



Methylene blue adsorption capacity and coherent isotherm model of commercial activated carbon

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Abstract

The use of dyes that pollute the earth and adversely affect the health of human beings is increasing day by day and it is important to remove them from nature. Methylene blue (MB), which is one of the most commonly used dyestuffs that affects natural life negatively, was the subject of this study. In this study, the adsorption mechanism of commercial activated carbon to remove MB was investigated. In the study, activated carbon was added to MB solution taken in different concentrations and the results were applied to ten different adsorption isotherms which are Langmuir, Freundlich, Temkin, Harkins-Jura, Halsey, Dubinin- Radushkevich, Janovich, Redlich-Peterson, Henry's and Hills. Adsorption isotherm graphs were drawn with the obtained results and constant parameters in adsorption isotherm equations were calculated through graphics. The adsorption mechanism was examined according to the correlation coefficients (R^2) from the graphs obtained. The highest correlation coefficients were calculated as 0,9993 and 0,9954 from Redlich-Peterson and Langmuir isotherm equations respectively. Adsorption isotherm was coherent Langmuir and maximum adsorption capacity was determined 208,3 mg/g.

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1. Introduction

With the increasing world population, the need for the industrial sector was increased day by day. This need has contributed to development of industrial sector. With the developing industry, many products were produced and many wastes occurred from these products. Among these wastes, the materials that pollute the nature the most and affect human health are chemical materials. Chemical materials have entered our lives in many areas such as drugs, cleaning products and dyes. Paints find uses in many areas being manufactured textile, food, cosmetics, paper and paint. The waste and unused paints cause environmental pollution. Environmental pollution can be handled under three main group as soil, air and water pollution. These dyestuffs are mostly mixed with our soil or water and cause contaminations in drinking water. These pollution occurring in drinking water negatively affect human life. Besides, these pollutions adversely affect aquatic lives and cause food shortages due to the use of these creatures as food products. It is important to prevent these impurities and to clean the increasing pollution [1-3].

Dyestuffs usually have a complex and durable structure and are generally divided into 3 groups as anionic, cationic and ionic. MB is a type of cationic dye with a heterocyclic aromatic structure. MB generally uses such as paper, silk, wool leather, ink, cotton dyeing and photocopy paper production. Even very low concentration of MB causes intense colouring in the water. Due to the colour intensity, sunlight cannot pass through the water and aquatic microorganisms harm because of the lack of sunlight. Besides, although MB is not a completely toxic dye, it has negative effects on humans and living when exposed for a long time. General effects which have seen in humans can be listed as vomiting, dizziness, skin diseases, etc. [4-7].

There are many processes to remove dyes from the aqueous environment. These processes are generally chemical, biological and photocatalytic oxidation as well as coagulation-flocculation, filtration, adsorption, membrane separation and ion-exchange. Adsorption is the most used process among them. This technique is frequently used because it is an economical, efficient, easy to apply and effective process [4,8].

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Adsorption is generally defined as the attachment of atoms, ions or molecules in one phase to another phase. Although liquid-solid phase is widely used in general, it is also used in phases such as liquid-liquid, liquid-gas and gas-solid systems. Clay, zeolite, bentonite, polymers and activated carbon are generally used as adsorbents in the adsorption process. It is important that the adsorbent used has high adsorption capacity, high surface area, easy accessibility and easy produce. Activated carbon is referable to other adsorbents in terms of high surface area, easy processing and easy availability compared to other adsorbents. In addition, activated carbons can be obtained commercially or easily synthesized. Activated carbon synthesis is generally obtained from agricultural wastes in recent years. Agricultural wastes are preferred because of their abundant easy access, low cost, easy storage and most importantly, their high carbon content [9-11].

The aim of this study is to determine methylene blue adsorption capacity and coherent isotherm model of commercial activated carbon. Also, it will be possible to compare other synthesized activated carbon samples in the literature and advantages and disadvantages of commercial activated carbon will be determined.

2. Materials and Methods

In this study, MB was used as the dye. MB is a cationic dye with the formula $C_{16}H_{18}N_3S$ and a molecular weight of $373,9 \text{ g mol}^{-1}$. The molecular structure of MB has dimensions of 1.43 nm in width, 0.40 nm in thickness, and 0.61 nm in depth [12]. The formula is shown in Figure 1.

