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Application of Acaciatortilis Shuck as of Low-cost Adsorbent to Removal of Azo Dyes Reactive Red 198 and Blue 19 from Aqueous Solution

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Background & Aims of the Study: Textile wastewaters are very various in chemical qualities. In other words, these industries are one of the largest producing dyes compounds; therefore their treatment is so complex and difficult. The aim of this study was to evaluate the potential removal of Reactive Red 198 and Blue Reactive 19 from synthetic solution by Acaciatortilis shuck as a naturally absorbent.

Materials & Methods: The dyes samples with concentration (10, 50 and 500 mg/L) was prepared in laboratory. Next step was to determination of optimum pH among (pH solutions of 2, 4, 6, 8 and 10) and optimum adsorbate dosage with mesh 30-60 and 60-100 among (0.5, 1, 2, 3 and 4 g/L).Sampling in times 10, 20, 40 and 80 minute was done from the supernatant and was filtered (through 0.45 µm); finally the dye concentration was obtained by Spectrophotometer device. Adsorption isotherms of dyes Acaciatortilis shuck can were determined and correlated with common isotherm equations such as Langmuir and Freundlich models.

Results: The highest efficiency of dye removal from dyes solutions in pH 4, 10 min contact time and 2 g/L adsorbate dosage and using mesh 60-100 was for both Reactive Red 198 and Blue 19. Also the adsorption rate of reactive dyes was increased by increasing of initial dye concentration. The efficiencies for Reactive Red 198 and Blue 19 were 77.54% and 77% respectively. Adsorption isotherms was examined by Freundlich and Langmuir isotherm that finally showed the Freundlich multe-layer isotherm has better accordance with dates.

Conclusions: The results illustrate Acaciatortilis shuck is effective adsorbent and can be used as an available and inexpensive adsorbent to removal of textile dyes from effluent.

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Background

Textile industries are the major industries of each country. Water consumption is very high in this industry. Water consumption in the industry, according to the 200 to 500 liters per kilogram of product has been reported. The main characteristic of this type of industrial waste is usually colored and they are the major

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sources of environmental pollution (1). Colored wastewater in various industries such as textile and dyeing, pharmaceutical, food, cosmetics and health products, paper, tannery industries such as are produced. Most colors are used in the manufacturing of synthetic dyes (2,3). Synthetic dyes are often of high molecular weight aromatic structures. The aromatic structure to stabilize them, and break them out is difficult. Reactive dyes (one of the most widely used textile dyes) can react with the immune system and are trying to make a critical state (4,5). Synthetic colors chlorinated water may increase. Aesthetic problems, reduced water quality, lack of light penetration to the lower layers, the effect on the solubility of gases, toxic substances produced by their color or their failure to bring water to exist. In addition to these colors, is toxic to plants and animals, aquatic life and threaten humans and has the potential dangers to human carcinogens and mutagen (6,7). Azo dyes contain 60 to 70 percent of all colors production (about 700,000 tons annually) containing one or more azo-N = N-bond and dyeing because it was cheap and simple, widely used for dyeing fibers are groups of colors are mainly macromolecules that are not easily broken (8, 9). Bit color recommended standard by the World Health Organization (WHO) TCU by 15 points is the maximum acceptable limit of 5 units (10). Coagulation and flocculation, ion exchange, filtration, ultra filtration, advanced oxidation processes, electrochemical treatment and adsorption methods for color removal from aqueous solutions are applied (11,12,13). In order to protect human health and the environment, exploitation of cheap and economical methods for the removal of dyes from textile wastewaters and paint manufacturing plant, is the importance in the developing countries (14,15). Among these methods, adsorption process in terms of low cost, simple design, ease of operation and lack of sensitivity to the most appropriate technique for the removal of toxic pollutants in industrial wastewater to the color and quality of the reuse, is a considered (15,16). Even a strong adsorbent is activated carbon for toxic compounds. Activated carbon has a regular structure and crystal. Today activated carbon is one of the most materials used for drinking water and industrials wastewater treatment but it is expensive and requires specialized personnel, so its use is limited (17,18,19). This has led researchers to consider the use of alternative adsorbents that are economical and yet effective. In this study, the Acaciatortilis shuck used as an available and new sorbent for the removal of dyes. This plant is Mimozase family and one of the most common tree species in arid and semi-dry areas that has a wide geographical distribution in the world, including North Africa, the Arabian Peninsula and southern parts of Iran including Sistan-Baluchestan and Hormozgan. This plant is drought tolerant and has great endurance in the face of floods. This plant is a food source for animals and it is used for firewood and shelter is perfect for nomads. Research on fruit and tree branch in Hormozgan Agricultural and Natural Resources Research Center shows that the young branches is containing 6.16 % crude protein, 4.2% crude fat, 3.18 % crude fiber, 8.6% ash, 72% Ca and 21% of P. (20,21).

