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Kinetic and Thermodynamic Study of the Elimination of Remazol Black on Activated Carbon Based on *Ricinodendron heudelotii* Shells

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Abstract

Activated carbon made from the shells of *Ricinodendron heudelotii* was used to remove the remazol black dye in aqueous solution. The results of the characterization of this carbon revealed that it is microporous, with a basic global surface (0.337 mmol/L) and a specific surface of 612 m^2/g. The prepared carbon therefore has excellent adsorbent properties. Kinetic and thermodynamic studies were carried out to describe the adsorption mechanism of remazol black on this carbon. It appears from this study that the pseudo-second-order kinetic model is the best suited to describe this adsorption phenomenon with an equilibrium time of 200 min. The adsorption equilibrium study revealed that Langmuir and Freundlich models can help to describe the adsorption process. We note that the optimum pH and optimum mass for the removal of 20 mg/L of remazol black are 3 and 0.25 g, respectively. This carbon made it possible to eliminate more than 98% of the remazol dye in aqueous solution. The thermodynamic study revealed that the adsorption is of the physisorption type, spontaneous and endothermic.

Keywords

Activated Carbon, *Ricinodendron heudelotii*, Dye Removal, Optimization, Adsorption Isotherm

1. Introduction

Indispensable to all forms of life, water is the element that promotes public health and socio-economic development [1]. Human activity in the world is the

cause of several forms of pollution, in particular water pollution, which continues to grow. Among these activities, the textile industry, in addition to using a large quantity of water, releases huge amounts of dye into the natural environment [2]. Indeed, after use, this water is discharged into the natural environment with or without adequate treatment. These discharged colored waters are pollutants affecting the soil and especially the aquatic environment with the risk of many diseases including cancers [3] [4]. The treatment or even the elimination of traces of dyes before any rejection in nature is essential. Several physical, chemical and biological methods have been developed to treat dye-laden effluents. Among these methods we can cite coagulation-flocculation, biodegradation, membrane filtration, chemical oxidation, ozonation, ion exchange, electrochemical methods, adsorption [5] [6] [7]…

Adsorption is by far the most favorable technique used for dye removal due to its ease of design, efficiency and relatively simple operation [8]. The principle of adsorption is essentially based on the use of a biomaterial as an adsorbent in order to trap the large molecules of pollutants, hence the need to find an effective and affordable adsorbent.

Various dyes were removed by adsorption on activated carbon including methylene blue [9], methyl orange [10], Rhodamine B [11] and indigo carmine [12]. The control of the kinetics of adsorption and the thermodynamic parameters are necessary for the control of the phenomenon of adsorption of an adsorbent on an adsorbate in order to optimize the study and to think of the use of the process on a large scale.

In previous works, we prepared activated carbons based on the shells of *Ricinodendron heudelotii*. The results showed the adsorbent capacities of this carbonaceous material [13].

The main objective of this study is to study the elimination of remazol black on activated charcoal based on the shells of *Ricinodendron heudelotii*. It will be a question of determining the physico-chemical characteristics of the prepared carbon then of carrying out the kinetic and thermodynamic studies of the elimination of this azo dye on this prepared carbon.

2. Material and Methods

2.1. Preparation of Activated Carbon

The preparation of the carbon was done by the method of chemical activation with the activating agent, sodium hydroxide (NaOH). To do this, the *Ricinodendron heudelotii* shells (**Figure 1**) were washed with tap water and then with distilled water to rid them of all impurities. Subsequently, the clean shells are dried in an oven at 105˚C for 24 hours. They are then crushed to have grains of dimension 3 mm. The ground shells are impregnated with 2 M NaOH in the proportions of 100 g for 200 mL of 2 M NaOH. The hermetically sealed mixture is maintained for 24 hours with stirring (1500 rpm). Once the activation step is complete, the mixture is filtered and baked at 105˚C for 1 hour. The impregnated

Figure 1. Picture of *Ricinodendron heudelotii* shells.

and steamed shells were recovered in a quartz crucible to be calcined in a Nabertherm muffle furnace at 500˚C for one hour (60 min). The carbon obtained is taken to a desiccator for complete cooling, and then weighed. It is then washed to neutral pH, and then steamed at 105˚C for 1 hour. We thus obtain activated carbon based on *Ricinodendron heudelotii* (ACRH).

2.2. Characterization of Activated Carbon

- Analysis of physical surface properties of carbons included determination of total specific surface area, total pore volume and pore size distribution. The pore properties of carbon were measured by adsorption of nitrogen at 77 K in an automatic adsorption system (micromeritics tristar II). Before the adsorption of the nitrogen, the carbon samples are treated under vacuum at 300˚C for 4 hours in order to eliminate the adsorbed gases such as water vapours. The acid functions were determined according to Boehm's method [14]. The protocol is as follows: 1 g of carbon is brought into contact with 50 mL of basic solution of $NaHCO_3$, Na_2CO_3, and NaOH with constant stirring for 72 hours. The mixture is then filtered. 10 mL of the filtrate is dosed with 0.1N hydrochloric acid solution. This method makes it possible to classify surface groups of acid type: strong carboxylic acids (-COOH), medium carboxylic acids (-COO-) and weak carboxylic acids (-OH).

For the determination of basic type of surface groups, 50 mL of a 0.1 N hydrochloric acid solution is brought into contact for the same time with 1 g of carbon. The filtrate is finally titrated with a 0.1N sodium hydroxide solution in the presence of phenophthalein. The quantification of the acid and basic groups was carried out using the following formula:

$$\text{Surface functions}\left(\frac{\text{meq}}{\text{g}}\right) = \frac{n_{reacts}}{m_{AC}} \times 100 \quad (1)$$

n_{reacts}: Quantity of matter (acid or base) which reacts

m_{AC}: Mass of activated carbon (g)

- The determination of the pHpzc was carried out by introducing a volume of 50 mL of NaCl solution (0.01 M) into reactors containing 0.15 g of ACRH. to be analyzed. The pH of each reactor is adjusted (variation of values between 2 and 12, in steps of 1) by adding 0.1 M NaOH or HCl solution. Stirring is maintained for 48 hours using a multi-stirring system, at a temperature of 25˚C. The content of these reactors is then filtered, then the final pH of each mixture is then measured. $\Delta pH = pH_f - pH_i$ is calculated and the graph $\Delta pH = f(pH_i)$ is drawn. The pHpzc is defined as being the point of intersection of the curve with the abscissa axis.
- The scanning electron microscope allowed us to visualize the morphological structure of activated carbon. The device used is Hirox SH-4000.
- The ash content was estimated by referring to the ASTM method (American Standards Technology Method) [15]. A mass m of each carbon sample is placed in a crucible and then deposited in the muffle furnace at 650˚C until there is no more detectable mass loss. It happens after 3 h.