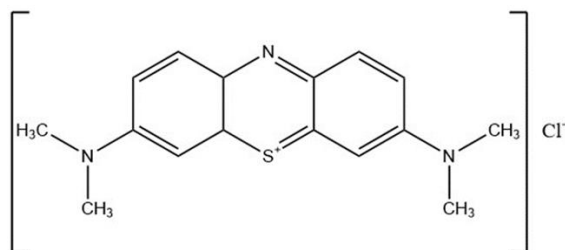


Figure 1. Methylene Blue

In this study, MB (ISOLAB C.I.52015, Turkey) was weighed 1 gram and added into a 1 litre volumetric flask and the total volume was completed to 1 litre. In this way, obtained a stock solution of 1000 ppm. The stock solution was diluted into 250ppm, 300 ppm, 350 ppm and 400 ppm solutions. The diluted solutions were taken into 100 ml flasks and 0,1 grams of active carbon was added. The mouths of the flasks were covered with paraffin and mixed for 24 hours in a miprolab brand mixer at 195 rpm. The highest peak value was determined by scanning the MB solutions in the

Agilent brand Cary-60 UV-VIS device between 200-800 nm as shown in Figure 2.

2.1. Adsorbance

The activated carbon used in this study was obtained commercially. Activated carbon is a product with code KIM-AC / 01CP / 091006 belonging to Kimetsan company (Turkey).

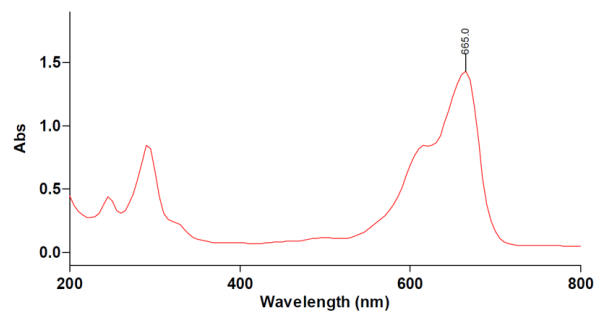


Figure 2. UV-Vis scanning of MB solution

2.2. Isotherm equations

2.2.1. Langmuir isotherm

Langmuir isotherm equation (Equation 1) was derived in 1918 by Irving Langmuir based on some assumptions. According to Irving Langmuir, adsorption takes place in a single layer and allows the calculation of the adsorption capacity. It also assumes that there is no interaction between the adsorbed molecules and that the entire adsorption field has the same energy. At the end of the adsorption process, the energy and adsorption rate is constant. In short, the maximum adsorption capacity is reached in the equilibrium state and the surface is covered in a single layer.

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

According to this equation, C_e/q_e versus C_e is plotted. $1/q_m$ gives the slope, $1 / K_L \times q_m$ gives intercept. In the equation, C_e : The amount of MB remaining in the solution at the end of the adsorption (mg/L), q_e : The amount of MB adsorbed on the activated carbon (mg/g), b : Langmuir constant, Q_0 : The maximum amount of adsorbed substance forming a monolayer surface [13].

2.2.2. Freundlich isotherm

Freundlich isotherm equation (Equation 2) was derived in 1906 by Herbert Max Finley Freundlich. According to H.M. Finley Freundlich, the areas on the adsorbent have heterogeneous energy and may differ in the adsorption areas. In addition, adsorption takes place not in a single layer, but in a multi-layer.

$$q_e = K_F \times C_e^{\frac{1}{n}} \quad (2)$$

The equation that occurred by linearization of Equation (2) is given in Equation 3.

$$\log q_e = \log K_F \times \frac{1}{n} \log C_e \quad (3)$$

$\log C_e$ versus $\log q_e$ is plotted according to the Freundlich equation. The slope is $1/n$ and the intercept gives the $\log K_F$ value. C_e : The amount of MB remaining in the solution at the end of the adsorption (mg/L), q_e : The amount of MB adsorbed on the activated carbon (mg/g), K_F shows the experimental adsorption capacity (1/mg), n adsorption density (without units) [14].

