



Study of Adsorption for Fast Green and Malachite Green Dyes on the Activated Surface

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Abstract

Unfortunately, pollution represents the major dangerous silent weapon in the world now a day, especially after technological development. Water contamination takes a high percentage of the distributed pollutant over the earth, and so many efforts have been made to remove such contamination. Type of dyes also was used in this study to calculation thermodynamic process .,Dyes which is colored materials that can bind in some way to the materials and give them bright colors. These dyes are considered pollutants of water and have toxic effects on humans. Studying the adsorption of fast-green and malachite-green dyes from their aqueous solutions on the surface of charcoal derivative from the kernels of Iraqi dates activated with citric acid as an economic method. Several parameters were studied to investigate the most optimum condition that improves the adsorption process. The adsorption study carried out on the Activated Charcoal derivative from Iraqi Date Cores which act Chemical adsorption, The results obtained after conducting several effects showed the contact time (90 minutes for fast green, and malachite green 30 minutes), the acidity function, as well as the different temperatures (290 to 320 K)
Key word: Adsorption, Fast green, Malachite green, Activated charcoal, Adsorbent, surface ,acid

1.Introduction

The Dyes (pigments) are good adsorbent surfaces can be classified according to their structure, functional group, or ion charge when dissolved in water into :

Chemical classification: Chemical dyes are classified into several classes, according to the chromophore group.

Classification according to the charge of the ion into ionic dyes and non-ionic dyes.

The decomposition of dyes is used in the textile industries, as dyes are one of the largest and most important organic compounds used in chemical industries in the world. Most dyes are made to be environmentally resistant, such as sunlight, so their presence in water cause

Pollution is one of the problems facing humans and the environment, especially after the technological development that accompanies life contemporary . Pollution occurs in its various forms, whether it is air, water, or soil pollution as a result of the presence of some substances, harmful organic and inorganic or due to the increase or decrease in the proportions of some basic compounds in the environment different than natural proportions to it,

this happens as a result of human interventions or due to some natural phenomena . Humans are the primary cause of water contamination. It is the product of human acts taken in order to improve one's self. These might be viewed as one of the different actions that contribute to pollution that man participates in. Pollution is mostly caused by population increase, industrialization, and agricultural activities. Overcrowding in metropolitan places exacerbates the problem. The main contaminants in agnatic environments are agricultural, residential , and industrial wastes. The most significant contaminant of freshwater is sewage .The techniques that have been used in addressing this problem are Adsorption on plant surfaces as well About some physical chemistry techniques such as ozone saturation reverse (Osmosis), silica gel, and others Adsorption is one of the most important of these technologies due to its high efficiency

Adsorption

It is the phenomenon of gathering a gaseous or liquid substance in the form of molecules, atoms, or ions of a substance certain substance called an

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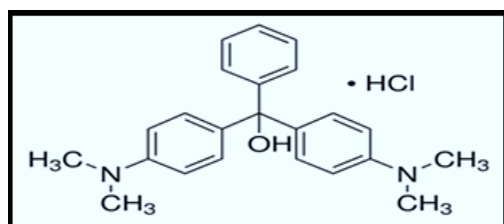
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adsorbate on the surface of another porous solid material which an adsorbent and the bonding is between the molecules of the material characterized by the active sites of the adsorbent surface either through Vander Waals forces. the weak sites are called physical adsorption or through the formation of chemical bonds with sites, it is called chemical adsorption. adsorption is one of the efficient techniques it is so widely used that hardly any industry today is devoid of a unified treatment and purification of wastewater before it is released into the environment by adsorption. Some of the commercially available materials are specifications that qualify them for use as good adsorbents, such as charcoal activator. Adsorption can be classified according to the type of the bond and its strength between the molecules of the adsorbent and the adsorbent material and the heat that accompanies the adsorption process to:

Physical adsorption: It is known as Adsorption Vander Waals (Physisorption). It is the physical attractive forces between the adsorbing surface and the molecules adsorbed on the surface. Very similar to the process of condensation, which may cause the formation of multiple layers of the adsorbent material on the surface of the adsorbent material

Chemical adsorption : It is called Chemisorption for short, which is a chemical force between the adsorbed atoms or molecules and the adsorbing surface. This type of adsorption occurs under certain conditions on a particular surface and may not occur on a surface Another surface when the same conditions are available, or on the same surface when the appropriate conditions change, that is, it has privacy. (Specificity) This type of adsorption is accompanied by much higher thermal changes than physical adsorption .

