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Utilization of Olive Kernel Ash in Removal of RB19 from Synthetic Textile Wastewater

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Abstract

The colored wastewater of textile industries has toxic and stable material in the environment. Several methods for removal of synthetic dyes was investigated that adsorption is most effective between these methods. This study was performed experimentally and in laboratory scale. The adsorption capacity of dye is related to pH of solutions, initial dye concentration, adsorbent dose and contact time. The concentrations of dye were determined by UV-Vis spectrophotometer at 592nm. To analyze the equilibrium data was used from the Langmuir, Freundlich and Temkin isotherms models and pseudo-first-order, pseudo-second-order and Elovich kinetics models. The results indicated that the increase in adsorbent dose led to increasing of the removal efficiency. According to the results, adsorption efficiency was increased with decreasing of pH and increase in reaction time. Increasing of initial dye concentration from 10 to 50 mg/L in present of 0.5g/100mL of adsorbent, pH=4 and at contact time of 180 min led to decrease of removal efficiency from 96% to 93.7%. In addition, the Langmuir isotherm model had good fit with obtained results. The adsorption kinetics followed the pseudo-second-order model. The results show that the olive kernel ash is a natural and cheap adsorbent that can be used for the removal of reactive blue 19 dye from textile wastewater. In addition we can utilize of this sorbent for other organic pollutants.

Key words: Desertification, surface adsorption, olive kernel ash, reactive blue 19 dye, isotherms, kinetics, textile wastewater.

1 Introduction

The dyes are a group of complex organic materials that resulted of textile industry different stages [1]. According to a survey conducted in 2009, the dye production and consumption in the world is seven hundred thousand tons [2, 3]. The colors used in textile industry divided to anionic (acidic, direct, reactive), cationic (all basic dyes) and nonionic (dispersive dyes) groups [4]. The discharge of colored wastewaters from textile industry performance into the receiving waters caused the reduction of light penetration and vision, eutrophication and reduction of photosynthesis process in aquatic plants and algae and finally affects on environment [5, 6]. These wastewaters have carcinogenic and mutagenic properties and can cause allergies and skin problems [7, 8]. To reduce colored effluents discharged into watercourses, the government of Taiwan adopted the Effluent True Color Standard in 1998. The true color discharge limit is 400 American Dye Manufactures Institute (ADMI) units. The adopted analytical method is the ADMI Tristimulus Filter Method (3 wavelength (WL) method), and the 31 WL ADMI method might be also adopted as an alternative for color value measurement [9].

A variety of technologies including adsorption, chemical reduction, precipitation, coagulation, enzymatic oxidation, microbial degradation, photochemical degradation, ozonization, rivers osmosis, ion exchange, and membrane processes have been studied for the dye remediation process [10-14]. Nevertheless, additional research and broader validation are still needed to solve this effluent problem for field application with cost effective techniques that are also environmental friendly. Nowadays, adsorption process has been proven as one of the best treatment technologies for the hazardous compound removal from aqueous solution [15, 16]. In addition, the regeneration of the adsorbent may be possible with economical operation, because adsorption is generally reversible. Activated carbon is the most widely used adsorbent for removal of dyes; however it is little expensive for the field application. Therefore, environmental

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researchers are looking for new and low-cost adsorbents for field application with cost-effective.

Due to improper performance of the treatment units and the nature of dyes, about 50% of reactive dyes, 8-20% of dispersive dyes and 1% of the pigment dyes may be lost directly into wastewater stream [17]. Reactive dyes there are in a hydrolyzed state in the effluent of dye bath or wash water, a form that cannot be reused in the dyeing process. Due to the importance of this class of dyes, reactive blue 19 dye was chosen as model dye [18].

