



Research Paper

Removal of Acrylic dye Blue-5G from Aqueous solution by Adsorption on Activated Carbon Prepared from Maize cobs

El-Sayed G.O.*, Aly H.M., Hussien S.H.M.

Chemistry Department, Faculty of Science, Benha University, Benha, EGYPT

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Abstract-The adsorption of acrylic dye Blue-5G (B-5G) from aqueous solution by activated carbon prepared from Maize cobs was studied. The activated carbon had been prepared by chemical activation with H₃PO₄. The adsorption was carried out in batch system. Different experimental parameters like initial pH, contact time, initial concentration, adsorbent dose, stirring rate and temperature on the adsorption of (B-5G) were optimized. Langmuir and Freundlich isotherm models were used to illustrate the experimental isotherms and isotherm's constants. The pH of dye solution on the range of 8-10 was found favorable for the removal of (B-5G) at equilibrium time (90 min) and adsorbent dose 0.1g with stirring rate (200 rpm).

Keywords: activated carbon, blue-5G, adsorption, Langmuir, Freundlich, isotherm.

Introduction

The wastewaters discharged from textile industries are the most pollutant wastes both as volume and waste composition. The dangerous wastes cause the biggest ecological problems^[1]. Color is the first contaminant to be recognized in wastewater and has to be removed before discharging into waterways or land. The treatment of spent textile dyeing wastewater by traditional methods has proven to be ineffective for many wastewater treatment facilities. Additional textile treatment methods; such as combinations of biological, physical, and chemical methods including coagulation/flocculation^[2], electrochemical oxidation^[3] and adsorption^[4], reverse osmosis^[5], ozone^[6] and oxidative/reductive chemical processes^[7]; are all techniques that can be used for treating textile wastewater. However, many of these technologies are cost prohibitive, especially when applied for treating large waste streams. Consequently, adsorption techniques seem to have the most potential for future use in industrial wastewater treatment^[8] because of their proven efficiency in the removal of organic and mineral pollutants and for economic considerations. The most widely used adsorbent for this purpose is activated carbon. Many types of agricultural residues are used for preparing activated carbon used for dye removal from industrial water.

The aim of the present study was to study the ability of an activated carbon to remove an acrylic dye B-2G from aqueous solution. Different parameters which affecting adsorption were studied and the optimum conditions for effective removal of the dye were determined. The biosorption equilibrium was modeled by using the Langmuir isotherm model and isotherm constants were evaluated at different temperatures.

Materials and solutions

Acrylic Dye solutions

The acrylic dye used throughout this study was Blue-5G (B-5G). The dye was obtained from the market as a commercial sample used for dyeing acrylic fibers. It was used as received without any treatment. The chemical structure of the dye is shown in Fig.1.

Preparation of activated carbon (AC)

Maize cop had been washed with distilled water and dried in a drier for 2 h at 105 °C until a constant weight was reached. It was then ground in a ball mill and sieved to particle sizes ranging from 0.3 to 0.5 mm. The sieved raw material was then activated by chemical treatment with concentrated H₃PO₄. The activation was carried out by pyrolysis in a muffle furnace in absence of air at 500°C for 2 h and then the sample was soaked in the acid (2 carbon:1 acid) for 24

h. After decantation, the sample was washed with distilled water several times and then dried in an oven at temperature of 110 °C overnight. The activated carbon (AC) was kept in a closed container till use.

Preparation of the investigated solutions

Stock solutions of the dye was prepared by taking appropriate weight of this dye in 250 ml round bottom flask and completed to (1000 ml) with distilled water and stored for later use. Analytical grade reagents were used for preparation of the dye solutions. In all experimental work, distilled de-mineralized water was used. All the chemicals used in the study were of analytical reagent grade and all experiments were performed at room temperature (25±1 °C).

Adsorption studies

The batch mode was used to study the adsorption of acrylic dye (B-5G) on AC. Sorption studies were conducted in a number of plugged Pyrex glasses Erlenmeyer containing a definite volume (25 ml in each case) of solutions of the dye of desired concentrations (2.5-50 mg/L). For the studies, 0.1 g of adsorbent was treated with 25 ml of dye solution. The flasks were stirred at a 200-rpm constant stirring rate for 2 hr to ensure equilibrium was reached. Samples (2 ml) were taken before mixing the sorbent and the dye solution and at pre-determined time intervals. The dye solution was separated from the sorbent by centrifugation at 5000 rpm for 5 min.

