Thermodynamics and Kinetic study of Bismarck Brown R Dye Adsorption from Aqueous Solution using Sewage Sludge

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Doi 10.29072/basjs.202026

Abstract

Synthetic wastewater and other untreated water must be treated to allow for drainage into rivers and re-use later. Adsorption focuses on the importance of means of extracting synthetic and inorganic contaminants from water. Research for products that are available and inexpensive to use as adsorbents. Sewage sludge can be used to eliminate the dye Bismarck brown R. the impact of different empirical determinants such as contact time, temperature, solution pH, the weight of adsorbents, initial dye concentration, and ionic strength were implemented. The adsorption experiments involved all these equilibrium adsorption isotherms and kinetics. The implementation of Langmuir and Freundlich isotherms was examined at various temperatures, and the Langmuir isotherm showed better suitability with experimental results. the thermodynamic determinants showed that the adsorption was an automatic practicability)ΔG was negative), the positive value of (ΔH) was investigated endothermically and (ΔS) was positive value (random), The kinetic information c shows that the adsorption of Bismarck brown R with activated sewage sludge belongs to the pseudo-second-order kinetics.

Article inf.

Received: *13/6/2020*

Accepted

22/7/2020

Published

31/8/2020

Keywords:

Adsorption, Bismarck brown R, Sewage sludge, Thermodynamic and Kinetic.

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1. Introduction

Pollution is the addition of undesirables foreign matter to the environment as a result of enormous industrial development and modernization[1], Synthetic dyes are one of most important materials that are widely used in many industries such as textiles [2], leather [3], paper [4], cosmetics [5], printing [6], pharmaceutical and food industries [7], and may generate large amount of aqueous colored effluents and causes an important environmental problems faced the human in last century[8]. The presence of very small amounts of dye in water (< 1ppm for some dyes) causes aesthetic worsening and decrease the solubility of dissolved oxygen , water transparency, and sunlight permeability , affecting aquatic life and the food chain [9]. Since some dyes are toxic and carcinogenic, their disposal in to waste water without advanced treatment causes environmental problems and damages the ecosystem [10]. Therefore, their removal became a vital task. Various methods such as sedimentation , ion exchange [11] , flotation, electro chemical [12] , chemical precipitation [13] , coagulation [14] , and flocculation [15] , reverse osmosis [16] , adsorption [17] etc. have been investigated for the removal of dyes from aqueous solutions [18] Adsorption seems to be the most effective technique that has been successfully employed for dye removal from waste water [19]. The production of activated carbon from waste water sludge means a significant saving in the cost of raw material. This material may be considered suitable for the production of activated carbon if it is decomposed in thermal conditions under controlled conditions or with some chemical treatment, given considerations such as the carbonic composition of the wastewater sludge and its high volatile portion quality, multiple scientific studies have been undertaken to demonstrate the viability of this conversion [20] [21]. In this present study, we investigated the adsorption of Bismarck brown R dye on the surface sewage sludge. The system variables studied include sorbent dose, contact time, initial concentration of the dye, pH, FTIR analysis, ionic strength and temperature.

2. Materials and Methods

2.1 Materials

2.1.1 Bismarck brown R dye (BBR)

Bismarck brown R (BBR), is a diazo dye with molecular formula: ($C_{21}H_{24}N_8.2HCl$) was procured from sigma Aldrich and the molecular weight of 461.39 $g.mol^{-1}$,IUPAC name (5,5-

[(4-methyl-m-phenyl) bis (azo)] bis [toluene 2,4-diamine] di hydrochloride) , the chemical structure of (BBR) is shown in figure (1) . BBR dye solution was prepared by dissolving 0.1gm of dye in 1L . The 100 mg/l after which dilute to the Concentration needed. The wavelength at maximum absorbance (λ_{max}) was restriction by Scanning and noticed to be 456.5 nm .

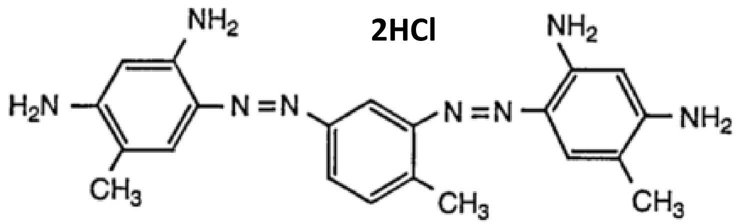

Figure 1: Chemical structure of Bismarck Brown R [22] .

2.1.2 Activated sewage sludge

Activated carbon prepared from sewage sludge was used as Adsorbent. The sewage sludge was collected as slurry from the settlers of Hamdan waste water treatment facility in Basra city .The sludge was washed many times by DW used to remove solid materials . Under sunshine the sludge was dried and then soaked for an hour in the furnace at 600℃ and then crushed and sieved to a particle size of 75 µm and then used.

2.1.2.1 Activated sewage sludge characterization

To explain the properties of particles (e.g., surface morphology and particle size) at the subsequent powder sludge, the powder texture was detected by the scanning electron microscope (SEM) type chemical function of groups is investigated by FT-IR devices Shiatsu. use KBr bead process of 25 ºC, The spectra have been recovered in the wave number spectrum from 400 to 4000cm$^{-1.}$

2.2 Adsorption study of Bismarck Brown R on Activated sewage sludge

Batch equilibrium studies have been carried out to determine the optimal pH, contact time and equilibrium isotherms. A series of 100 ml stopper conical Flasks were used by adding 0.5 g of sludge particle size of (75 µm) to 50 ml of 100 mg/l of Bismarck brown R dye.

The tests were performed at 25 ºC after Soft shaking for time period at 140 rpm . The sample was centrifuged and the concentration of the dye determines using (UV-VIS) spectroscopy (UV-1200 spectrophotometer). The removal efficiency and the equilibrium up take of BBR dye Was determined by equation

$$Q_e = \frac{V_{sol.}(C_\circ - C_e)}{m} \text{ X } 100 \text{ -----}(2)$$

Where $C_\circ$ is concentration of Bismarck Brown R dye before the adsorption (mg/L), C_e is concentration of Bismarck Brown R dye at equilibrium (mg/L) , m (g) weight of activated sewage sludge and $V_{solution}$ volume of solution of Bismarck Brown R dye in liter, Q_e is the equilibrium adsorption capacity (mg/g).

2.3 Kinetic study of Bismarck Brown R on Activated sewage sludge

Kinetic of Bismarck Brown R on sludge were done with (0.5 g) of the sludge powder converted in to 100 ml screw cupped conical flask holding 50 ml of 100 mg/l of Bismarck Brown R dye, Diverse specimens were shaken At (140 rpm) at (10, 20, 35 and 45ºC) by using isothermal water-bath shaker. and the mixture was carried out at taken out at various periods of time, The mixture was centrifuged The amount of the adsorbed dye is then determined spectrophotometrically at 456.5nm, and Approximated via eq (2).