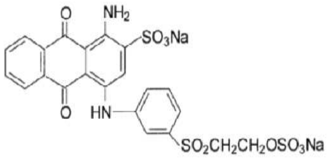
According to the international olive council, the annual production of olive oil in the world at the year 2012 was more than 3 million tons, translating to approximately 15 million tons of olive cakes as byproducts .The materials resulting from olive stones carbonization have been successfully used as sorbents for a wide variety of pollutants in aqueous solutions [19, 20]. The aim of this study is to evaluate the efficiency of olive kernel ash as a new sorbent for the removal of reactive blue 19 dye.

2 Materials and Methods

2.1 Materials

Reactive Blue 19 dye (RB19) was obtained from Dye Star Company. Other used materials were obtained from Merck Company. Properties and chemical structure of the RB19 dye have been presented in table 1[21].

Table 1. Basic properties of the reactive blue 19 dye

Chemical formula	$C_{22}H_{16}O_{11}N_2S_3Na_2$
Commercial name	Remazol Brilliant Blue R
Class	Azo
Molecular weight (g/mol)	626.5
λ_{max}	592
Molecular structure	

2.2 Preparation of biosorbent

Olive kernel were separated and then washed with distilled water to remove impurities and surface adhered particles and then dried in the sunlight. Washed Olive kernel transformed to ash by electric oven during 2 hours in temperature 700 °C. After changing to the ash, adsorbent was grinding and then pulverized by ASTM standard sieves with the range of 40 to 60 meshes (0.25- 0.4 mm particles).

2.3 Procedure and analysis

Batch adsorption studies were performed in the initial dye concentrations of 10, 25 and 50 mg/L, adsorbent dose of 0.05-1.5 g/100mL, contact times of 10-180 min and pH of 4-10. All experiments were run twice and a good reproducibility of the procedures was obtained. The pH of solutions was adjusted with NaOH and H_2SO_4 (0.1N) using pH meter Mi 151. The mixing of solution was carried out by mechanical shaker (INNOVA 40R, England) with 120 rpm at 20°C. After shaking, all solutions were filtered through 0.45 µm membrane filter paper (Sartorius, Germany) and the filtrate was analyzed. The residual concentrations of dye in the samples were detected by UV-Visible spectrophotometer (Optima SP-3000 Plus model, Japan) at 592 nm [22, 23]. The amount of adsorbed RB19 dye was calculated by the difference of the initial and residual amount in the solution. The adsorption capacity (q_e) of RB19 dye in mg/g at time t and the percentage of removed RB19 dye (%R) in solution were computed using Eq (1) and Eq (2), respectively:

$$q_e = \frac{(C_o - C_t)V}{M} \quad (1)$$

$$R\,(\%) = \frac{(C_0 - C_t)}{C_0} \times 100 \quad (2)$$

Where C_o and C_t are the dye concentrations in mg/L initially and at a given time t, respectively. V is the volume of the dye solutions in mL; M is the adsorbent dose in g [3, 4].

2.4 Isotherms models for the adsorption

The results also were evaluated by various adsorption isotherms including the Langmuir, Freundlich and Temkin isotherms. Langmuir equation is represented in the linear form as follows:

$$\frac{1}{q_e} = \frac{1}{q_{max} \times bC_e} + \frac{1}{q_{max}} \quad (3)$$

The essential characteristic of the Langmuir isotherm can be expressed by the dimensionless constant called separation coefficient (RL) and is calculated by the following equation:

$$RL = \frac{1}{1 + bC_0} \quad (4)$$

The value of separation factor (RL) indicates the nature of adsorption process. RL values indicate the type of isotherm to be irreversible (RL=0), favourable (0<RL<1), linear (RL=1) or unfavourable (RL>1) [21].

The Freundlich isotherm expressed by equation (5):

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \quad (5)$$

The 1/n values indicate the type of isotherm to be irreversible (1/n=0), favourable (0<1/n<1), and unfavourable (1/n>1) [21].

