Adsorption Study For Removal Of Direct Red 89 and Basic Blue 41 Dyes From Aqueous Solutions Using Sawdust As a High Efficient Adsorbent

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ABSTRACT

Local wood sawdust, as a surrogate for the current expensive adsorbents of removing Direct red 89 (DR89) and Basic Blue 41(BB41) dyes from aqueous solution was investigated. concerning the factors affected on the adsorption efficiency such as contact time, initial dye concentration, adsorbent dosage and pH of solution have been studies. Results showed that the adsorption capacity of DR89 and BB41 dyes was increased with the increase of the initial dye concentration and the optimum dye concentration was 10 mg/L for each dye. The contact time required to reach equilibrium was 40 and 10 minute for DR89 dye and BB41 dye, respectively. Was the optimum pH 2 for adsorption of DR89 dye and pH 10 for adsorption of BB41 dye. Adsorption isotherm data showed that adsorption can be fitting to both Freundlich and Langmuir isotherm. The results of thermodynamic parameters study showed that the adsorption process of both dyes onto sawdust surface are spontaneous. The enthalpy values ΔH found that adsorption was endothermic for DR89 dye adsorption and exothermic for BB41 dye.

دراسة الامتزاز لإزالة الصبغتين Direct Red 89 و Basic Blue 41 من محاليلهما المائية باستخدام نشارة الخشب كمادة مازة عالية الكفاءة

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الملخص

تمت دراسة قابلية نشارة الخشب المحلي، كمادة مازة بديلة عن المواد الممتزة المكلفة لإزالة الصبغتين Direct Red 89 و Basic Blue 41 من محاليلهما المائية. اما بخصوص العوامل المؤثرة على كفاءة الامتزاز مثل زمن التماس، وتركيز الصبغة الأولي، جرعة المادة المازة والدالة الحامضية ،فقد أظهرت النتائج ان سعة الامتزاز لكلا الصبغتين DR89و BB41 تزداد مع زيادة التركيز الأولي للصبغة وكان التركيز الامثل لكلا الصبغتين هو 10 ملغم/ لتر، وكان الزمن اللازم للوصول الى الاتزان هو (40 دقيقة) بالنسبة للصبغة DR89 و (10 دقائق) بالنسبة للصبغة BB41. وكان افضل pH هو 2 لامتزاز الصبغة DR89 و 10 لامتزاز الصبغة BB41. ان نتائج الامتزاز تم تطبيقها على كل من معادلة فروندليتش ولانكماير واظهرت النتائج ان الامتزاز ملائم لكل من ايزوثيرم فروندليتش ولانكماير. كذلك بينت نتائج الدراسة الثيرموديناميكية أن الامتزاز لكلا الصبغتين على سطح نشارة الخشب هي عملية تلقائية، ومن خلال قيم الانثالبي ΔH وجد ان الامتزاز هو ماص للحرارة بالنسبة للصبغة DR89 لكن باعث للحرارة بالنسبة للصبغة BB41.

1. Introduction

Human is facing in the current era a big problem that needs concerted efforts to reduce it and treated namely, environmental pollution and increases the problem seriously. Human who has the clear role in this increased risk, through the various activities that made human life-threatening, and the pollution of the environment and his close relationship with the population expansion in the world (Tsai, W.T., et al. 2006). Some liquid wastes from industrial production operations may be hard to purify by conventional sewage treating technology, because of the complex structure for some of their components. The textile industry is a big polluter. It wastewater is the most environmentally harmful, attributed to the dyes found there (Demirbas, O., et al. 2002).

The discharge of coloured wastes such as pigments into rivers waters causes harm to the environment where they are toxic to aquatic organisms (Al-Kaim A. F. 2007). Large quantities of Basic and Direct dyes are utilized by the textile industry. Dyes is the most contaminant found in the wastewater, and it must be removed before disposal into water bodies. The removal of dyes from wastewater is often most important than the removal of other colorless substances (Simphiwe P. B. et al. 2012).