The ash content is obtained by the relationship:

$$\text{Ash content} = \frac{m_i - m_f}{m_i} \times 100 \quad (2)$$

where, m_i and m_f are respectively the mass of charcoal before and after firing.

2.3. Characteristics of the Adsorbate

Remazol Black (RB), also known as RB5, is a synthetic azo dye that comes in the form of a black crystalline powder. It is used in the textile field for coloring synthetic fibers and also in scientific research due to its unique properties. The properties of remazol black are summarized in **Table 1**.

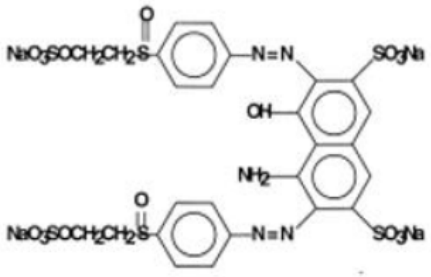
2.4. Kinetic study of Remazol Adsorption

For this study, 0.2 g of activated carbon is brought into contact with 20 mL of remazol black solution of different concentrations C_0. The flasks are placed on magnetic stirrers and stirred at regular time intervals. After these times, the mixtures are removed and filtered. The initial solution and the filtrate are then assayed by UV spectrometer. A given time corresponds to a residual concentration C_r. The amount of remazol black adsorbed is calculated from the following equation:

$$q_t\,(\text{mg/g}) = \frac{C_0 - C_t}{m} \times V \quad (3)$$

where, C_0 and C_t are respectively the initial and residual concentrations at a time t of remazol black in mg/L, V, the volume of the solution of remazol black in L, m, the mass of activated carbon in g and q_t the quantity of remazol black adsorbed

Table 1. Characteristics of remazol black.

Common name	Remazol black
IUPAC name	CI reactive black 5 Tetrasodium;4-amino-5hydroxy-3,-bis[[4-(2-sulfonatooxyethylsulfonyl) phenyl] diazenyl]naphthalene-2,7-disulfonate
Family	Azo dye
Formula	$C_{26}H_{21}O_{19}N_5S_6Na_4$
Structure	
Molar mass	999.8 g/mol
λ_{max}	593 nm
Solubility	82 g/L in water at 293 K

at a specific time in mg/g.

2.5. Influence of Physicochemical Parameters

pH is an important parameter in dye removal processes by adsorption. By fixing the concentration of the solution, the mass of adsorbent, the equilibrium temperature, and by carrying out the study until the equilibrium time, let us vary the pH of the solution. The pH is adjusted using sulfuric acid solution (H_2SO_4) or soda (NaOH) to obtain solutions whose pH varies from 2 to 12.

The percentage of elimination is determined according to the formula:

$$\%\text{Removal} = \frac{C_0 - C_r}{C_0} * 100 \quad (4)$$

where C_0 and C_r are respectively the initial concentration before treatment and the residual concentration after treatment.

As for the pH, we will proceed in the same way, keeping constant all the parameters except one, whose influence on the elimination of remazol black by the activated carbon will be studied. Thus, the influence of the mass of carbon used, the initial concentration of remazol black and the temperature of the solution was studied.

3. Results and Discussion

3.1. Characteristics of Activated Carbon

3.1.1. SEM Image

The SEM analysis allowed us to know the morphological structure of the ACRH.

The analysis result is shown in Figure 2. We observe an irregular distribution of pores on the surface of the carbon. These wide open pores are explained by a high temperature treatment of the carbon during the complex activation process. Some authors have proven that the use of alkali metal hydroxides such as NaOH or KOH as activating agents promotes the development of pores and the specific surface of carbon [16].

Moreover, this porous structure of the ACRH informs us about the quality of the material in terms of adsorption. Indeed, the presence of a large number of micropores would considerably increase the adsorption capacity of carbon [17].

3.1.2. Specific Surface and Porous Volume

The specific surface and the pore volume are important parameters for evaluating the quality of activated carbon. The higher the specific surface and the pore volume, the greater the adsorbent capacity of the adsorbent. They are determined by adsorption-desorption of dinitrogen at 77 K.

Figure 3 illustrates the adsorption-desorption isotherm of nitrogen at 77 K obtained after analysis at the BET of the ACRH.

In Figure 3 we observe the adsorption isotherm which is similar to two types of the IUPAC classification; type I for low relative pressures between 0 and 0.3 and type IV for intermediate and high pressures.

In the first part, of type I, we observe a strong increase in the shape which tends to stabilize during a long relative pressure, characteristic of microporous material. The slope observed at intermediate and high type IV pressures indicates the transition from microporosity to mesoporosity, in other words the coexistence of microporous and mesoporous materials [18]. In addition, the presence of an H3-type desorption hysteresis loop at relative pressure > 0.6 indicates capillary condensation of the adsorbate at the level of the mesopores. The results provided by the adsorption-desorption isotherm of our carbon are in agreement with the result obtained by the SEM analysis.

