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Dynamic Sorption of Alizarin Red S by Fixed Bed Activated Carbon

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Dynamic Sorption of Alizarin Red S by Fixed Bed Activated Carbon

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Keywords: alizarin red S, norit GCA830, fixed bed, adsorption capacity.

I. Introduction

Due to the rapid urbanization and rapid growth of industrialization, large quantities of waste containing pigments and dyes are discharged into the receiving aquatic environment.It is expected that the dyeing industry is responsible for the release of 100 tons of dyes per year into the environment, contaminating rivers and springs [1]. Of these dyestuffs, 5–10% is lost in industrial effluents, and consequently, wastewater treatment is one of the biggest problems we face today [2]. In fact, due to ineffective treatment processes, the presence of these dyes will have a negative impact on environmental life in general [3]. Knowing that synthetic dyes are not biodegradable and toxic in nature; their presence in the aquatic ecosystem hinders the growth of biota and affects the food web [4]. In addition, long term human exposure to dye pollutants increases the risk of tumours, cancers, cerebrovascular and lung diseases [5].

Nowadays, several industrial sectors use dyes in their product such as leather, textiles and food processing industries, there is a primary concern of decolorization and treatment of wastewater [6]. In addition to adsorption which has been widely used, there are various methods of physical and chemical treatment for organic dyes. The most important treatment processes are coagulation and flocculation [7], photodegradation, biosorption, oxidizing agents, membrane and ultrafiltration. The advantages, disadvantages and limitations of each technique have been widely studied by many researchers [8-10]. However, those technologies are costly, less effectiveand lack adaptability to a wide range of dyes present in the wastewaters. As a technique obeyed by environmental rules, capable of eliminating a wide range of contaminants even at very low concentrations [11], the adsorption processes presents the most preferable method for the purification of water from synthetic dyes [12, 13].

The adsorption studies are generally divided into two parts. The first is the batch adsorption. It has the advantage to provide useful data and parameters on the adsorbent and the adsorbent-adsorbent mass transfer mechanism such as the maximum adsorption capacities and the kinetic constants as well as the type of the isotherm inducing the type of kinetics controlling the adsorption process [14, 15]. The second technique is that of continuous adsorption, for which the effluent to be treated flows over a bed of active charcoal or other adsorbent placed in a fixed bed. This process is also necessary for access to practical operational information [16-18]. During the adsorption on a fixed bed, the columns can be placed in series or in parallel. In this area, small column studies are started to simulate the potential performances of adsorption, and then the results obtained will be extrapolated in large scale columns or reactors [19,20].

In addition to the kinetic parameters, the adsorption capacity of activated carbon in a fixed bed is of great practical importance because it makes it possible to evaluate the efficiency of the fixed bed with respect to the elimination of the contaminants [21]. This was the subject of several studies. Different methods have been used to access this parameter. We distinguish among others, those who use the curve of breakthrough [22,23]. In addition the BDST model and the Clarks model are frequently used to access in this parameter [14].

In the present work, we have carried out an experimental study for testing the ability of Norit

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activated carbon to remove the Alizarin Red S as an example of anionic dye. Our study is realised for static regime in Batch reactor. However series of columns is used in dynamic regime. The adsorption capacity of activated carbon in static and dynamic is calculated by using different methods for different operating conditions.

II. Material and Method

a) The Adsorbent

In present study we used activated carbon NORIT GCA830; the main characteristics are extracted from Norit Degital Library is shown in Table 1 [24].

Table 1: main characteristics of NORIT GCA830 activated carbon

Specific surface (m^2/g)	1000
Molasses Index	210 min
Iodine number (mg/g)	920 min
Moisture conditioning (%)	2 max
Mesh size(%) :	
- Greaterthan 8mesh (2.36mm)	8 max
- Lessthan 30 mesh (0.6mm)	4 max
Abrasion number (AWWA)	75 min
Apparent density (mg/l)	0.51
Effective size (mm)	0.86
Density after backwash and drainage ($g.cm^{-3}$)	0.45

b) The Adsorbate

Our study focused on adsorption of Alizarin Red S dye by NORIT GCA 830 in batch and fixed bed reactor. Alizarin Red S (sodium alizarin sulphonate, CI 58005) is an antraquinone.