Aims of the study: The aim of this study was to survey of application of Acaciatortilis Plant shuck to removal of the dye Reactive Red 198 (RR-198) and Reactive Blue 19 (RB-19) from aqueous solutions. The effect of various parameters such as initial dye concentration, pH, contact time, and the size and adsorbent dose isotherms were studied.

Materials & Methods

The dyes used in this study were Alvan Sabet Company product and other chemicals used in the experiments were prepared by Germany's Merck product. Chemical structure of Reactive Red 198 and Blue 19 dye used is presented in Table 1.

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This study is an applied research which is carried out experimentally and in laboratory scale in Chemical laboratory environment of Zahedan University of Medical Sciences. Acaciatortilis plant shuck used in the experiments, in fact known as agricultural waste, was prepared in the city of Iranshahr Located at the Sistan-Baluchistan Province. The for preparation adsorbent, seeds were isolated on the shuck. Shucks were washed several times with distilled water to remove dust and other foreign particles. The cleaned shucks to a constant weight were dried for 12 h at 60°C in the oven. The dried shucks were crushed with food processor (Moulinex, France) and sieved using Octagon sieve (OCT-DIGITAL 4527-01) to various mesh sizes 30-60 and 60-100 to be tested for their adsorbance capacities. Finally the biomass was stored in plastic bottles for further use. Experiments will be carried out discontinuously in to 1000 ml beakers. Wastewater samples containing dye in different concentrations (10, 50 and 500 mg/L) will be prepared as synthetic in the laboratory. The materials used are manufactured by Merck Company, Germany. In the first stage with constant dye concentration, contact time and adsorbent dose, pH optimum is determined among the pHs of 2; 4; 6; 8 and 10. In the next stage with constant dye concentration, contact time and pH, the optimum adsorbent is determined among the amounts of 0.5, 1, 2, 3, 4 g/L. Optimum adsorbent dose of 2 meshes will be used between 30-60 and 60-100. In all experiments the samples will be mixed for 80 min at the speed of 100 rpm by orbital shaker. At intervals 10, 20, 40 and 80 min, 50 mL sample are taken from the supernatant solution. Taken samples are smoothed by the filter 0.45 mm until the dye concentration of remaining Reactive Red 198 and Blue 19 is reported the wavelength of 518 and 592 nm respectively, using UV-Vis Spectrophotometer (Thermo Spectronic, USA; model GENESYS 2), in accordance with the procedures set forth in the book Standard Methods for examination of water and wastewater (10). The final pH of the solution is measured to determine the rate of pH change. The pH of the adsorbate solution was adjusted by adding 0.1 N H_2SO_4 or 0.1 N NaOH. In this study, the Langmuir and Freundlich used to describe the relationship between dye and adsorbent and the amount of dye adsorbed at equilibrium time (q_e (mg/g)) and the percentage of removed dye (R%) are determined by equations (1) and (2) ,respectively (15):

$$R(\%) = \frac{C_o - C_t}{C_o} \times 100 \qquad (1)$$

Where C_o and C_t (mg/L) are the initial dye concentration and concentration at time t, respectively.

$$q_e = \frac{(C_o - C_t)V}{W} \qquad (2)$$

where q_e (mg/g) is the equilibrium adsorption capacity, C_e is the dye concentration at equilibrium, V (L) is the volume of solution and W (g) is the weight of adsorbent.

