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Assessment of Adsorption properties of *Acacia concinna* (Shikakai) for Removal of Acid Black 234 from Aqueous Solution

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Abstract This present study was performed for the sorption of acid black 234 from water with the help of *Acacia concinna* powder and find out the greatest operating conditions for sorption of acid black 234 on *Acacia concinna* powder process at the laboratory scale. Several parameters such as initial dye concentration, sorbent dose and contact time were applied. The kinetic data obtained from different batch experiments were analyzed using both pseudo first-order and pseudo second-order equations. Results revealed pseudo second order model accurately explains the data because of high R^2 value (0.9997). Elovich equation was also applied to study the kinetics, results endorsed that pseudo second order is the one to explain the kinetics of current process. Equilibrium study also performed to the sorption data by changing initial concentration of acid black 234 and removal was calculated to be 10-80 mgL^{-1}. It was concluded that Langmuir isotherm accurately explains calculated data because of maximum value of R^2 (0.9922).

Keywords Acid black 234, Shikakai powder, Kinetics, Isotherms

1. Introduction

For the existence of life, water is essential. A healthy life is possible if we drink pure water which is a main component of our wellbeing. Water is a basic requirement and an essentials need for every household activity [1]. Out of all the water reservoirs 3% fresh water is available and from it 0.01% is available for humans. Water Air, and land are being polluted by the physical, chemical and biological pollutants [2]. That's why it is imperative that the water must be of good quality. The water cannot be used again for the same purpose without treatment after has been used once for industrial purpose [3]. Various origins of water pollution include metals, industrial waste, dyes, pesticides and other solvents et cetera [4]. Our health and environment are being disturbed by polluted water. Polluted water causing deaths of millions people and different diseases [5].

Synthetic dyes are extensively used in various industries such as leather, paper, textile, printing, cosmetics, food, paint, petroleum, solvent, pigments, rubber, pesticide, wood, plastic, pharmaceutical industry and preserving chemicals and have received much attention for several decades because of release of these dyes into water sources contaminate and pollute the water [6]. There are many issues such as water, air and soil contamination as well as noise pollution is connected to Textile industry [7]. There is a thoughtful warning to the aquatic ecosystem as a result of dye pollution [8]. Textile effluent is a reason for critical measure of ecological debasement and human ailments [9]. Acid dyes are anionic, water soluble dyes which are applied as wool, nylon and silk over fibers at near boiling points using neutral to acid dyes baths [10].

Acacia concinna, belonging to the sub-order Mimoseae of the family Leguminosae, known as "Shikakai" in Hindi. The plant is found throughout India and Pakistan in tropical areas [11]. A decoction of the pods checks biliousness. It is also an expectorant, emetic and a purgative. Its decoction is useful for hair wash, growth of hair, killing of lice and removing of dandruff. The leaves are being used as substitute for tamarind in cooking and are acidic in nature. A paste (chutney) made of tender leaves, salt, tamarind and chillies is given in bilious affections such as jaundice. An infusion of the leaves is used in malarial fever; it checks flatulence and is a mild laxative [12].

Different treatment strategies including, physical, physiochemical and chemical processes have been researched for treating color bearing effluents. Every one of these strategies has diverse color expulsion capacities also have capital expenses and working rates. Coagulation, flocculation sedimentation, Floatation, Equalization and homogenization, Fenton reaction and Ozone oxidation et cetera are some methods for treating dye effluents [13]. Latest reviews showed that lethal and unmanageable natural mixes incorporating colors in wastewater can be eradicated by the most developed method i.e biosorption [14]. Biosorption is a procedure that uses natural and regular materials as adsorbents and this procedure has been considered by various researchers as a choice framework to standard methodologies for heavy metals and hues ejection from waste water. Usually the sorption of dyes is achieved from the aqueous solution, where the motivating force for adsorption is very sturdy and makes the process fast [15]. Hence, there is a mounting concentration in finding less expensive and easily available materials for the dye removal from industrial effluents [16]. It could be determined that a few of existing adsorbents have exposed the constrained capability to remove the working dyes, whereas others have the aptitude of potential development and application was noticed [17].