Figure 1 shows its chemical structure molecule:

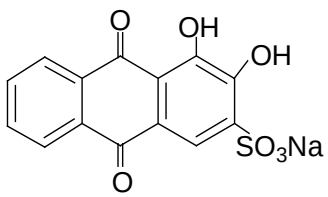


Fig. 1: Chemical structure molecule of RAS

The mains property of RAS is resumed in Table 2.

Table 2: Characteristic of RAS

Number (CI)	58005
Class	anthraquinone
Molar Wight (g)	342.267
Ionisation	acide
Ebullition temperature (°C)	430
Fusion temperature (°C)	287-289

c) Batch equilibrium studies

The study of the adsorption of RAS by the activated carbon NORIT GCA830 in a static condition was carried out according to the following experimental protocol:

1000 ml of RAS aqueous solution of variable initial concentration are stirred with 50 mg of NORIT GCA830 of particle size between 0.4 and 0.63 mm at ambient temperature. After a given stirring time, samples were taken and then centrifuged at 3000 rpm for 5 minutes. The supernatant is analyzed using a spectrophotometer (HATCH DR / 2000).

d) Fixed-bed adsorption experiments

For the experiments under dynamic conditions, we carried out experiments of adsorption in fixed bed by using the prototype given in Figure2:

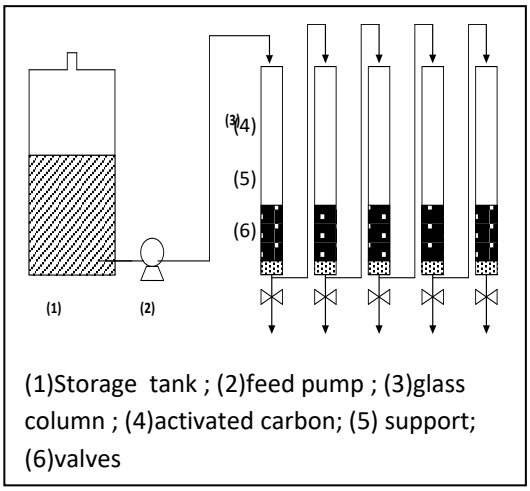


Fig. 2: Fixed bed adsorption set up

Each column of inner diameter of 1.2cm, contains a given weight of active carbon of particle size of 0.4 to 0.63 mm, corresponding to cumulative height respectively of 3.3, 5.3, 7.3, 9.3 and 11.3cm. The inlet solution dye concentration is 25 mg / l to 75 mg /l and its flow rates are 7, 15, 22.5 and 30ml/s.

III. Results and Discussion

a) Batch equilibrium studies

In order to determine the adsorption parameters of RAS on NORIT GCA830, the sorption isotherms of

Langmuir and Freundlich isotherms in their linearized form were determined. We obtain right lines whose regression coefficient ($R^2 > 0.98$).

The values of Langmuir parameters such as the maximum adsorption capacity q_m and of the equilibrium constant K_l and their of Freundlich (K_F and n) are presented in Table **3**.

We can be seen that the n value is between 2-10 the rung indicates a good favourability of sorption [13]. According of applicability of both Freundlich and Langmuir isotherms, we deduce that both heterogeneous surface and monolayer adsorption conditions exist under the actual studied process of sorption