2.2.3. Temkin isotherm

Temkin isotherm assumes that there are adsorbent-adsorbate interactions. Very low and high concentration is ignored according to the model. In this model, the heat of adsorption decreases linearly. In other isotherms, this decrease is given logarithmically. The nonlinear equation of the temkin isotherm is given in equation 4.

$$q_e = \frac{RT}{b_T} \ln(K_T C_e) \quad (4)$$

The equation that occurred by linearization of Equation (4) is given in Equation 5.

$$q_e = b_T \ln K_T + b_T \ln C_e \quad (5)$$

According to this equation, q_e versus $\ln C_e$ is plotted. The slope gives b_T while the intercept gives the value of $b_T \ln K_T$. According to this equation, q_e : MB adsorbed on activated carbon (mg/g), C_e : Indicates the amount of MB remaining in the liquid phase (mg/L). b_T is calculated from the value of RT/b . R is the ideal gas constant, T (°K) is the temperature and b is the temkin constant. K_T is the binding energy [15].

2.2.4. Harkins-Jura isotherm

According to Harkins-Jura isotherm model, there are micro and mesopores in the structure and multi-layer adsorption is available. Isotherm equation is given in equation 6.

$$\frac{1}{q_e^2} = \frac{B_H}{A_H} - \frac{1}{A_H} \log C_e \quad (6)$$

According to this equation, $\frac{1}{q_e^2}$ versus $\log C_e$ is plotted. Slope gives $1/A_H$, intercept gives B_H/A_H values. C_e : The amount of MB remaining in the liquid phase in equilibrium (mg/L), q_e : The amount of MB adsorbed on the activated carbon (mg/g), A_H , B_H are the isotherm constants [16].

2.2.5. Halsey isotherm

Halsey equation (Equation 7) is based on multi-layer adsorption with the adsorbate having micro and mesopores.

$$\ln q_e = \frac{1}{n} \ln k - \frac{1}{n} \ln C_e \quad (7)$$

According to this equation, $\ln q_e$ versus $\ln C_e$ is plotted. The slope is $1/n$, intercept is $\frac{1}{n} \ln k$. C_e : the amount of MB remaining in the liquid phase (mg/L), q_e : the amount of MB adsorbed on the activated carbon (mg/g), k value is the isotherm constant [16].

2.2.6. Dubinin- Radushkevich (D-R) isotherm

It is an equation derived based on the micropore volume filling theory put forward for gas phase adsorption. In this model, the adsorption is assumed to occur with the multilayer model. Generally, this model is used to calculate adsorption energy. Dubinin-Radushkevich isotherm equation is given in equation 8. Potential energy and adsorption energy calculation equations are given in equations 9 and 10, respectively.

$$\ln q_e = \ln q_m - K' \varepsilon^2 \quad (8)$$

$$\varepsilon = RT \ln \left(1 + \frac{1}{C_e}\right) \quad (9)$$

$$E = \frac{1}{\sqrt{2K'}} \quad (10)$$

$\ln q_e$ against the ε^2 (square of the potential energy) is plotted. q_m : theoretical adsorption capacity (mg/g) and from the intercept and slope of this line, the constants of q_m and K are found, respectively. R : gas constant (8.314 J mol⁻¹ K⁻¹), T : absolute temperature (°K), K' : Isotherm constants related to adsorption energy, ε : potential Polanyi. The average adsorption energy (E) help us predict the adsorption mechanism. If the E value is 8-16 kJ mol⁻¹, the adsorption process is characterized by ion exchange. If $E < 8$ kJ mol⁻¹, adsorption can be considered as physical, if $20 < E < 40$ kJmol⁻¹, adsorption can be considered chemical [17].

2.2.7. Janovich isotherm

According to the Janovich isotherm, Langmuir assumes that the isotherm is valid in adsorption. But it is assumed to be important in the interactions between the adsorbate and the adsorbent, which are not in the Langmuir isotherm. The Janovich equation is given in equation 11.

$$q_e = q_m (1 - e^{-K_j C_e}) \quad (11)$$

$$\ln q_e = \ln q_m - K_j C_e \quad (12)$$

The equation that occurred by linearization of Equation (11) is given in Equation 12.

$\ln q_e$ versus C_e is plotted. Slope gives value K_j . q_m : theoretical adsorption capacity (mg/g), q_e : amount adsorbed to equilibrium (mg/g), C_e : amount remaining in equilibrium solution (mg/L), K_j is Janovitch's constant [18].