The different methods of treatment for the removal of colors and dyes from wastewater suggested are filtration, deposition, coagulation, membrane separation process, oxidation and adsorption (Hakan D., et al., 2008). In these methods, adsorption process has been clear to be an appropriate and efficient method to remove dyes (Vadivelan and Kumar, 2005; Jumasiah et al., 2005). Adsorption process is a technique often used for water purification, and involves in the accumulation of particles or molecular species of gas, liquid, or dissolved solid on the surface of adsorbent. It occurs between two phases, like gas-liquid, liquid-liquid, liquid- solid or gas-solid, interfaces (Walter and Weber, 1972; Abbas H. S. and Waleed M. A., 2013). Adsorption process is a widely used for the wastewaters treatment due to the increas of environmental requirements (Robert M. C., 1989). Activated carbon is mostly used as an adsorbent in adsorption process, but it is relatively expensive (Chiou et al., 2003).

The wood structure is very porous and has a very high surface area that would allow easy access aqueous solutions to the components of cell wall. In addition, it has been appeared that break of wood into very finer particles increases the sorption efficiency (Rowell, 2005). Sawdust has received the great attention of the scientists due to both useful aspects for environment, by using it as a very low cost adsorbent as well as using it as disposal of waste sawdust from Carpentry workshops. "Sawdust from pine contains cellulose, lignin, and tannins or other phenolic compounds which are active ion exchange compounds" (Shukla A. et. al. 2002).

In this work, the ability of local wood sawdust to remove Direct Red 89 (DR89) and Basic Blue 41 (BB41) dyes from aqueous solution by adsorption process in different conditions has been studied. In addition, adsorption isotherms and

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thermodynamic parameters at equilibrium conditions were studied as well.

2. Materials and Methods

2.1. Materials

A. Adsorbent: Local wood sawdust is collected from carpentry workshops in Al-Hilla city. It was washed with large amounts of distilled water several times to remove the soluble impurities and dust, then it left to be dried at room temperature overnight. The dried sawdust was grounded and sieved to a required particle size was (50) µm.

B. Adsorbate: The Direct Red 89 (DR89) and Basic Blue 41(BB41) dyes used in this study was obtained from Hilla textile factory, The chemical structures of both dyes are given in Figure (1).

To prepare a dye solution, 1 gm of each dye powder was dissolved in 1L of distilled water to obtain solution with 1000 mg.L^{-1} concentration. Then diluted to obtain required concentrations range (10-100) mg.L^{-1} to use it in the experiments. The maximum absorbance wave length (λ_{max}) for (DR89) and (BB41) dyes were 495 nm and 605 nm respectively, It was measured using optima UV- visible spectrophotometer.