3. Results and Discussion

3.1 Characterization of the Activated sewage sludge

3.1.1 Fourier Transform Infrared Spectra (FT-IR)

FT-IR devices type Shimadzu (400-4000 cm^{-1}) was used for the detection of functional groups in the Activated sewage sludge. Spectra FTIR as shown in fig (1) Signify that strong band appears at (3435 cm^{-1}) resulting from the stretching vibration of the hydroxyl groups. The absorption band at (1660 cm^{-1}) belongs to carbonyl groups. The spectrum also showed peaks confined between (1400-1600 cm^{-1}) belongs to the groups of (C=C) at the aromatic rings the absorption band at (1120-1153 cm^{-1}) represent the adsorption of the Si-O group. The spectrum also showed absorption band at (1037 cm^{-1}) belongs to the group of (P-H) of phosphine compound and another band at (877 cm^{-1}) belongs to (S-O) group. These chemical groups represent effective sites where the adsorption process occurs because of its negative charge, which gives these groups the ability to from chemical bonds or physical bonds because the contain an electronic pair or an electrostatic charge that you can share to from a physical link[23].

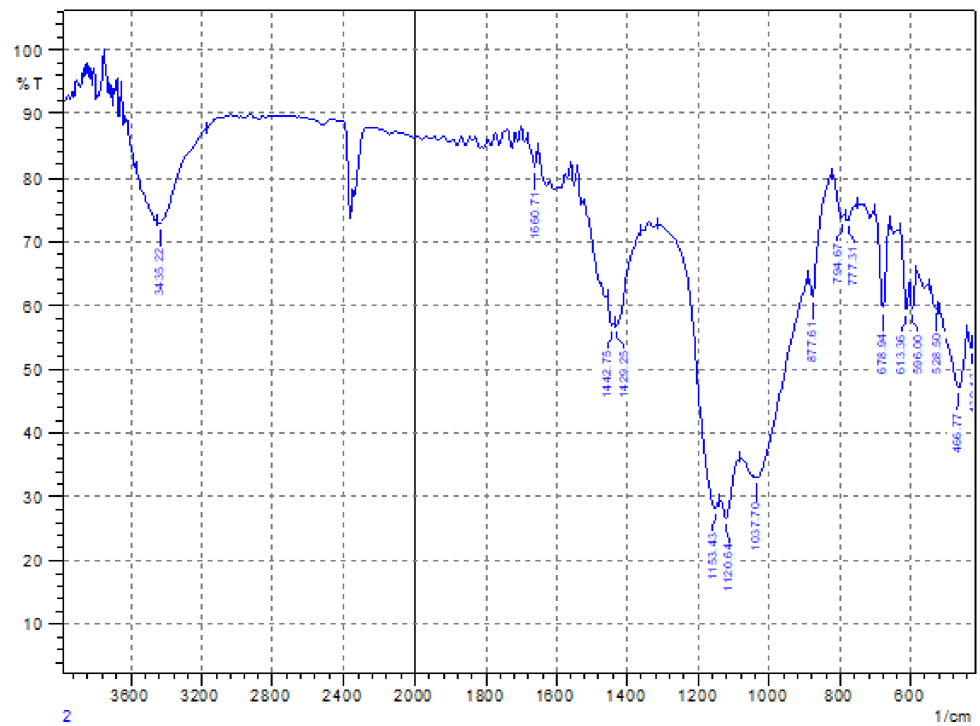

Figure 2: FT-IR of sewage sludge.

3.1.2 Scanning Electron Microscope (SEM)

Scanning electron microscopy is a species of electron microscope that creates sample pictures by use a concentrated electron beam to scan the surface. The electrons communicate with the atoms in the specimen, delivering specific signals providing specific data on the surface topography and the sample; compounding as it gives complete information about the surface topography and the composition of the prepared adsorbent material. The SEM plays a fundamental role in understanding the nature of adsorbents. The morphology of sewage sludge were investigated using SEM.

It appears through the images of the scanning electron microscope of Figure (3) of the surface of the sewage sludge before the adsorption process. It is granular and has an irregular distribution and contains many pores, or effective adsorption sites, and these pores allow dye to penetrate through during the adsorption process.

As shown in Figure (4) after the adsorption process, a layer is formed on the surface of the sewage sludge by the dye Bismarck brown R It works on the adsorption sites by confining itself to the active groups that existed before adsorption.

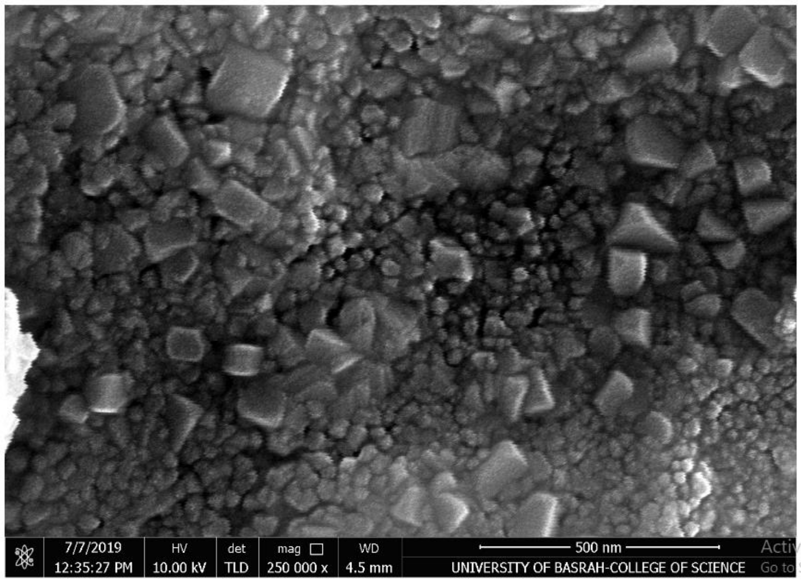

Figure 3: SEM image of sewage Sludge before BBR adsorption .