To our knowledge, the use of Shikakai as a biosorbent for the sequestration of acid black 234 (C.I.30027) from water has not been examined. The current study focuses on the acid black 234 removal properties of Shikakai powder, this powder is made as a desecrate biomass and has no significant marketable worth. Solution initial dye concentration varies the biosorption process, for maximum dye biosorption the amount of biosorbent and contact time were increased. The acid black 234 biosorption distinctiveness of Shikakai were interrelated with the biosorption capacities, communication mechanisms via biosorption isotherms and kinetic modeling.

2. Experimental

Biosorbent Preparation

The biomass, *Acacia concinna* (shikakai) fruit was collected. After the collection, the fruit of shikakai was washed and grinded into powder. It was wash carefully with tap water to get rid of dust and sand particle then dried the wet powder of shikakai under shade so that its chemical structure should not disturb in sunlight. After drying, the powder was washed again with distilled water to remove remaining impurities and dried it again. The dried powder was sieved to get equal sized particle of biomass. Bath sorption experiments were performed at room temperature. 1000mg/L Stock solution was prepared by adding 1g of Acid Black 234 dye in 1000mL of measuring flask by using distilled water. Then a standard solution of 20mg/L was prepared directly from stock solution by taking 2mL of stock solution in 100mL measuring flask and added distilled water up to the mark.

Procedure of Biosorption

The biosorption studies for valuation Acacia concinna (shikakai) of for the removal of Acid black 234 from the aqueous solutions was showed with the help of batch biosorption procedure. UV-VIS spectra were obtained by scanning the dye solution in range of 350-700nm.A calibration curve was drawn by making the variable concentration of dye solution from 10-100mg/L. The experiments of biosorption was performed using 50 mL of initial acid black 234 dye concentration (10mgL-1 to 80 mgL-1) with fixed shikakai biosorbent dosage (0.05–0.5 g) kept in a 250 mL stoppered conical flasks and restless in a thermostatic shaking incubator at the 120 rpm for an appropriate contact time (10–80 minutes). The samples were withdrawn, at a predetermined time intervals for remaining analysis of acid black 234 concentrations determined with the help of a double beam UV–visible spectrophotometer.

In order to measure the percentage sorption of acid black 234 and sorption capacity for all the values of acid black 234 (qe) following equations can be used.

$$\text{Percentage Sorption} = \left[\frac{C_i - C_f}{C_i}\right] \times 100 \quad (1)$$

$$q_e = \left[\frac{C_i - C_f}{m}\right] V \quad (2)$$

Where Ci denotes the acid black 234 concentration before sorption process, Cf denotes the concentration of acid black 234 after sorption process, qe represents the sorption capacity of acid black 234, V represents the volume and m is the mass of shikakai powder in gram. FTIR spectra was recorded before and after the adsorption.

3. Results and Discussion

Absorbance of 20mg/L solution was noted by UV-Visible spectrophotometer (from 350nm to 750nm) and found out the λ_{max} that was 610nm and then proceed at λ_{max} as shown in figure 1(a) and correlation coefficient R^2 value was 0.9791 nearly equal to unity as shown in figure 1(b). The FTIR studies of biosorbent and dye was performed as shown in figure 2. Fourier transmission infrared spectroscopy used to study functional materials as they performed vital role in the adsorption process. Therefore, FTIR analysis was performed before and after dye and biosorbent absorption to determine the participation of these functional groups [18]. The FTIR spectrum before and after adsorption was recorded in region of 400-500cm^{-1}. There was definite peak shift after the adsorption which is indicative of adsorption of acid black 234 dye on acacia fruit bio sorbent.A strong band at 3,291 cm^{-1} was attributed to either –OH or –NH group, and a band at 2,919 cm^{-1} was assigned to C–H stretching variation [19]. There was the peak at 1,738 cm^{-1} corresponding to C=O stretching variation [20]. After the adsorption the major peaks were shifted, the difference in peaks before and after are given in table.