Results

3.1. Effect of initial pH

H^+ ions have a high potential in absorption to result is influenced pH of ambient on the rate possess and extent of absorption and for any particular system to be tested, determine the best conditions in terms of pH. Results of color tests to determine the pH optimum under certain conditions (concentration of both colors 50 mg/L, contact time of 30 minutes and adsorbent dose 1 g/L) is shown in Fig.1. It can be seen that with increase in pH from 2 to 10 color removal of Reactive Red 198 from 75.21 to 67.24% and the dye Reactive Blue 19 reduced from about 69.17 to 50.85%. In other words, the amount of dye removal efficiency is

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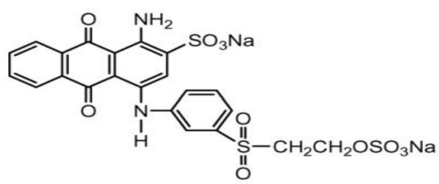
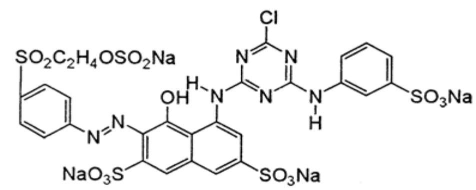
Table 1) Some features of RR-198 and RB-19 dye (22)

Features	Reactive Blue 19(RB-19)	Reactive Red 198(RR-198)
Chemical structure		
Molecular formula	$C_{22}H_{16}O_{11}N_2S_3Na_2$	$C_{27}H_{18}ClN_7Na_4O_{15}S_5$
The scientific name of dye	Reactive Blue 19	Reactive Red 198
Molecular weight	626.5g/mol	967.5 g/mol
Solubility in Water	70 (gL^{-1})	75(gL^{-1})
λ_{max}	592 nm	520 nm

higher at acidic pH. It also reduces the amount of adsorbent capacity increases for both colors. The optimum pH was adjusted to 4 times the next steps.

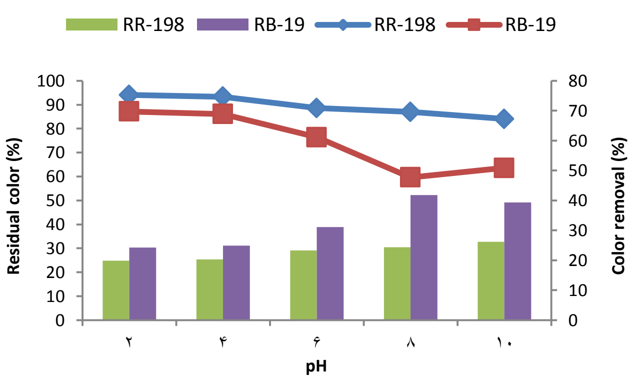


Figure 1) Effect of pH on adsorption of the dye by Acaciatortilis plant shuck. Conditions: 50mg/L concentration of dye, 30 minutes contact time, 1g/L adsorbent dosage

3.2. Effect of adsorbent dose

Fig.2. shows the results of experiments to study the effect of adsorbent dose. That is observed for both dyes with increasing adsorbent dose From 0.5 g/L to 2, in the dye concentration 50 mg/L, increased absorption but absorption rate has dropped with the increase of adsorbent dose 2-4 g/L.

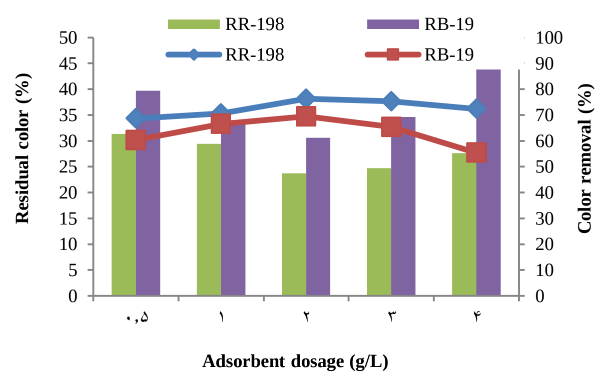


Figure 2) Effect of adsorbent dosage on adsorption of the dye by Acaciatortilis plant shuck. Conditions: 50 mg/L Concentration of dye, 30 minutes contact time, pH 4

3.3. Effect of contact time and dye concentration

Contact time in the adsorption process is a critical variable So that the Removal efficiency and absorption are linked with contact time**.**the stage was examined in certain circumstances, a different mesh between 30-60 and 60-100, pH

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equal to 4, adsorbent dosage 2 g/L in the amounts of colors10, 50 and 500 mg/L and contacts times 10, 20, 40 and 80 minutes. The results of the test are shown in Fig.3 and 4. Results indicate that in mesh 30-60 and 60-100, color of concentration 10 mg/L, with time passage dropped from 10 to 80 minutes. In result color removal efficiency increases, but the concentration of the dyes 50 and 500 mg/L in the first times to the border 20 minutes, the color removal efficiency is rising that with time passage from 20 minutes to 80 minutes reversed and reduced remove the colors. As reduction efficiency is observed more in dye concentration 500 mg/L.