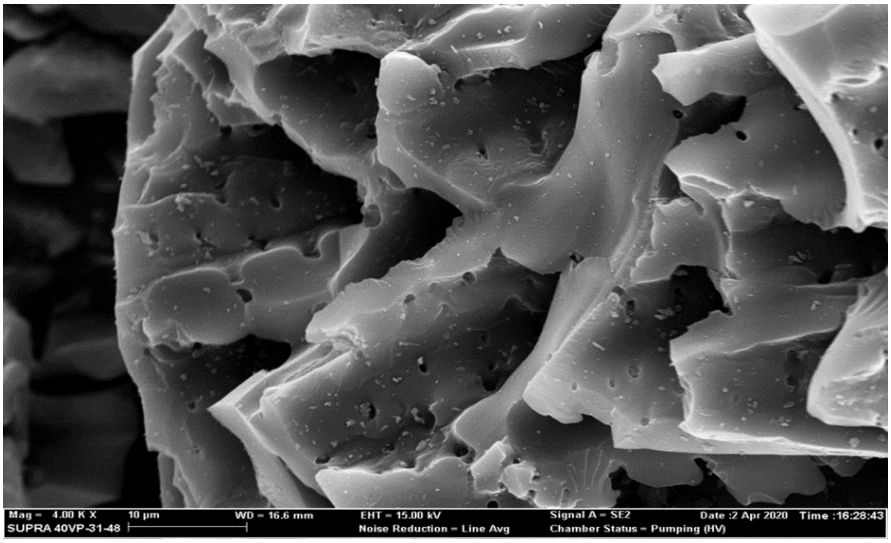


Figure 2. Morphological structure of ACRH.

The specific surface and the pore volume obtained for this carbon are recorded in Table 2.

The specific surface of the carbon in our study is greater than 500 m^2/g. This relatively high value makes our carbonaceous material a good adsorbent. In addition, the pore volume has a high value indicating the microporous structure of the prepared carbon [19]. This result is in agreement with the SEM image obtained previously.

The ash rate obtained for the prepared carbon is 4.2% (<10%), reflecting good preparation of the material given the low rate of volatile matter within it [19].

The comparison of the specific surface of the carbon of our study with other surfaces of carbon determined by the method of BET is presented in Table 3. In view of the values, we can say that this precursor of *Ricinodendron heudelotii*

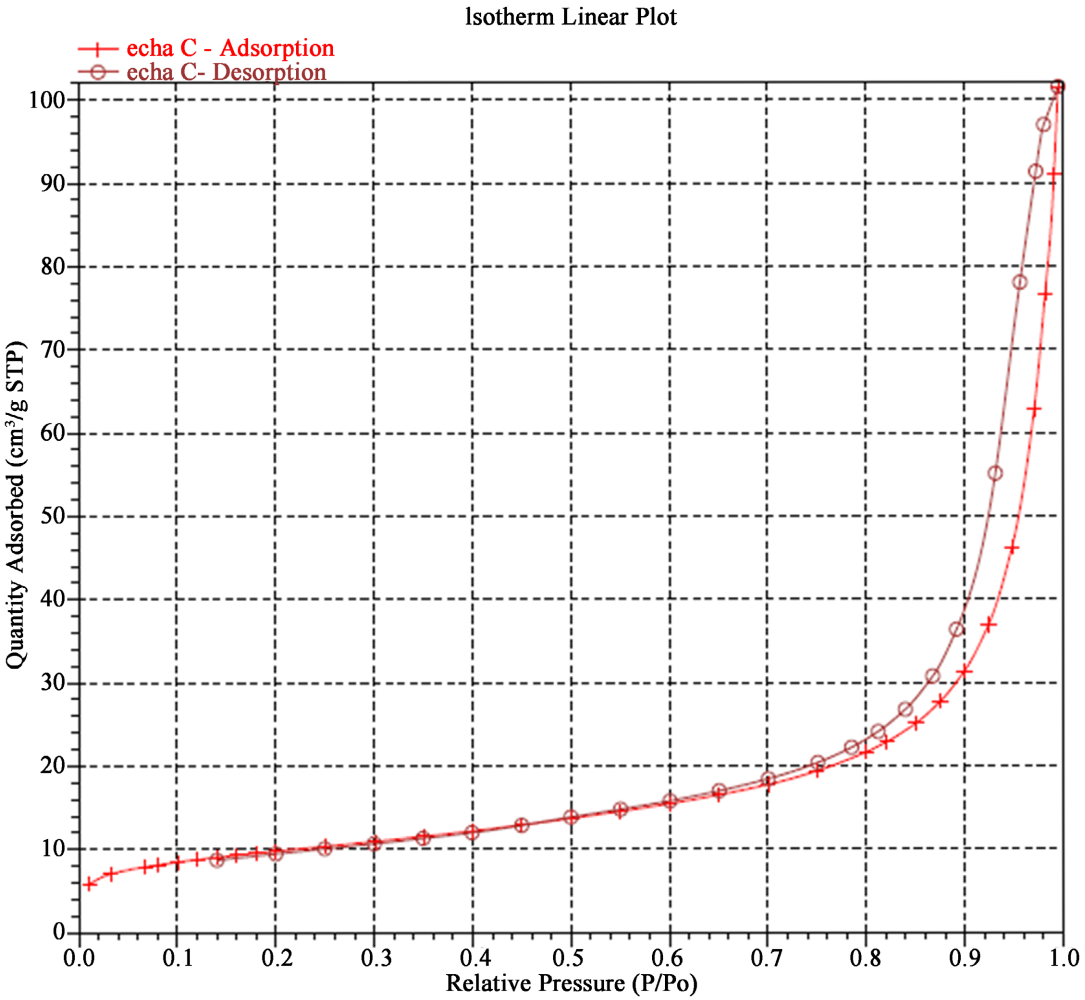


Figure 3. Adsorption-desorption isotherm of nitrogen at 77 K of the ACRH.

Table 2. Important physico-chemical parameters of the adsorbent.

Adsorbent	Specific surface S_{BET} (m^2/g)	Porous volume (cm^3/g)	Ash content (%)
ACRH	612.41	0.465	4.2

provides good quality activated carbon.

3.1.3. Determination of Surface Functions

The surface functions were determined by Boehm's method. This test allowed us to obtain the different functional groups on the surface of the carbon. The results of this experiment are presented in the following **Table 4**.

According to the results provided by the table, the activated carbon has surface acid functions of the lactone (-COO) and phenol (-OH) type, but also basic functions.

The total surface of the carbon is generally basic.