Table 3: Langmuir and Freundlich parameters for RAS Adsorption

Langmuir			Freundlich		
K_l (l/mg)	q_m (mg/g)	R^2	K_F	n	R^2
0.13	384.6	0.9978	78.90	2.55	0.9835

b) Dynamic sorption

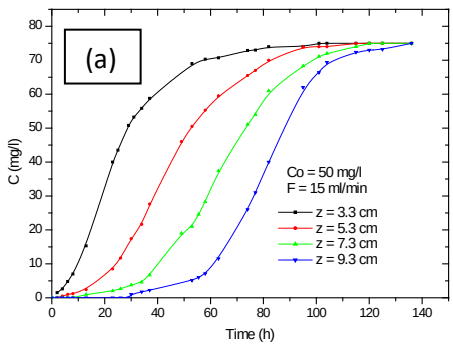
i. *Breakthrough curve*

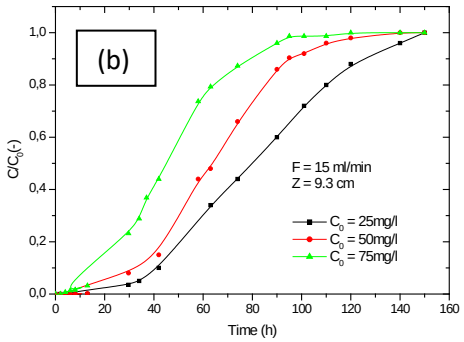
To obtain these curves, we followed, as a function of the time, the evolution of the concentration of RAS effluents at the columns exit. The curves are represented for different height of the absorbent bed (Figure **3.(a)**) and for three concentrations (Figure **3.(b)**). The influence of flow rate on the breakthrough curve is given in Figure **3.(c).**

It can be seen that, these curves are characterized by two particular points

- The breakthrough time t_b: corresponding to the leakage concentration C_b of $0.1C_0$, which it is chosen arbitrarily, as the extreme value of the RAS solution concentration at the outlet of column.
- The saturation time t_s: corresponding to the saturation of fixed bed activated carbon, where $C_s = C_0$.

According the results illustrated in above curves, the evolutions of breakthrough and saturation times are function of studied parameters. From Figure **3(b)**, we can be seen that the breakthrough and saturation times are all the shorter as the height of the bed is short. However they are shorter as the initial concentration and flow rates are high (see Figure **3(a)** and **(c)**).





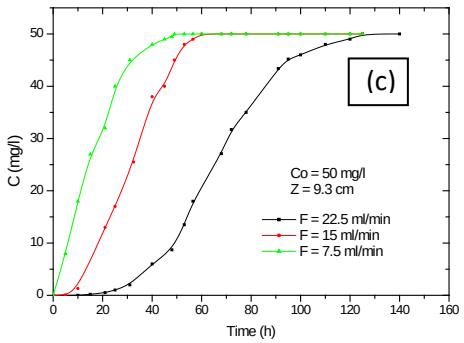


Fig. 3: Breakthrough curves for**(a)**different bed height, **(b)**different inlet concentration, **(c)** different flow rate

In order to understand the evolution of the breakthrough and saturation time as a function of the studied parameters, we have plotted in Figure **4 (a)** and **(b)**, those variations respectively versus bed height and the reverse of flow rate. It can be seen, the increase in t_b and t_s is directly proportional to the bed height (Z), but inversely proportional to the flow velocity. However, no relationship could be established to describe the evolution of t_b and t_s the initial concentration.