2.2.8. Redlich-Peterson isotherm

Redlich-Peterson isotherm is a mixture of two different isotherms, Langmuir and Freundlich. Although the adsorption mechanism is a mixture, it assumes that adsorption takes place in a multi-layer. This isotherm equation is given in Equation 13.

$$\ln \frac{C_e}{q_e} = \beta \ln C_e - \ln A \quad (13)$$

$\ln \frac{C_e}{q_e}$ versus $\ln C_e$ is plotted. The slope gives β and intercept gives the value of $\ln A$. C_e : amount remaining in equilibrium solution (mg/L), q_e : amount adsorbed in equilibrium (mg/g), β and A are Redlich-Peterson constant [19].

2.2.9. Henry's isotherm

It is known as the most basic adsorption isotherm. This isotherm is generally valid at very low concentrations. It increases the adsorption capacity of the adsorbate by increasing the surface area. Henry's isotherm equation is given in equation 14.

$$q_e = K_{HE} X C_e \quad (14)$$

According to this equation, q_e versus C_e is plotted. From the slope of the graph, the Henry constant K_{HE} is found. In the equation, q_e : Amount adsorbed in equilibrium (mg/g), C_e : Amount remaining in the solution in equilibrium (mg/L) [19].

2.2.10. Hills isotherm

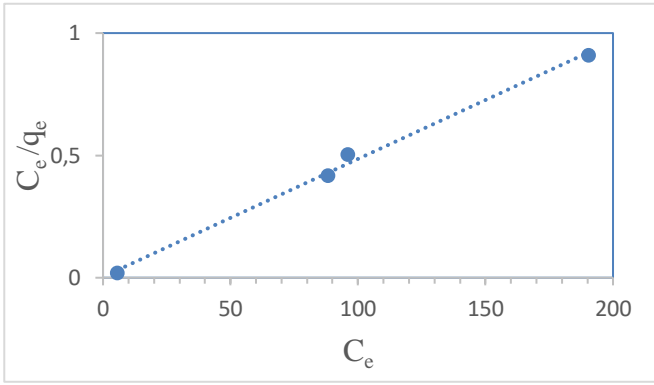
According to Hills isotherm model, homogeneous adsorbates are bound to adsorbents. Therefore, it affects other active sites on the surfaces of adsorbates and effect adsorption. The adsorption equation derived in this direction is given in equation 15.

$$\log \left(\frac{q_e}{q_{SH} - q_e} \right) = n_H \log C_e - \log K_D \quad (15)$$

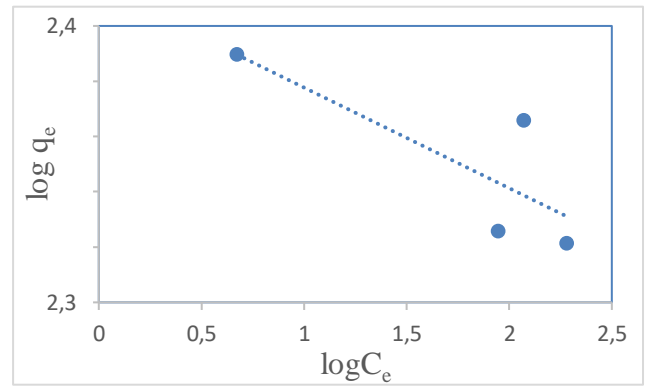
$\log \left(\frac{q_e}{q_{SH} - q_e} \right)$ versus $\log C_e$ is plotted. In the resulting graph, n_H is found from the slope and K_D from intercept. In the equation, K_D and n_H hills constant, q_e : adsorption amount of activated carbon (mg/g), C_e : Amount remaining in the solution in equilibrium (mg/L) [18].