3.1.4. pH of Zero Charge (pHpzc)

pHpzc is a good indicator of the chemical and electronic properties of functional groups [23]. Thus, when pHpzc >pH, the adsorbent surface is positively charged, while when pHpzc < pH, the adsorbent surface is negatively charged. The pHpzc result is given in **Figure 4**. The value of pHpzc is 8.1, suggesting that the character of ACRH is basic. When the pH of the solution is lower than the pHpzc, the surface functional groups of the carbon are protonated by an excess of protons H^+ of the solution, then the adsorbent is said to be an adsorbate attractor and is negatively charged. On the other hand, if the pH of the solution is greater than the pHpzc, the surface functional groups will be deprotonated by the presence of OH^- ions in the solution [24]. Consequently, the support attracts any positively charged adsorbate and promotes the adsorption of cationic dyes, by increasing the electrostatic forces between the negative charge of the adsorbent and the positive charge of the dye. Thus the adsorption of cationic molecules on this carbon in aqueous medium would be favored if the pH is greater than 8.1 and for pH < 8.1 it is the anionic molecules which would be favored. This pH value agrees with the basic character of the surface of the material.

3.2. Adsorption Kinetics

3.2.1. Equilibrium Time

The purpose of this study is to determine the time required for carbon to reach

Table 3. Comparison of specific surface areas of various carbons.

Carbon	Specific surface (m^2/g)	Reference
ACRH	612.41	This study
Grain Sorghum activated carbon	182 - 508	[20]
Activated carbon from olive husks Olive-seed waste	367 - 506	[21]
Activated carbon made from corn cobs	600 - 700	[22]

Table 4. Concentrations (mmol/L) of acidic and basic groups of activated carbon.

Carboxylic	Lactone	Phenolic	Total acid surface	Total basal area
0.000	0.011	0.042	0.053	0.337

adsorption equilibrium for remazol. This equilibrium time was determined from the curves expressing the quantity of dye eliminated as a function of time at different concentrations by AC at ambient temperature (25˚C).

The result is shown in Figure 5.

We find that the amount of adsorbed dye increases with concentration and time.

Indeed, the higher the concentration, the more molecules are available to be adsorbed.

This adsorption is highly time-dependent and takes place in several phases.

For all given concentrations, adsorption of remazol occurred rapidly in the first moments. A large amount of remazol was eliminated during the first 20 minutes of the contact time. This short time is due to a high affinity between the dye and the adsorbent. Indeed, a large number of vacant sites were available on the surface of the activated carbon [9].

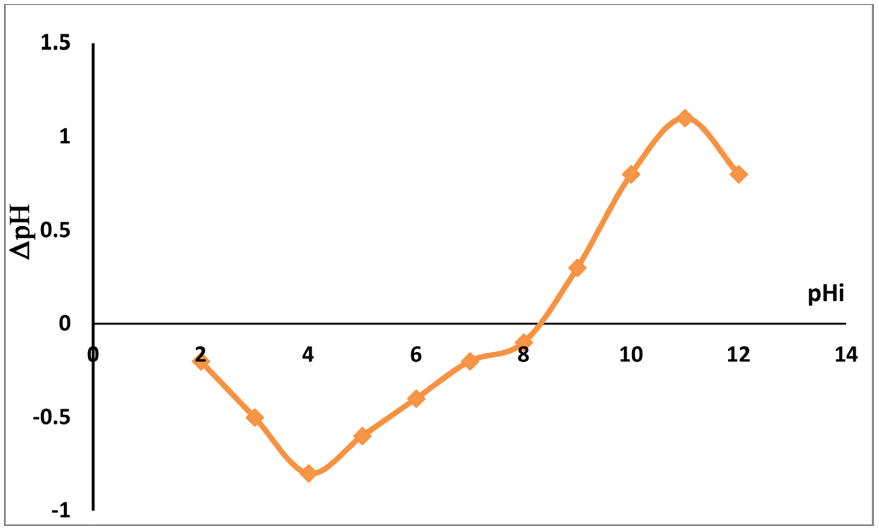


Figure 4. Curve of pHpzc determination for ACRH.

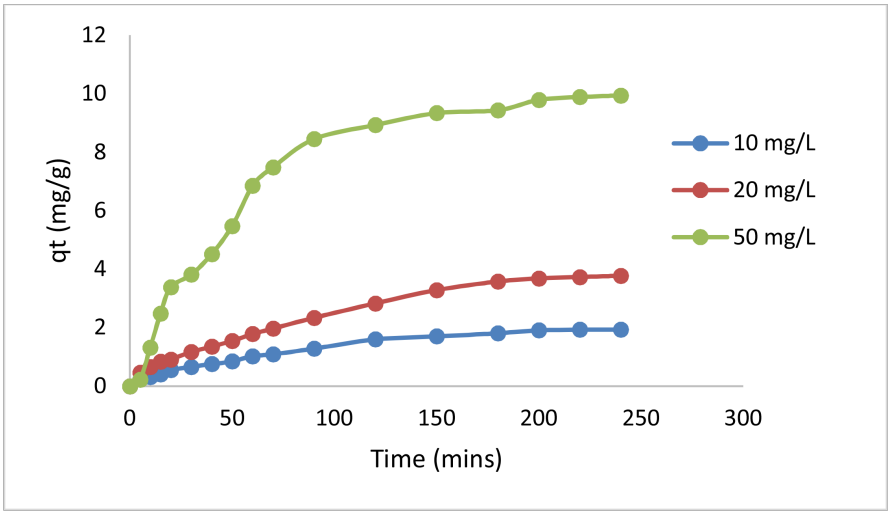


Figure 5. Evolution of the amount of dye eliminated as a function of contact time.

After 20 mins the growth is more or less rapid. The remaining vacant surface sites have become difficult to access and are becoming saturated. Equilibrium is reached after 200 min because the amount of dye adsorbed hardly varies beyond this time. The dye concentration does not influence the adsorption equilibrium time. Thus 200 min will be taken as equilibrium time for the study of the adsorption isotherms of RB5 on the ACRH.

These results are in agreement with other work on the elimination of remazol on other carbonaceous materials [25].

3.2.2. Application of Kinetic Models

In order to better explain the adsorption kinetics, we used three models to interpret the experimental data as judiciously as possible. These are the pseudo-first-order, pseudo-second-order, and intraparticle diffusion models.

Figure 6 is obtained by applying the pseudo-first order kinetic model to the experimental data of the adsorption of RB on activated carbon. The values of the various parameters obtained are given in Table 5. The linear regression coefficients suggest that this model does not apply very well to the adsorption of RB on the studied carbon. These results are confirmed by the difference between the values of $q_{e,exp}$ and $q_{e,cal}$.