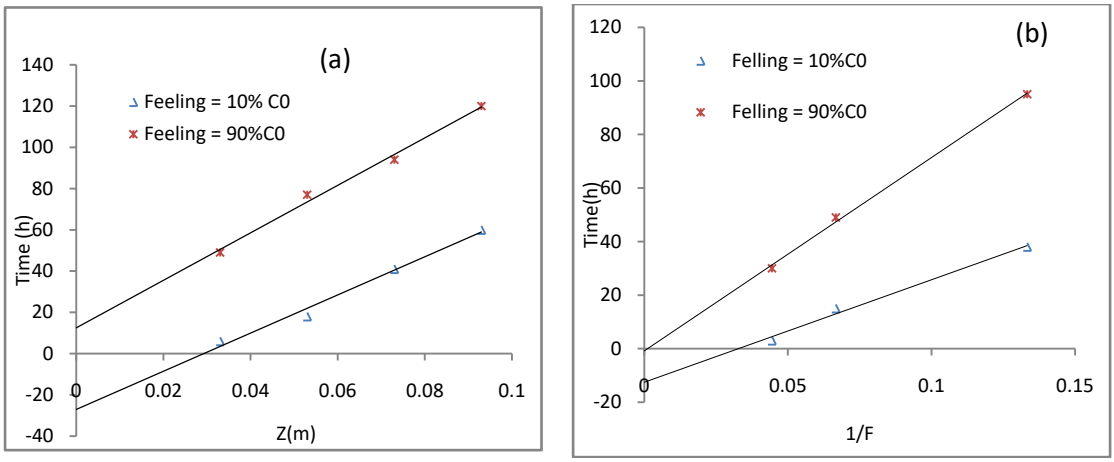


Fig. 4: Breakthrough and saturation times versus **(a)** bed height **(b)** the reverse of flow rate.

An important result can be deduced from these curves is that the time difference (t_b-t_s) is independent of the bed height (Figure **4 (a)**). Result means that the adsorption zone height (where the concentration varies between 0 and C_0) is independent of the bed height. On the other hand, when the flow rate increases the time difference (t_b-t_s) decreases (Figure**4 (b)**).

ii. *Assessment of adsorption capacity*

Based on the breakthrough curves, the amount of RAS adsorbed by activated carbon in the fixed bed is calculates by using the following equation [25] :

$$q = \frac{F}{m}\int_0^{t_s}(C_0 - C_s)\,dt \qquad (1)$$

Where F is the volumetric flow and m is the weight of activated carbon.

The integral in Eq. (1) is equal to the area includes the ordinate axis and the breakthrough curve, where it's calculates by using the Matlab software. Values of adsorption capacity calculated from the breakthrough curves at different experimental conditions are grouped in Table 4.

Table 4: Adsorption capacity of fixed bed for different operating conditions

Bed height (m)	Flow rate (ml/min)	Inlet concentration (mg/l)	Adsorption capacity (mg/g)
0.033	7.5	75	360
0.053			380
0.073			382
0.093			385
0.053	7.5	25	222.3
		50	347.6
		75	380
0.053	7.5	50	347.6
	15		318.7
	22.5		276

It is found that the adsorbed amount depends of the bed height, the initial concentration and the flow rate. As the bed height increases the liquid residence time in the bed increases, and therefore the molecules of RAS diffuse deeply into the adsorbent grains. This results in an increase in the adsorbed amount which reaches its maximum value of 380mg / g when the bed height exceeds about 0.08m. Increasing inlet concentration results to increase its gradient in the liquid film. Consequently, an improvement of the solute diffusion from the liquid phase to activated carbon grains. This results in an increasing of adsorbed amount. However, the adsorbed amount decreases, when the flow rate increases (Figure 5). In fact, an increase in flow rate results in a reduction in the thickness of the liquid film around grain, which has the effect of improving external diffusion, hence adsorption. But at the same time there is a reduction in the

residence time and the molecules of solute doesn't penetrate enough within the adsorbent grains. In sum, a decrease in the adsorbed amount is observed when the flow rate increases.

It should be noted that at zero flow velocity, the adsorbed amount value is about 385 mg / g (see Figure 5). It is equal to that determined by the Langmuir model.