The Temkin isotherm equation is given as:

$$q_e = B\ lnA + B\ lnC_e \quad (6)$$

In these equations, q_e and q_{max} are the dye amount adsorbed per mass of adsorbent at equilibrium time and the maximum adsorption, respectively (mg/g). The equilibrium concentration is C_e (mg/L), b (L/mg) is Langmuir constant and K_f (mg/g) is related to the adsorption capacity and n (g/L) is related to the intensity of adsorption in the Freundlich isotherm model. In The Temkin isotherm equation the constant B is related to the heat of adsorption (kJ/mol) and A is the equilibrium binding constant (L/mg) corresponding to the maximum binding energy [22-24].

2.5 Kinetic models for the adsorption

The adsorption kinetics evaluated by the pseudo-first-order, pseudo-second-order and Elovich kinetic models. The pseudo-first-order reaction equation was used for the adsorption of liquid/solid system on the basis of solid capacity. The pseudo-first-order kinetic expressed by equation (7):

$$\frac{dq_t}{dt} = k_1 (q_e - q_t) \qquad (7)$$

Where q_e and q_t (mg/g) refer to the amount of dye adsorbed at equilibrium and at time t, respectively. By integrating of this equation, the linear relationship (8) is obtained:

$$ln\left(1 - \frac{q_t}{q_e}\right) = -k_1 t \qquad (8)$$

In the pseudo-second-order model, adsorption follows the second-order chemisorptions. The pseudo-second-order kinetic model given as equation (9):

$$\frac{dq_t}{dt} = k_2 (q_e - q_t)^2 \qquad (9)$$

By integrating the above equation, the linear relationship (10) is obtained:

$$\frac{t}{q_t} = \frac{1}{k_2\, q_e^2} + \frac{1}{q_e} t \qquad (10)$$

Elovich equation is one of the most useful models for describing activated chemisorptions. Elovich kinetic model can be expressed as follows:

$$q_t = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln t \qquad (11)$$

Where α is the initial adsorption rate in (mg/g.min), and β (g/mg) is the desorption constant related to the extent of the surface coverage and activation energy for chemisorptions [25-29].

3 Results

3.1 Adsorbent dose influence on RB19 dye removal

The removal of dye by olive kernel ash at different adsorbent doses (0.05-1.5 g/100 mL) for the dye concentrations 10, 25 and 50 mg/L at pH=7 is investigated (Figure 1). According to the results, with increase in the adsorbent dose, removal efficiency increased. So that by increasing of adsorbent dose from 0.05 to 1.5 g/100mL for dye concentration of 10 mg/L, dye removal reached from 53.6% to 98%. The removal efficiency for concentrations of 25 and 50 mg/L was reached from 45.3% to 95.27% and from 32.8% to 89.26%, respectively. Nevertheless, adsorption capacity decreased from 10.27 to 0.65 mg/g (C_0=10 mg/L), from 22.65 to 1.59 mg/g (C_0=25 mg/L) and from 32.8 to 2.98 mg/g (C_0=50 mg/L). Since that increasing of adsorbent dose from 0.5 to 1.5 g/100 mL had little effect on removal efficiency, so optimum dose for other experiments stages was considered 0.5 g/100 m