The dye content in the test flask was evaluated by measuring its absorbance at λ_{max} (632nm) value characterizing the dye by using a JascoV-530 (UV-Vis) spectrophotometer (Japan). The different tested effects were contact time, pH of the solution, ionic strength, stirring rate, adsorbent dose, initial dye concentration and temperature. The flasks with their contents were shaken for different adsorption times (5, 10, 20, 30, 45, 60, 90 and 120 min) at the temperatures of 25, 35 and 45 °C with different initial concentrations (2.5, 5, 10, 20, 30 and 50 mg/L) with different adsorbent dose ranging (0.05, 0.1, 0.15, 0.2 and 0.25 g). The effect of pH was investigated at 25°C. The initial pH (1.5, 3.5, 6.5, 9 and 11.5) of solutions were adjusted using 0.1 M HCl and 0.1 M NaOH solutions. The effect of salinity on the dye uptake was carried out by adding different quantities of KCl to the dye solutions with concentrations ranging (4, 12, 20 and 28 g/l) and this performed before adding the adsorbent AC then the solution had been treated by a fixed amount of AC (0.1g). The concentration of B-5G in the supernatant solution before and after the adsorption was determined with 1 cm light path quartz cells. Also the effect of stirring rate was conducted at rates of (0, 200 and 400 rpm). Uptake values were determined as the difference between the initial dye concentration and that in the supernatant. The data obtained in batch mode studies was used to calculate the equilibrium dye adsorptive quantity.

$$q_e = \frac{(C_0 - C_e) v}{m}$$

where q_e is the amount of dye adsorbed onto per unit weight of beer yeast in mg/g, v the volume of solution treated in L, C_0 the initial dye concentration in mg/L, C_e the equilibrium dye concentration in mg/L, and m is the dry weight of the adsorbent in g. The percent of dye removal was evaluated from the equation:

$$\% \text{ Removal} = \frac{(C_0 - C_e)}{C_0} \times 100$$

Results and discussion

Effect of contact time

Equilibrium time is an important parameter for an economical wastewater treatment system. For a given substance to be effective as an adsorbent of organic dyes, its adsorption rate must be fast and quantitative. The effect of contact time on the amount of dye adsorbed on the AC was investigated at 25 ml of dye solution of initial concentration of 20 mg/L at room temperature. The system was subjected to an agitation speed of 100 rpm for 90 min. Fig. 2 shows a rapid adsorption of B-5G on AC at the initial stages of the adsorption and equilibrium was attained within about 60 min. Such uptake indicates a high degree of affinity towards dye molecules via chemisorptions^[9]. After the rapid uptake, the capacity of the adsorbent became exhausted and the adsorption would be replaced by the transportation of dye from the external sites to the internal sites of the adsorbent particles. Therefore, the uptake rate began to drop down, which can be explained by intra-particle diffusion model^[10]. The equilibrium was attained, the removal percent was evaluated as 52.0%.

Effect of pH on efficiency of dye removal

The pH of the dye solution plays an important role in the whole adsorption process and particularly on the adsorption capacity. The variation in adsorption capacity in the studied pH range was largely due to the influence of pH on the surface adsorption characteristics of the AC. The observed relatively low adsorption rate of the dye on AC at acidic pH values may be because the surface charge became positively charged, thus making (H⁺) ions compete effectively with dye molecules causing a decrease in the amount of dye adsorbed. As the pH of the adsorption solution was increased, the negative charges on the surface increased. This would attract the positively charged functional groups located on the dye^[11]. The effect of pH on the dye removal onto AC is shown in Fig. 3. As shown in Fig. 3, the dye removal percent on AC increases from about from 33% to 92% when pH increases from 1.5 to 10.5. Similar observations were reported for adsorption of dyes indicating that the adsorbent has a net positive charge on its surface^[12]. The effect of pH on the adsorption capacity of AC activated carbons is in agreement with the previously reported results for adsorption of different chemicals from aqueous solutions by sunflower oil cake activated carbon^[13], apricot stone activated carbon^[14], coir pith carbon^[15], hazelnut husk activated carbon^[16], walnut shell activated carbon^[17], hazelnut shell^[18], different

bagasse pith activated carbons^[19], *euphorbia rigida* activated carbon^[20].