The application of the pseudo-second-order kinetic model on the experimental data of the adsorption of RB on ACRH. is presented in Figure 7. Parameter values are listed in Table 5. It can be seen that all the linear correlation coefficients are greater than 0.98. This model is therefore applicable. Moreover, the quantities adsorbed calculated by this model are closer to the quantities adsorbed experimentally. The applicability of this model suggests that the limiting step in the adsorption process of RB on this carbon could be chemisorption [26]. Adsorption rate constants (k_2) decrease with increasing dye concentration. This is

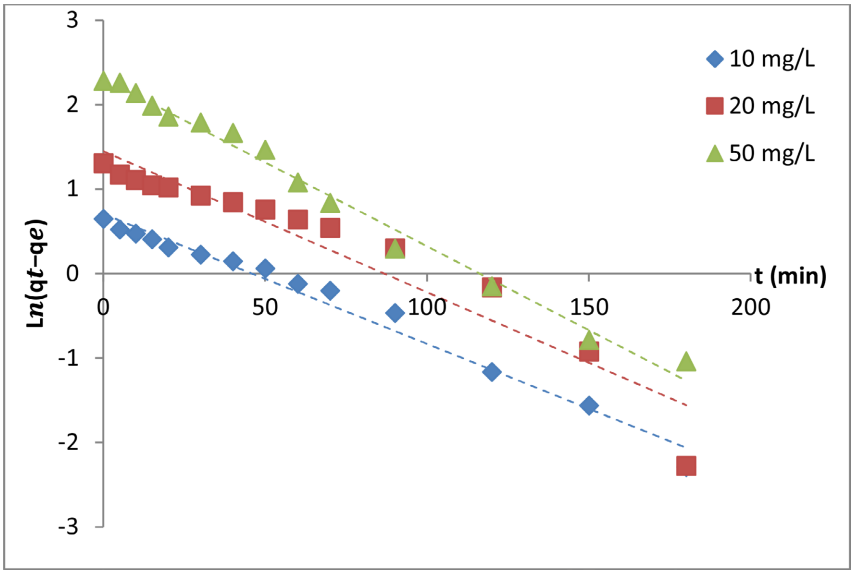


Figure 6. Graphical representation of the modeling of the adsorption kinetics of RB by application of the pseudo-first-order model.

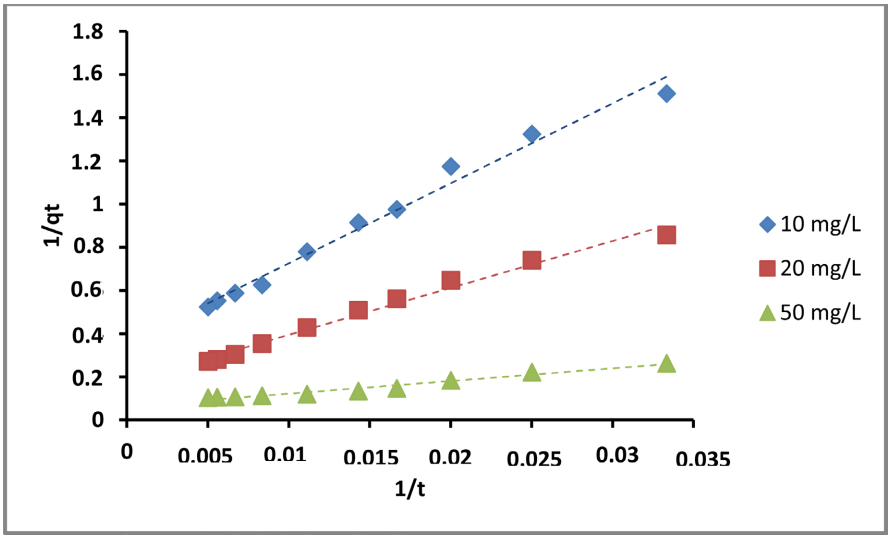


Figure 7. Graphical representation of the modeling of the adsorption kinetics of RB by application of the pseudo-second-order model.

Table 5. Values of the parameters obtained from the kinetic models of the adsorption of RB on activated carbon as a function of the initial concentration.

Kinetic models and parameters	Initial concentration		
Pseudo-first order	10 mg/L	20 mg/L	50 mg/L
k_1	0.0154	0.0167	0.0199
$Q_{e,cal}$ (mg/g)	2.81	5.63	15.67
R_1^2	0.9808	0.9155	0.9886
Pseudo-second order			
k_2	0.0096	0.0015	0.0007
$Q_{e,cal}$ (mg/g)	2.02	4.27	10.10
R_2^2	0.9843	0.9854	0.9904
Diffusion intraparticulaire			
K_i	0.1418	0.2773	0.7242
C	0.0739	0.2928	0.4635
R_i^2	0.9896	0.9861	0.9522
$Q_{e,exp}$ (mg/g)	1.92	3.73	9.90

explained by the increased competition on the adsorption sites. On the other hand, this competition decreases on the active sites of the adsorbent for low concentrations. This result is in agreement with the adsorption of Remazol Black on corn cobs [12] and sugar cane [27] carbons.

Intraparticle diffusion step in the adsorption process, we plotted the curve $q_t = f\left(t^{1/2}\right)$ and then determined the parameters obtained from this graph. The values are recorded in **Table 5**. It is noted that R_i^2 is between 0.95 and 0.98.

This model could be applicable. Thus, the diffusion of the dye in the pores of the carbon would be part of the adsorption mechanism but it is not kinetically determining. The pseudo-second-order model is the best describing the kinetics in this study.

3.3. Adsorption Isotherm

The study of the adsorption of remazol black on activated carbon was carried out at different temperatures (25˚C, 30˚C, 40˚C and 50˚C). The quantity of remazol adsorbed at equilibrium per mass of carbon is given by Figure 8. It can be seen that for all temperatures, the quantity of remazol adsorbed increases with the concentration at equilibrium. These isotherms seem to be type I according to the IUPAC classification [28], justifying the microporous aspect of the carbon used. This result is in agreement with the characteristics of activated carbon.

To study the adsorption phenomenon, 2 models were used, namely the Freundlich model and the Langmuir model.

