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Kinetic and Thermodynamic Study the Adsorption of Brilliant blue Dye on Peel of Plant.

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Abstract : In the present work, it was used low-cost adsorbent (almond peel and sunflower seed shell) which activated by H₂SO₄ and HCl to removal Brilliant blue dye from aqueous solution. Adsorption studies were carried out to find the effect of different parameters such as: (contact time, adsorbent doses, pH, and temperature effect). The results in this experiment showed that the adsorption of brilliant blue dye at initial concentration (30 ppm) was rapid in the first of contact time until reached to the equilibrium time at (40, 30)min for almond peel activated by H_2SO_4 and HCl respectively and (35, 40)min for sunflower seed shell activated by H_2SO_4 and HCl respectively. The percentage removal of brilliant blue from aqueous solution was about 95% with almond peel activated by H_2SO_4 and 97% with almond peel activated by HCl. But the percentage removal for dye with sunflower seed shell activated by H₂SO₄ and HCl about 97%. This percent removal of Brilliant blue was at the equilibrium time for all the adsorbent also at the optimum doses which were 0.8g for almond peel activated by H_2SO_4 and HCl but 0.9g for sunflower seed shell activated by H_2SO_4 and 0.6g for sunflower seed shell activated by HCl. The maximum value to percent removal of Brilliant blue from aqueous solution was in the acidic pH. The adsorption kinetics was analyzed by using the pseudo – first and second order models and the results showed that the adsorption kinetics were more accurately described by a pseudo - second order model. Thermodynamic parameters such as the change of free energy, enthalpy and entropy were also evaluated. The results indicated that the adsorption of the Brilliant blue dye onto the (almond peel activated with H_2SO_4) is endothermic, decrease disorder, spontaneity of the adsorption process. But the adsorption of the Brilliant blue dye onto the (almond peel activated with HCl) was an endothermic, decrease disorder and no spontaneity of the adsorption process. The adsorption of the Brilliant blue dye onto (sunflower seed shell activated with H₂SO₄ and HCl) were an exothermic process, increase disorder and spontaneity of the two adsorption process.

Keywords : Adsorption, Brilliant blue, almond peel, sunflower seed shell.

Introduction

Environmental pollution due to industrial effluents is the major concern because of their toxicity and threat for human life and the environment. The discharge of industrial effluents to the water bodies has raised much concern because of potential health hazards associated with the entry of toxic components into the food chains of humans and animals. Synthetic dyes are extensively used for dyeing and printing in a variety of industries¹. The removal of colour from industrial effluents has targeted attention over the last few years, not only because of its toxicity, but mainly due to its visibility². Dyes are widely used in many industries such as food, textiles, rubber, paper, plastics and so on. About over 7.10⁵ to 10.000 different commercial dyes and pigments are produced annually all around the world. It has been estimated that about 10-15% of these dyes is lost during the dyeing process and released with the effluent³. The discharging of these dyes into water

resources even in small amounts can affect aquatic life and the food chain. Dyes can also cause allergic dermatitis and skin irritation. Some of them have been reported to be carcinogenic and mutagenic for aquatic organisms⁴. The Brilliant Blue molecule is a weak acid organic molecule with polar and non-polar components, which may lead to complex sorption behavior. Its negative charge stems from sulfonic acid groups⁵. It is used as a common food additive to color confectionary and dairy products. It is carcinogenic, causes reproductive and neurological disorders, severe allergies, anaphylactic reactions including rashes, swelling and trouble in breathing in human being. Even behavioral convulsion, gastrointestinal tumors, blood-lymphoma have also been found in rodents⁶. Brilliant Blue dye is a acidic dye belongs to triphenylmethane class of dye. Triphenylmethane dyes are those dyes in which a central carbon atom is bonded to two benzene rings and one p-quinoid group (chromophore)⁷. The auxochromes are - NH2, NR2 and -OH⁸. Triphenylmethane dyes are used extensively in many industries such as food, textiles, rubber, paper and plastics.⁹ Very few techniques for the removal of Brilliant Blue FCF from industrial waste water have been studied because of its complex structure. Reports exist on the removal of this dye through electrochemical, photochemical and adsorption¹⁰.