Malachite green (MG), a basic dye, is commonly used for coloring reasons in the silk, wool, cotton, leather, and paper industries. It is also used to treat parasites, fungal infections, and bacterial infections as a medicinal agent. MG dye, despite its widespread usage, has hazardous qualities that have been linked to cancer, mutagenesis, teratogenesis, and respiratory toxicity. As a result, it is critical to remove MG from wastewater before it is discharged into the environment .Figure (1-2) .



Figure(1):Structure of malachite green

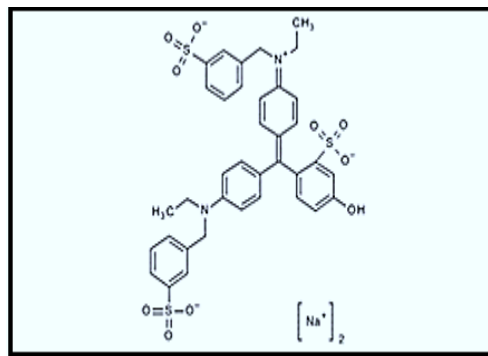


Figure (2): Structure of fast green

Types of Adsorbents

Activated carbon, synthetic zeolites, and polymers are the three forms of adsorbents used on a significant basis. Each kind of adsorbent has properties that make it useful in certain applications but ineffective or uneconomic in others.

A-Activated Carbon

It was the first adsorbent to be used and is still the most used today. It is made from a range of resources, including coal, wood, and coconut husks, via high-temperature steam pyrolysis. Because of its large specific surface area (usually 800 to 1,400 square meters per gram) and the existence of tiny holes of varying sizes, the resultant carbon has exceptional adsorption properties (typically between 2 and 500 angstroms). The activated carbon generated has different properties depending on the kind of material employed .

B- Zeolites

Zeolites have a crystalline structure that is consistent in size and shape, with high specific surface areas and tiny, uniform pores. Because its holes capture molecules of specified sizes while allowing other molecules to flow through, zeolites are frequently referred to as "molecular sieves."

C- Dyes Assorbent and Polymers

Dyes compounds act colored materials that can bind in some way to the materials to be dyed and give them bright colors so that not affected by washing, light, oxygen, acids, and bases contained in their molecules it contains groups called chromophores that are, and other combinations called (auxochromes) of an acidic or basic nature that intensify and fix the color. Textiles, rubber, plastics, printing, leather, cosmetics, and other industries utilize dyes to color their goods. As a consequence, they produce a large quantity of colorful effluent

Polymer adsorbents are granules or beads comprised of strongly crosslinked synthetic polymers that create a matrix of tiny holes and large surface areas. Polymer adsorbents may be used to regulate a broad spectrum of volatile organic compounds (VOCs)

Methodology and Material:

The adsorption study carried out on the Activated Charcoal derivative from Iraqi Date Cores., Iraqi Date Cores, Citric acid, Sodium hydroxide, and Sodium nitrite were purchased from CDH, India, nitric acid and hydrogen chloride was obtained from Himedia (India)

Preparation of the Adsorbent Surface

To obtain carbonated charcoal from the Iraqi date core, the core was washed several times with distilled water and then allowed to dry at a temperature of (100 °C) for two hours before being placed in an oxygen-free oven for two hours at a temperature of (500 °C), after which the charcoal was crushed thoroughly. It is sifted through a sieve with a diameter of (0.1 micrometers), after which it is washed five times with distilled water and dried for two hours at a temperature of (50 °C), as shown in Figure (3).