Table 1: FTIR spectral characteristics of *Acacia concinna* fruit biosorbent before and after adsorption (cm^{-1})

Before adsorption	After adsorption	Groups
3291	3323	Bonded O-H stretching
2920	2929	C-H stretching
1738	1718	C=O stretching
1617	1624	N-H out of plane
1366	1328	C-O stretching
1216	1219	C-N stretching
1024	1051	C-O stretching

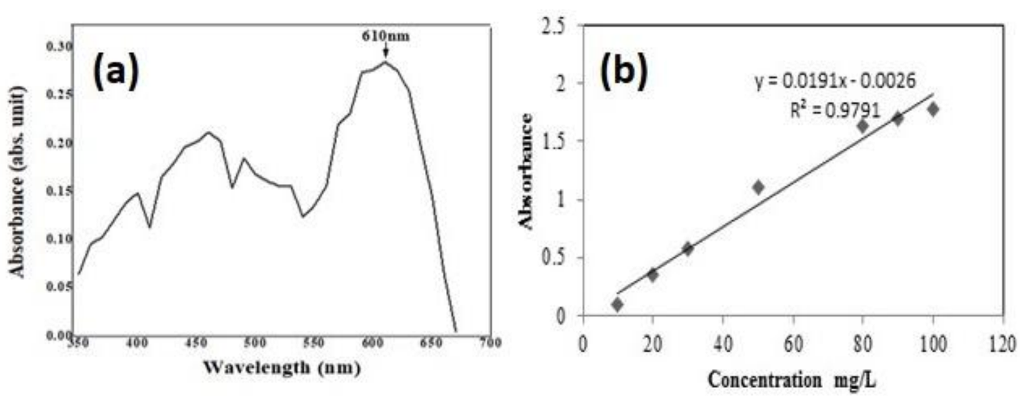

Figure 1: (a) UV-VIS spectrum for Acid Black 234 (b) Calibration curve to find correlation coefficient

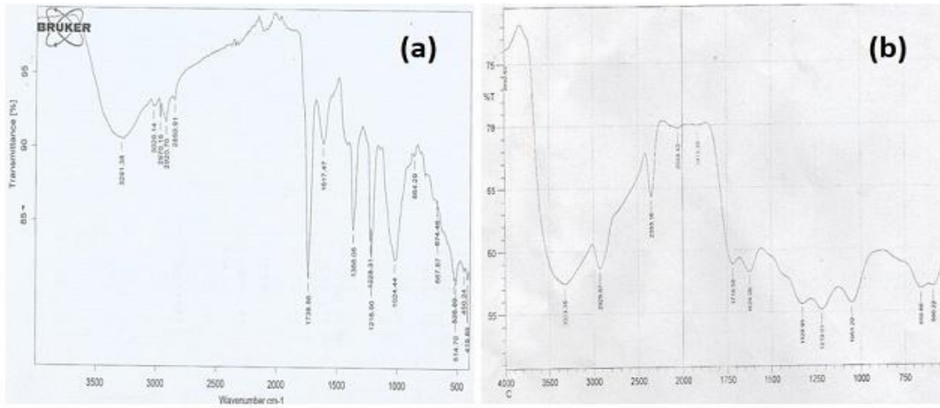

Figure 2: FTIR spectrum of Biosorbent (a) before removal of dye (b) after removal of dye

Effect of Biosorbent Dose

The dosage of adsorbent has also an effect over adsorption capacity. By increasing the amount of sorbent, increase in the adsorption capacity is observed. Samarghandy et al. reported that sorption of Reactive Black 5(RB5) by different dosage of biomass (potato peel waste) 0.2 and 1g has adsorption efficiency i.e. 82.21 and 95.06% respectively. They elaborated it as we increase the biomass the more active sites are available so the adsorption efficiency is increased [21]. Adsorption equilibrium is attained at 0.4g/L Maximum sorption efficiency was decreased from 38.92-7.43mg/g by increasing the dosage of biomass from 0.5-4g/L [22]. The effect of biomass weight was tested by taking different amount of biomass in culture tubes and added 50mg/L prepared solution of Acid Black 234. After shaking and filtration the absorbance of initial solution and shaken solution was noted by UV Visible spectrophotometer at λ_{max} (610nm).The results obtained from the graph (plotted weight verses absorbance) showed that with increase of biosorbent weight, the adsorption capacity or percentage removal also increased and after reaching a certain level it became almost constant. Dye removal increases from 32% to 73% as the biosorbent weight increased from 0.05g to 0.5g which can be credited to an increase in surface area of sorbent and availability of more active sites as shown in figure no. 3b [23].