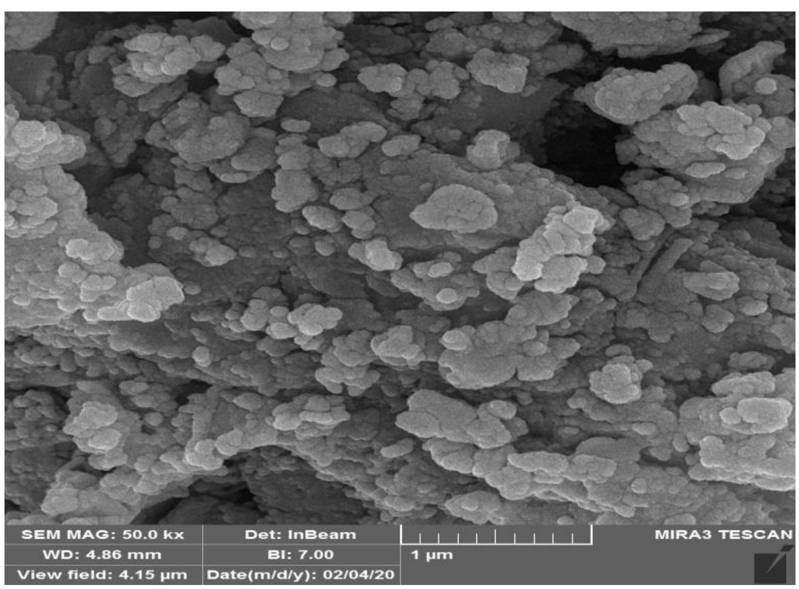

Figure 4: SEM image of sewage sludge after BBR adsorption .

3.2 Factor Affecting Adsorption Process

3.2.1 Effect of contact time

The removal of Bismarck brown R dye by adsorption on Activated sludge was analyzed as a functionality of contact time at different period (1/4-24 hr.) at (25ºC) and percentage removal signed contra time (fig 5) indicates the adsorption average of BBR onto sludge which is high at the start and becomes steady after (4hr). The removal percentage of Bismarck brown R dye on Activated sludge are checked 86.73%

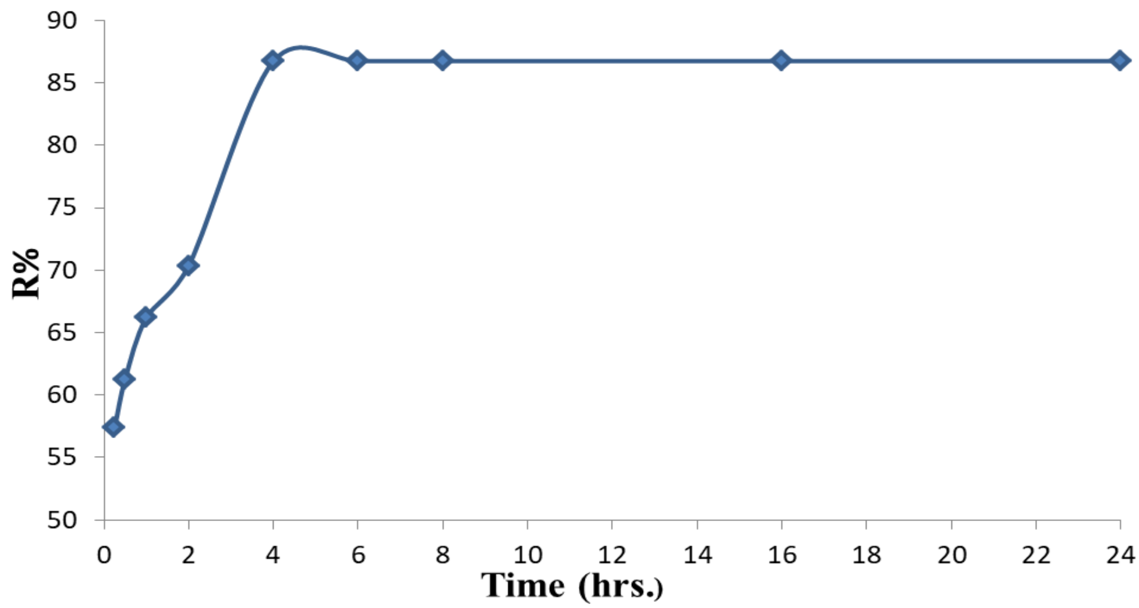

Figure 5 : Effect of contact time of BBR dye with sludge .

3.2.2 Effect of pH on Adsorption of BBR dye

The pH of the dye solution is plays a key role in the capacity of adsorption where it influences both the degree of ionization of the dye and the surface properties of the adsorbent [24],in this study the pH effect on the adsorption of the dye was analyzed while the initial dye concentration, contact time , temperature and sludge concentration were fixed (50 mg/l), (4 hrs), (25 ºC), (0.5 g) respectively. It studied the impact of pH on dye adsorption with in pH extent (2-11). The pH was modified with the addition of a minimal volume (0.1 M) of HCl and (0.1 M) NaOH , The results obtained showed that the removal percentage decreased from 96.60 % to 73.75 % for BBR

Removal when pH accelerates between 2 and 11 (see fig 6) , At acidic solution the BBR molecules exist in cationic form , Consequently, the adsorption capacity increases result to the propensity of the BBR dye to agglomerate on the adsorption surface rather than to bind with solvent molecules, whereas The adsorption capacity in the base medium decreased result to an increase in the negative cheery of the dye molecule and electrostatic repulsion occurs between dye molecules and the adsorbent surface. [25].

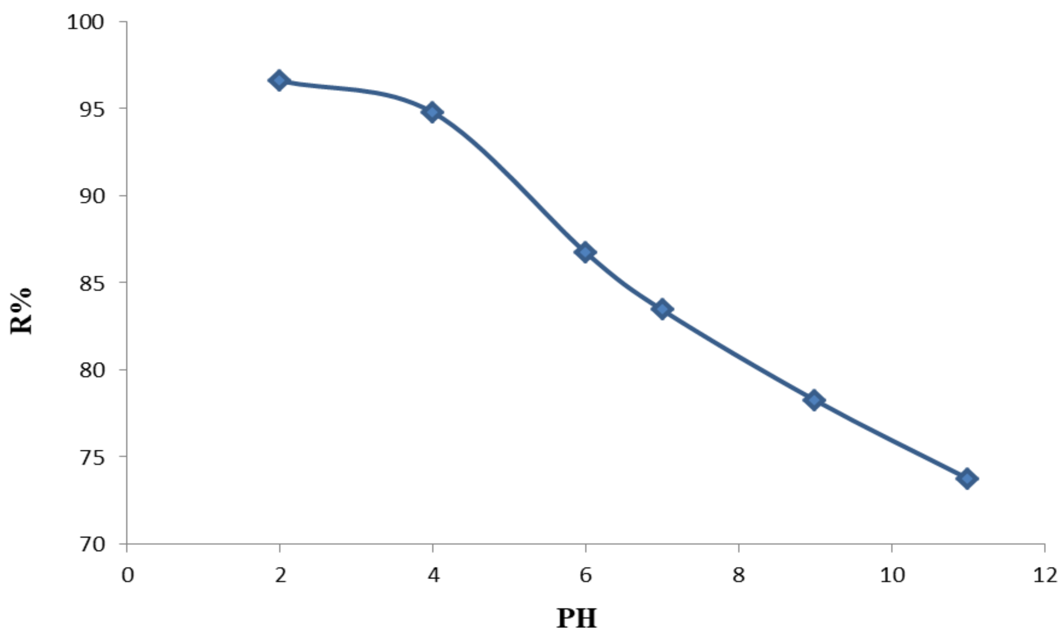

Figure 6: Effect of pH solution on adsorption of BBR dye with sludge .