Effect of ionic strength on efficiency of dye removal

Generally, various salts and metal ions exist in dye containing wastewater. The salts lead to high ionic strength, which may affect the dye adsorption onto adsorbents. The influence of salinity on dye sorption by AC was investigated to determine solution chemistry effects. Different values of KCl (4, 12, 20 and 28 g/L) were added to dye solutions with a constant dye concentration (20 mg/L). The effect may be varied for different dyes and adsorbents. Generally, with increasing the salinity, there is usually a little decrease in dye removal percent which can be attributed to presence of sorption sites of different affinities. As shown in the Table 1, there is a slight decrease in dye adsorption with increasing the salinity by adding KCl to the dye solution at equilibrium for adsorption onto AC. This could be attributed to the competitive effect between dye ions and cations from the salt for the sites available for the biosorption process. And, with the ionic strength increasing, the activity of B-5G and the active sites decreases, so the adsorption amount of B-5G decreases^[21]. Such behavior was detected in the literature^[22, 23].

Effect of stirring rate on efficiency of dye removal

The effect of the stirring rate on the dye removal is shown in Fig. 4. The stirring speed was varied up from 0 to 400 rpm, keeping other parameters constant. The results clearly show that there was a definite improvement in the dye removal efficiency with the increase of the stirring rate from 0 to 400 rpm. Depending upon the degree of agitation of the fluid particle system, the rate of adsorption is controlled either by film diffusion or pore diffusion. At lower agitation speeds, the fluid film around the particle is thicker and the film diffusion seems to be rate limiting step. The adsorption kinetics is influenced by low mass transfer of adsorbate to the internal surface of particle. It is likely that at higher agitation speeds, the film diffusion increases to a maximum value and pore diffusion thus becomes the rate controlling step^[24]. This behavior is observed in other works dealing with the dye removal by adsorption^[25].

Effect of adsorbent dose on efficiency of dye removal

The solid/solution ratio is an important factor determining the capacity of a sorbent in a batch sorption study. The effect of sorbent dosages on the percentage removal of (B-5G) has been shown in Table 2 for AC. It followed the predicted pattern of increasing percentage sorption as the dosage was increased. This is probably because of the resistance to mass transfer of dye from bulk liquid to the surface of the solid, which becomes important at high adsorbent loading in which the experiment was conducted. It can be clearly seen that the removal of (B-5G) increased with increasing the amount of AC. However,

the amounts of adsorbed dye per unit weight (q_e) of the AC decreased with increasing the solid/solution ratio. The removal of the dye increased when the dosage was changed from 2 to 10 g/L. As expected, at constant initial concentration of dye, increasing the sample dose provides a greater surface area and larger number of sorption sites and hence enhancement of dye uptake^[26]. The primary factor explaining this characteristic is that adsorption sites remain unsaturated during the adsorption reaction whereas the number of sites available for adsorption site increases by increasing the adsorbent dose^[27].

Effect of initial dye concentration on efficiency of dye removal

The effect of the initial dye concentration on the dye adsorption rate was investigated in the range (5–50 mg/L) at room temperature without changing the initial pH of the medium. The results presented in Fig. 5 show that the percentage of removal decreased with increasing initial dye concentration for AC. The lower uptake at higher concentration resulted from an increased ratio of initial adsorption number of moles of the dye to the available surface area; hence fractional becomes dependent on initial concentration. For a given adsorbent dose the total number of available adsorption sites is fixed thereby adsorbing almost the same amount of adsorbate, thus resulting in a decrease in the removal of adsorbate corresponding to an increase in initial adsorbate concentration (saturation of the adsorbent). Similar results were also reported by other researchers^[28].

Effect of temperature on the adsorption capacity of AC

The adsorption processes clearly showed that the adsorption capacity for AC increased with increasing the temperature from 25 to 45 °C. Fig. 6 shows the relation between % dye removal and the temperature by using AC. An increase in adsorption capacity with increasing temperature denotes that some kind of endothermic chemical interactions may be taken place during adsorption process^[19]. Additionally, the temperature has two major effects on the adsorption process. Increasing temperature is known to increase the rate of diffusion of the adsorbate molecules across the external boundary layer and in the internal pores of the adsorbent particle, owing to the decrease in the viscosity of the solution. In addition, changing temperature will change the equilibrium capacity of the adsorbent for a particular adsorbate. The increasing temperature may increase the tendency of disaggregation and so the uptake of the monomers of B-5G.