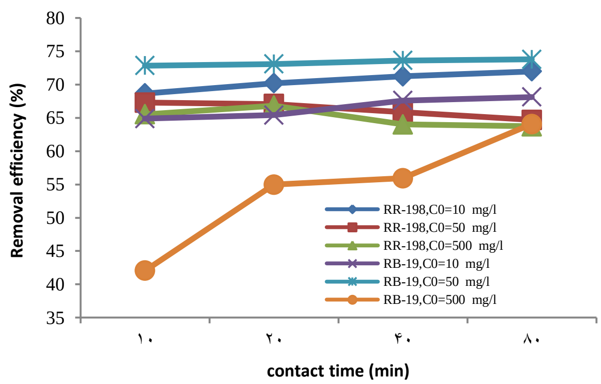

Figure 3) Effect of contact time and initial dye concentration on adsorption of the dye by Acaciatortilis plant shuck. Conditions: pH 4, 2 g/L adsorbent dosage, mesh size 30-60

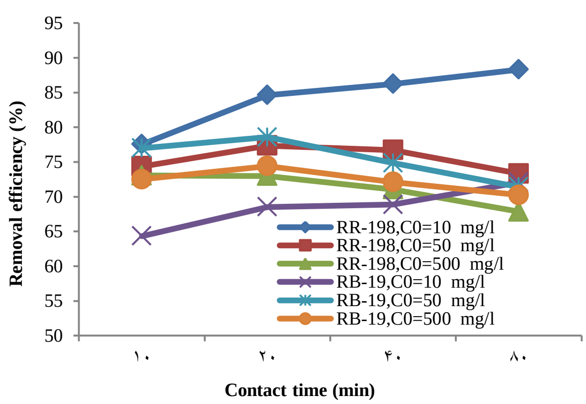

Figure 4) Effect of contact time and initial dye concentration on adsorption of the dye by Acaciatortilis plant shuck. Conditions: pH 4, 2 g/L adsorbent dosage, mesh size 60-100

4.3. Effect of adsorbent size

Sorbent particle size has an important influence on synthetic adsorption. Results of contact time with the adsorbent particles range in size from 30 to 60 and 60 to 100 were tested. Observe as the particle size decreases the amount of adsorption increases. This happens when the mass of adsorbent is stable due to larger external surface area available on the smaller particles. It is evident from Fig. 3 and 4 with different concentrations of dye uptake of both sorbent particle size decreased from 30-60 to 60-100, 10% had increased.

5.3. Adsorption isotherm

Among the important features that should be considered in studies of the adsorption of pollutants onto the absorber, the absorber adsorption isotherm is used. In this study the experimental isotherm data fit the Langmuir and Freundlich models and their relationships are given in equations 3 and 4 is used (15). Data consistent with regression coefficients of isotherm models, model selection is one of the best ways is absorbed. Table 1 show that The values of correlation coefficients is observed, the test results of both color RR-198 and RB-19, respectively, with correlation coefficients of 0.84 and 0.81 for the mesh to mesh 100-60 and 60-30 respectively 0.83 and 0.82, Freundlich models are a good fit (Fig. 5 to 8). Therefore, the isothermal data fitted the Freundlich model both types of dye.

$$\frac{C_e}{q_e} = \frac{1}{K_l\,Q_m} + \frac{C_e}{Q_m} \qquad (3)$$

$$\ln q_e = \ln K_f + \frac{1}{n}\,C_e \qquad (4)$$

where K_L is the Langmuir adsorption constant (l/mg) and Q_m is the theoretical maximum adsorption capacity (mg/g), K_f (l/mg) and n are isotherm constants indicate the capacity and intensity of the adsorption, respectively. Also the Langmuir constant (K_L) can be used to

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determine applicability of the adsorbent for the removal of RR19 and RB19 by using R_L dimensionless as follows (25):

$$R_l = \frac{1}{(1 + K_l\, C_o)} \qquad (5)$$

If the R_L values are between 0 and 1, the adsorption is favorable, the R_L values is larger than 1, the adsorption is unfavourable, the R_L values is equal with 1, the adsorption is linear and the R_L values is equal with 0, the adsorption is irreversible (25). The calculated value of R_L was found to be 0.047- 0.185 at 22 ± 20°c from Eq. (5) and is given in Table 2. The result indicates that the adsorption of RR198 and RB19 ions onto Acaciatortilis plant shuck is favorable.