Figure: (3): (A) Iraqi date Cores, (B) Activated charcoal

Preparation of Standard Solutions and Calibration Curves

This study carried out in chemistry department – Collge of Education-Kufa Univ. Iraq) .,The standard stock solution of fast green and malachite green were prepared at a concentration of (100 ppm), by dissolving a weight of (0.1gm) in (1000 ml) of distilled water. After that, different concentrations of fast green dye were prepared from (3 to 30 ppm), while in the case of malachite green, it was prepared from (2 to 20 ppm). The spectrum was taken for both dyes using a UV-visible spectrophotometer to obtain the lambda max in which the greatest absorption was utilized

using distilled water as a blank. Then, the absorbance was measured for each of the ten samples by a UV-visible spectrophotometer to determine the calibration curve of the dyes. (Fast green and malachite green).

Effect of Contact Time

This study was performed at a temperature of 290 K between activated charcoal and the dye, the volume of all samples was 20 ml with a concentration of 30 ppm for fast green dye, while for malachite green at a concentration of 20 ppm and the weight of activated charcoal (0.2 g) and at different times interval (0.5, 1, 1.5, 2, 2.5, 3 h). After that, the samples are placed for both dyes in a shaking water bath and then the samples were filtered with filter paper, and then the filtrated absorbance was measured by UV-visible spectroscopy

Effect of Surface Weight on Adsorption of Fast Green and Malachite Green

The effect of weight surface on the adsorption capability was investigated to calculate the best contact time for Fast-green and Malachite-green dye at a constant temperature of (290) degrees Celsius with various weights (0.05, 0.1, 0.15, 0.2, 0.25, 0.3) gm, and volume (20ml). The prepared samples were placed in a shaking water bath for 90 min. and 30 min. for Fast-green and Malachite-green dye respectively, and then filtered with filter paper, and the absorbance was measured by a spectrometer (UV-vis).

Effect of Acid Function (pH)

This study was done to measure the acidity function (pH) by taking six different concentrations of the highest concentration concerning the two dyes (Fast-green and Malachite-green), using a pH meter to measure the pH (2, 4, 6, 8, 10). In the next step, a fixed weight from the surface was added to each of these samples, and then measured the absorbance with a UV-visible spectrophotometer, all these studies (Effect of Acid Function (pH), Adsorption Isothermal, Effect of Surface Weight, Effect of Contact Time, and other studies) carried out in chemistry department –Collge of Education-Kufa Univ, Iraq)

Effect of Zero Point

To find the surface charge on which the adsorption process takes place, this study was done by taking six samples of a volume of 20 ml of both dyes, with a concentration of (30ppm) for Fast Green dye and a concentration of (20 ppm) for Malachite Green dye, respectively. Then add to each of the six samples for each of the two dyes a fixed weight of the surface (activated charcoal) in the amount of

(0.05 gm), then add 10 ml of sodium nitrite solution and leave it for 24 hours, after that, the absorbance was recorded and from the knowledge of the initial and final pH, the surface charge is calculated .

$$\Delta\text{pH}=\text{pH}_f-\text{pH}_i \quad \dots\dots\dots(1)$$

pH_f = After Adsorption, pH_i = before adsorption

Adsorption Isothermal

Ten samples were prepared from each of the two dyes, Fast-green, at concentrations of (3-30 ppm), while Malachite green dye, at concentrations (2 - 20 ppm), respectively, and 20 ml of each concentration of both dyes, was added to the surface with a weight of (0.05gm) These samples were placed in a shaking water bath and after a specified period of time (Fast green for 90 min, Malachite green for 30 min.), then the samples were filtered with a filter paper. Absorbance measurement in a UV spectrophotometer at the corresponding max of each of the two dyes and detect the concentration after adsorption (c_e) through the calibration curve by the following equation

$$q_e = (c_0 - c_e) \frac{Vsol}{m} \quad 2)$$

Where q_e : The amount of adsorbent (mg/gm)

c_0 : The initial concentration of the dye (mg/l)

c_e : Concentration at equilibrium for the dye (mg/l)

$Vsol$: The total volume of the adsorbent, m : The weight of the adsorbent (gm)

Result and Discussion

Calibration Curve for Fast Green and Malachite Green Dyes

The maximum absorption wavelength was obtained after scanning of each dye, and it was found to (637 for fast green and 617 for malachite green) as declared in Figure (4). Meanwhile, the calibration curves of mentioned dyes were constructed using the double-spectrum ultraviolet and visible spectroscopy at the specified λ_{max} for both dyes as shown in figure (5).