3.2.3 Effect of initial dye concentration

The affect of initial concentration of Bismarck brown R dye was analyzed by different initial concentration of dye from (10-100 mg/l) at (25ºC) .The results showed that the percentage of dye adsorption decreased with initial concentration raised .The number of free adsorption sites reduces hence the removal percentage decreases by increasing the initial dye concentration [26]. The figure (7) revealed that the C_o (mg/l) was the ideal initial concentration of BBR.

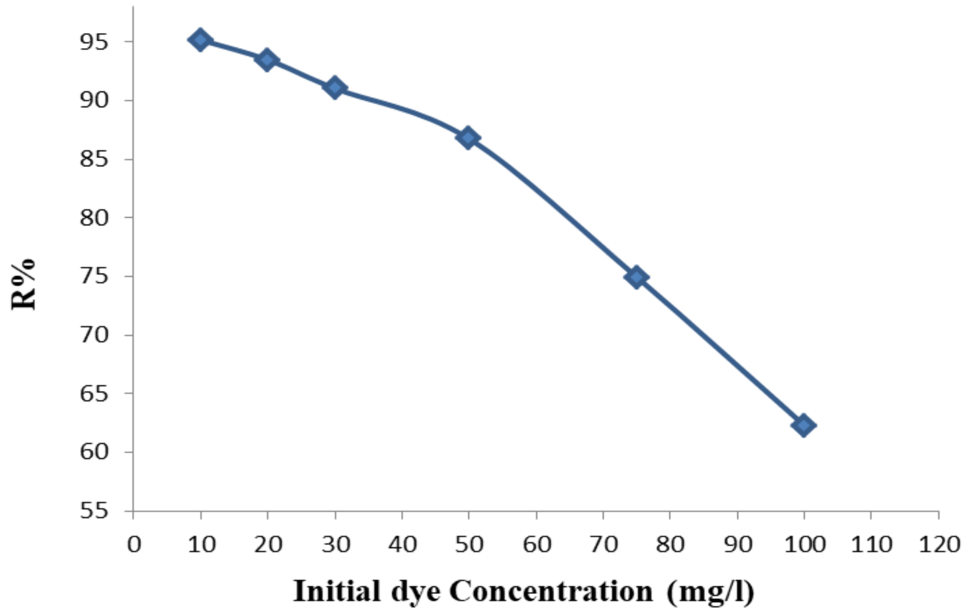

Figure 7 : Effect of initial dye concentration on adsorption of BBR dye with sludge.

3.2.4 Effect of Adsorbent Dose

The effect of different sludge weights on BBR percentage removal at contact time (4 hrs) was examined by the sludge amount range from (0.1-1.5 g) in 50 mg/l BBR solution. figure (8) shows an raised in percentage removal of Bismarck brown R dye with raising of activated sludge amount and that was because large surface area requires more adsorbent sites by increasing the sludge weight [22]

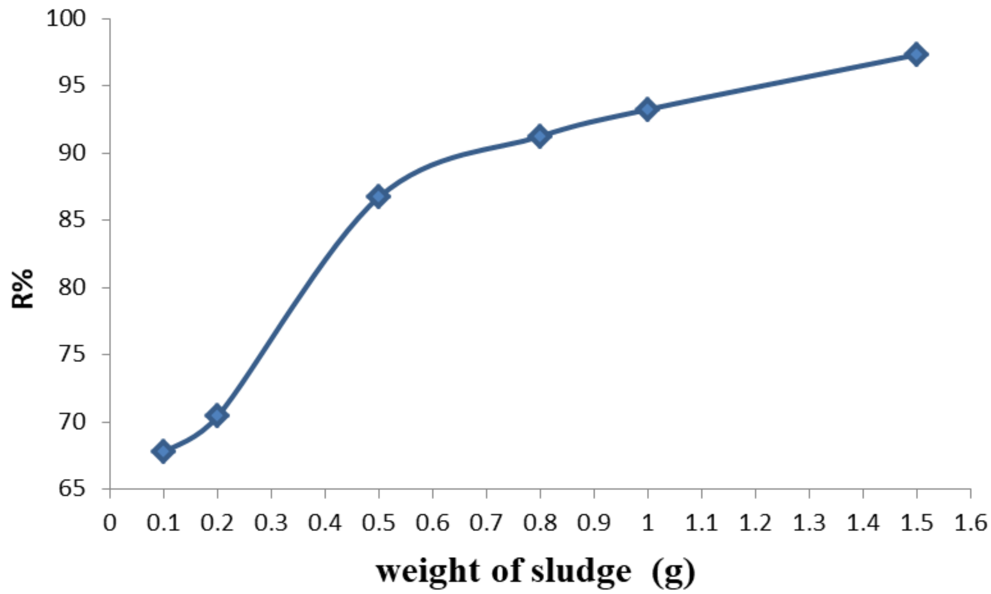

Figure 8: Effect of adsorbent dose on adsorption of BBR dye with sludge.

3.2.5 Effect of the ionic strength on the adsorption of BBR with sludge

The effect of ionic strength on adsorption of the dye was studied by the addition various concentrations of salts (NaCl, $CaCl_2$, Na_2SO_4) to the solution. The concentration of the salts was kept in the range of 0.01 to 0.08 M. As show in (figure 9), increasing the ionic strength of the solution caused decrease in adsorption capacity of the dye this could be attributed to the competition phenomenon between the dye molecules and the ions for the same sites of the adsorbent. On the other hand , ionic atmosphere may be progressively for med around BBR molecules with increased salts concentration and result in the reduction of BBR adsorption onto sludge [27] .