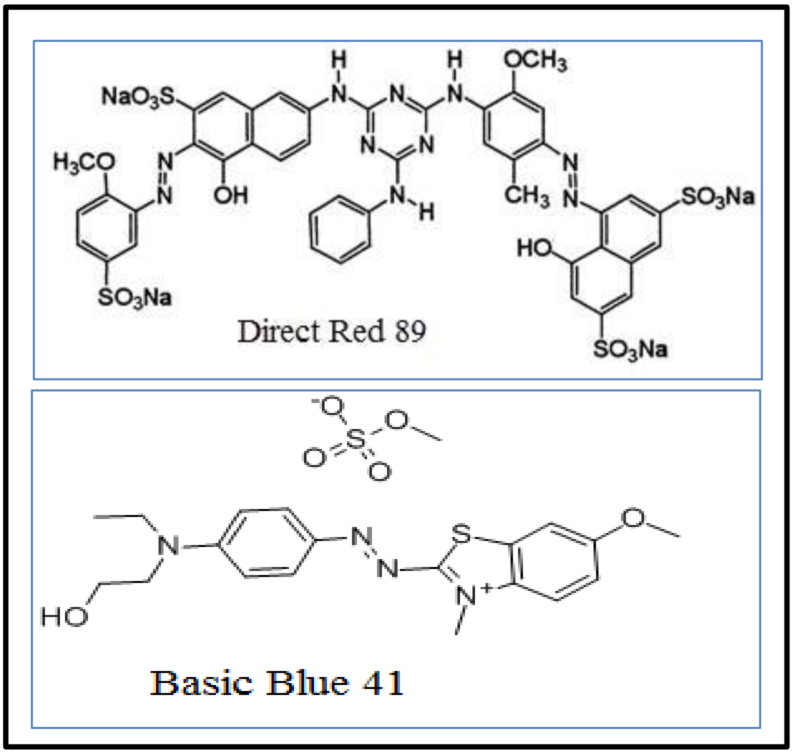

figure (1): Chemical structure of dyes

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2.2. Methods

Batch experiments were implemented by shaking (25) mL of dye solution (10) mg.L^{-1} concentration with (0.2) gm of sawdust powder (50) µm particle size in 100 ml conical flasks at room temperature for a predetermined contact time of (40) min for (DR89) dye and (10) min for (BB41) dye. After that, centrifuged at 3000 rpm for 10 minutes. The concentration of each dye in the aqueous phase was determined spectrophotometrically by UV spectrophotometer at (λ_{max}) for each dye. The adsorption capacity (q) of each dye by sawdust adsorbent (mg/g) was calculated using the following equation (Vanderborght M. and Van Grieken E. 1977):

$$q = \frac{V(C_i - C_e)}{w} \quad \ldots\ldots\ldots\ldots (1)$$

Where: C_i is the initial concentration of dye (mg/L), C_e is the concentration of dye at equilibrium (mg/L), V is the volume of dye solution (L) and w is weight of the sawdust (g).

3. Results and Discussion

3.1. Adsorption isotherm

Adsorption isotherms are necessary for the characterization of how dyes concentration will interact with the surface of adsorbent and helpful to improve the adsorbent surface for the removable dyes. Adsorption isotherms, are one of the basic requirements to understand adsorption systems(Ozer and Dursun, 2007). The adsorption capacity of dye on wood sawdust surface is plotted as a function of equilibrium concentrations at room temperature, shown in Figure (2). It is clear that dyes can be readily removed from the polluted water by sawdust powder.

To characterize the adsorption of dyes on adsorbent, the equilibrium data for this study were analyzed using, Freundlich and Langmuir isotherm models.

The Freundlich isotherm is an empirical model, applies to heterogeneous surfaces and involve a multilayer adsorption, with clear interaction forces between the adsorbed molecules. This model suppose that when the adsorbate concentration increases, the adsorption process also increases, and accordingly, the sorption energy dramatically decreases due to completion of the sorption sites of the adsorbent. The general linear form of the Freundlich model is (Faust, S. D. and Aly, 1987; Vijayakumar G., 2012):

$$\ln q_e = \ln K_{f} + {}^{1}/_{n} \ln C_e \quad \ldots\ldots\ldots\ldots (2)$$

Where: q_e is the amount of adsorbate at equilibrium (mg g^{-1}), C_e is the equilibrium concentration of adsorbate (mg L^{-1}), K_f is the Freundlich constant (Indicates the relative adsorption capacity) and n is constant (refers to the adsorption intensity). The values of K_f and $1/n$ can be obtained from the intercept and slope of the graph of ln q_e versus ln C_e. Linear relationship of Freundlich isotherm shows in Figure (3). If value of $1/n < 1$ it denote a natural adsorption. If $n = 1$ indicates that the distribution between the two phases are not dependent on the concentration. while if $1/n > 1$ denote cooperative adsorption (Mohan S., and Karthikeyan J. 1997). Freundlich isotherm constants were calculated in Table (1), from this data it is deduced that the Freundlich isotherm was a more fit to the adsorption of DR89 dye than the BB41 dye.

Langmuir isotherm suppose that there is a limited number of effective sites which distributed homogeneously on the adsorbent surface. These effective sites have the same familiarity for adsorption of a unimolecular layer, and the interaction forces between molecules of adsorbed was very slight. The general linear form of the Langmuir model is (Langmuir, I., 1918).

$$\frac{1}{q_e} = \frac{1}{Q_o} + \frac{1}{Q_o K_L C_e} \quad \ldots\ldots\ldots\ldots\ldots\ldots \ (3)$$