Effect of Contact Time

In the literature it has been reported that biosorbent (potato peel waste) has sorption efficiency of 89.14% in early 60 minutes for RB5. After that there was no steep increase in the sorption efficiency upto 120 minutes but there was constant increase in sorption efficiency. At 120 minutes there is an equilibrium which has been established [21]. In this study, the effect of contact time was tested by shaking the culture tubes, containing 50mg/L prepared solution of Acid Black 234 and biosorbent weight, for different times. After shaking and filtration the absorbance of initial solution and shaken solution was noted by UV Visible spectrophotometer at λ_{max} (610nm). The results from the plot (time verses absorbance) showed that the time of contact with biosorbent is inversely proportional to absorbance and directly proportional to removal of colour. Adsorption or percentage removal of dye colour increases with increase of time of contact and the optimized time was 10 to 80 minutes. In present work the maximum % removal occur at 60 minutes and further became constant.

Effect of Concentration of Acid Black 234

The effect of contact time was tested by varying the concentrations of Acid Black dye solution. The culture tubes containing 0.3g biosorbent and these solutions were put on shaker. After shaking and filtration, the absorbance of initial solution and shaken solution was noted by UV Visible spectrophotometer at λ_{max} (610nm). Maximum percentage removal occurred with 10mg/L which was 76.76%. Results, obtained from plot (concentration verses %age removal) also showed that by increasing concentration, percentage removal will decrease and vice versa. (Figure 6).

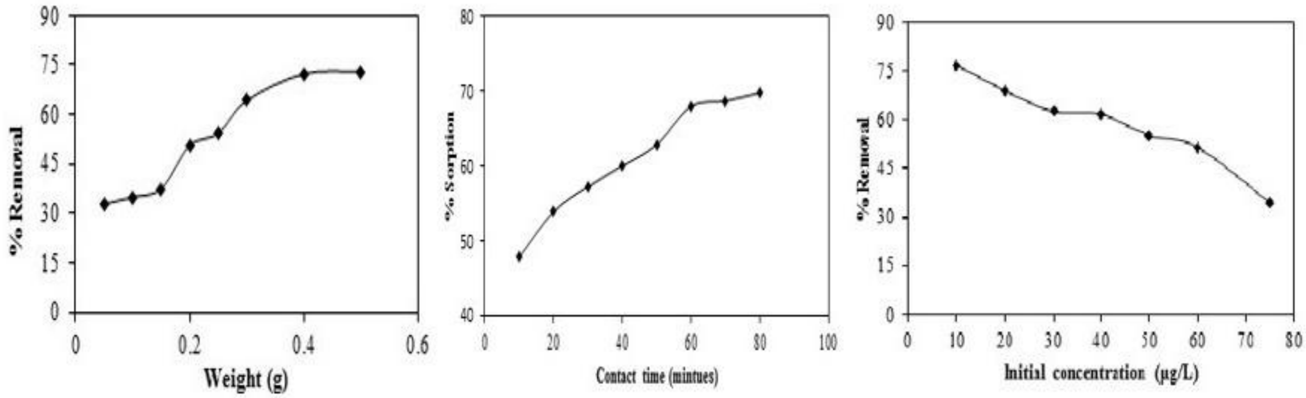

Figure 4: Effects of parameters on the removal of Acid Black 234 dye.(a) biosorbent dose (b) contact time(c) concentration

Kinetic Study

In order to investigate the biosorption kinetics of Acid Black 234, two kinetic models i.e first order and second order were used in this experimental study.

The equation for Pseudo-first-order kinetics is following

$$\log\ (q_e - q_t) = \log q_e - \frac{K_1 t}{2.303} \qquad (3)$$

Where q_t and $q_e$are amount adsorbed per gram at time t and at equilibrium respectively and k_1 is the first order rate constant (min^{-1}) for biosorption practice. The rate constant k_1 and equilibrium biosorption capacities $q_e$obtained from the slope and intercept respectively. It is a straight line equation interpreting the adsorption process. Values of correlation coefficient R^2 was 0.9869. The value of adsorption capacity q_e was very low as compared to experimental values moreover graph is giving negative value of slope (shown in figure 4a), displaying that the biosorption kinetics for whole process did not follow the first order kinetics.