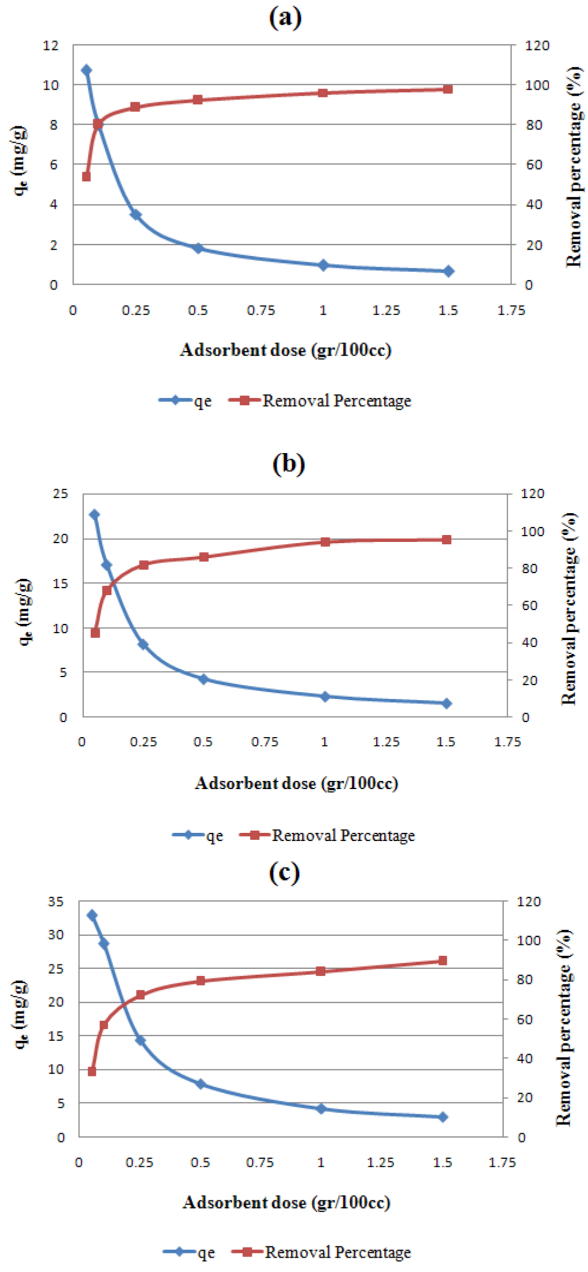


Figure 1. Effect of adsorbent dose on the adsorption capacity and removal percentage of RB19 dye (a: 10 mg/L, b: 25 mg/L, c: 50 mg/L)

3.2 pH influence on RB19 dye removal

The results of the pH effect (4 to 10) on reactive blue 19 dye removal by olive kernel ash, at 20 ̊C and concentrations of 10, 25 and 50 mg/L of RB19 dye, are shown in Figure 2. In this stage, adsorbent dose and contact time were 0.5 g/100mL and 24 h, respectively. The results of this study showed that as the pH of solution was decreased, the adsorption capacity (q_e) and adsorption efficiency increased.

By increasing of pH from 4 to 10, RB19 dye adsorption capacity (removal efficiency) decreased from 2 mg/g to 1.74 mg/g (from 100% to 87%), 4.8 mg/g to 3.8 mg/g (from 96% to 76%), 9.54 mg/g to 6.6 mg/g (from 95.4% to 66%) for dye concentrations of 10, 25 and 50 mg/L, respectively. The results showed that the maximum adsorption capacity of RB19 dye was at pH 4.

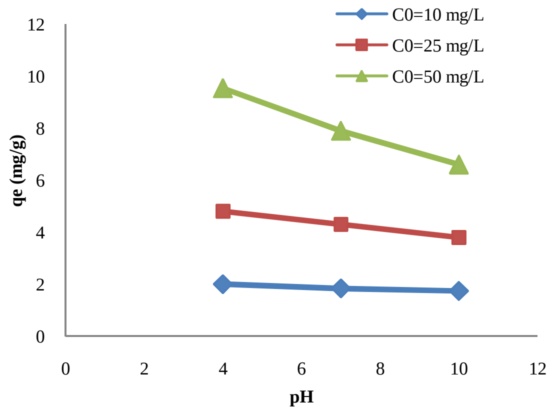


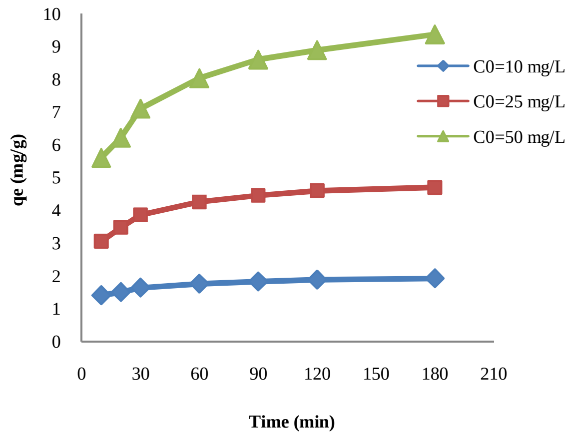
Figure 2. The effect of pH on the adsorption capacity of RB19 dye