Where: K_L (L mg^{-1}) and Q_o (mg g^{-1}) are Langmuir constants related to energy of adsorption and maximum adsorption capacity respectively. A graph of $1/q_e$ against $1/C_e$ results in a straight line with a slope of ($1/Q_oK_L$) and an Intercept ($1/Q_o$), Figure (4). The basic features of the Langmuir isotherm can be described by a constant named the separation factor R_L, which is defined as (Weber, T. W. and Chakkravorti R. K. 1974):

$$R_L = \frac{1}{(1 + K_L C_o)} \quad \ldots\ldots\ldots\ldots \ (4)$$

Where: C_o is the initial concentration of dye (mgL^{-1}) and K_L is the Langmuir constant (mgL^{-1}), and R_L values refer to the nature of adsorption. Values of ($R_L > 1$) denote the adsorption is unfavorable, if ($R_L = 1$) indicates linear, ($0 < R_L < 1$) indicates favorable adsorption while ($R_L = 0$) mean irreversible. From the data computed in table 1 the R_L values for DR89 dye and BB41 dye are 0.495 and 0.135 respectively, ie ($0 < R_L < 1$) denote the adsorption was favourable for Langmuir isotherm.

Table (1): Freundlich and Langmuir Isotherm Parameters for the adsorption of DR89 and BB41 dyes onto sawdust surface at 298 K.

Isotherm models	***Parameters***	***Dyes***	
		DR89	BB41
Freundlich	K_f (mg/g)	1.149	5.394
	1/n	0.747	1.631
	n	1.338	0.613
	R^2	0.989	0.851
Langmuir	Q_o (mg/g)	11.641	3.435
	K_L (L/mg)	0.102	0.637
	R^2	0.999	0.863
	R_L	0.495	0.135

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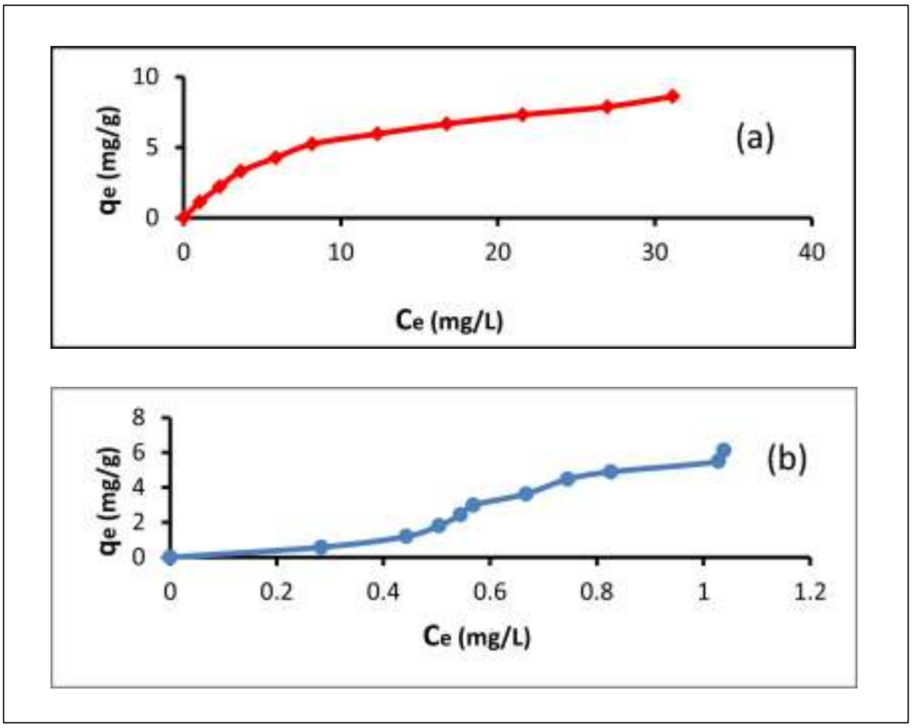

Figure (2): adsorption isotherm of (a) DR89 dye and (b) BB41 dye onto sawdust surface at 298 K.

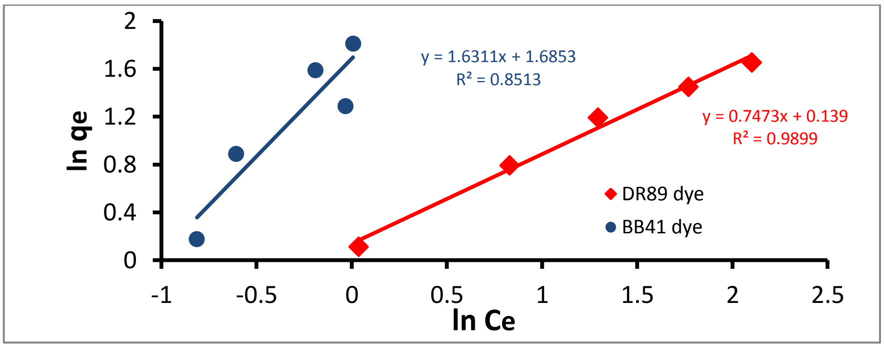

Figure (3): Freundlich isotherm of DR89 dye and BB41 dye onto sawdust surface at 298 K.