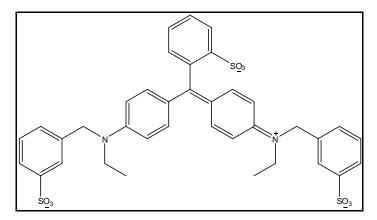


Fig. 1.Structure of Brilliant blue dye.

The present work was undertaken to explore the feasibility of finding a low cost effective adsorbent, almond peel and sunflower seed shell. for the treatment of Brilliant blue dye from aqueous solution as a function of temperature, dosage, contact time and pH by batch mode adsorption studies and to report the applicability of various kinetic models for the chosen dye by the adsorbent in a controlled system.

1. Materials:

2.1 Adsorbent

The sunflower seed shell (S.S.S) and almond peel (A.P) were washed thoroughly with distilled water for several times to remove the surface adhered particles and this material was then simmer with distilled water at polling point then filtered, this process was repeated to get rid of all colored or soluble materials then filtered and dried. The washed materials were then oven dried at 80°C for 2 h. The dried materials were ground in the grinder then sieving through 500µm-sieve.Then part of this material treated with (1N) HCl and other part treated with (1N) H₂SO₄ then dried in oven at 100°C for 2 h then used as adsorbent.¹¹

2.2 Adsorbate

Brilliant blue (B.B)dye (molecular formula: $C_{37}H_{34}N_2O_9S_3$; Molecular Weight 746.87g/mol, 99%) supplied by Merck company. Its acidic dye soluble in water. Dye has maximum absorption at 610 nm.

2. Experimental:

Put 25 ml of the 30 ppm dye solution in ten conical flask. Then, different adsorbent dose was added in each flask (0.1-1)g (particle size 500 μ m). All the flasks were kept inside the shaker COMPENSTATE with citenco motors F.H.P. England at 500 rpm. After 60 minutes, the flasks were withdrawn from the shaker and the dye solutions were separated from adsorbents by using centrifuge ALL-PRO corporation at 4000 rpm for 10

minutes. The absorbance of all the solutions measured by using UV-Visible spectrophotometer double beam Shimadzu UV 1650 PC Japan.⁽¹²⁾With same method the effect of pH studied. The pH of each flask was adjusted in the range of (2-10) with dilute HCl (0.05 M) and NaOH (0.05 M) solution via pH meter -Hanna instrumentals.⁽¹³⁾The effect of contact time studies in different time by the same method above.¹⁴The effect of temperaturehas beenaccomplished with taking three degrees (35, 45, and 55)°C and the effect of temperature on adsorption has been studied in each degree.¹⁵The adsorption mechanism of B.B dye onto the adsorbent was studied using pseudo-first-order and pseudo-second-order kinetic equations. The pseudo-first-order kinetic equation is given as: ¹⁶

$$Log (qe - qt) = Log qe - \frac{K_1}{2.303}t$$
 (1)

where qe and qt (mg/g) are the amount of B.B dye adsorbed on the adsorbent at equilibrium and at time t (min), respectively. K_1 (1/min) is the pseudo-first-order rate constant. The pseudo-second-order kinetic equation is described as:¹⁷

$$\frac{1}{qe-qt} = \frac{1}{qe} + K_2 t \tag{2}$$

where K_2 (g/mg min) is the rate constant and qe and qt (mg/g) are the amount of B.B dye adsorbed on the adsorbent at equilibrium and at time t (min), respectively. ⁽¹⁸⁾ Spontaneity of a process can be determined by thermodynamic parameters such as enthalpy change (Δ H), free energy change (Δ G) and entropy change (Δ S). The temperatures used in the thermodynamic study were 308, 318 and 328 K. The thermodynamic parameters were calculated based on the following equations:

$$\Delta G = -RT ln K_f \tag{3}$$

$$\frac{\Delta G}{T} = \frac{\Delta H}{T} - \Delta S \tag{4}$$

where K_f is the equilibrium constant, R is the universal gas constant (8.314 J/mol K), and T is the temperature (K). The percent removal (%*Rem*.) has been calculated as:¹⁹

$$\% Rem. = \frac{A^{\circ} - A}{A^{\circ}} * \mathbf{100}$$
 (5)

Where, A° = initial dye Absorbance and A = final dye Absorbance.