3.3 Initial concentration and contact time influence on RB19 dye removal dye

To determination of effects of contact time and initial dye concentrations, contact times of 10, 20, 30, 60, 90,120 and 180 min were studied. In this stage, initial dye concentrations 10, 25 and 50 mg/L, pH =4 and adsorbent dose of 0.5 g/100mL were considered. The obtained results presented in Figure 3.

The results of this study showed that with increasing reaction time, efficiency of adsorption increased. The results show that removal efficiency for concentrations of 10, 25 and 50 mg/L, by increasing of contact time from 10 to 180 min were increased from 70.2% to 96%, from 61% to 94% and from 56% to 93.7%, respectively. According to presented results in Figure 3, the adsorption of reactive blue 19 dye by olive kernel ash at 180 min reached to equilibrium.

According to the results, adsorption capacity increased with increasing of dye initial concentration from 10 to 50 mg/L. adsorption capacity of dye for contact time of 180 min and initial dye concentrations of 10 and 50 mg/L were 1.92 mg/g and 9.37 mg/g, respectively.



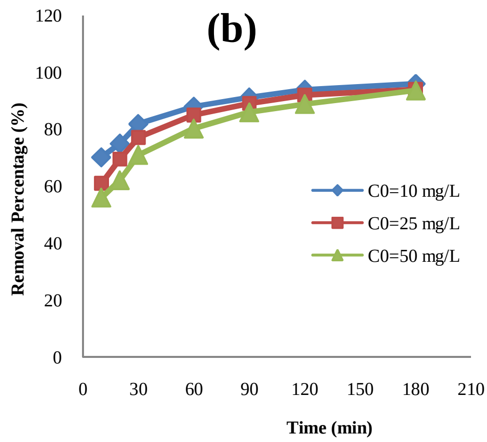


Figure 3. Effect of initial dye concentration and contact time on (a) adsorption capacity and (b) removal percentage of RB 19 dye

3.4 Adsorption isotherms of RB19 dye

The results also were evaluated by various adsorption isotherms including the Langmuir, Freundlich and Temkin isotherms (Figure 4 and Table 2).

According to the results, the data was not compliance with temkin isotherm model because of the correlation coefficients for this isotherm was not high. The absorption data from the point of view correlation coefficient rate had concordance to Langmuir and Freundlich isotherms, respectively. But according to the results that presented in table 2, values of 1/n for initial dye concentrations were higher than 1 that confirms the adsorption of reactive 19 dye onto olive kernel ash not concordance to Freundlich isotherm. The RL amounts in Langmuir isotherm for concentrations of 10 and 25 mg/L were 0.018 and 0.001 that are lower than 1 and shows the results fit with Langmuir isotherm mode.

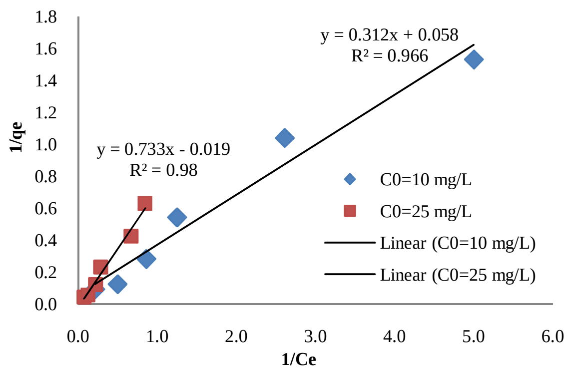


Figure 4. Langmuir isotherm model for RB19 adsorption on olive kernel ash

3.5 Adsorption kinetics of RB19 dye

Adsorption kinetics study also was carried out with the pseudo-first-order, pseudo-second-order and Elovich kinetics (Figure 5 and Table 3). The results showed that adsorption of dye onto olive kernel ash fitted with pseudo-second-order kinetic with the correlation coefficient (R^2) values higher than 0.99.