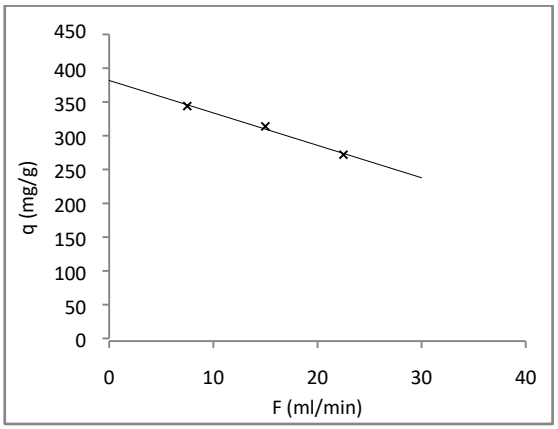


Fig. 5: Absorption capacity versus inlet flow velocity

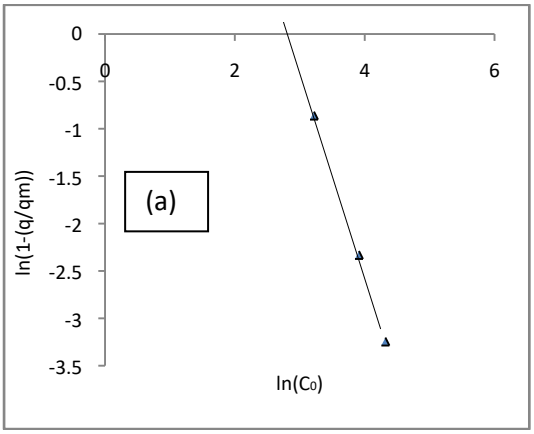
As we have seen above, the adsorption capacity of the fixed bed is a function of the inlet concentration C_0, the flow rate F and the bed depth Z. It can be written according to Danny et al. [26] in the form:

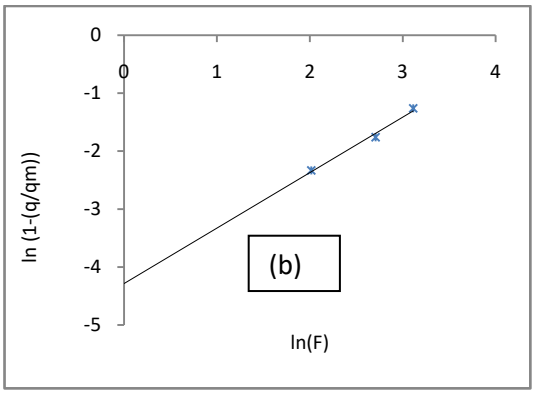
$$1-\frac{q}{q_m}=w.C_o^a.F^b.(S.Z)^c \qquad (2)$$

Where: q_m = 385mg/g is the maximum absorption capacity.

And S is the column section.

The coefficients a, b and c are the slopes of line plotted in Figure 6 (a), (b), (c). The ordinates at the origin are respectively $C_1 = w.F^b.(S.Z)^c$; $C_2 = w.C_0^a.(S.Z)^c$ and $C_3 = w.C_0^a.F^b$, from which we deduce the mean value of w.





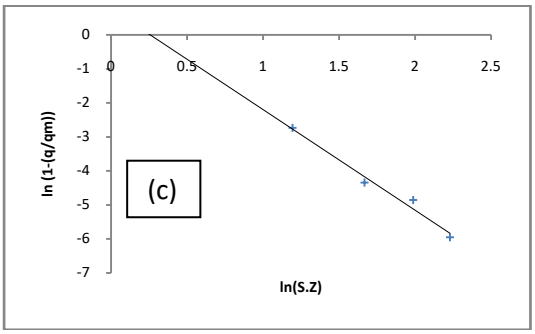


Fig. 6 (a), (b), (c): Determination of Equation 2 parameters

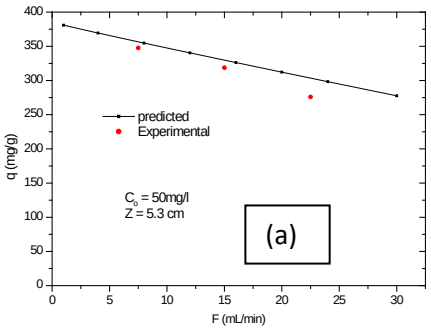
The adsorption capacity of Norit GCA830 in fixed bed for the RAS is given by the following expression:

$$1-\frac{q}{q_m}=5000\left(C_o^{-2.17}.F^{0.96}.(SZ)^{-2.96}\right)$$

It can be expressed as

$$q=385\left(1-5000\left(C_o^{-2.17}.F^{0.96}.(SZ)^{-2.96}\right)\right) \qquad (3)$$

Figures. 7 (a), (b), (c)shows a good correlation between the experimental values and those calculated from Equation (3).