Table 2) Freundlich and Langmuir is other models parameters at 22 ± 2°c

		Langmuir isotherm				Freundlich isotherm		
Type color	**Mesh size**	**R_L**	**R^2**	**Q_m (l/mg)**	**K_L (l/g)**	**R^2**	**1/n**	**K_f**
RR-198	30-60	0.185	0.81	20.49	0.088	0.84	0.40	2.34
RB-19	30-60	0.065	0.76	15.72	0.286	0.81	0.41	2.56
RR-198	60-100	0.108	0.8	19.46	0.164	0.83	0.50	3.96
RB-19	60-100	0.047	0.76	11.30	0.401	0.82	0.41	3.24

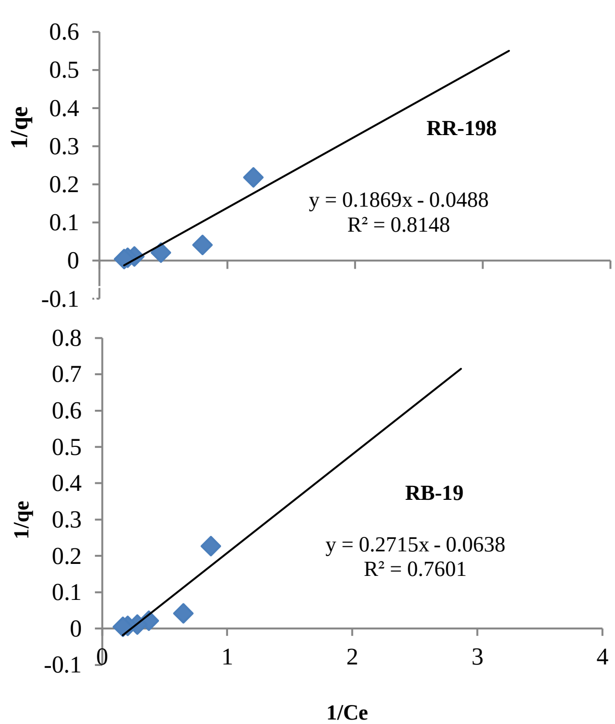

Figure 5) Langmuir adsorption isotherms of RR198 and RB19 on Acaciatortilis plant shuck to mesh size 30-60

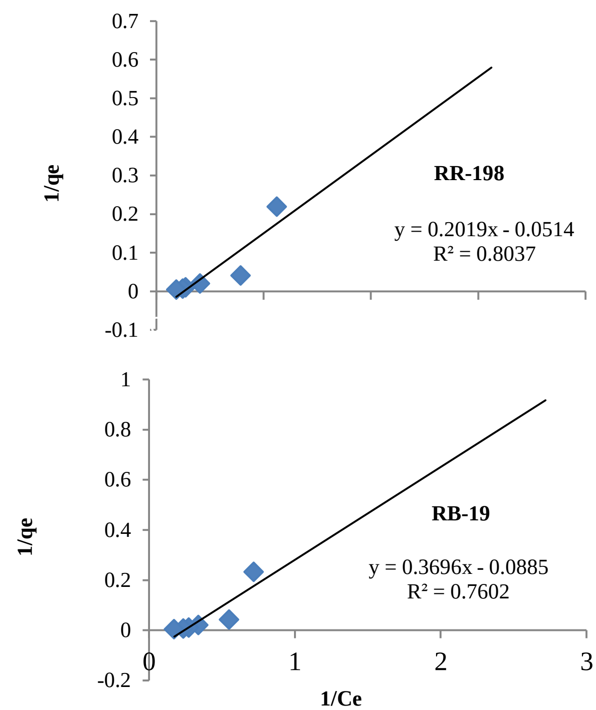

Figure 6) Langmuir adsorption isotherms of RR198 and RB19 on Acaciatortilis plant shuck to mesh size 60-100