3. Results and Discussion

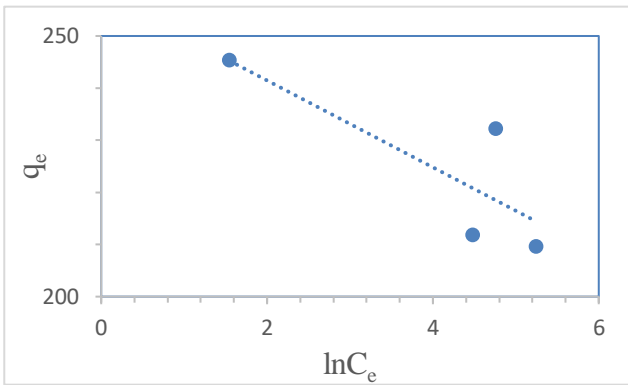
Adsorption is similar to an equilibrium reaction. When the solution is brought into contact with a certain amount of adsorbate, the concentration of the adsorbed material in the solution decreases until it reaches equilibrium with those on the adsorbent surface. After the adsorption balance is established, the concentration of the adsorbed substance in the solution phase remains constant. The amount of adsorbed that can be retained by an adsorbent is a function of the concentration and temperature of the adsorbent. Generally, the amount of material adsorbed is determined as a function of the concentration at constant temperature. The result function called adsorption isotherm is obtained by plotting the amount of solute adsorbed, in unit adsorbent weight, against the solute concentration remaining in the solution in equilibrium at constant temperature. Adsorption isotherms are essential for our understanding of how adsorption occurs. From the isotherms drawn using adsorption equations, it can be predicted how adsorption takes place. According to correlation coefficients in the adsorption isotherms drawn, it is decided which isotherm suits and how the adsorption takes place. If the correlation coefficient (R^2) has a value between 0 and 1, this indicates a positive relationship between the two variables. In other words, as the value of one variable increases, the other tends to increase. How clearly this relationship can be demonstrated depends on the value of R^2 . If the value of R^2 is close to 1, the relationship between the two variables can be demonstrated well. However, as the value of R^2 approaches 0, it can be said that there may be only a weak relationship between the two variables or that the available data (or the sensitivity of the data) are insufficient to reveal such a relationship. At this point, it means that the adsorption mechanism of the isotherm is applied to a correlation coefficient close to 1. In this study, adsorption isotherms were drawn in order to learn the relationship between two variables. The adsorption isotherms drawn are given in Figure 3.



