbed by the MRHA.



**FIGURE 6.** Effect of shaking rate on the adsorption of BG dye by MRHA adsorbents (Experimental conditions: initial BG dye concentration of 200 mg/L, adsorbent dosage of 2 g, contact time of 1 h, and room temperature)

### Effect of contact time

**FIGURE 7** illustrates the effect of contact time on the BG removal efficiency. The contact time or the time the MRHA remained in the solution was varied from 15min to 120min. The other operating parameters such as the initial BG dye concentration, adsorbent dosage, shaking rate, and temperature were maintained at 200mg/L, 2g, 150rpm, and room condition, respectively. The result shows that BG removal efficiency increased linearly with increasing contact time from 15 to 60min. For the first 15min, about 77% of BG dye was successfully removed, after which, the process began to approach equilibrium slowly and after 90min, the BG removal efficiency started to plateau out.



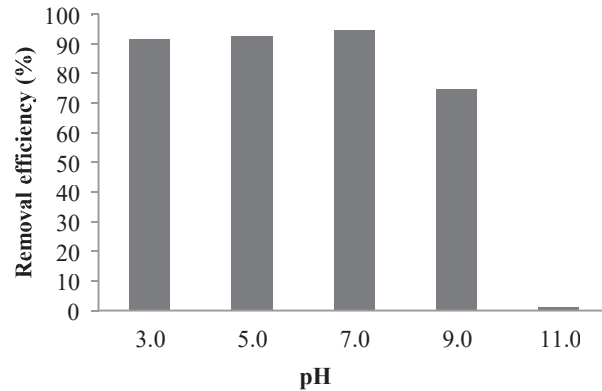
**FIGURE 7.** Effect of contact time on the adsorption of BG dye by MRHA adsorbents (Experimental conditions: initial BG dye concentration of 200mg/L, adsorbent dosage of 2g, shaking rate of 150rpm, and room temperature)

### Effect of pH

**FIGURE 6** shows the effect of pH on the BG dye removal efficiency. The pH of the dye solution was varied from 3 to 11. The result indicated that the MRHA sample was effective in removing the BG dye from acidic aqueous medium. The removal efficiency increased from 91.51% to 94.64% as the pH of the solution was increased from 3.0 to 7.0. The maximum efficiency of 94.64% was achieved at pH 7.0. This is similar to the previously reported achievement using red clay adsorbent [57] in which, the optimum efficiency of 97% was achieved at pH 7.0. The optimum efficiency at pH 7.0 was also observed previously with kaolin [58] and poly(acrylic acid) hydrogel composite [59]. In this work however, the removal efficiency decreased gradually to 77% as the solution became more basic (pH 9.0). MRHA failed to remove BG dye from highly alkaline solution at pH 11. It appears that the electrostatic attraction and repulsion between the BG molecules and MRHA played an important role. It was suggested that the electrostatic attraction was influential in acidic solution in which, the positively charged surfaces of the adsorbent became effective



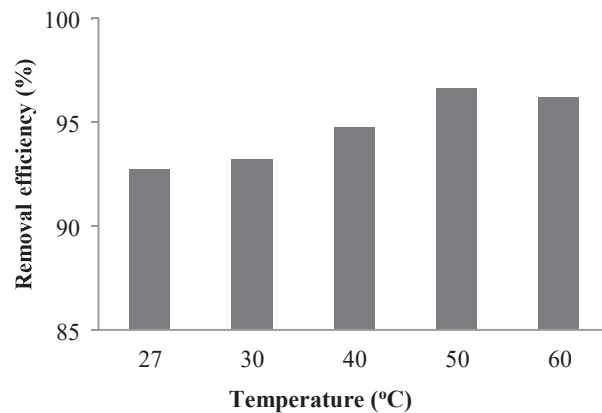
in attracting the negatively charged BG dye molecules [60]. The positively charged surfaces of the adsorbent became the most effective when the solution was neutral since the negatively charged BG dye molecules could concentrate on the solid surface positive charges. At higher pH of alkaline solution however, there would be an increase in the quantity of negatively charged sites on the MRHA, which did not benefit the dye molecules adsorption. Nevertheless, there was an exception when chemically activated carbon (CAC) was used [61]. The CAC was reportedly independent of electrostatic force since the BG dye removal efficiency remained at >97% despite pH increase from 2.0 to 10.5.