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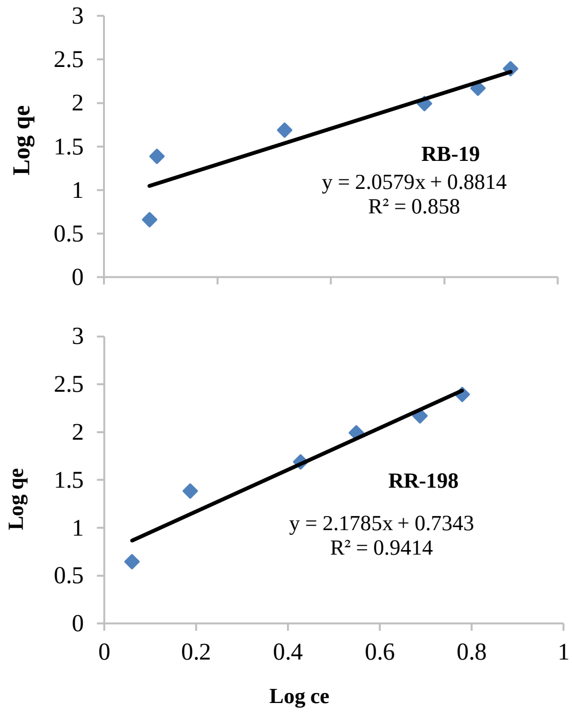

Figure 7) Freundlich adsorption isotherms of RR198 and RB19 on Acaciatortilis plant shuck to mesh size 30-60

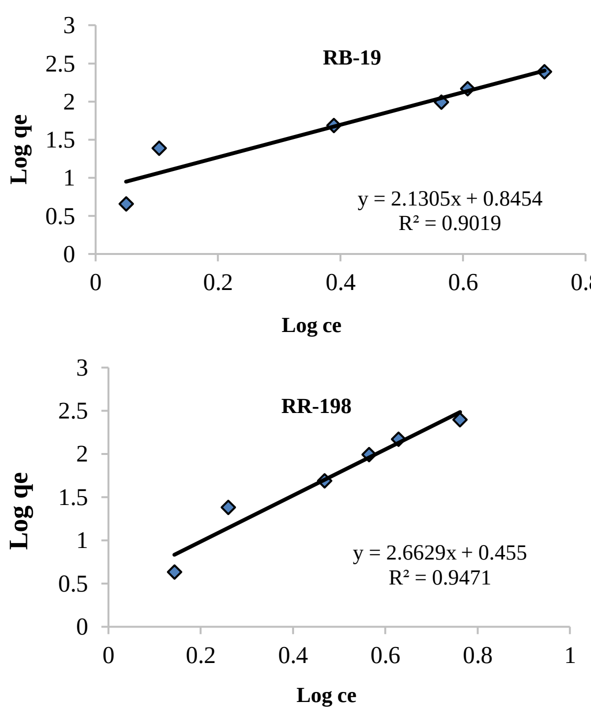

Figure 8) Freundlich adsorption isotherms of RR198 and RB19 on Acaciatortilis plant shuck to mesh size 60-100

Discussion

As seen in Fig.1. the pH of the solution plays an important role in the system, And with increase in pH from 2 to 10 color removal of reactive Red 198 from 21.75 to 24.67% and the dye Reactive Blue 19 from about 17.69 to 85.50% reduced. Reduction in both color removal efficiency increases or decreases due to changes in pH levels decreased H^+ and OH^- ions and positive ions on the adsorbent surface increases described (22,23). With decreasing pH of increase places with a positive charge will so these are places due to electrostatic attraction tends to absorb color (RR-198 and RB-19). These results are in line with the results of the Shu el al study about the removal of Black1dye using UV/H_2O_2 process, Powell et al study about of anionic and cationic dyes adsorb on the ash and Sariglu et al study about removal of methylene blue by volatile biosolids (24,25,26). In another study Amin about removal of reactive dye onto activated carbons prepared from sugarcane bagasse pith and Santhy et al about Removal of reactive dyes from wastewater by adsorption on coir pith activated carbon Similar observations have been reported indicating that the carbon has a net positive charge on its surface and with increasing pH the decreased dye adsorption capacity. As the pH of the adsorption solution was lowered, the positive charges on the surface increased. This would attract the negatively charged functional groups located on the reactive dyes (27,28). Impact studies of particle size, information important for achieving efficiency absorbing and provides maximum absorption. As expected, the amount of surface area available for adsorption on a solid surface with a constant mass of adsorbent particle size, are different. Adsorption capacity is directly related to the total level of exposure and to non-porous adsorbent with a particle diameter has an inverse relationship. The presence of a large number of smaller particles