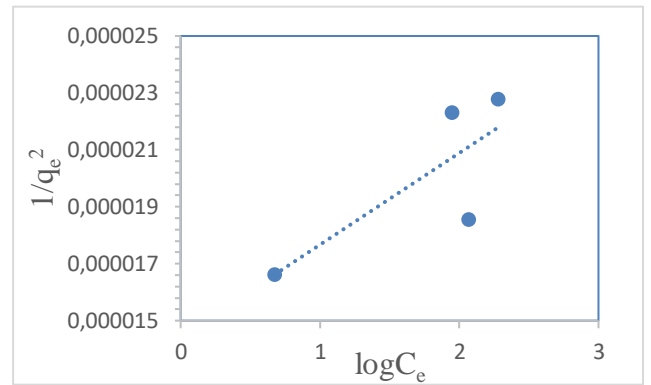
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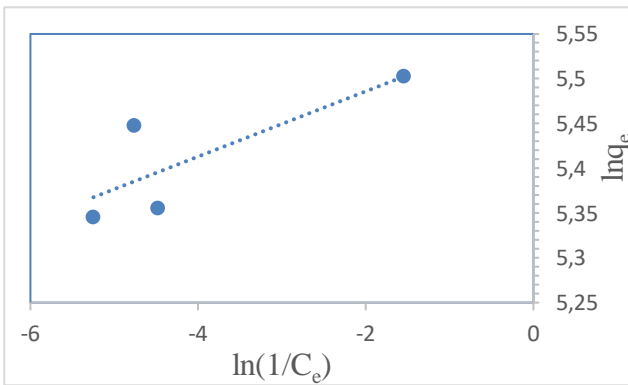
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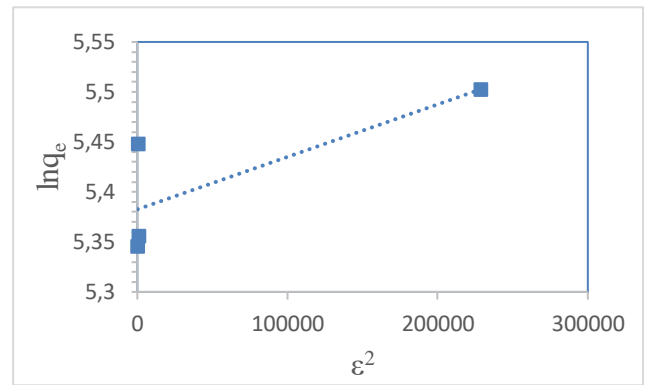
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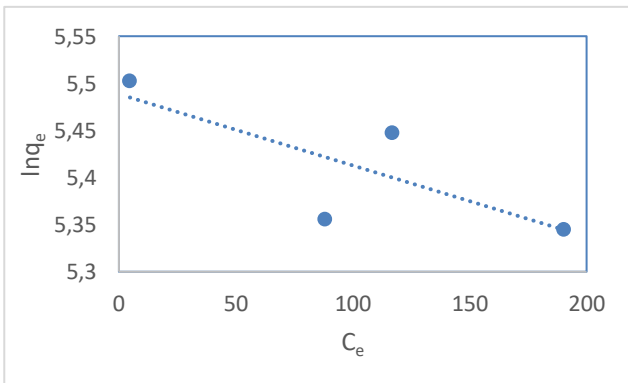
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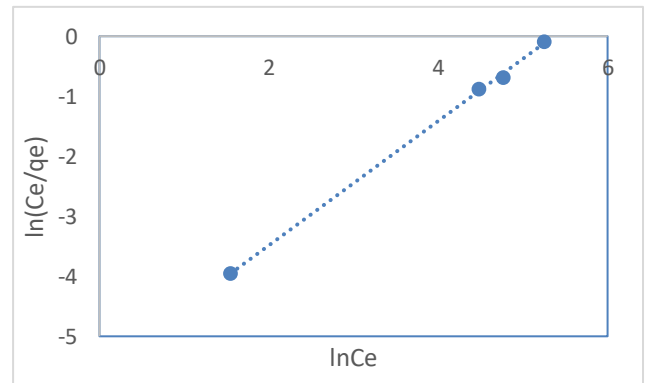
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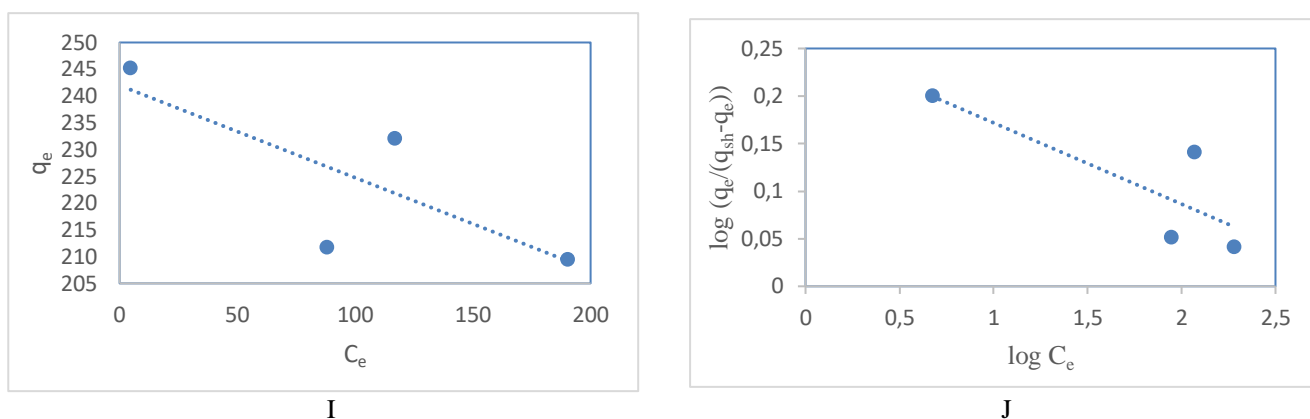


Figure 3. A. Langmuir Isotherm B. Freundlich Isotherm C. Temkin Isotherm D. Harkins-Jura Isotherm E. Halsey Isotherm F. Dubinin- Radushkevich Isotherm G. Janovich Isotherm H. Redlich-Peterson Isotherm I. Henry Isotherm J. Hills Isotherm

Using the correlation coefficients (R^2 values) obtained from the isotherm graphs shown in Figure 3, their applicability was evaluated and the R^2 values obtained for each isotherm are given in Table 1. In addition, the

constants of each isotherm were determined by using the slopes and intercept of the graphics in Figure 3. These constants are also given in Table 1.