**FIGURE 8.** Effect of pH on the adsorption of BG dye by MRHA adsorbents (Experimental conditions: initial BG dye concentration of 200mg/L, adsorbent dosage of 2g, contact time of 1h, shaking rate of 150rpm, and room temperature)

### Effect of temperature

The effect of temperature on the BG removal efficiency is illustrated in **FIGURE 9**. The experiments were conducted at five different temperatures ranging from 27 to 60 C, while keeping the other parameters constant. It was observed that the BG dye adsorption removal increased from 92.74% to 96.65% as the temperature was raised from 27 to 50 C. The removal efficiency decreased slightly to 96.20% with further increase in the temperature from 50 to 60 C. The optimum BG dye adsorption efficiency of 96.65% was found to be at 50 C. The operating temperature did have a significant effect on the adsorption process since it influenced the diffusion of dye molecules at the external boundary layer and inside the adsorbent pores. The result indicated that BG adsorption was endothermic at 27 to 60 C during which, the mobility and dispersion level of BG dye molecules improved the degree of interaction between BG dye molecules and MRHA. The process changed slightly into exothermic process at 50 to 60 C, during which the increase in temperature was not favorable, thus decreasing removal efficiency was observed.



**FIGURE 9.** Effect of temperature on the adsorption of BG dye by MRHA adsorbents (Experimental conditions: initial BG dye concentration of 200 mg/L, adsorbent dosage of 2 g, contact time of 1 h, and shaking rate of 150 rpm)

## MRHA adsorption characteristic

The adsorption isotherms of BG dye molecules with MRHA sample were determined in order to understand their adsorption characteristic. The characteristic of BG dye adsorption into MRHA sample was compared with the three well known adsorption isotherm models namely, Langmuir, Freundlich and Temkin. The calculated Langmuir, Freundlich and Temkin isotherm constants and the corresponding coefficient of determination are tabulated in **TABLE 1**. The Langmuir isotherm model assumes that the homogeneous formation of monolayer adsorbate occurs at the outer surface of the adsorbent, while Freundlich isotherm assumes that multilayer adsorption occurs inside the heterogeneous surfaces of the adsorbent. Temkin isotherm on the other hand, assumes that there is a linear decrease in the adsorption heat and that the adsorption process corresponds to the uniform distribution of binding energies. In other words, Temkin isotherm suggests that there is strong adsorption, equivalent to the level of chemical adsorption, taking place between the adsorbate and adsorbent.

The value of  $R_L = 0.0085$  indicates that the adsorption was favorable. The coefficient of determination,  $R^2$  of  $>0.8$  suggests that all the three phenomena described by the Langmuir, Freundlich and Temkin isotherm models might occur in the adsorption of BG inside the MRHA micro structures. However, Temkin isotherm model described the adsorption behavior of BG the best, based on the highest coefficient of determination presented in the table. The result indicated that there was strong interaction between BG dye molecules and the surface of MRHA adsorbent due to the uniform distribution of binding energies in the latter. The Temkin isotherm constant and the Temkin constant related to the heat of adsorption were 1.167L/mg and 0.385J/mol, respectively. These values are considered to be significant in promoting the surface binding of BG dye molecules onto the high energy sites of MRHA.

**TABLE 1.** Summary of isotherm model constants and coefficient of determination for BG dye adsorption onto MRHA adsorbent

Langmuir isotherm	0.131	5.29	0.0085	0.803
Freundlich isotherm	41.73	0.009		0.807
Temkin isotherms	1.167	0.385		0.811

## CONCLUSIONS

MRHA was prepared, characterized and applied for the removal of synthetic BG dyes in aqueous medium. The highly porous MRHA sample with fractured and elongated cell walls around the pores demonstrated its potential for BG dye removal from industrial effluent. BG molecules were thought to form homogeneous monolayer formation along the MRHA micro pore, whereas multilayer formation occurred inside meso and macro pores. The BG molecules adhered to the MRHA pore surfaces strongly as suggested by the Temkin isotherm model.

The removal efficiency of BG by MRHA was found to increase and decrease depending on the degree and variation of the operating parameters being investigated. Decreasing the initial BG dye concentration, increasing the MRHA dosage and contact time led to the increase in BG removal efficiency. The shaking rate of 150rpm was acceptable in order to establish the surface binding of BG dye molecules onto the high energy sites of the adsorbent so that more than 90% removal efficiency could be achieved. The maximum BG dye removal efficiency of 96.65% was achieved at pH 7.0, initial dye concentration of 200mg/L, adsorbent amount of 2g, shaking rate of 150rpm, contact time of 60min, and temperature of 50 C.

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