4. Results and discussion.

4-1 Effect of Adsorbent Dose on the adsorption of B.B dye:

The effect of adsorbent dose (A.P and S.S.S) on the removal of B.B dye was studied with adsorbent dose varying in the range (0.1-1)g at initial concentration (30 ppm) is illustrated in figure (2).

From this figure, it is concluded that adsorbent weight for the best adsorption is $(0.8 \text{ g for A.P with } H_2SO_4 \text{ and with } HCl \text{ but } 0.9 \text{ g for S.S.S with } H_2SO_4 \text{ and } 0.6 \text{ g with } HCl)$ as beyond this dose, there is not much increase in the adsorption. With increase in the adsorbent dose the percentage removal of B.B dye increase. That attributed to increased adsorbent surface area and availability of more adsorption site.⁽¹²⁾

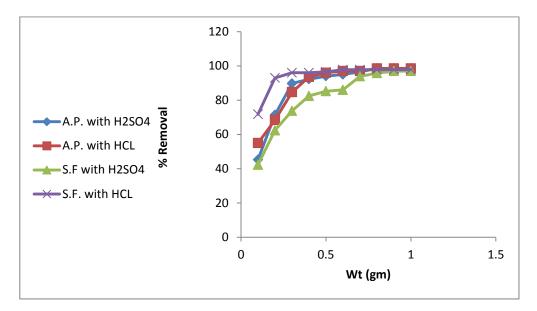


Figure (2): Effect of adsorbent weight on adsorption of B.B dye.

4-2 Effect of pH solution on adsorption B.B dye:

The pH of solution has been identified as the most important parameter affecting dye adsorption onto the adsorbent. The adsorption of B.B dye on (A.P and S.S.S) was studied in the pH range (2-10), The removal percentage of B.B dye exhibit in the figure (3).

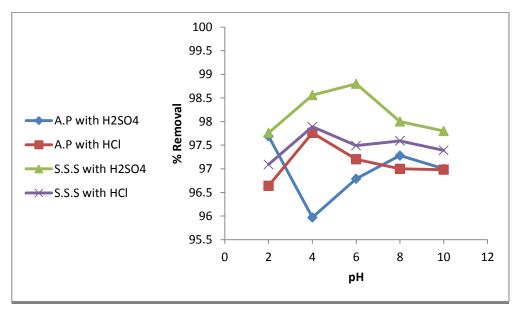


Figure (3): pH effect on adsorption of B.B dye.

The maximum adsorption of dye onto A.P with H_2SO_4 at the pH(2), A.P with HCl at the pH(4) and S.S.S with H_2SO_4 at the pH (6), S.S.S with HCl at the pH (4). At lower pH, the removal of dye is more favorable due to the presence of carbonyl and hydroxyl functional groups on the surface of adsorbent. In acidic condition, the functional groups on the adsorbent surface polarize thereby creating electrostatic interaction along side with hydrogen bonding and van der walls interaction. This increases the electrostatic attraction between the anionic group (SO₃⁻) of the dye and protonated groups of A.P and S.S.S an increase in dye adsorption. However, the lower dye adsorption within a higher pH solution may be due to the abundance of hydroxide ions (OH⁻) and the ionic repulsion occurring between the anionic dye molecules and the negatively charged surface of the A.P and S.S.S. The optimum pH is frequently reported in the literature to be around pH acidic medium for anionic dye adsorption.²⁰

4-3 Effect of Contact Time on adsorption of B.B dye:

The effect of contact time can be seen from figure (4) for the B.B dye. It is clear that the extent of adsorption is rapid in the initial stages and becomes slow in later stages until the maximum adsorption of B.B dye onto adsorbent (A.P and S.S.S) was observed. It can be estimated that the equilibrium is reached after (40,30 min. for A.P with H_2SO_4 and HCl respectively and 35,40 min. for S.S.S with H_2SO_4 and HCl respectively), as there is not much increase in the percentage removal after this time period. It is basically due to saturation of the activate site which does not allow further adsorption to take place.¹⁴

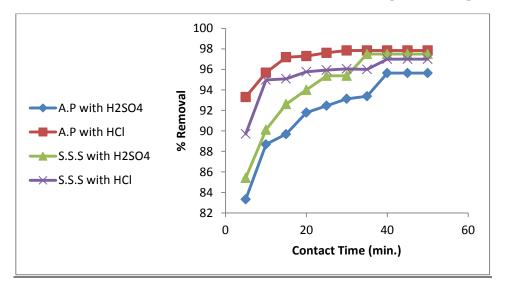


Figure (4):Effect of Contact Time on adsorption of B.B dye.

4-4 Effect of temperature on adsorption of B.B dye:

The temperature effect was investigated by applied three temperature degrees $(35, 45, 55)^{\circ}$ C at the following conditions, the best pH of adsorbents, and less than 500 µm diameter granules of the(0.1-1)g weight of adsorbent, with adsorbate of initial concentrations (30) ppm. The results are shown in figures (5),(6),(7) and (8).

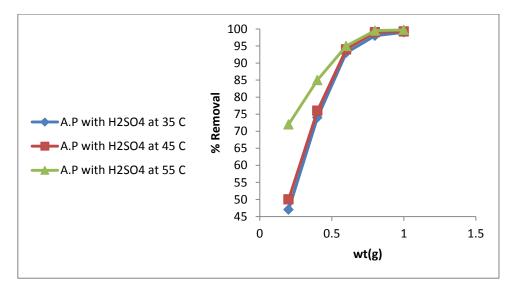


Figure (5): Effect of temperature on adsorption of B.B dye with A.P activated by H₂SO₄.

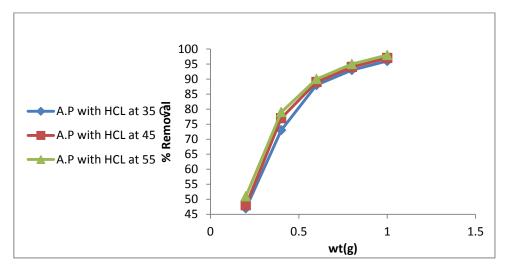


Figure (6):Effect of temperature on adsorption of B.B dye with A.P activated by HCl.

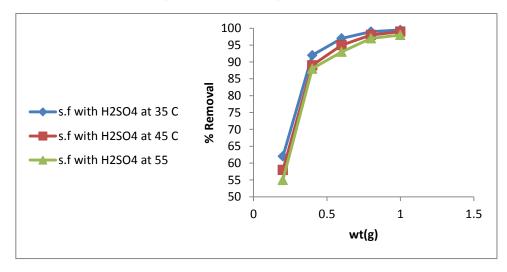


Figure (7):Effect of temperature on adsorption of B.B dye with S.S.S activated by H₂SO₄.

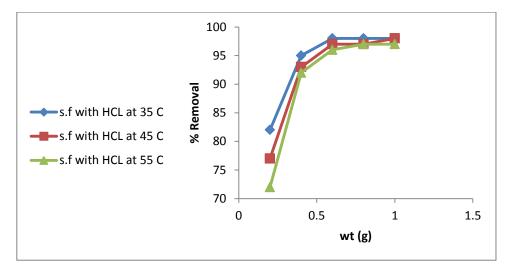


Figure (8): Effect of temperature on adsorption of B.B dye with S.S.S activated by HCl.