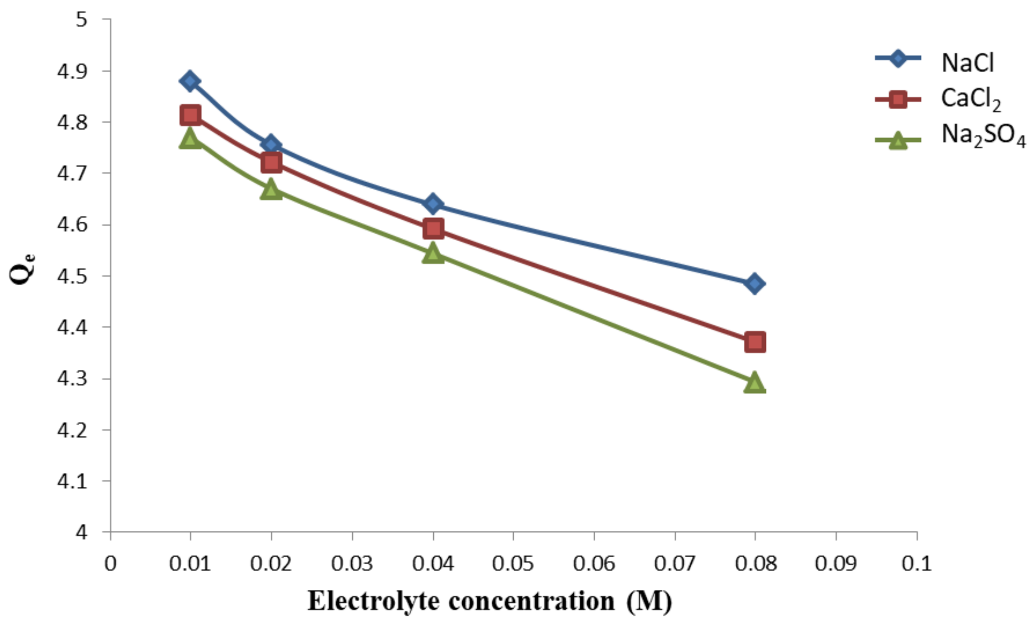

Figure 9: Effect of ionic strength on adsorption of BBR dye with sludge.

3.3 Adsorption isotherm

Bismarck brown R dye adsorption from aqueous solution on Activated sludge was studied at various temperatures (10,20,35and 45 ºC) the general shape of dye adsorption isotherm are show in figure (10) Where quantity of activated sludge adsorbed. (Q_e) was plotted as an equilibrium concentration function (C_e) at (10, 20 ,35 and 45 ºC) .

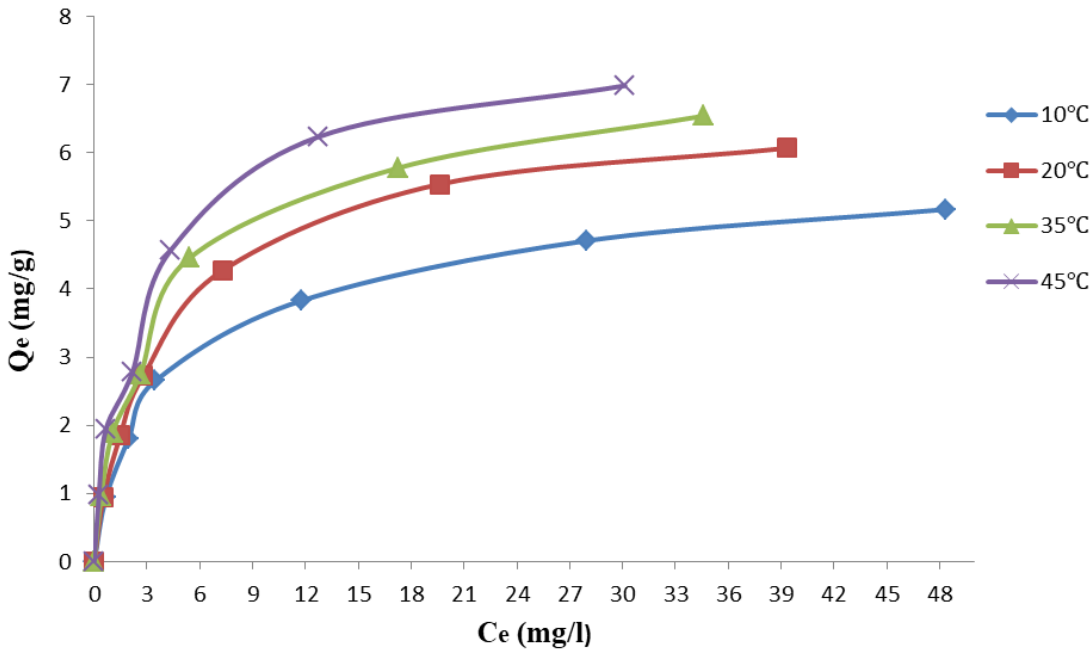

Figure 10: Adsorption isotherm of Bismarck brown R dye with sludge at different temperatures

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The adsorption isotherm type of BBR dye on sludge corresponding to (L2 type) in the classification Giles. This type, indication of horizontal orientation of adsorbed in which the adsorbed molecules are parallel to the adsorbent surface [28] The experimental adsorption data were extended to both the theatrical equation Langmuir isotherm and theoretical equation Freundlich. The results of Langmuir (eq.3) and Freundlich (eq.4) were extended.

$$\frac{C_e}{Q_e} = \frac{1}{Q_m b} + \frac{C_e}{Q_m} \text{......(3)}$$

Where Q_m is the maximum adsorption capacity (mg/g) and b is related to the adsorption energy.

$$\log Q_e = \log K_f + \frac{1}{n} \log C_e \text{.......(4)}$$

Where K_f is a function of the adsorption capacity and n is the intensity of adsorption.

Table 1: Results of application of Langmuir and Freundlich isotherm on the system studied.

	Temp. (°C)	Langmuir			Freundlich		
		Q_m (mg/g)	b (l/g)	R^2	K_f	n	R^2
BBR	10	5.531	0.244	0.9982	1.331	2.591	0.9469
	20	6.627	0.267	0.9996	1.510	2.321	0.9495
	35	7.086	0.307	0.9978	1.720	2.348	0.9519
	45	7.543	0.391	0.9964	2.031	2.399	0.9557

The isotherm Langmuir and Freundlich was extended on experimental data on the adsorption of BBR dye on sludge by plotting (C_e /Q_e) versus (C_e) and (log Q_e) versus (log C_e) Consequently, (Figures 11 and 12). The results of table (1) show that the value of (Q_m) increased with increasing in the temperature because the adsorption was endothermic, The results also indicate that the Langmuir isotherms are best suited on this system than the Freundlich as seen by the linear (C_e / Q_e) versus (C_e) relationship (Figure 11).