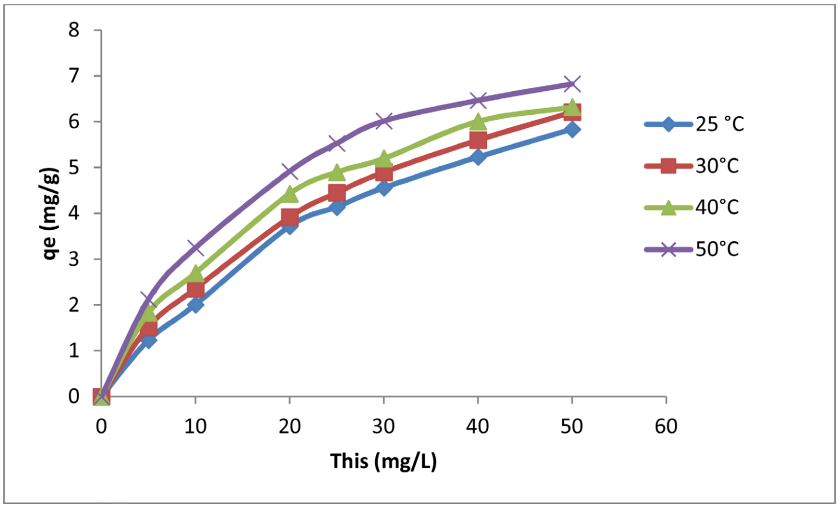


Figure 8. Adsorption isotherms of remazol black on activated carbon.

3.3.1. Langmuir Model

The Langmuir model is commonly used for the study of the adsorption of adsorbates on activated carbons [9] [12]. Langmuir's equation is:

$$q_e = \frac{q_m b C_e}{1 + b C_e} \tag{5}$$

where b designates the Langmuir thermodynamic constant linked to the free energy of adsorption, q_e (mg/g) the quantity of solute adsorbed per unit mass of adsorbent at equilibrium and q_m (mg/g) the quantity of solute adsorbed per gram of solid required to cover the surface of the adsorbent with a monomolecular layer. The maximum adsorption capacity C_e (mg/L) represents the residual solute concentration at equilibrium.

Several linear forms of the Langmuir model are proposed in the literature. One of the most used forms is:

$$\frac{1}{q_e} = \frac{1}{bq_m}\frac{1}{C_e} + \frac{1}{q_m} \tag{6}$$

The Langmuir model was applied by representing the curve $\frac{1}{q_e} = f\left(\frac{1}{C_e}\right)$ (Figure 9). The values of the parameters obtained from the graph are grouped together in Table 6. The linear correlation coefficients are all greater than 0.98. The Langmuir model can therefore be used to describe the adsorption of remazol on this activated carbon for all temperatures. The adsorption of remazol on this activated carbon would be of the monolayer type. The maximum quantity of remazol black adsorbed by 0.1 g of HRAC for a concentration of 20 mg/L depends on the temperature. It is greater than 8.4 mg/g.

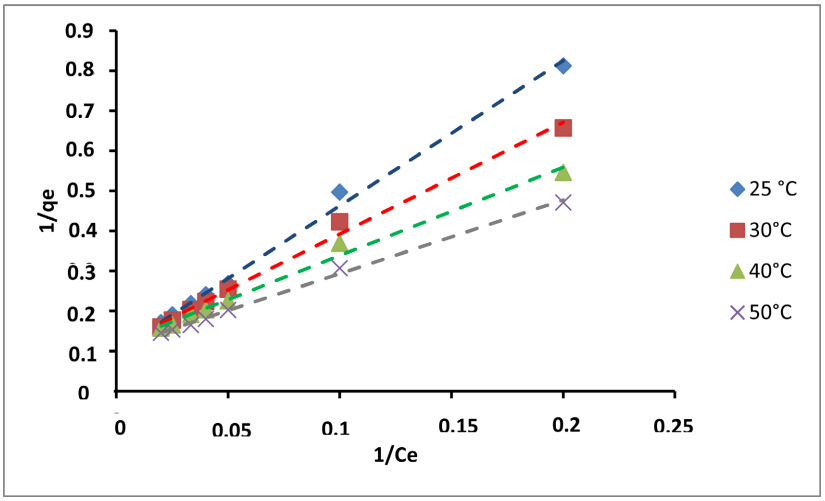


Figure 9. Application of the Langmuir model to the remazol adsorption of carbon on activated carbon.

3.3.2. Freundlich Model

The Freundlich isotherm is a semi-empirical model also used to describe the adsorption of a dye on a porous material. Considering that the dynamic equilibrium takes place between the concentration of the adsorbent in the liquid phase and the quantity q_e (mg/g) adsorbed, the Freundlich equation is written [29]:

$$q_e = K_F C_e^{1/n} \tag{7}$$

C_e, K_F and n are Freundlich constants with:

- C_e (mg/L), the concentration of the adsorbate at equilibrium in the residual solution;
- K_F, there adsorption capacity in L/mg;
- $1/n$, the surface adsorption intensity.

The expression shows that as the adsorbate concentration increases, the concentration of the adsorbate on the surface of the adsorbent increases. The linear form of the Freundlich model is:

$$\ln q_e = \ln K_F + \frac{1}{n}\ln C_e \tag{8}$$

The application of the linear form of the Freundlich model made it possible to draw the curve $\ln q_e = f(C_e)$ (Figure 10). The values of the parameters of this curve are grouped in Table 6.

The correlation coefficients are greater than 0.98 and $1/n > 0.5$. Adsorption is favorable for all concentrations. The Freundlich model can be used to describe the adsorption of remazol black on this carbon, reflecting the heterogeneity of the surface of this carbon as observed with the SEM analysis.