Adsorption Isotherms

The adsorption isotherms are used to indicate how the adsorption molecules distribute between the liquid phase and the solid phase at equilibrium. The analysis of the isotherm data is important to find the suitable model^[29].

Langmuir isotherm

Langmuir's isotherm model suggests that uptake occurs on homogeneous surface by monolayer sorption assuming uniform energies of adsorption onto the surface and no transmigration of adsorbate in the plane of the surface. The linear form of Langmuir isotherm equation is represented by the following equation:

$$\frac{C_e}{q_e} = \frac{1}{bQ_0} + \frac{C_e}{Q_0}$$

where q_e is the amount adsorbed at equilibrium time (mg/g), C_e is the equilibrium concentration of the adsorbate ions (mg/L), Q_0 and b are Langmuir constants related to maximum adsorption capacity (monolayer capacity) and energy of adsorption, respectively. When C_e/q_e is plotted against C_e , a straight line with slope $1/Q_0$ and intercept $1/bQ_0$ is obtained (Fig. 7), which shows that the adsorption of dye follows Langmuir isotherm model ($R^2=0.9991$). Values of Q_0 and b were calculated from the intercept and slope of the linear plot and are presented in Table 3. The essential characteristics of the Langmuir equation can be expressed in terms of a dimensionless separation factor, R_L , defined as^[30]:

$$R_L = \frac{1}{(1 + bC_0)}$$

where C_0 is the highest initial solute concentration, b is the Langmuir's adsorption constant (L/mg). The R_L value implies the adsorption to be unfavorable ($R_L > 1$), linear ($R_L=1$), favorable ($0 < R_L < 1$) or irreversible ($R_L=0$). The values of R_L was found to be <1 (Table 3) which confirmed that AC is favorable for adsorption of B-5G under experimental conditions used in this study.

Freundlich isotherm

The Freundlich isotherm^[31] is the earliest known relationship describing the sorption equation. This fairly satisfactory empirical isotherm can be used for non-ideal sorption that involves heterogeneous sorption and is expressed by the following equation:

$$q_e = K_F C_e^{1/n}$$

The equation may be linearized by taking the logarithm of both sides:

$$\ln q_e = 1/n \ln C_e + \ln K_F$$

where K_F and n are Freundlich constants, n giving an indication of how favorable the adsorption process is and K_F (mg/g)(L/mg) $^{1/n}$ is the adsorption capacity of the adsorbent. K_F can be defined as the adsorption or distribution coefficient and represents the quantity of dye adsorbed onto adsorbent for a unit equilibrium concentration. Fig. 8, shows a straight line with a slope of $1/n$ and an intercept of $\ln K_F$ when $\ln q_e$ is plotted against $\ln C_e$. The slope $1/n$ ranging between 0 and 1, is a measure of adsorption intensity or surface heterogeneity, becoming more heterogeneous as its value gets closer to zero^[32]. K_F and $1/n$ were calculated and recorded in Table 3. The results suggest that Langmuir isotherm is more suitable for B-5G

adsorption compared to Freundlich isotherm ($R^2=0.9525$).

Conclusion

The results of different experiments showed that activated carbon derived from maize cobs has an ability to adsorb B-5G dye from aqueous solutions. Different variables, such as contact time, adsorbent dose, solution pH and temperature influenced the adsorptive quantity. The sorption process was found to be pH dependent and the optimum pH is > 10 . The adsorption process has nearly reached equilibrium in 90 min. The experimental data of adsorbing disperse 2BLN dye are fit well to Langmuir isotherm model more than Freundlich model and the maximum adsorptive quantity of activated carbon was 2.32 mg/g according to Langmuir model. The adsorptive process is endothermic and greatly affected by temperature.