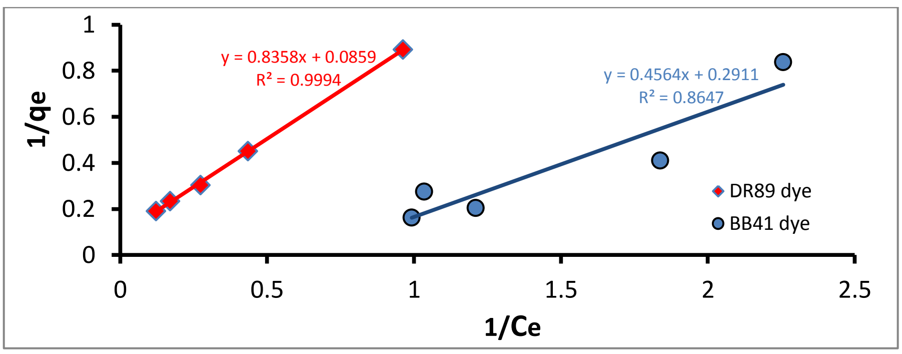

Figure (4): Langmuir isotherm of DR89 dye and BB41 dye onto sawdust surface at 298 K.

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3.2. Factors affecting on adsorption

3.2.1. Determination of equilibrium contact time

The effect of equilibrium contact time on removal of dyes was studied. The results were displayed in Figure (5), the removal of dyes was increased remarkably in the beginning with the contact time increases, but it progressively decrease with time even up to equilibrium. The rapid increases in adsorption in the beginning may be due to a large number of effective sites of adsorption which available on surface but after a passage of time, the residual effective sites are hard to be occupied. This is because of the disharmony between the solute particles of the bulk and solid phases (Idris M.N., 2011).

1.1.1. Effect of solution pH

The adsorption of anionic (DR89) and cationic (BB41) dyes at pH solution range between (2 -10) was studied. The solution was adjusted by using few drops from 0.1N of (HCl or NaOH) solution to obtain the required pH. The results was shown in Figure (6). There was slight decrease in the adsorption of (DR89) dye with the increase in pH solution, while slightly increase in the adsorption of (BB41) dye with the increase in pH solution. This behavior may be due to dyes become in an ionic form when dissolve in aqueous solutions. Therefore, adsorption capacity of dyes was affected by the charge on the surface of adsorbent, which in turn is influenced by pH solution (Abd El-Aziz A. S. et al. 2013). At lower values of pH, the adsorbent surface becomes positively charged and so an electrostatic attraction increases between the negatively charged anionic direct dye and the positively charged adsorbent surface (El-Nemr A., et al. 2009). While repulsion was increased between the cationic basic dye and the positively charged adsorbent surface.

1.1.1. Effect of adsorbent mass

The effect of adsorbent dosages on the removal percentage of (DR89 and BB41) dyes was studied by using sawdust dosages range (0.05-0.3) g with initial dye concentration of 10 mg/L at normal pH. Figure (7) shows the removal percentage of dyes adsorbed with different amounts of adsorbent. It can be noted that increase the removal percentage of each dye in this study with increase adsorbent dosage even up to a certain extent and then it stay constant. This increase in adsorption can be due to more effective adsorption sites available due to increase surface area of adsorbent which leads to enhanced adsorption process (Karthikeyan G., et al. 2005).