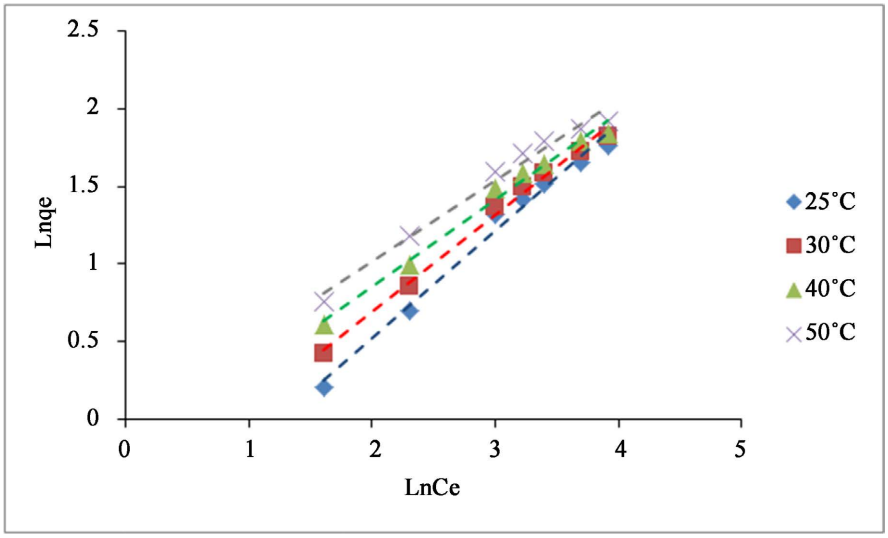


Figure 10. Application of the Freundlich model to the adsorption of remazol black on activated carbon.

Table 6. Parameters of the Langmuir and Freundlich models.

	Langmuir model			Freundlich model		
	R^2	q_m (mg/g)	B	R^2	K	$1/n$
25˚C	0.9953	9.92	0.0278	0.9869	0.4106	0.6943
30˚C	0.9935	8.74	0.04108	0.9934	0.5698	0.6253
40˚C	0.9898	8.41	0.0540	0.9862	0.7647	0.5603
50˚C	0.9969	9.09	0.0599	0.9799	0.8962	0.5239

3.4. Influence of pH and Carbon Mass on Remazol Adsorption

3.4.1. Effect of Solution pH

The pH is an important parameter in the study of the adsorption phenomenon because it affects both the chemical properties of the dye and the active sites found on the surface of the adsorbent [30]. The influence of pH on the adsorption of remazol by activated carbon was studied in a pH range between 2 and 12. The results are presented in Figure 11. The amount of dye adsorbed is greater in an acid medium (95.1% at pH = 3; 96.3% at pH = 3). The rate of removal is almost constant in the range of pH 2 to 3. As the pH increased from 3.0 to 11.5, the percent dye removal decreased by 76%. The optimum pH for the adsorption of

remazol on this carbon is acidic, *i.e.* in the pH range between 2 and 4. This result is in agreement with the results of the characterization of the carbon and the properties of the dye. Indeed, RB is a negatively charged anionic dye in aqueous solutions. It adsorbs better on a material if the surface of this material has a positive charge highlighting the electrostatic interactions [30].

For the activated carbon in this study, the electrostatic interaction occurs for pH < 8.1.

At a pH higher than the pH of zero charge and in particular in a very basic medium, the adsorption of remazol is not favored.

This result is consistent with those obtained by adsorption of remazol black B on biomass-based activated carbon [31].

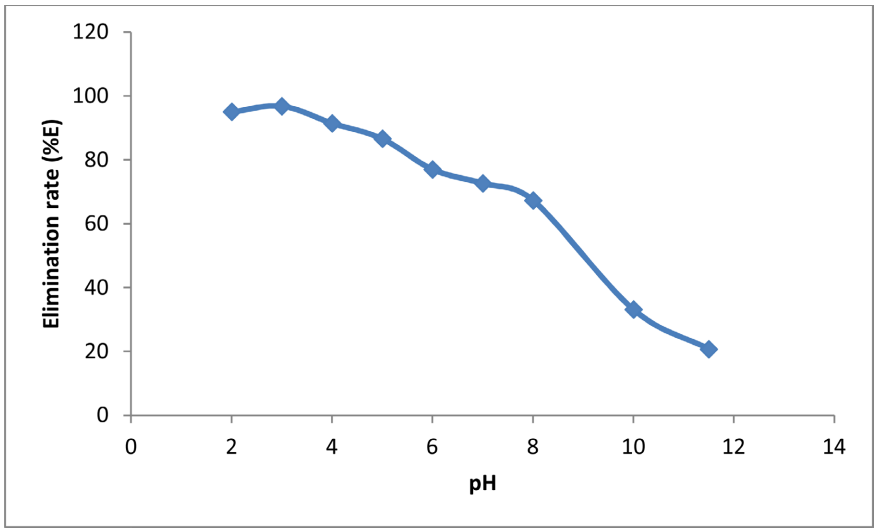


Figure 11. Influence of pH on the adsorption of remazol black.

3.4.2. Effect of Carbon Mass

To judge the effectiveness of remazol in adsorbing on the prepared carbon, we studied the influence of the variation in the mass of the carbon on its adsorption. **Figure 12** shows the adsorbed quantity of remazol as a function of the mass of carbon. Thus, we observe an increase in the elimination rate from 88% to 98% with the mass of carbon from 0.1 g to 0.25. This increase is explained by the availability of more active sites with the increase in the mass of carbon. The dye removal rate appears to be constant for carbon masses of 0.2 g to 0.3 g. Beyond 0.3 g, the dye elimination rate is almost constant because there would be fewer molecules to adsorb on the active sites, the number of which has increased. The optimum mass of carbon prepared to remove 20 mg/L of remazol black in a volume of 25 mL is 0.2 g.

3.5. Thermodynamic Study of the Adsorption of Remazol on AC

The main criterion for knowing the nature of the adsorption phenomenon is the determination of the thermodynamic parameters. For the determination of the free enthalpy (ΔG), the enthalpy (ΔH) and the entropy (ΔS) of adsorption of

remazol black on activated carbon, the following equations were used:

$$\ln\left(\frac{q_e}{C_e}\right) = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{T} \tag{9}$$

$$\Delta G^\circ = -RT\ln\left(\frac{q_e}{C_e}\right) \tag{10}$$

where q_e (mg/g) is the quantity of RB adsorbed at equilibrium, C_e (mg/L) is the concentration of RB at equilibrium, T is the temperature and R (8.314 J/mol·K) is the ideal gas constant.