Pseudo second order equation describes the capacity of solid surface for removal of acid black 234. Linear form of equation is

$$\frac{t}{q_t} = \frac{t}{q_e} + \frac{1}{K_2 q_e^2} \qquad (4)$$

k_2 is the rate constant for second order model. The plot of t/q_t and t for different time intervals gave a straight line. The rate constant k_2 and equilibrium biosorption capacities q_e obtained from the slope and intercept respectively. The rate constant k_1 and equilibrium biosorption capacities $q_e$obtained from the slope and intercept respectively. (figure 4b)

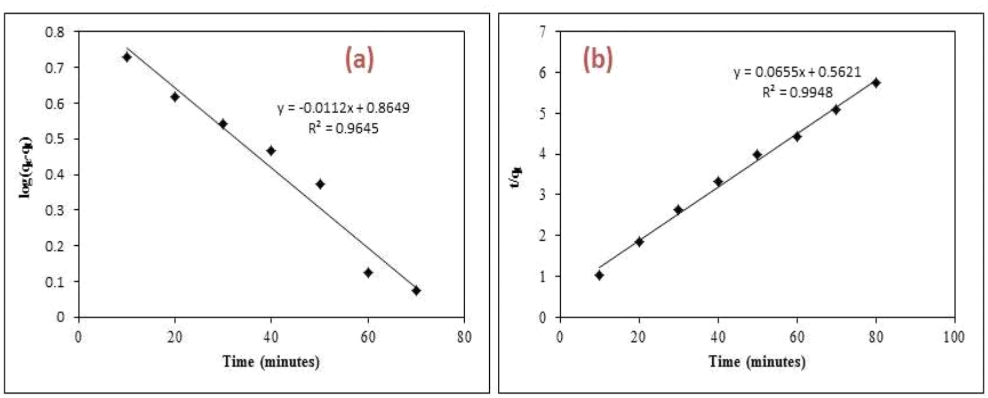

Figure 4: Biosorption kinetic study of Acid black 234 (a) First order (b) Second order

Intraparticle Diffusion Model

Diffusion of sorbate species into the pore of adsorbent is explained by intraparticle diffusion model and considers this step as the rate determining step. This model describes a direct association between the sorption of sorbate species and $t^{1/2}$. By only applying the pseudo-first order ad pseudo-second order we can't find mechanism of diffusion model which explains the intraparticle diffusion. The model is generally shown by the following equation:

$$q_t = K_{int}\, t^{1/2} + C \quad (5)$$

Where K_{int} (mol L^{-1} $min^{-1/2}$) is the rate constant for intraparticle diffusion and C is intercept. According to this model, if the line of the plot of the adsorption versus $t^{1/2}$ of time capacity q_t passes through the origin in any adsorption process, intraparticle diffusion could be the only controlling step. On the other side if line does not pass through the origin then some other processes will be involved alongside with this model. Some other steps are involved along with the intraparticle diffusion model [24].

Elovich Equation

Kinetics of heterogeneous surface by Chemisorption is explained by Elovich equation. The simple form of this equation is given under here:

$$q_t = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln(t) \quad (6)$$

qt is the amount of imidacloprid sorbed (mg g^{-1}) in a time (t), α (mg g^{-1} min^{-1}) is the initial sorption rate and extent of surface coverage is expressed as β (g mg^{-1}). Coefficients of Elovich equation are shown graph [25].