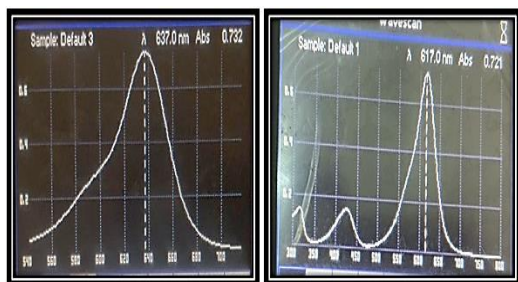
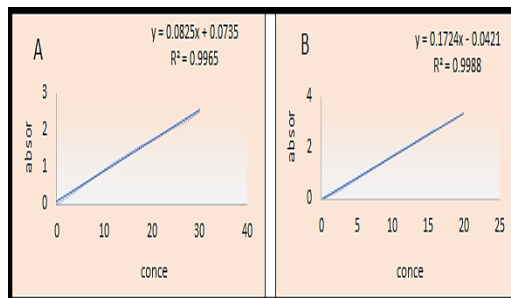


Figure.(4):UV-visible spectra of (A)Fast green and (B)malachite green in distil water pH (7)



Figure(5)A-Calibration curve of fast green, (B) Calibration curve of malachite green

The effect of contact time

To find out the best contact time between the dye and the activated surface derived from Iraqi date kernels, this experiment was conducted and the results showed according to the calculations that the best time for contacting Fast green dye was 90 min., while the best contact time between the Malachite green dye and the surface according to the results was 30min. The amount of adsorption (Q_e) is high because the active sites on the surface are not occupied and thus the highest adsorption of the dye will occur on the active sites on the surface. After that, the adsorption capacity will decrease due to the occupation of these sites, that is, it reaches the state of saturation with time. The adsorption is low and at the specified time of contact with this dye, the adsorption capacity will rise and the active sites of the activated surface will be occupied , Figure(6) demonstrate this effect

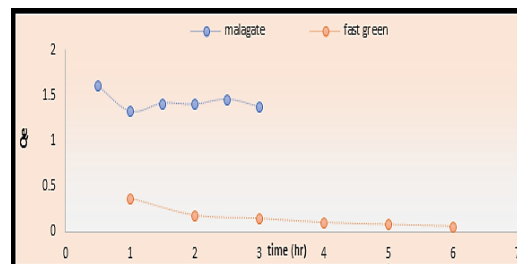


Figure (6) Effect of contact time for adsorption of 20 mg / L of dyes on Activated Carbon from Iraqi Waste Date Cores at 290k and pH =7

The Effect of Surface Weight on Adsorption Dyes

The adsorption process on activated carbon does not depend on the surface area of the carbon only but also depends on the chemical composition, which varies according to the raw material used, as well as the conditions for its preparation and activation. Adsorption is a process that has a positive relationship with an increase in the percentage of porous structures in the adsorbents in general and activated carbon in

particular. Figure (7) represents the relationship between the surface, the weight of adsorption, the amount of adsorbents mg/g at 290 °C, and the acidity function= 7, similar outcome was obtained (Kushwaha AK et al 2014).