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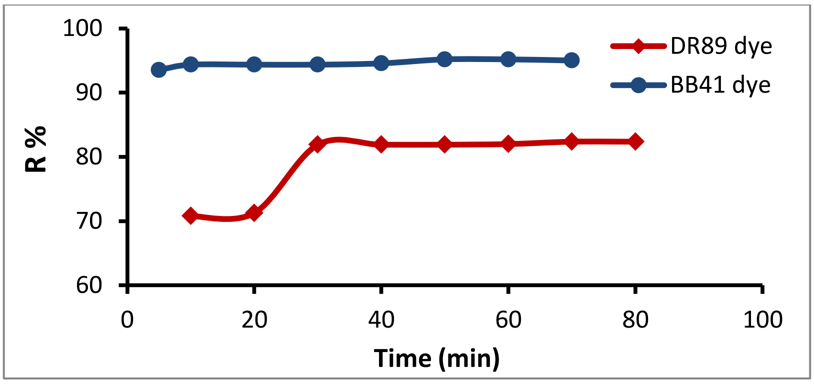

Figure (5) : Effect of equilibrium contact time on the removal percentage of DR89 dye and BB41 dye onto sawdust surface at 298 K.

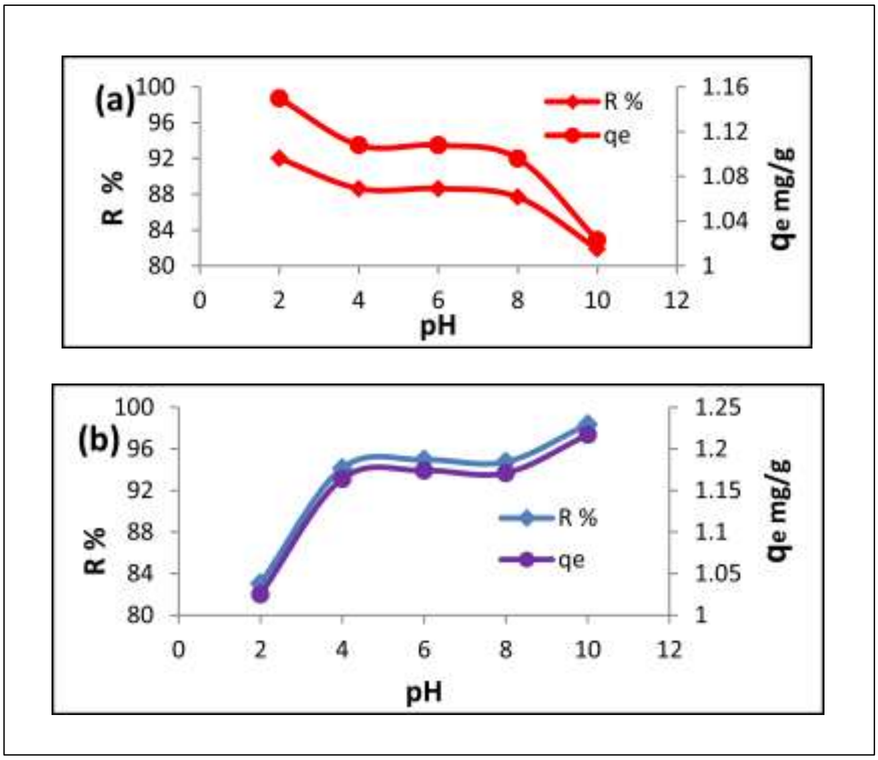

Figure (6): Effect of pH solution on removal percentage and adsorption capacity of (a- DR89 dye and b- BB41 dye) onto sawdust surface at 298 K.

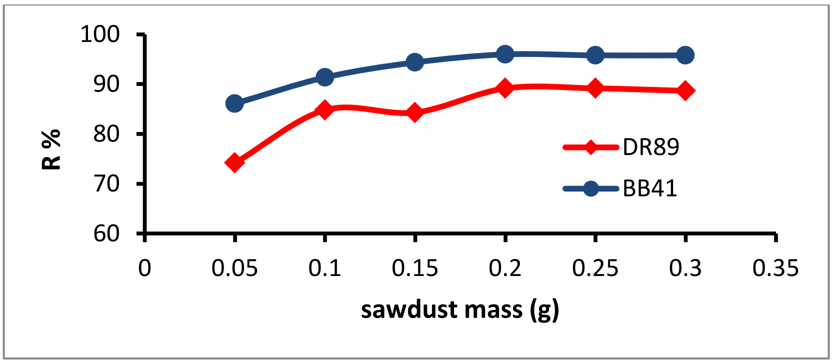

Figure (7): Effect of sawdust mass on the removal percentage of DR89 dye and BB41 dye at 298 K.