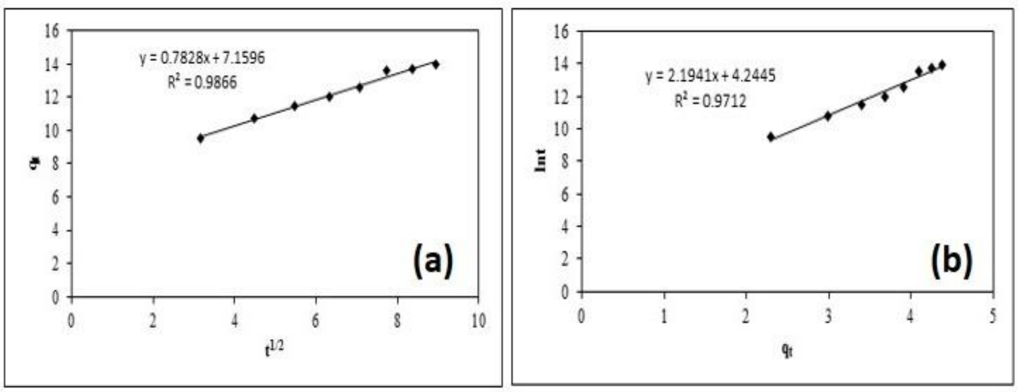

Figure 5: (a) Intraparticle diffusion model (b) The Elovich model

Table 2: Adsorption Kinetics coefficients of Acid Black234 on *Acacia concinna*

Model	Acephate
Pseudo-first-order kinetic	
q_e (mg g^{-1}) (exp)	14.928
q_e(mg g^{-1}) (cal)	7.32
K_1 (min^{-1})	0.025
R^2	0.9645
Pseudo-second-order kinetic	
K_2 (g mg^{-1} min^{-1})	7. 89×10^{-3}
q_e (mg g^{-1})	15.03
R^2	0.9948
Intraparticle diffusion	
K_{int}(mg g^{-1} $sec^{1/2}$)	0.7828
C	7.1598
R^2	0.9866
Elovich	
α (mg g^{-1} min^{-1})	8.0798
β (g mg^{-1})	0.4557
R^2	0.9712

To calculate the constant parameters, the particular linear equations were used and are summarized in Table 1. Based on the correlation coefficient (R^2), pseudo-second order is one of the kinetic models to explain the kinetics of the process because R^2 of this model is higher as compared to other models. Furthermore, the value of qe calculated from pseudo-second order was nearer to the experimental qe which suggests that pseudo-second order well interpreted the sorption data of acid black 234 onto *Acacia concinna* (Shikakai) [18].

Equilibrium studies for sorption of acid black 234

The adsorption isotherm is critical data demonstrating how adsorbate particles are disseminated between the solid phase and fluid when the adsorption procedure achieves equilibrium. It is very important to know about the equilibrium of system for finding the utmost sorption capacity of *Acacia concinna* for the dye in solution. Equilibrium data are vital necessities for adsorption systems and adsorption models, which are used for mathematical description of the adsorption equilibrium of the dye [26].

The experimental equilibrium adsorption data was inspected by using Langmuir and Freundlich adsorption isotherm models.

Langmuir isotherm

This isotherm explains the occurrence of sorption on a homogeneous surface in the form a monolayer, and the sorbate molecules have no interaction with each other [27]. This isotherm model may be represented in the following linearized form:

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \qquad (7)$$

Where K_L (L mg^{-1}) indicates an association between binding sites energy and affinity, while q_m (mg g^{-1}) denotes maximum sorption capacity. Langmuir isotherm has a very important parameter called dimensionless constant (R_L) which suggests whether the removal of the Acid black 234 onto *Acacia concinna* (Shikakai) is favorable or not and can be evaluated from the following equation:

$$R_L = \frac{1}{[1+K_L C_o]} \qquad (8)$$

In the aforesaid equation, K_L represents Langmuir isotherm constant (L mg^{-1}), while C_o is the initial concentration of acid black 234 (μg mL^{-1}). The nature of the removal process of acid black 234 can be investigated from the R_L value. If the value of R_L is < 1 then the process will be favorable and if it is >1 then the process will be unfavorable, while for linear sorption the value of R_L is equal to one [28]. The values of R_L are < 1 which indicates that the removal of acid black 234 onto *Acacia concinna* (Shikakai) is favorable process.

Freundlich Isotherm

This isotherm describes that sorbent surface consists of sorptive sites having different energies for the interaction of sorbate molecules and may be represented in the following linearized form:

The linear Freundlich isotherm is given by equation

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \qquad (9)$$

Where KF measures the sorption capacity (mg g^{-1}), q_e is the quantity of acid black 234 removed per unit mass of *Acacia concinna* (Shikakai) powder (mg g^{-1}) and n measures the sorption intensity and may help in finding the favorable nature of sorption. The value of n for favorable sorption is >1, while for unfavorable sorption it is < 1 while for a linear sorption its value is equal to one.