Table 2. Adsorption isotherms parameters of the reactive blue 19 dye

Isotherm models	Constants	Initial dye concentration (mg/L)	
		10	25
Langmuir	q_m (mg/g)	3.21	1.364
	b (L/mg)	5.379	38.579
	R_L	0.018	0.001
	R^2	0.966	0.98
Freundlich	K_f (mg/g)(L/mg)$^{1/n}$	2.841	1.363
	1/n	0.98	1.117
	R^2	0.962	0.975
Temkin	K_T (L/mg)	3.432	8.633
	B (mg/g)	4.525	2.203
	R^2	0.873	0.894

4 Discussion

4.1 Adsorbent dose influence on RB19 dye removal

Optimum adsorbent dose determined the amount of removed dye. According to results of present study by increasing adsorbent dose, removal efficiency increased. Because of the increased removal efficiency with increasing adsorbent dose is an increase in the adsorbent surface and more availability of adsorption sites [21]. The results indicated that increasing adsorbent dose led to decreasing of adsorption capacity. This reveals that the instantaneous and equilibrium sorption capacities of RB19 are functions of the olive kernel ash dosage [28]. Ghaneian et al. (2011) in their study on the efficacy of cuttlefish bone powder in the removal of reactive blue 19 dye concluded that by increasing adsorbent dose, the removal efficiency of dye increases. These researches were investigated adsorbent doses of 0.2-1 g/100ml. In their study, optimum adsorbent dose was determined to be 0.4g/100ml [21]. The adsorption of basic dye onto sepiolite, fly ash and apricot shell activated carbon was investigated by kargozoglu et al. (2007). These researchers' findings were consistent with the results of this study [7].

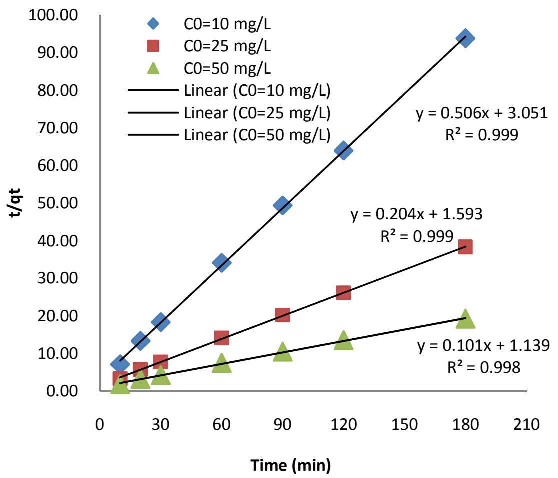


Figure 5. Pseudo-second-order kinetic for RB19 adsorption on olive kernel ash

4.2 pH influence on RB19 dye removal

The results indicated increasing adsorption capacity of RB19 dye with decreasing pH. The reason is the electrostatic attraction between negatively charged dye's anions and positively charged adsorbent's surface. At low pH, more protons are available, so electrostatic attraction between the dye molecules and adsorbent surface is increased and causes an increase in adsorption capacity [30]. The removal of reactive blue 19 dye using chitosan/oil palm ash composite beads cross-linked studied by Hasan et al. (2008). These researchers stated that dye adsorption at acidic conditions is very higher than neutral or alkaline conditions [31]. Bayramo˘glu et al. (2006) in their study about adsorption of reactive blue 4 dye using fungi concluded that the removal efficiency decreases with increasing pH. In this researcher's study the maximum dye biosorption was observed at pH= 3 [30].