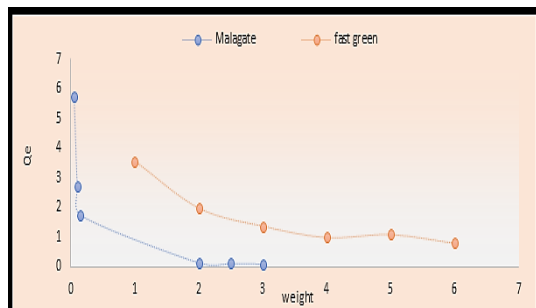
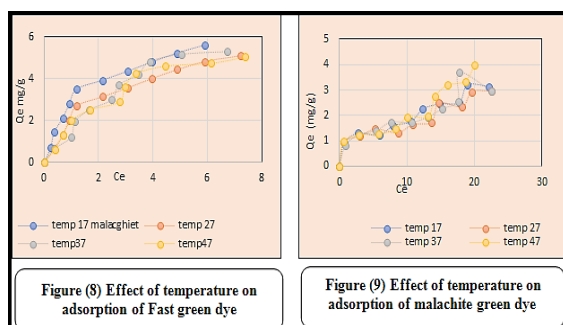


Figure.(7) Effect of Weights for adsorption of 20 mg /L dyes on Activated Carbon from Iraqi Waste Date Cores at 290k and pH =7

Effect of Temperature.

The effect of temperature is an important factor in carbon-activated charcoal practical applications. Also, the temperature is an important parameter in the study of adsorption to know the extent to which temperature affects the efficiency of adsorbents. Experiments were carried out at different temperatures. The effects of temperature in adsorption were studied on each of the dyes (Fast green and Malachite green) on the surface of the coal of Iraqi date cores in the thermal range (290 to 320 K) as shown in the figure below for each of the two dyes, respectively. The obtained indicate that adsorption of Fast green dye and malachite green dye, respectively, on the surface of charcoal derivative from Iraqi date cores increases with increasing temperature at the limits (310 and 320 K) as revealed in :



Thermodynamics of Absorption

As declared in figure (10), the thermodynamic behavior of Fast Green and Malachite Green adsorption on charcoal activated was evaluated using the following equations .

$$\Delta G = -RT \ln K_c$$

$$\Delta G = \Delta H - T\Delta S$$

$$\ln K_c = -\Delta H/RT + \Delta S/R$$

Where ΔH is the enthalpy, ΔS is the entropy, and ΔG is the free energy change where enthalpy and entropy are obtained by drawing $\ln X_{eq}$ versus $1/T$.

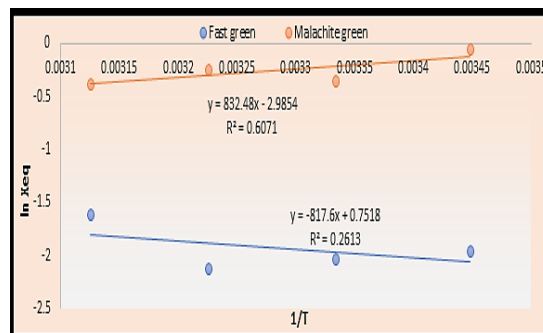


Figure (10) Thermodynamic process at 290k for adsorption of 20 mg / L of dyes on Activated Carbon from Iraqi Waste Date Cores at pH =7

Adsorption Isotherm Model Study

There are many varieties of adsorption isotherms, including Freundlich, Langmuir, Temkin, and Harkins-jura isotherms, the latter two of which are the most often utilized. Any of these might be employed for the best fitting of the data according on the kind of data supplied. In general, an adsorption isotherm is a graphical representation that shows the relationship between the amount of adsorbate adsorbed on the surface of an adsorbent and the quantity of adsorbate adsorbed on the surface of an adsorbent. Depending on the criteria, there are two forms of isotherms (Kumar Shetty M et al 2021).

Langmuir models

The Langmuir isotherm model shows that the adsorption occurs on homogeneous surfaces of the adsorbents. That is, in this model, the adsorption is of a single-layer type, and the experiments were conducted at different temperatures (290 to 320 K) with different initial concentrations for each of the two dyes (fasts green and malachite green.) in a successive manner, Figure (11) below illustrates these outcomes.

$$C_e / Q_e = 1 / Q_m * C_e + 1 / Q_m * 1 / K \quad \dots\dots(6)$$

where C_e (mg/L) is the equilibrium MG concentration, q_e (mg/g) is the mass of MG and FCF adsorbed at equilibrium per, q_m (mg/g) is a concentration standard linked to the adsorbent's monolayer adsorption capacity, and K_L (L/mg) is the Langmuir constant related to adsorption efficiency of solute. Figure 1 shows a straight-line plot of C_e/q_e vs C_e with slope equal to C_e/q_e and

intercept equal to $(1/q_m)$ $(1/K_L)$ (11). K_L , q_m , R_L , and the linear correlation coefficient values, R^2 , are presented in Table (1).