Freundlich rate constant K_f is evaluated from intercept in graph. The values of 1/n obtained from slope is less than unity which indicates that considerable adsorption take place at low concentration but the increase in adsorption becomes less significant at higher concentrations and vice versa (figure 7b). The data obtained from isotherm studied were tested for the applicability of above two models of isotherms. Values of parameters and correlation coefficients nearly equal to unity in both models are shown in graph. Both models are showing good adsorption on *Acacia concinna* but Acid Black 234 satisfied more to Freundlich isotherm model having R^2 0.9922 for adsorption on *Acacia concinna* as compared to Langmuir R^2 0.9567.

Table 3: Evaluation of adsorption constants at different isotherm models

Model	Acid black 234
Freundlich isotherm	
K_F (mg g^{-1})	0.651328
N	1.778094
1/n (g L^{-1})	0.5624
R^2	0.9922
Langmuir isotherm	
a_L(L mg^{-1})	0.2426
K_L (L g^{-1})	0.183621
Q_o(mg g^{-1})	4.122012
R^2	0.9567

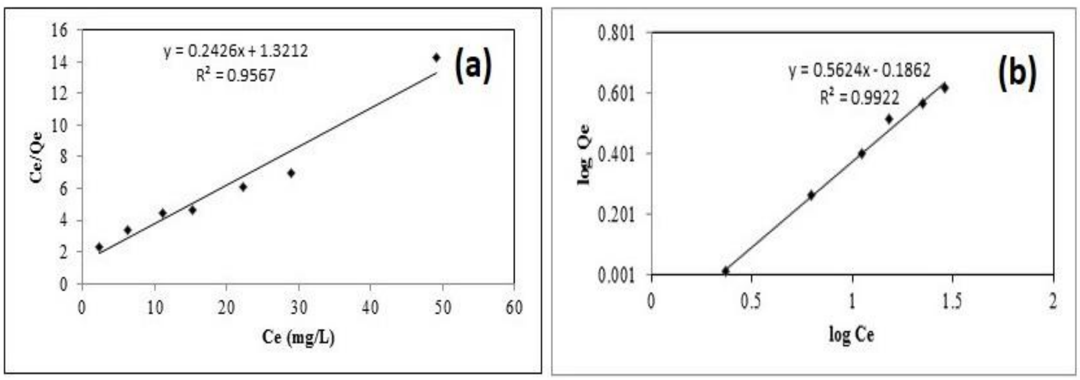

Figure 7: Adsorption isotherm for sorption of acid black dye by (a) Langmuir isotherm (b) Freundlich isotherm

Conclusion

The purpose of this effort was to investigate the probable utilization of biomass *Acacia concinna* (shikakai) powder available in low price in Pakistan, as adsorbent for the removal of dyes especial Acid Black234 used in textile and leather industries. Adsorptive removal of dyes on selected biomass can be taken as effortless, profitable and substitute to the modern exclusive methods. The removal of Acid Black 234 from wastewater using grounded *Acacia concinna* has been investigated under different experimental conditions in batch mode. The tentative fall outs have revealed that for decolorization of textile waste waters adsorption was a valuable process and *Acacia concinna* powder (adsorbent) is suitable for the sorption of Acid Black 234. It's an environmental affable substance. The Adsorption studies have showed that weight of biomass, contact time and concentration of dye solution played a significant role in effecting the capacity of biomass and the best dye removal was effective for the contact time of 10 to 80 minutes, dye concentration of 10-80 mgL^{-1}, adsorbent dosage of 0.05 to 0.5g, at room temperature. There adsorption increased with the increase in biosorbent weight and contact time and with increase of concentration of dye the results were opposite. Adsorption capacity q_e however increased with increase of dye concentration. The sorption of Acid Black 234 on *Acacia concinna* (shikakai) powder was found to follow both Langmuir and Freundlich isotherm, however perfectly suggesting the possibility of heterogeneous distribution of active sites on the surface of the sorbent and second order kinetics is the best fit. The introduced adsorption system on shikakai powder seems both very economical and efficient for the removal of acidic and reactive dyes such as acid black 234 from aqueous solutions.

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