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with larger surface area available for adsorption systems provides for the removal of dyes (29-31). The amount of dye adsorbed increased 10% for a decrease in adsorbent particle size from 30-60 to 60-100. The higher adsorption with smaller adsorbate particles could be attributed to the fact that smaller particles provided a larger surface area. The result showed that there was a gradual increase of adsorption with the decrease in particle size. These results are similar to results obtained by Gong et al in a study about removal of cationic dyes from aqueous solutions by adsorption onto peanut shell, that the particle size of the adsorbent as an important parameter in the adsorption of dyes studied (32). Effect of initial dye concentration showed that the color removal efficiency decreases with increasing the color value of 10 to 500 mg/L. Thus it can be said that for a specified amount of an adsorbent, adsorption sites is constant, and the available active sites on the adsorbent surface further reduced over time. Thereby increasing the amount of dye،adsorption is decreased and as well as increasing the dye concentration can increase the repulsive forces between the dye molecules, and the repulsion caused by adsorption onto the adsorbent is prevented (33). In a study conducted by Entezari and et al as Fast and efficient removal of Reactive Black 5 from aqueous solution by a combined method of ultrasound and sorption process, has been shown that by increasing dye concentration, the removal efficiency decreases due to the number of fixed positions over a specified amount of adsorbent is cited (34,35). In another study, Wang and et al shown that the absorption basic dyes on zeolite, color removal efficiency decreases with increasing concentration of color (36).

Equilibrium data, commonly known as adsorption isotherms, are basic requirements for the design of adsorption systems. In order to discover the adsorption capacity of Acaciatortilis Plant shuck, the experimental data points were fitted to the Langmuir and Freundlich isotherm equations and the constant parameters of the isotherm equations were calculated. The Langmuir equation is valid for monolayer adsorption onto a completely homogenous surface with a finite number of identical sites and with negligible interaction between adsorbed molecules (37). Fig. 5 and 6 shows the Langmuir plots for adsorption of RR198 and RB19 at different mesh size. The value of Q_m and K_L constants and the correlation coefficients for Langmuir isotherm are presented in Table 2. The isotherms of all adsorbents were found to be linear over the whole concentration range studies and the correlation coefficients were high (R^2 > 0.76) as shown in Table 2. The values of Q_m decrease with mesh size increase (38). The Freundlich isotherm is derived by assuming a heterogeneous surface with a non-uniform distribution of heat of sorption over the surface (39,40). The linear plot at each mesh size indicates that adsorption of RR198 and RB19 dye also follows Freundlich isotherm (Fig.7 and 8). Table 2 shows the Freundlich adsorption isotherm constant and correlation coefficients. The value of 1/n for Freundlich isotherm was found to lie between zero and one, indicating that RR198 and RB19 dye is favorably adsorbed by Acaciatortilis Plant shuck at all meshes size studied. The results of isotherm studies showed that Freundlich isotherm equation is at the relatively high correlation coefficient. Thus we can say that the removal of RR-198 and RB-19 dye follows the equation of Freundlich isotherm. In a study conducted by Entezari and et al Maximum absorption is achieved equal 3.91 mg/g and obeys the Freundlich model (33).

In this study the adsorption of Reactive Red 198 and Reactive Blue 19 by Acaciatortilis shuck has been investigated. This study examined the conditions affecting the adsorption process by shell Acaciatortillis, and was determined that the most effective way of

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changing the color removal was achieved. The experimental results showed that adsorbent prepared from Acaciatortilis shuck was a suitable adsorbent for removal of reactive dyes from synthetic aqueous solution. In batch studies, the adsorption increased with an increase in solute concentration and adsorbent dose in 0.5 to 2 g/L. Dye solution was decolorized in relatively short time (10 min). Removal of dyes was higher at the acidic pH range. Also the results of study showed that the absorption follows the Freundlich model and shows a multi-layer adsorption and could be economically feasible than conventional methods. However, more research should be done for practical scale. According to R_L of adsorbent that was prepared, Acaciatortillis as a natural absorbent ideal for Reactive Red 198 and Reactive Blue 19 dye was determined. It can be concluded that this adsorbent can be used as an effective agent for the removal of reactive dyes from textile and dyeing wastewater.

Footnotes

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