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1.1.2. Effect of initial dye concentration

The effect of initial dye concentration on the removal efficiency of dye was studied, by combining 0.2 g of sawdust with different dye concentrations ranging (10-100) mg/L for DR89 dye and (10-50) mg/L for BB41 dye at room temperature. The results shows in Figure (8) that the removal percentage of dyes decreased with increasing initial dyes concentration, While adsorption capacity increased with increase of the dye concentration. The influence of the initial dye concentration depends on the relationship between the effective sites available on a surface of adsorbent and the dye concentration. "At low concentration there will be unoccupied effective sites on the adsorbent surface and when the initial dye concentration increases, the effective sites required for adsorption of the dye molecules will be lacking" (Kannan and Sundaram 2001). The optimum dyes concentration was identified as 10 mg/L in this study.

3.3. Thermodynamic parameters

The effect of temperature on adsorption processes is an important factor. The thermodynamic parameters were determined by study the adsorption of both dyes at different temperatures range of (15-60 Cº) at an initial dye concentration of (10 mg/L) onto 0.2 g of sawdust. The enthalpy (ΔH°), entropy (ΔS°) and the Gibbs free energy (ΔG°) were calculated from the following Equations (Moradi O. et al., 2010; Mehrizad A. et al., 2012):

$$\ln K_d = -\frac{\Delta H^\circ}{R}\left(\frac{1}{T}\right) + \frac{\Delta S^\circ}{R} \quad \ldots\ldots\ldots\ldots\ldots (5)$$

$$K_d = \frac{q_e}{C_e} \quad \ldots\ldots\ldots\ldots\ldots (6)$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad \ldots\ldots\ldots\ldots\ldots (7)$$

Where: K_d is the equilibrium constant, R is the gas constant (8.314 J/mol.K) and T is the temperature of solution at (K).

The slope and intercept from the plot of (ln K_d) versus (1/T) in equation (5) was calculated ΔH° and ΔS° respectively as it is clear in Figure (9) and listed in Table 2.

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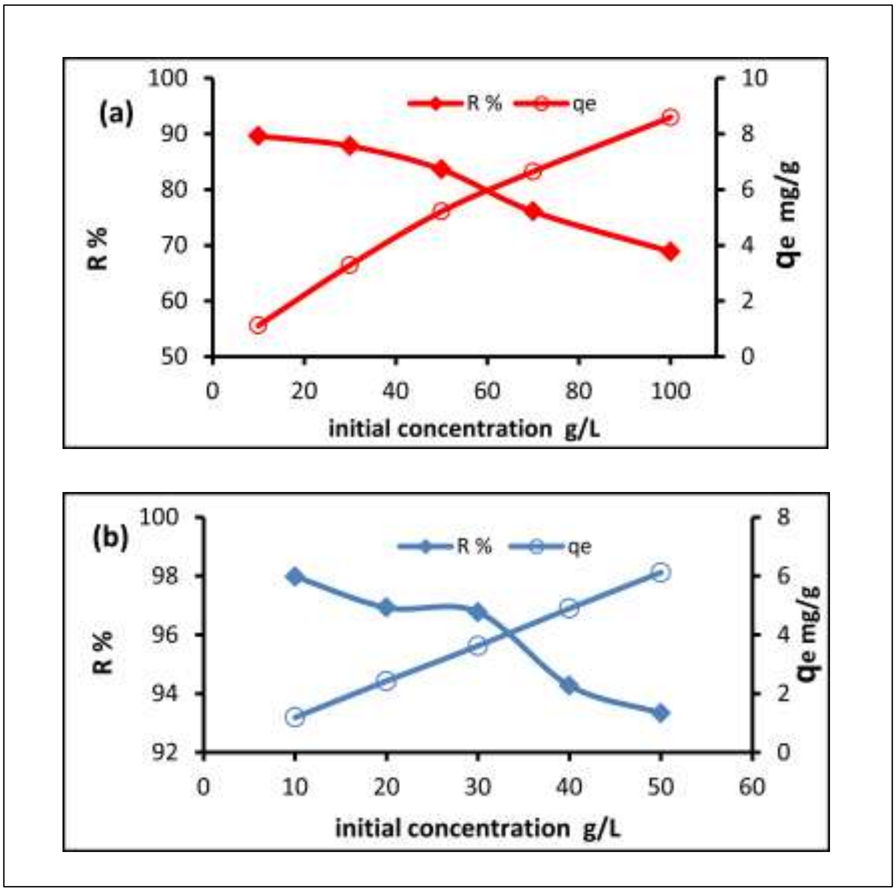