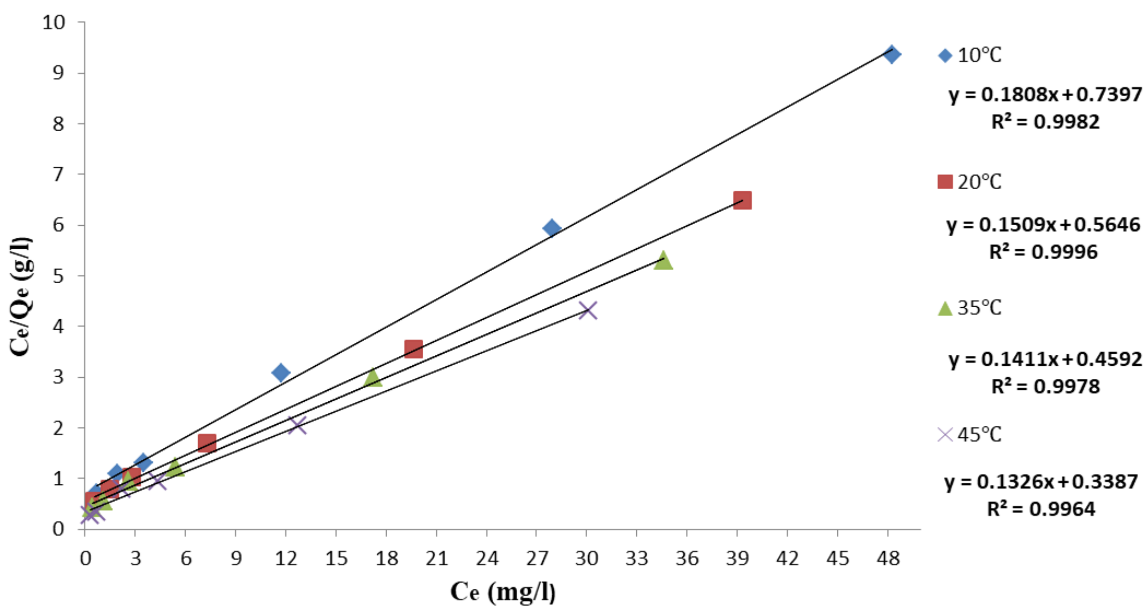

Figure 11: Linear from of Langmuir isotherm of BBR on sludge at different temperature.

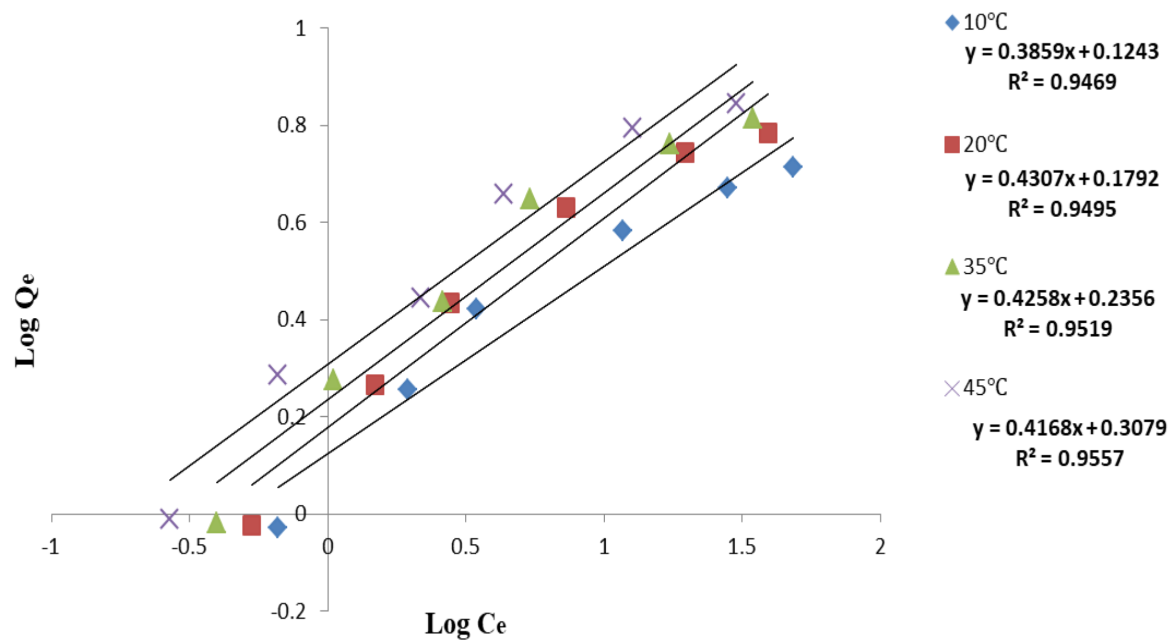

Figure 1): Inear from of Freundlich isotherm of BBR dye on sludge at different temperature.

3.4 Thermodynamic study

Temperature's effect on adsorption of BBR dye onto sludge was studied in the range of (10-45ºC) using various initial concentrations, The thermodynamic parameters of Gibbs energy change (ΔG) enthalpy change (ΔH) and entropy change (ΔS) can be used to approximate the efficiency of the adsorption process using the following equations.

$$\Delta G = -RT \, Ln \, K \ldots\ldots\ldots\ldots (5)$$

$$K = C_{Soild} / C_{Liquid} \ldots\ldots\ldots\ldots (6)$$

$$Ln \, k = \Delta S/R - \Delta H/RT \ldots\ldots (7)$$

Where (ΔG) is the Gibbs energy change ($KJ.mol^{-1}$), K is the equilibrium constant , C_{solid} is the solid phase concentration of equilibrium (mg/l), C_{liqued} is the liquid phase concentration of equilibrium (mg/l), T is the temperature in kelvin and R is the gas constant (0.0083 $KJ.mol^{-1}.K^{-1}$). ΔH ($KJ.\ mol^{-1}$) and ΔS ($J.mol^{-1}.K^{-1}$) can be estimated from the slope and intercept of eq. (6), Consequently,. The thermodynamic parameters for the temperature ranges studied are mentioned in table (2).

Table 2: Thermodynamic functions for adsorption of studied dye.