Table 3. Adsorption kinetics parameters of the reactive blue 19 dye

Kinetic models	Constants	Initial dye concentration (mg/L)		
		10	25	50
Pseudo first -order	q_e (mg/g)	1.77	1.707	4.599
	K_1 (1/min)	0.011	0.016	0.017
	R^2	0.961	0.980	0.992
Pseudo second-order	q_e (mg/g)	1.976	4.902	9.901
	k_2(g/mg.min)	0.0731	0.0261	0.009
	R^2	0.999	0.999	0.998
Elovich	α (mg/g.min)	34.004	11.51	7.808
	β (g/mg)	5.348	1.695	0.734
	R^2	0.986	0.983	0.991

4.3 Initial concentration and contact time influence on RB19 dye removal dye

The results showed that with increasing initial dye concentration, adsorption capacity increases. Cause of the increased adsorption capacity with increasing initial dye concentration is probability collision and contact between adsorbent and adsorbate. With increasing initial concentration of reactive blue 19 dye, the removal efficiency decreased due to constant of the number of adsorption sites against increasing adsorbate molecules number which this result compliance with the findings of other researchers. The results of Hassan et al. (2008) study showed that by increasing initial RB19 dye concentration from 50 to 500 mg/L, the removal efficiency, decreased [31].

The obtained results indicated that the removal efficiency increases with time. According to the results, the adsorption was fast at the initial stages of the contact period, and then it became slower near the equilibrium because large number of vacant surface sites are available for adsorption during the initial stage but with lapse of contact time, the remaining vacant surface sites are difficult to be occupied due to repulsive forces between the solute molecules on the solid and bulk phases [32]. According to results presented in Figure 3, adsorption of reactive blue 19 dye by olive kernel ash reached equilibrium at 180 minutes. In Ehrampoush et al. (2011) study on application of eggshell as a biosorbent in the removal of reactive red 123 dye founded that with increasing contact time, adsorption capacity increased and adsorption equilibrium reached after 120 min [24].

4.4 Adsorption isotherms of RB19 dye

The isotherm models are the most important information which represents adsorption mechanism, surface properties and described adsorption experimental data [3, 31]. Data analysis for selection of the best isotherm model determined by linear regression analysis and comparison of the correlation coefficient (R^2) [24]. The results showed that the adsorption of reactive blue 19 dye by olive kernel ash follows Langmuir isotherm. According to the results of Moussavi et al. (2009) study, adsorption of reactive blue 19 and reactive red 198 dye by MgO nanoparticles followed Langmuir isotherm (R^2>99) [3]. Similar results by Xue et al. (2009) also have been reported [33]. Tsai et al. (2009) in their study on the removal of methylene blue dye by waste aquacultural shell powders concluded that adsorption data are in accordance with Langmuir and Freundlich adsorption isotherm [34].

4.5 Adsorption kinetics of RB19 dye

Kinetics models are suggested for clarify of absorption mechanism and evaluation of adsorbent performance that depends on adsorbent physical and chemical properties and mass transfer process. According to results of the present study, adsorption data fit better with the pseudo-second order kinetic. Hasan et al. in their study on adsorption of reactive blue 19 dye using cross-linked chitosan/oil palm ash composite beads founded that the dye adsorption kinetic had better concordance with pseudo-second order kinetic with the maximum correlation coefficient [31]. Moussavi and Mahmoudi (2009) also achieved the same results [3].

5 Conclusions

Textile industries wastewater containing significant amounts of color that are often toxic, resistant to biodegradation and persistent in the environment; Thus eliminating color should be considered. The present study results showed that increase in pH and initial concentration of reactive blue 19 dye caused removal efficiency decreased. While the increase in adsorbent dose and contact time led to increase of removal efficiency. Langmuir adsorption isotherm had a better match with adsorption data. The results showed that the data fitted the pseudo-second-order kinetic. According to the results, olive kernel ash is an appropriate and effective adsorbent for the removal of RB19 dye. The ability of Olive kernel ash as adsorbent in the removal of other pollutants should be investigated. If this adsorbent can remove dye as an indicator organic pollutant, so will be able to removal of other organic contaminants.

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