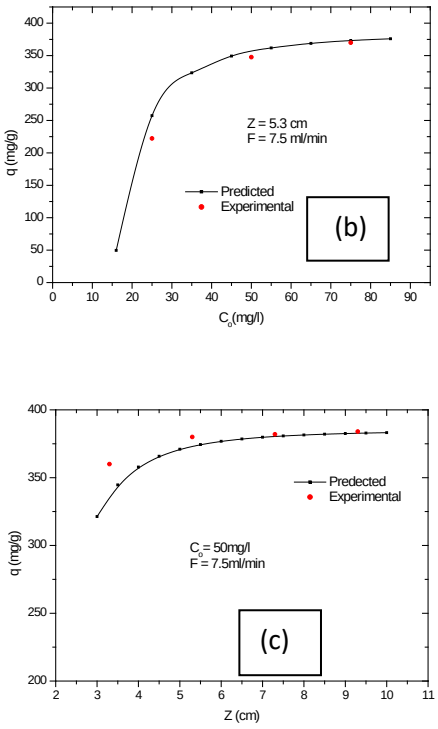


Fig. 7 (a), (b), (c) : Comparison of experimental and calculated absorption capacity for **(a)**different flow rate, **(b)** inlet concentration, **(c)**bed height

iii. *BDST model*

The present model, proposed by Hutchins [27], yields the breakthrough concentration C_b to the breakthrough time.

$$t_b = \frac{q}{C_0 u_0} Z - \frac{1}{kC_0} \ln\left(\frac{C_0}{C_b} - 1\right) \qquad (4)$$

Where u_0 is the flow velocity (m/s)

According to the Equation **(4)**, the curve $t_b = f(z)$ is a right line called Bed Depth Service Time (BDST). Its slope is $m_x = \frac{q}{C_0 u_0}$; allowed to calculate the absorption capacity when C_0 and u_0 are defined. In addition it can define the velocity of absorption zone migration v, where it's the reverse of slope m_x.

The ordinate at the origin $C_x = \frac{1}{kC_0} \ln\left(\frac{C_0}{C_b} - 1\right)$ makes it possible to calculate the kinetic constant k. When C_0 and C_b are known.

In addition the thickness of mass transfer zone Z_0 is defined as the length at $t_b = 0$, it given by the following equation:

$$Z_0 = \frac{U_0}{qk} ln\left(\frac{C_0}{C_b} - 1\right) \qquad (5)$$

BEST model present many advantageous such as it reduce the experiments duration. In fact, it is sufficient to determine the breakthrough time, corresponding to the breakthrough concentration C_b ($C_b = 0.1C_0$), at different bed height. The curves BDST plotted at different operating conditions shown in figure **8 (a), (b)** are linear ($R > 0.988$).

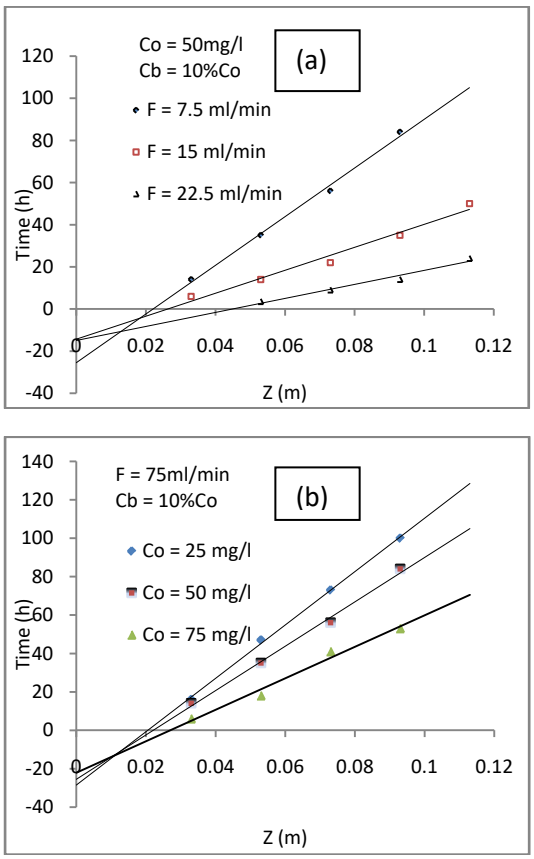


Fig. 8: Curves BDST at **(a)**different flow rate and **(b)**different inlet concentration

As it's shown in **Figure 8(a)**, the variation of inlet concentration leads to the variation of the slope m_x, the ordinate at the origin C_x and the thickness of mass transfer zone Z_0. In addition, increasing the flow rate, results in a decrease in the slope m_x and the ordinate at the origin C_x, Contrary to the thickness of mass transfer zone Z_0 (Figure **8(a)**).