Figure (8): Effect of initial dye concentration on removal percentage and adsorption capacity of (a- DR89 dye and b- BB41 dye) onto sawdust surface at 298 K.

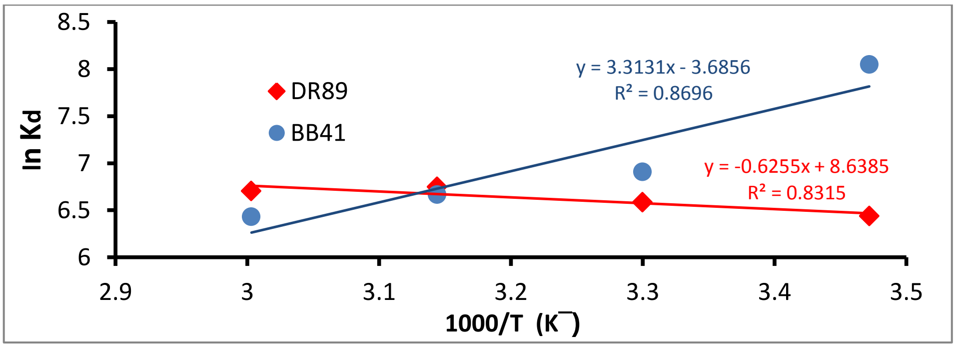

Figure (9): A plot of (ln K_d) versus (1/T) for DR89 dye and BB41 dye onto sawdust surface

Table (2): Thermodynamics parameters for adsorption of DR89 dye and BB41 dye onto sawdust surface

T(K)	DR89 dye			BB41 dye		
	ΔHº (KJ/mol)	**ΔGº** (KJ/mol)	**ΔSº** (J/mol.K)	**ΔHº** (KJ/mol)	**ΔGº** (KJ/mol)	**ΔSº** (J/mol.K)
288	5.200	-15.41	71.820	-27.545	-18.72	- 30.642
303		-16.56			-18.26	
318		-17.63			-17.80	
333		-18.71			-17.34	

The values ΔGº are negative for adsorption both dyes, indicates that the adsorption process of Both dyes onto sawdust surface are spontaneously and thermodynamically favorable. "The decline in the negative values of ΔGº with the improvement in temperature decreases the driving force at the adsorption process. Accordingly, the improvement in temperature has negative impact on the dyes sorption process" (Shokry H., et al. 2014). The values of ΔH° are positive value for DR89 dye adsorption while negative value for BB41 dye adsorption, this indicated that the sorption for DR89 dye was endothermic, but sorption was exothermic for BB41 dye adsorption. The endothermic adsorption process, may be attributed to increasing the mobility of the molecules of dye. An addition, increasing the number of effective sites for the adsorption with temperature rising (Senthilkumaar et al. 2006). The positive value for ΔS° showed that increased randomness at the solid-liquid, interface through adsorption process of DR89 dye onto the sawdust surface.

2. Conclusion

Based on the results above, sawdust can be used as a very efficient and cheap adsorbent for the dyes removal from Industrial Wastewaters. The results showed a short time required to reach adsorption equilibrium for removal of both dyes, adsorption capacity of DR89 and BB41 dyes was found to be increased with the increase of the initial dye concentration. The adsorption of DR89 dye on sawdust was favorable at pH 2 (which is acidic medium) while the adsorption of BB41 dye on sawdust was favorable at pH 10 (which is basic medium). Adsorption isotherm data showed that adsorption can be fitt to both Freundlich and Langmuir isotherm. There are two environmental benefits for this research, first that sawdust can be used as an inexpensive adsorbent with high efficiency for dyes removal from industrial wastewaters. The second benefits was considered as an appropriate method to disposal of the sawdust waste.

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