Table 1. Constant parameters and correlation coefficients

Langmuir		Freundlich		Temkin	
Q_0 (mg/g)	208,3	K_F (L/mg)	259,47	$-b_T$ (J/mol)	8,33
b (L/mg)	1,26	n	27,47	$K_{TX} \cdot 10^{-14}$ (L/g)	3,51
R^2	0,9954	R^2	0,6503	R^2	0,665
Harkins-Jura		Halsey		Janovich	
$-A_H \cdot 10^{-4}$	33	$-n$	0,17	q_m (mg/g)	241,89
$-B_H$	3,3	k	1,238	$K_J \cdot 10^{-4}$ (L/g)	8
R^2	0,6214	R^2	0,6503	R^2	0,5934
Redlich-Peterson		Henry		Hills	
A (L/g)	259,48	$-K_{HE}$	0,172	$-n_H \cdot 10^{-2}$	8
β	1,036	R^2	0,6006	K_D	1,8
R^2	0,9993			R^2	0,6703
D-R					
q_m (mg/g)	217,58				
$-K \cdot 10^{-7}$ (mol ² /j ²)	5				
R^2	0,6285				
E (kJ/mol)	1000				

According to Table 1, the correlation coefficients are listed as Redlich-Peterson > Langmuir > Hills > Temkin > Freundlich = Halsey > Harkins-Jura > Dubinin-Radushkevich > Henry > Janovich. When the correlation coefficients are examined, the closest isotherms to 1 are Redlich-Peterson and Langmuir isotherms. Therefore, since the correlation values of the adsorption mechanism are very close to each other, it can be assumed that it occurs according to Redlich-Peterson and Langmuir. When these two isotherm equations are examined, it is seen that the equations

have similar properties. The Redlich-Peterson equation is based on the Langmuir equation and is obtained by combining this equation with the Freundlich equation. This equation obtained is similar to Freundlich's at high concentrations, while low concentrations are similar to Langmuir equation [20].

Table 2 demonstrates adsorption capacity of the different kinds of activated carbon used for MB removal.

Table 2. Adsorption capacity of different activated carbon

Adsorbent	Raw Material	Activation	Chemical/Gas	MB Adsorption Capacity	Ref.
Commercial Activated Carbon	-	-	NiFe ₂ O ₄	167.45 mg/g	[22]
Activate Carbons	-	-	Silver Nanoparticles	416.66 mg/g	[23]
Activated Carbon	Walnut Shell	Physical Activation	CO ₂	174,81 mg/g	[24]
Activated Carbon	<i>Platanus Orientalis</i>	Chemical Activation	ZnCl ₂	138,88 mg/g	[25]
Activated Carbon	Almond Shell	Chemical Activation	ZnCl ₂	201.40 mg/g	[26]
Activated Carbon	Sucrose	Chemical Activation	KOH	1666.66 mg/g	[27]
Activated Carbon	Almond Shells	Chemical Activation	H ₂ SO ₄	131.58 mg/g	[28]
Activated Carbon	Peach Kernel Shells	Physical Activation	CO ₂	3,67 mg/g	[29]
Activated Carbon	Pistachio Shell	Physical Activation	CO ₂	93,45 mg/g	[30]
Commercial Activated Carbon	-	-	-	208,3 mg/g	This Study

4. Conclusions

MB adsorption has been studied on commercially obtained activated carbon and applied to 10 different adsorption isotherms to understand the adsorption mechanism. The adsorption mechanism was estimated by examining their suitability to the isotherm equations on the line of the obtained results. The suitability of the isotherm equations was determined according to the correlation coefficients (R^2) of the isotherms. The two isotherms with the highest correlation coefficients were Redlich-Peterson and Langmuir. Since the correlation coefficients of other isotherm graphs are very low, the adsorption mechanisms in which the equations are explained are not correct for this study. Since the Redlich Peterson equation has the highest correlation coefficient, it can be thought that the adsorption mechanism occurs in this way. However, since this equation consists of a mixture of two different equations which are Langmuir and Freundlich equations, it is necessary to investigate which isotherm the equation integrate. The linear state of the Redlich-Peterson equation is shown in equation 13. Since the constant value of β in this equation is approximately equal to 1, the equation is reduced to the Langmuir equation [18,21]. In this case, the adsorption mechanism in the study

integrate the definition of Langmuir. Adsorption took place in a single layer with a homogeneous adsorbent surface.

Conflicts of interest

The author declares that there is no conflict of interest.

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