Table.(1):Langmuir constants for Fast green and Malachite green on charcoal of date Cores

Dyes	Qm	Kl	R ²
Fast Green	3.93	0.11	0.78
Malachite Green	7.04	0.61	0.98

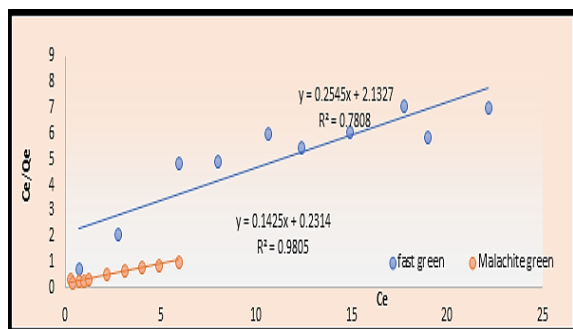


Figure (11): Langmuir isotherms at 290k for adsorption of 20 mg / L of dyes on Activated Carbon from Iraqi Waste Date Cores at pH =7

Isotherm Freundlich.

As shown in figure (12), the adsorption occurs on heterogeneous surfaces and are multi-layered. The adsorption equation of Freundlich was applied to both Fast green and Malachite green dyes on the surface of the date kernels within the thermal range (290 to 320 K) The Freundlich constants, represented by (n) , which express the adsorption intensity, were estimated. The amount of curvature of the curve that the surface is saturated with. and (K_f) which expresses the adsorption capacity of the surface, from the slope and intercept. When plotting $\ln Q_e$ against $\ln C_e$ and the constants are shown in Table (2) according to the following equation (Plaza Cazón J, et al 2012).

$$\text{Log } Q_e = \text{Log } K_f + 1/n \text{ Log } C_e \quad \dots\dots(7)$$

Where K_f and n are Freundlich Constants characteristics of the system, including the adsorption capacity and the adsorption intensity, respectively

Table.(2) Freundlich constants for fast green and Malachite green on charcoal of date cores

Dyes	K _f	n	R ²
Fast Green	0.89	2.77	0.84
Malachite Green	2.34	1.75	0.90

Harkin -Jura Adsorption Isotherm:

Another parameter in the investigation of isothermal behavioral of adsorption process depend

on the application of Harkin equation, the outcome is shown in figure (14),

$$1/q_e^2 = B/A - 1/A \log C_e \quad \dots\dots(9)$$

Where q_e denotes equilibrium adsorption and C_e denotes equilibrium concentration. The isotherm constant is B , and the Harkins-Jura isotherm parameter is A . It is hypothesized that multilayer adsorption occurs in a heterogeneous pore distribution in this isotherm⁽²⁵⁾.

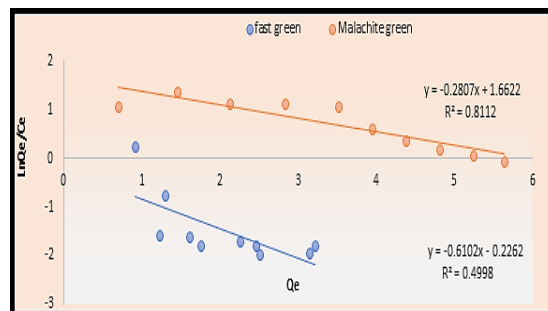


Figure (13) Elovch isotherms at 290k for adsorption of 20 mg / L of dyes on Activated Carbon from Iraqi Waste Date Cores at pH =7

Table (4):Harkin's constants for Fast green and Malachite green dye on date Cores charcoal