	C_o mg\l	Equilibrium constant k				$-\Delta G$ $KJ.mol^{-1}$				ΔH $KJ.mol^{-1}$	ΔS $KJ.mol^{-1}.K^{-1}$
		Temperature (kelvin)				Temperature (°C)					
		283	293	308	318	283	293	308	318		
BBR dye	10	14.197	17.939	24.125	36.313	5.866	6.746	8.066	8.946	19.038	0.088
	20	9.224	12.513	18.102	29.395	5.124	6.050	7.565	8.755	23.543	0.101
	30	7.643	9.795	10.511	12.805	4.623	5.133	5.898	6.408	9.810	0.051
	50	3.258	5.867	8.226	10.528	2.819	3.759	5.169	6.109	23.783	0.094
	75	1.685	2.822	3.350	4.904	1.241	2.011	3.166	3.936	20.550	0.077
	100	1.069	1.543	1.890	2.318	0.075	0.625	1.450	2.000	15.490	0.055

The plots of LnK opposite 1/T were found to be linear with a correlation coefficient (R^2 = 0.9643-0.9712)

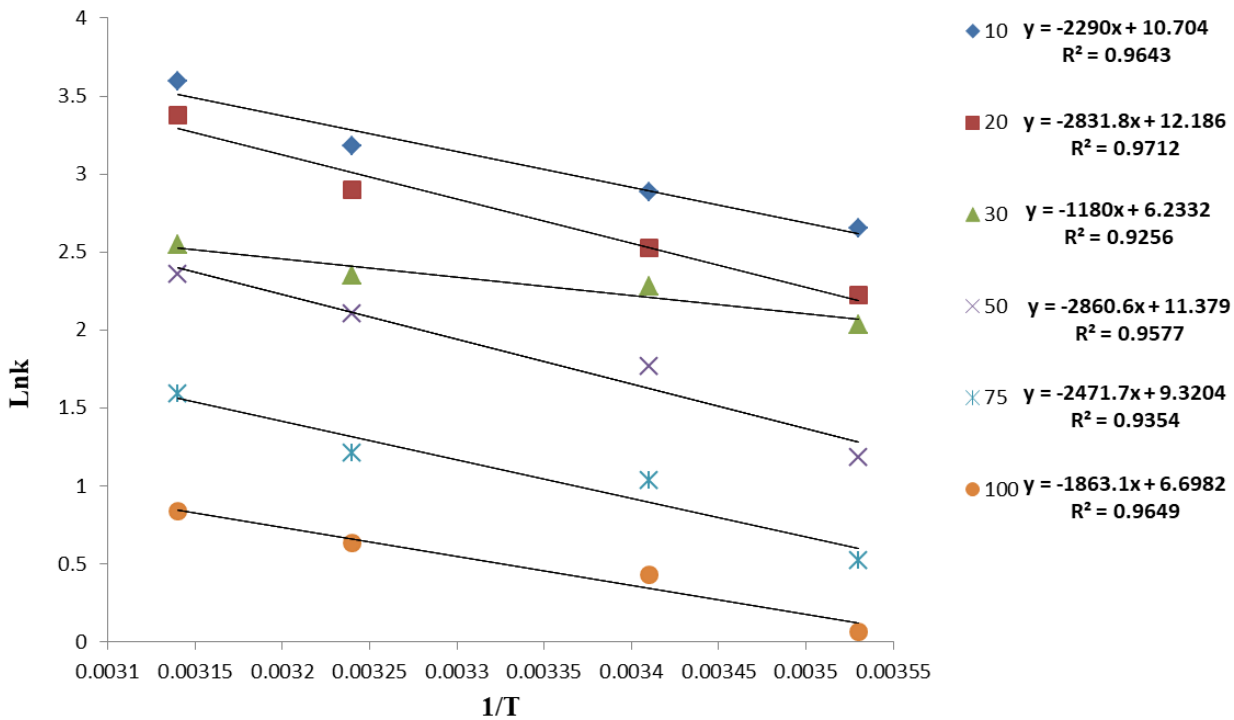

Figure 13: Plot of van, t Hoff relationship between lnK and 1/T.

Table (2) indicate , ΔG at all temperature were negative , Indicates the immediacy of dye adsorption in sludge [29].the positive value of ΔH indicates the endothermic nature of the process of adsorption , while the positive value of ΔS indicate increase in randomness of the solid /solution interface during the adsorption process[30].

3.5 Kinetics of adsorption

To clarify the adsorption process function, the kinetics of the adsorption data is analyzed using the first-order pseudo (Lagergren equation), and pseudo-second-order (Hoand McKay).Pseudo first -order rate equation is

$$\ln (Q_e - Q_t) = \ln Q_e - k_1 t \text{ ……………… (8)}$$

Where (Q_e) and (Q_t) (mg/g) are quantity of adsorbed dye on the activated sewage sludge at equilibrium and time (t) in (hour) accordingly, and (K_1) (hrs^{-1}) is the rate constant of the first order adsorption.

The implementation of the pseudo-first order rate equation (8), the plot of ln (Q_e-Q_t) opposite (t) given linear relationships whereby the calculated Capacity value and K_1 respectively are estimated from the slop and intercept. The obtained outcomes are seen in Table 3 and in Figure 14 .

The calculation pseudo-second order average is

$t/ Q_t = 1/K_2Q_e^2 + 1/Q_e (t)$ …………… (9)

Where (K_2) (g $mg^{-1}hr^{-1}$) is the rate constant of the second order adsorption.

The implementation of the pseudo-second order rate equation (9), the plot of (t\Q_t) versus (t) given linear relationships from which the calculated Q_e and K_2 are estimated respectively from the slope and intercept. The obtained outcomes are seen in table (3) and figure (15).

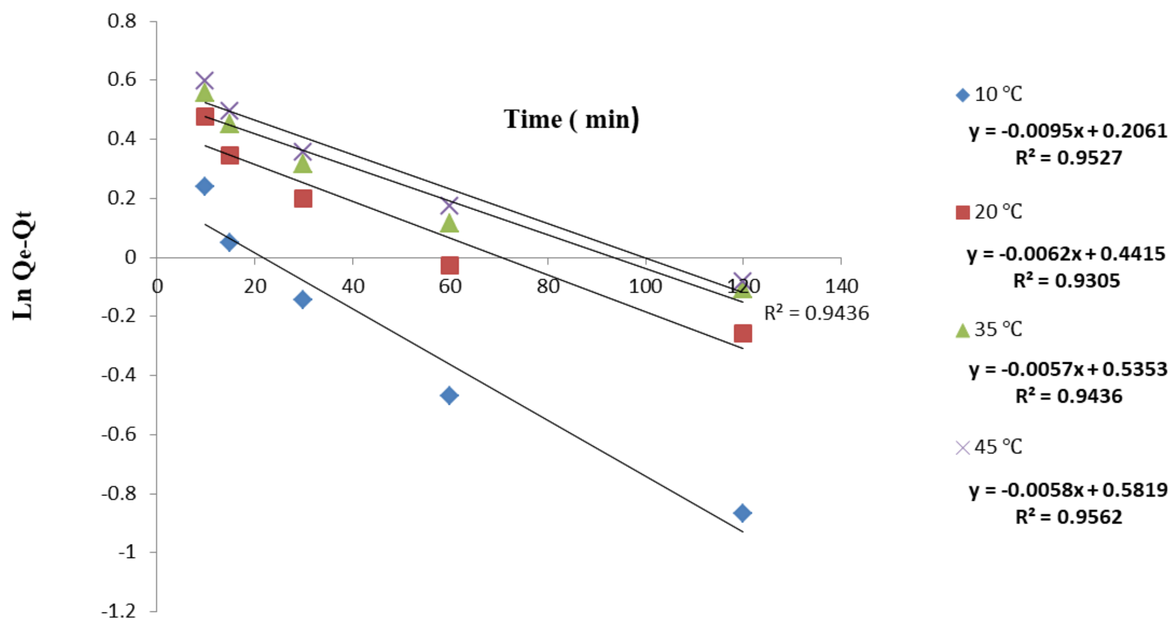

Figure 14): linear from of pseudo -first order kinetic model of Bismarck brown R dye on sludge at different temperature.