If m_x, C_x and Z_0 are defined from BDST curves, adsorption capacity q, velocity of migration of adsorption zone v and the kinetic constant k are defined too, their values are shown in Table **5** for the different operating conditions studied.

Table 5: Influence of initial concentration and flow rate on adsorption capacity q, velocity of migration of adsorption zone v and the kinetic constant k

C_0(mg/l)	u_0(m/h)	m_x(h/m)	C_x(h)	q(mg/g)	Z_0(m)	k(l/kg.s)	v(10^3 m/h)
25	2.92	1390	28.57	209.2	0.02	0.854	0.72
	5.84	680	18.04	204.7	0.026	1.353	1.47
	8.76	415	17.2	187.4	0.041	1.42	2.4
	11.68	270	12.4	162.6	0.045	1.968	3.7
50	2.92	1155	25.515	348	0.022	0.478	0.866
	5.84	545	14.385	328.4	0.026	0.848	1.834
	8.76	332.5	14.973	300.5	0.045	0.815	3
	11.68	167.5	11.093	268.1	0.050	1.1	5.97
25	2.92	1390	28.57	209.2	0.020	0.854	0.72
50		1155	25.515	348	0.022	0.478	0.866
75		830	22.29	374.8	0.027	0.365	1.2

As indicated in Table 5, the absorption capacity increases if we increase the input concentration and if we reduce the flow rate. It reaches its maximum value of 374 mg/g for the lower flow rate studied. For the thickness of mass transfer zone Z_0, it can be seen that it's proportional of inlet concentration and flow rate. In fact, an increase in the flow rate reduces the residence time of the solution in the bed and consequently the contact time solute-adsorbent. Thus the molecules don't penetrate enough within the adsorbent. This results in a widening of the height of adsorption zone.

Concerning the kinetic constant k, it can be seen that k increases as the flow rate increases. That because, the increase in the flow rate results in a reduction in the thickness of the film through which the diffusion becomes faster. With respect to the influence of the initial concentration, k decreases as C_0 increases. This evolution was also observed by Danny et al. [28] during the study of the adsorption of some metals in a fixed bed of activated carbon.

For the evaluation of the velocity of migration of the adsorption zone, it's clear that an increase in the flow rate or the inlet concentration leads to an increase in the velocity of migration and consequently the exhaustion of the bed and the saturation of the activated carbon, all the faster that u_0 and C_0 are higher.

IV. Conclusion

In the present work, our aim is to predict the adsorption capacity of fixed bed activated carbon Norit GCA830 to remove Alizarin Red S. From the experiments in Batch reactor the adsorption capacity determine using Langmuir model is of 385 mg/g. We are found that both model of Langmuir and Freundlich fit well our experimental results, but the Langmuir model is better.

For fixed bed adsorption process, the breakthrough curves have been plotted at various inlet dye concentration, bed height and flow rate. The obtained results show that both breakthrough and exhaustion times increases with the increase of bed height and the decrease of flow rate and inlet solution dye concentration. According the operating condition, the maximum absorption capacity, calculated from breakthrough curve area, is about 380mg/g. Using breakthrough curve, the absorption capacity of fixed bed is correlated in function of studied operating condition as following:

The adsorption capacity estimated using the BDST model increase with the inlet concentration where it achieved its maximum value of 374mg/g for a lower flow rate studied. In addition from BDST model, allowed to define the thickness of absorption zone it's between 0.02 to 0.05m. The constant kinetic and the velocity of migration of absorption zone estimated by BDST model are mostly influenced by the residence time.

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