Dyes	A	B	R
Fast green	-	-	0.94
Malachite green	1.36	0.77	
	-	-	0.50
	1.09	0.42	

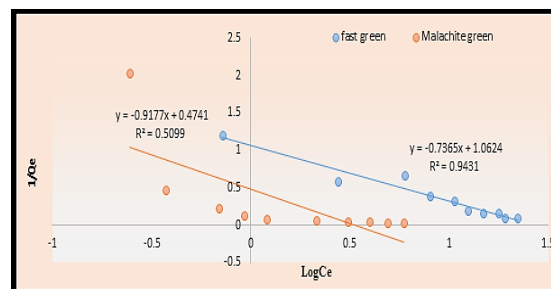


Figure.(14).Harkin-Jura isotherms at 290k for adsorption of 20 mg/L of dyes on Activated Carbon from Iraqi Waste Date Cores at pH =7

Temkin Isotherm

Moreover, Temkin parameter also describe the isothermal work of the adsorption process through the application of the following equation ⁽²⁶⁾:

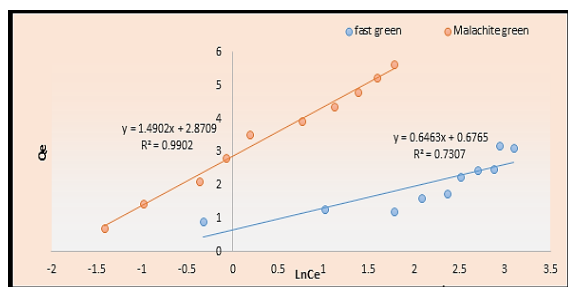
$$Q_e = B \ln A + B \ln C_e \quad \dots\dots(10)$$

A and B are the Temkin constants and can be determined by a plot of Q_e versus $\ln C_e$. The parameters, A and B , together with the correlation coefficient.

Table (5) Constants Temkin dyes Fast green and Malachite green on charcoal of date Cores

Dyes	KT	B	R
Fast green	2.84	0.64	0.73
Malachite green	6.86	1.49	0.99

The result of this investigation is seen in Figure (15), which illustrate that the adsorption process goes mostly with the physical type (Plaza Cazón J, et al 2012).

**Figure (16) :Temkin isotherms at 290k for adsorption of 20 mg / L of dyes on Activated Carbon from Iraqi Waste Date Cores at pH =7****Table.(6):Constants of Dubinin Isotherm for Fast green and Malachite green on charcoal of date Cores**

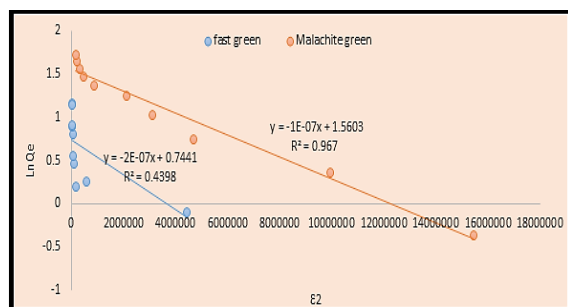
Dyes	Qm	B	R
Fast green	2.06	-2E-07	0.43
Malachite green	5.2	-1E-07	0.96

Dubinin Isotherm:

The Dubinin–Radushkevich isotherm is often used to explain the adsorption process onto a heterogeneous surface with a Gaussian energy distribution. The model has had a lot of success fitting data with high solute activity and a wide range of concentrations⁽²⁷⁾. The obtained result are revealed in figure (17)

$$\ln Q_e = \ln Q_m - B\epsilon^2 \quad \dots\dots(11)$$

$$\epsilon = R T \ln(1+1/C_e) \quad 12)$$

**Figure.(17):Dubinin isotherms at 290k for adsorption of 20 mg / L of dyes on Activated Carbon from Iraqi Waste Date Cores at pH =7****Conclusion**

The results showed that the activated charcoal derivative from the Cores of Iraqi dates is a good adsorbent for removing both fast-green and malachite-green dyes from their aqueous solutions, even if the dyes concentration was very low by (0.05). Moreover, from the analysis of the outcome and fitness to the isothermal equations, the mechanism of adsorption are physical type can be predicted.

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