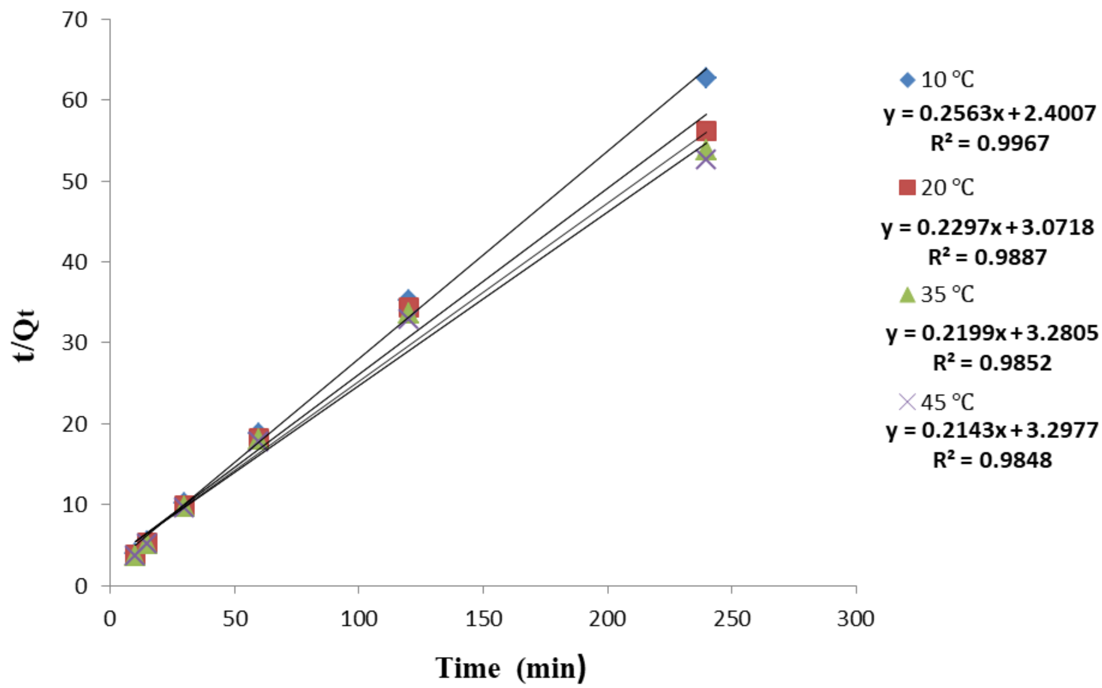

Figure (15): linear from of pseudo -second order kinetic model of Bismarck brown R dye on sludge at different temperature.

Table (3): First order and second order kinetic rate constant, calculated and experimental Q_e values and R^2

	Temp.(°C)	Pseudo-first order				Pseudo-second order			
		Q_e exp	Q_e Cal	K_1 min^{-1}	R^2	Q_e exp	Q_e Cal	K_2 min^{-1}	R^2
BBR dye	10	3.826	1.228	0.0094	0.9527	3.826	3.906	0.027	0.9967
	20	4.271	1.554	0.0062	0.9305	4.271	4.366	0.017	0.9887
	35	4.458	1.707	0.0057	0.9436	4.458	4.566	0.014	0.9852
	45	4.566	1.787	0.0058	0.9562	4.566	4.672	0.013	0.9842

The results shown in Table (3) indicate that Qe (exp) is smaller than that calculated experimentally Qe (Cal). For pseudo-first order equations, and the measured Q_e (Cal) equation for pseudo-second order equation. Values were very well accepted with the experimental Q_e

(exp) value and even the pseudo-second order kinetics value R^2 rather than the pseudo-first order kinetics values.

It is obvious that the adsorption of the analyzed dye on sludge adsorbent was better described by pseudo-second order kinetics that means that the adsorption process corresponds to the Kinetics of pseudo-second order [31].

4. Conclusions

The present study emphasize that the activated carbon prepared from sewage sludge was employed an adsorbent for the removal of Bismarck brown R dye from aqueous solution. The maximum percentage removal of (96.60 %) occurred at pH 2. The equilibrium data followed Langmuir isotherm modal. Adsorption kinetic was found to followed pseudo-second order kinetics rate expression. Thermodynamic study shows the spontaneous and endothermic nature of the adsorption system. It can be inferred that activated carbon produced from sewage sludge is a good low cost and high performance adsorbent for extracting dye from wastewater and can be used in a magnetically aided water treatment system.

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المستخلص

أن مياه الصرف الصناعي وغيرها من المياه الملوثة تحتاج الى معالجة قبل طرحها الى الانهار واعادة استخدامها مرة اخرى . أن الامتزاز هو احدى الطرق المهمة المستخدمة لأزاله الملوثات العضوية وغير العضوية من المياه , وفي هذا الدراسة تناول البحث عن مواد متوفرة محلياً ورخيصة الثمن واستخدامها كسطوح مازة , حيث استخدمت حمأة مياه الصرف الصحي كسطح ماز لأزاله صبغة البسمارك البني R وباستخدام طريقة الدفعات ز وتم دراسة العديد من العوامل التجريبية المختلفة التي تؤثر على عملية الامتزاز مثل زمن الاتزان ودرجة الحرارة والدالة الحامضية للمحلول ووزن المادة المازة والتركيز الابتدائي للصبغة والقوى الايونية ز وتضمنت دراسات الامتزاز دراسة ايزوثيرمات الامتزاز وحركية الامتزاز . وتم تطبيق ايزوثيرمات لانكماير وفريندلش وفي درجات حرارية مختلفة واظهرت الدراسة ان ايزوثيرم لانكماير اكثر انطباقاً وملائمة مع النتائج التجريبية . وتشير الدراسات الثرموديناميكية ان عملية الامتزاز هي عملية تلقائية (ΔG سالبة) وماصة للحرارة (ΔH موجبة) وعشوائية (ΔS موجبة) . ومن دراسة الحركية تشير الى ان عملية الامتزاز تتبع معادلة حركية المرتبة الثانية الكاذبة