The figures (5) and (6) exhibit an increase in the adsorption of B.B dye on the A.P that activated by H_2SO_4 , HCl respectively with increase the temperature that due to presence of pores on the surface of adsorbent and when temperature increasing that lead to increase the kinetic energy for dye molecules and enter inside the pores thus increase the adsorption with increasing the temperature.⁽²¹⁾The figures (7) and (8) exhibit decrease

the adsorption of B.B dye on the S.S.S that activated by H_2SO_4 , HCl respectively with increase the temperature that due to when temperature increasing that lead to increase the kinetic energy for adsorbing molecules on the adsorbent surface which may increase the probability of molecule desorbed.²¹

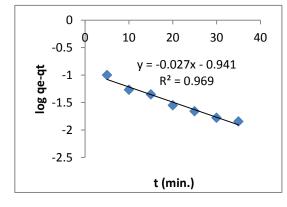
4-5 Kinetic of adsorption of B.B dye on A.P and S.S.S:

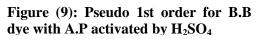
Adsorption kinetic studies are important in the treatment of aqueous effluents because they provide valuable information on the mechanism of the adsorption process.⁽²²⁾

From the figures (9), (10), (11), (12), (13), (14), (15) and (16) we conclude that pseudo- second -order is the best fitting kinetic model for adsorption B.B dye onto A.P and S.S.S because the regression correlation coefficient (R^2) of the pseudo second order is more linear when compared with that of the pseudo first order as show in table (1).The best fit of the second-order expression suggests that the chemisorptions mechanisms involved in the adsorption.¹¹

Table (1): the regression correlation coefficient (R²) of the pseudo 2nd , 1st Order.

Type of adsorbent	Pseudo 1st order R ²	Pseudo 2ndorder R ²
A.P activated by H_2SO_4	0.969	0.982
A.P activated by HCl	0.965	0.991
S.S.S activated by H_2SO_4	0.981	0.987
S.S.S activated by HCl	0.799	0.963





10

20

y = -0.0639x - 1.2427

 $R^2 = 0.9658$

30

0

-1

-3

-1.5 -2 -2.5

log qe-qt

-0.5

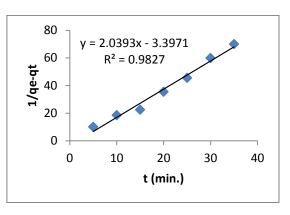


Figure (10): Pseudo 2ndorder for B.B dye with A.P activated by H₂SO₄

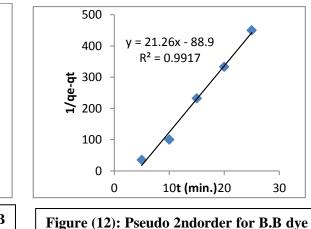
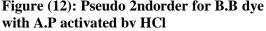
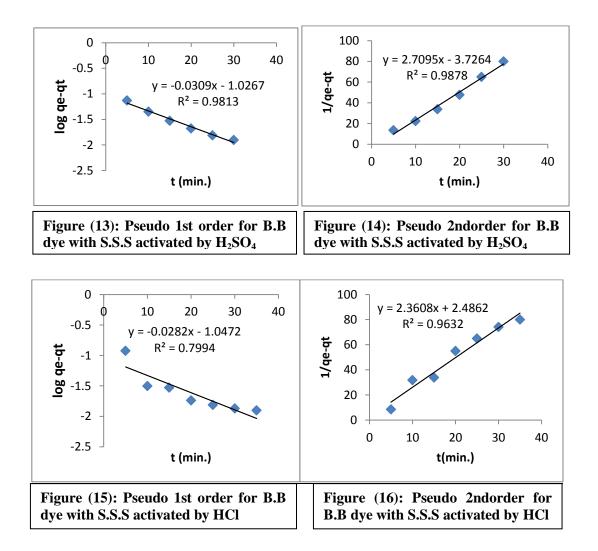


Figure (11): Pseudo 1st order for B.B dye with A.P activated by HCl

t (min.)