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Table 1. Effect of addition of KCl on B-5G dye removal by AC

KCl (g/L)	Dye Removal %
4	51.3
8	50.4
12	48.0
20	46.0
28	43.2

Table 2. Effect of initial AC dose on dye removal %

Adsorbing Dose (g/L)	Dye Removal %	Removal Efficiency q_e (mg/g)
2	36.0%	9.00
4	50.0%	6.25
6	66.2%	5.51
8	77.3%	4.83
10	86.0%	4.30

Table 3. Langmuir and Freundlich isotherm parameters for adsorption of B-5G on AC

Langmuir Constants			Freundlich Constants		
Q_0 (mg/g)	b (L/mg)	R^2	K_F	$1/n$	R^2
2.32	0.3216	0.9991	0.8203	0.2774	0.9525

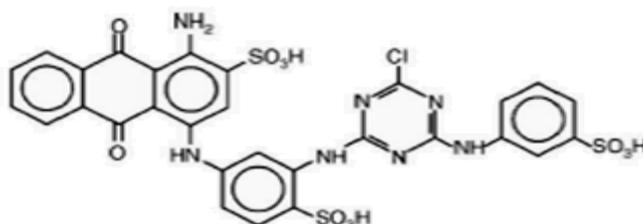


Fig. 1. Chemical Structure of Blue-5G

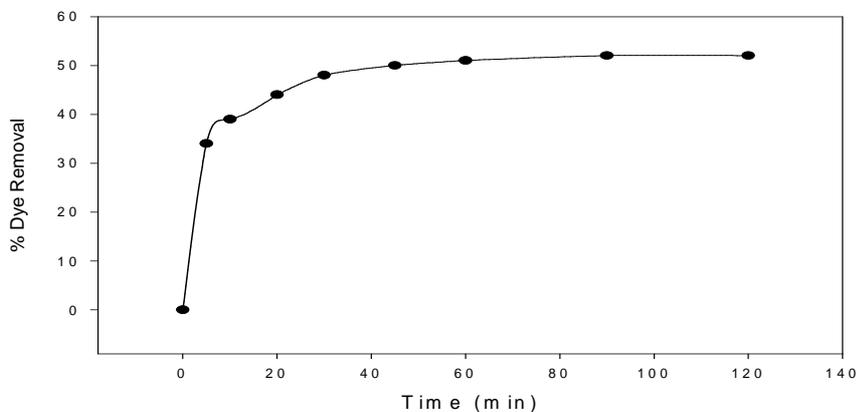