The curve $\ln\left(\frac{q_e}{C_e}\right)$ as a function of $1/T$ is represented in Figure 13. The exploitation of the data of these straight lines made it possible to determine the

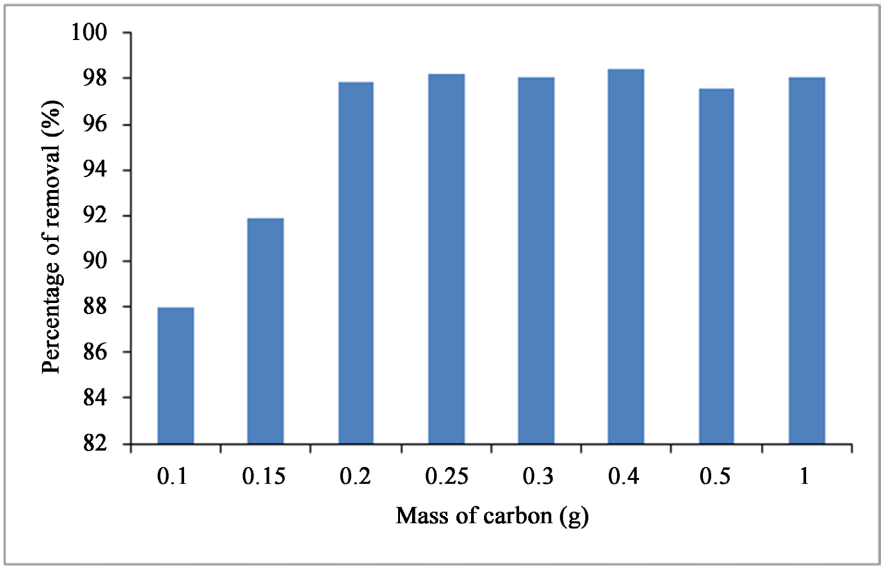


Figure 12. Influence of carbon mass on remazol black adsorption.

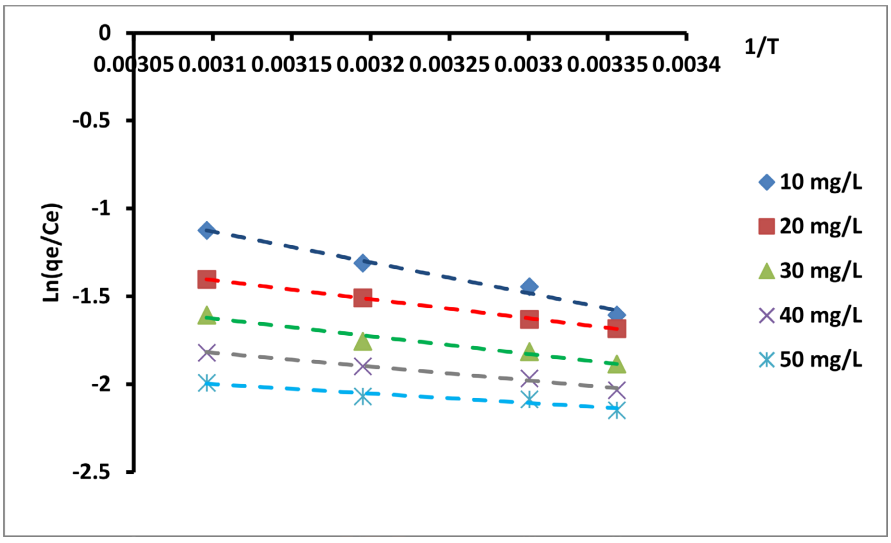


Figure 13. Effect of temperature on RB removal.

thermodynamic adsorption parameters grouped in Table 7. These values show that the maximum adsorption quantity of Remazol (*qm*) increases with temperature. The free enthalpy (ΔG°) is negative, which reflects the spontaneity of the adsorption phenomenon [27]. There is therefore a good affinity between activated carbon and RB.

Moreover, the positive and relatively low enthalpy values ($0 < \Delta H^{\circ} < 40$ kJ/mol) suggest that the process is endothermic and the adsorption is of a physical nature [30]. The positive value of entropy (ΔS°) could be related to an increase in disorder at the adsorbent/adsorbate interface which confirms the increased randomness within the solid-liquid interface [30].

4. Conclusion

In this work, remazol black adsorption experiments on activated carbon based on *Ricinodendron heudelotii* shells were conducted. The characterization results

Table 7. Values of the thermodynamic parameters of the adsorption of remazol black on activated carbon.

Concentration	Temperature (K)	ΔG° (kJ/mol)	ΔH° (kJ/mol)	ΔS° (J/mol·K)
10 mg/L	298	−13.640	17.572	35.824
	303	−14.402		
	313	−15.360		
	323	−16.246		
20 mg/L	298	−13.139	10.901	73.823
	303	−13.764		
	313	−14.569		
	323	−15.532		
30 mg/L	298	−12.947	10.136	70.036
	303	−13.296		
	313	−14.053		
	323	−14.784		
40 mg/L	298	−12.659	7.880	62.600
	303	−13.054		
	313	−13.735		
	323	−14.499		
50 mg/L	298	−11.794	5.394	54.704
	303	−12.147		
	313	−12.594		
	323	−13.204		

showed that the carbon prepared is microporous, basic and has a large specific surface (602 m^2/g). The kinetic study revealed that the equilibrium time of adsorption of the dye on this carbon is 200 min. This time is very little influenced by the concentration of the solution and the temperature of the medium. Also, the pseudo-second-order model better describes the adsorption phenomenon. Intraparticle diffusion, although not kinetically determining, is important in the process. This phenomenon of adsorption is influenced by the pH of the medium of the solution and the mass of the adsorbent. Adsorption is better at pH< pHpzc, in particular in an acid medium. The optimum values for the elimination of this dye are pH = 3 and mass = 0.2 g at ambient temperature for a concentration of 20 mg/L of remazol black with a volume of 25 mL. The maximum quantity of remazol black eliminated by this carbon is 9.92 mg/g with these values. The Langmuir and Freundlich models can be used to effectively describe the adsorption phenomenon of Remazol Black on prepared carbon. The thermodynamic study revealed that the adsorption phenomenon is spontaneous, endothermic and of the physisorption type. In addition, there is an increase in disorder at the solid-liquid interface.

Author Contributions

Kouakou Yao Urbain and Trokourey Albert defined and designed the work and the experiment. Kambire Ollo prepared the materials. Kone Yétchié Tchonrontcha and N'goran Sévérin carried out the experiences. Kouakou Yao Urbain and Kone wrote the manuscript. Kambire Ollo and Eroi N'goran Sévérin revised the manuscript critically.

All authors read and approved the final manuscript.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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