4-6Thermodynamic parameters of adsorption of B.B dye on A.P and S.S.S:

Table (2) shows the values for the thermodynamic parameters. The positive value for the enthalpy change, ΔH (4000, 3000 J/mol), indicates the endothermic nature of the adsorption, which explains the increase of B.B dye adsorption efficiency onto A.P as the temperature increased. But the negative value for the enthalpy change, ΔH (-10500, -5000 J/mol), indicates the exothermic nature of the adsorption, which explains the decrease of B.B dye adsorption efficiency onto S.S.S as the temperature increased.⁽²³⁾

The positive value for the entropy change, ΔS (31.11, 14.03 J/mol K), indicates that there is an increased disorder at the solid/liquid interface during B.B dye adsorption onto the adsorbent (S.S.S). But the negative value for the entropy change, ΔS (-10.26, -9.933 J/mol K), indicates that there is a decrease disorder at the solid/liquid interface during B.B dye adsorption onto the adsorbent (A.P).²⁴The negative value for the free energy change, ΔG , implies the spontaneity of the adsorption process, which does not require an external energy source for the system. But the positive value for the free energy change, ΔG , implies the no spontaneity of the adsorption process.²⁵⁻³¹

Type of adsorbent	Tem. °C	$\Delta \mathbf{G} \mathbf{J}.\mathbf{mol}^{-1}$	$\Delta \mathbf{H} \mathbf{J}.\mathbf{mol}^{-1}$	$\Delta S J.mol^{-1} k^{-1}$
A.P with H ₂ SO ₄	35	-106.15	3000	-9.933
	45	-243.55		
	55	-314.01		
A.P with HCl	35	772.54	4000	-10.26
	45	730.65		
	55	571.50		
S.S.S with H ₂ SO ₄	35	-648.70	-5000	14.03
	45	-456.65		
	55	-439.61		
S.S.S with HCl	35	-790.24	-10500	31.11
	45	-450.57		
	55	-157		

Table (2): Thermodynamic parameters of adsorption of B.B dye on A.P and S.S.S:

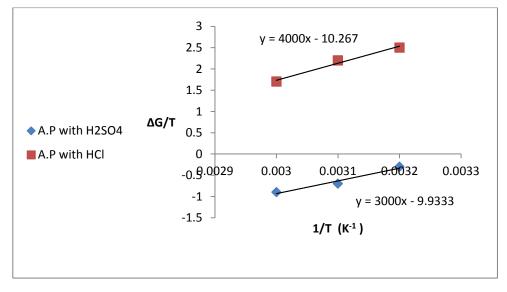


Figure (17): Van't Hoff plot for different temperature for AP with H₂SO₄ and HCl.

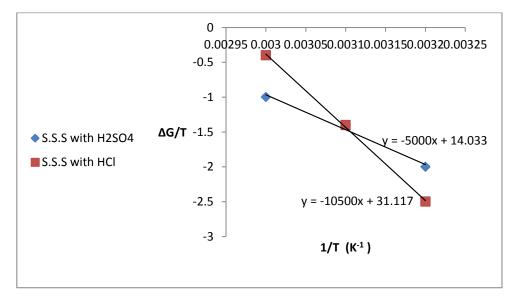


Figure (18): Van't Hoff plot for different temperature for S.S.S with H₂SO₄ and HCl.

5. Conclusion:

- 1. The adsorption of B.B dye was examined at different experimental conditions. The results corroborate that the adsorption of B.B dye onto almond peel that activated by H_2SO_4 and HCl increase with increase in dose of adsorbent, contact time ,temperatures and the best pH was (2,4) respectively.
- 2. The results for adsorption of B.B dye on the sunflower seed shell activated by H₂SO₄ and HCl shows that the removal percentage of B.B dye decrease with increase in the temperatures but the removal percentage of B.B dye increase with increase in dose of adsorbent, contact time and the best pH was (4,6) respectively.
- 3. The kinetic studies proved that the pseudo second order kinetic was the best applicable model.
- 4. Thermodynamic parameters showed that the adsorption process for B.B dye onto (almond peel with H_2SO_4 and HCl) an endothermic process, decrease disorder and spontaneity of the adsorption but almond peel with HCl non spontaneity. But the adsorption of the B.B dye onto (sunflower seed shell with H_2SO_4 and HCl) was an exothermic process, increase disorder and spontaneity of the adsorption process.
- 5. The present research work established that almond peel and sunflower seed shell were excellent low cost adsorbent for removal of B.B dye.

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