Fig. 2. Effect of time on percent of B-5G dye removal on AC

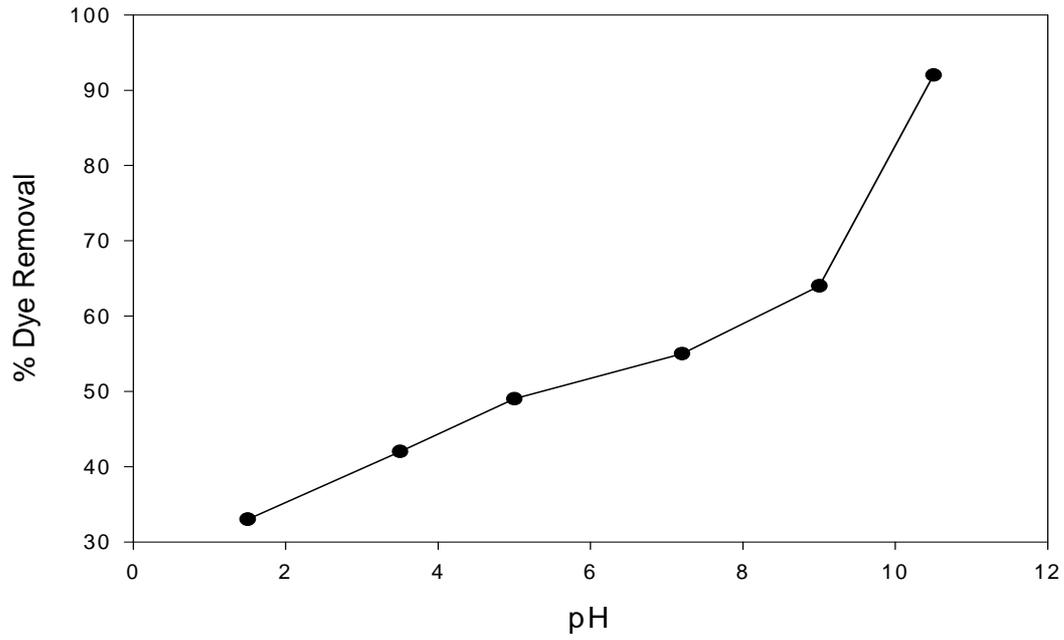


Fig. 3. Effect of pH on percent of B-5G dye removal on AC

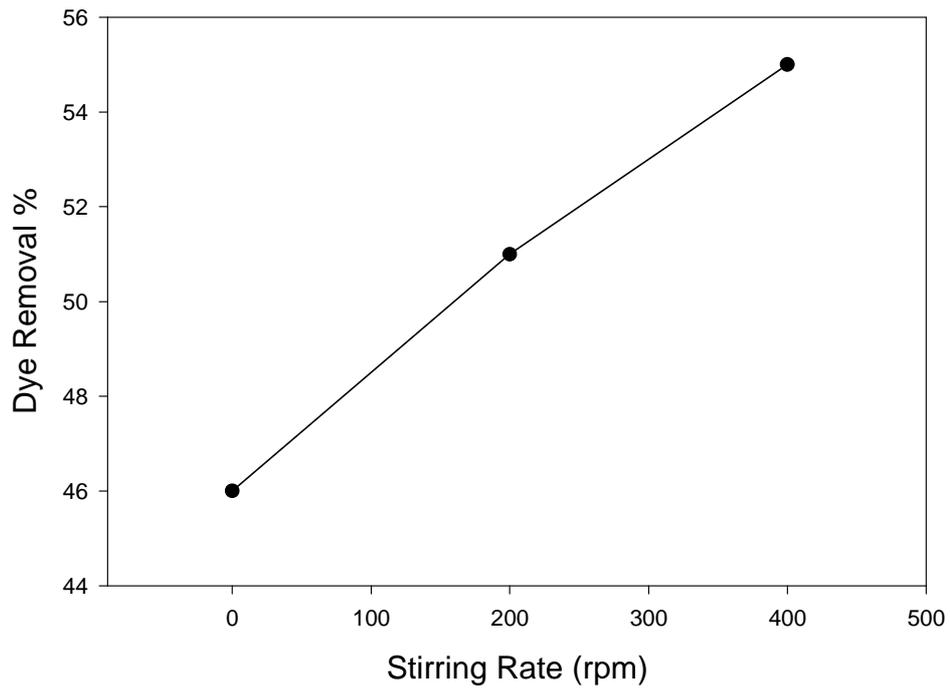


Fig. 4. Effect of stirring rate on percent of B-5G dye removal on AC

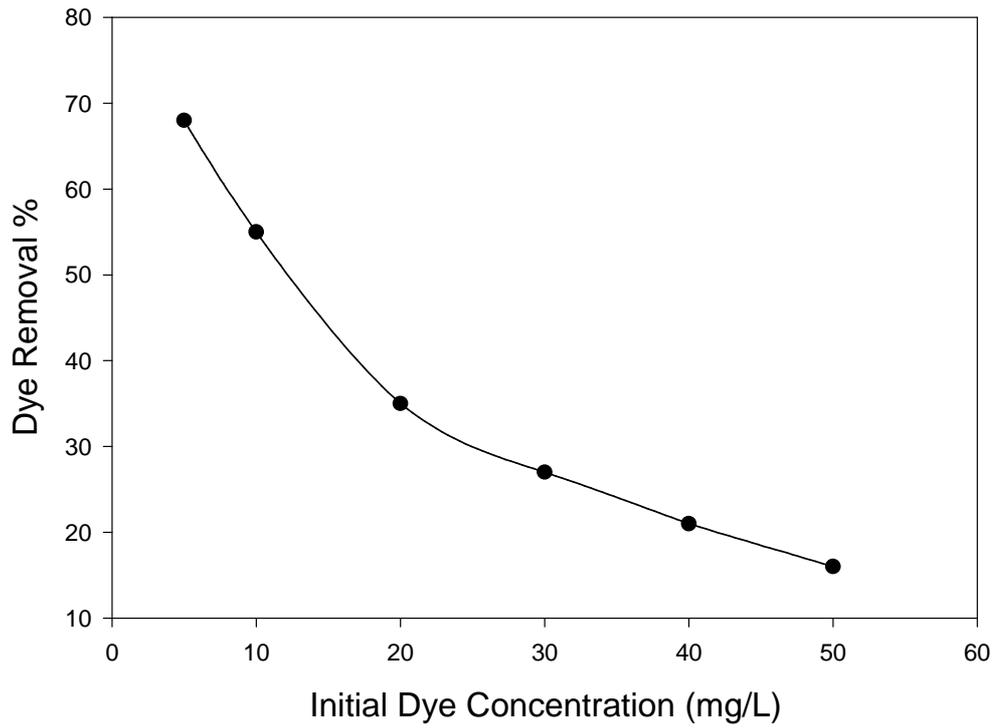


Fig. 5. Effect of initial dye concentration on percent of B-5G dye removal on AC

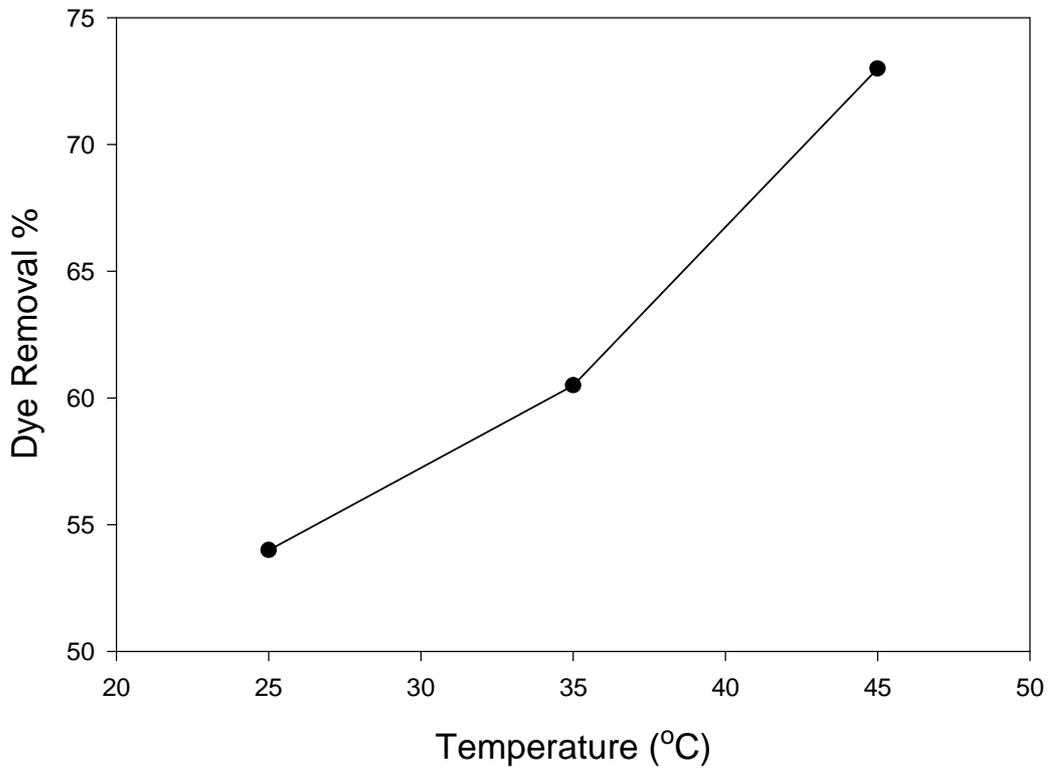


Fig. 6. Effect of temperature on percent of B-5G dye removal on AC

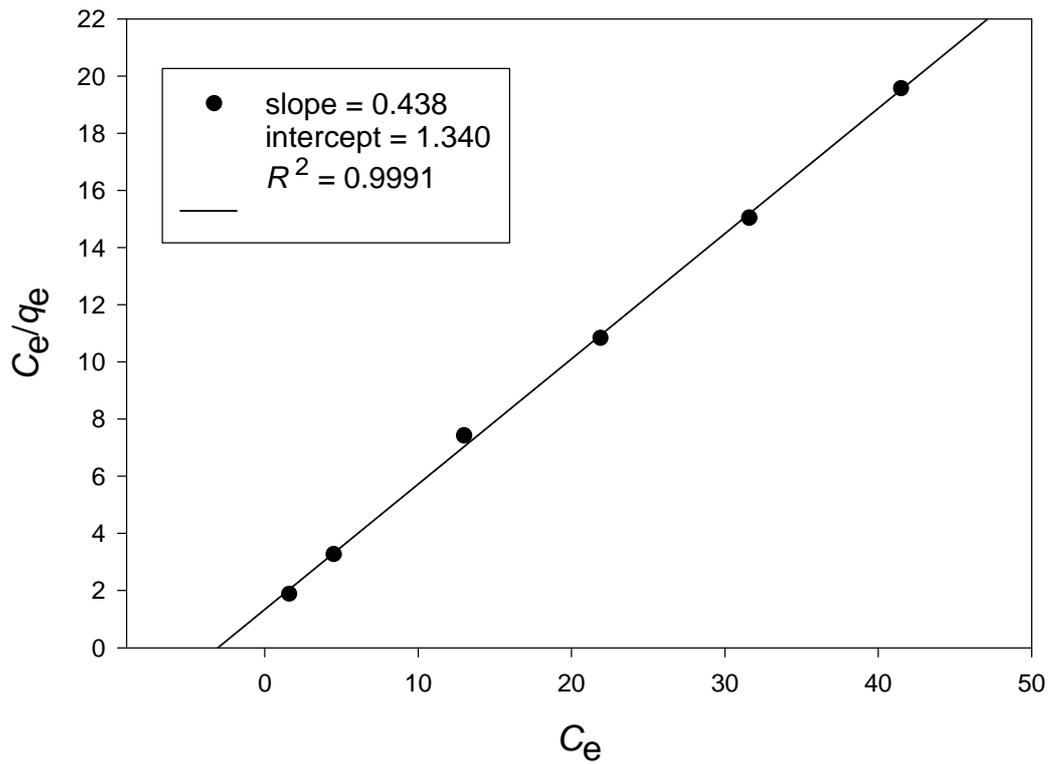


Fig. 7. Langmuir isotherm plot of B-5G dye adsorption on AC

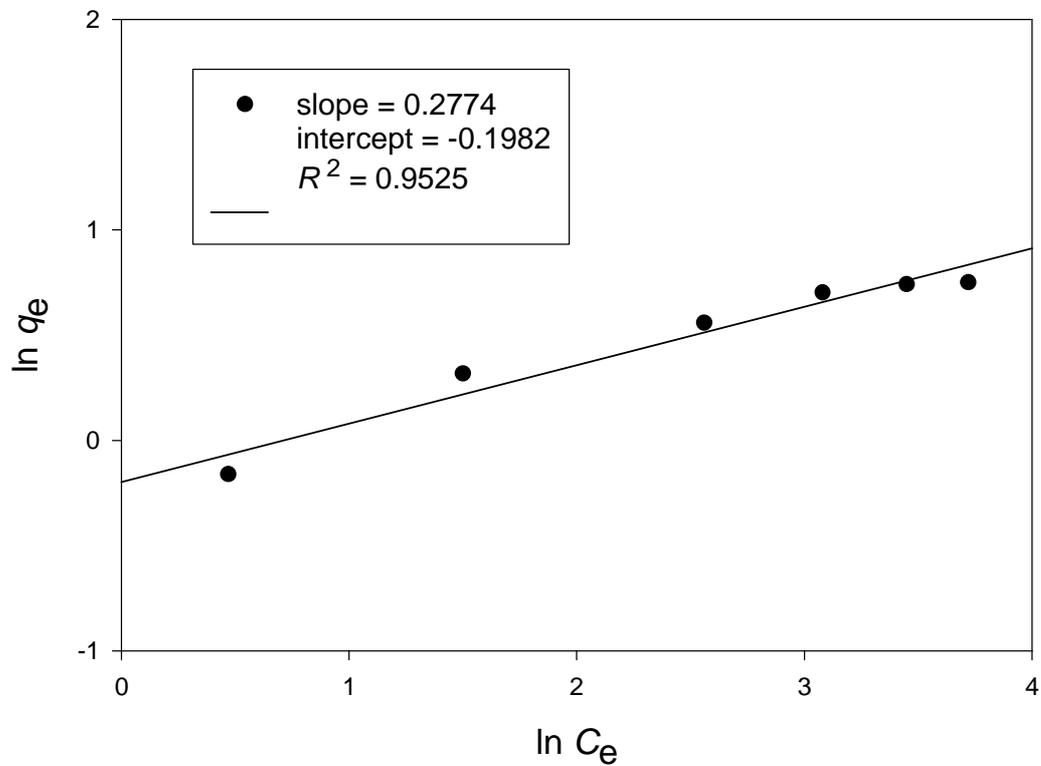


Fig. 8. Freundlich isotherm plot of B-5G dye adsorption on AC