Efficiency of Pistachio Tree Sawdust in Removing Reactive Red 11 from Contaminated Aquatic Environment

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Abstract

Introduction: Dyes are among the hazardous pollutants that are often found in fabric wastewater. These pollutants have a significant environmental detrimental effect on the ecosystem of aquatic plants and animals. They are also likely to alter the physical and chemical properties of water. Therefore, in present study, it was attempted to investigate the effect of pistachio tree sawdust on removing reactive red 11 (RR-11).

Materials and methods: The present study was conducted experimentally and on a laboratory scale. The impact of effective parameters such as initial dye concentration RR-11 (20-100 mg/L), pH of the solution (4-11), adsorbent amount (0.1- 0.6 g/L) and reaction time (5-100 min) were investigated in RR-11 dye removal efficiency using pistachio sawdust. Finally, isotherms and adsorption kinetics were examined and analyzed as well.

Results: The results have indicated that under optimal conditions including initial dye concentration of 40 mg/l, the solution of the pH being equal to 4, pistachio sawdust content of 0.6 g/l, and reaction time of 60 minutes, a removal efficiency of 99.2% is obtained. The adsorption process using sawdust showed a better agreement with second-order kinetics (R^2 = 0.9934) and Langmuir isotherm (R^2 = 0.9906); the maximum adsorption capacity was 52.7 mg/g.

Conclusion: According to the obtained results, pistachio sawdust can be used as a suitable adsorbent; it can replace the common expensive and costly adsorbents.

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1. Introduction

Textile industry is one of the largest consumers of water and producers of significant amounts of effluent. These effluents contain significant amounts of colored effluents of varying chemical quantity and quality. Annually, more than ten thousand types of synthetic dyes are produced in different parts of the world, widely used in various industries. The amount of dye production in the world is estimated to be 700000 to one million tons per year, which is used in various industries including cosmetics, leather, paper and textile industries [1,2]. Therefore, since about 40-50 million tons of textiles are produced annually in the world, the amount of effluent produced by the textile industry is estimated to be 4-10 billion cubic meters. The textile industry is one of the leading industries in terms of production diversity in raw materials and pollution load [3-5]. Azo dyes are the largest group of synthetic organic dyes; more than 2,000 azo compounds have been listed. Among azo dyes, given their high water solubility and low degradability, reactive dyes are known as the most problematic compounds in textile industry effluents [6, 7].

It is estimated that about 50% of the reactive dyes used in industry are converted into wastewater and its concentration in industrial effluent is about 10-200 mg/L. Discharging effluents containing pigments to aquatic environments such as lakes and rivers results in reduce light transmission, reduce dissolved oxygen, increased chemical oxygen required, and thereby disrupts the aquatic life. In addition, due to their high thermal and optical stability, dyes can remain in the environment for relatively long periods of time [8, 9]. Given their complex structure, dyes are often stable and resistant to biodegradation, which are often toxic; from a hygienic point of view, dyes have carcinogenic and mutagenic properties and can also cause allergies and skin problems. The presence of dye contaminants, even in amounts less than one milligram per liter, are visible and important in appearance [10-14]. Given the high volume of production of colored wastewater in the country and their adverse effects, it is necessary to develop and adopt appropriate scientific measures for the treatment of such wastewater. Physical, chemical, biological or combined methods and processes can be applied to remove and treat dyes from aqueous solutions. Given the stability of dyes against biodegradation, physical and chemical methods such as coagulation-clotting, adsorption, chemical oxidation, advanced oxidation and membrane processes are commonly used to remove them. Adsorption is one of the efficient methods in removing dye from textile wastewater where various adsorbents such as activated carbon, sawdust, chitosan, etc. are applied. Given its brightness, ease of use, optimal stability while being washed, and low energy consumption, reactive red 11 (RR-11) is among the most important factors that have made this dye widely used in the textile industry. Ghorbaniant et al (2010) attempted to remove methylene blue dye in a study using pistachio sawdust. In their study, they determined the maximum removal efficiency under optimized conditions including the initial dye concentration of 50 mg/L, the pH of the solution being equal to 8, the amount of adsorbent being equal to 0.14 g/L and the reaction time of 100 minutes, an efficiency of 99.7% was obtained. The study of these researchers showed that the adsorption process using pistachio sawdust can be used as a high efficiency economic adsorbent in textile wastewater treatment [5]. In a study conducted in 2021, Akbarpour et al used active carbon prepared from hard pistachio shells for removing nitrate from contaminated water. Their study showed that under optimized conditions, including initial nitrate concentration 50 mg/L^{-1}, the amount of adsorbent being equal to 0.2 mg/L, pH of 5, and a reaction time of 30 minutes, the carbon is capable of removing as much as 99.4% of the nitrate. They have also concluded that using activated carbon prepared from pistachio shells, in addition to proper management of pistachio shells, has significant efficiency and effectiveness in removing contaminants [16].

RR-11 is one of azo dyes and reactive dyes. Therefore, the present study aims at using garden waste (pistachio sawdust) as an adsorbent and to remove RR-11 dye, being one of the most widely used dyes in the textile industry. In this study, in addition to the characteristics of pistachio sawdust, it has been attempted to investigate the effect of various operational parameters such as initial dye concentration, solution pH, adsorbent amount, and reaction time, as well as adsorption kinetics and isotherms as well.

2. Materials and methods

The present study was conducted experimentally on a discontinuous scale to investigate the effect of initial dye concentration (20-100 mg/L), solution pH (4-11), adsorbent amount (0.1-0.6 mg/L) and reaction time (5-100 min) on the efficiency of RR-11 dye removal. All chemicals used in the study were purchased with a laboratory degree and with high purity from the representative of Merck Group (Germany). Pistachio tree branches were used to prepare sawdust as an adsorbent. The branches were collected and then completely dried in the open air and ground in the laboratory. They were then granulated and meshed using ASTM standard sieves with a mesh of 50 (300 microns). Then, to remove possible contaminants, the sawdust of the previous stage was boiled with 10% hydrochloric acid for half an hour, and then to remove the remaining acid, it was washed with distilled water, so that the silver nitrate test will be negative. Then it was dried in an oven for 24 hours at a temperature of 60-80 degree Celsius; the prepared adsorbent was then kept in a closed container to prevent contamination; This adsorbent was used for adsorption experiments. For the experiments, 250 ml Erlenmeyer flakes containing 100 ml of useful volume were used as the reactor. The amount of dye residue was measured at 522 wavelength using the UV/Vis spectrophotometer (Optizen-3220-Korea). The pH of the sample was measured and adjusted using the pH meter (Metrohm 827 pH lab-Switzerland). The temperature of the reaction medium was considered equal to 25 ° C in all experiments, and the mixing speed was 150 rpm. Finally, the removal efficiency was calculated using the following equation [17, 18].

$$(1) \quad \%\text{Removal} = \frac{C_0 - C_t}{C_0} \times 100$$

3. Results

Absorbent characteristics

The images obtained from the FE-SEM analysis of pistachio sawdust are shown in Figure 1. The structure of sawdust, as seen in the pictures, has different structures and shapes with uneven and rough surfaces.

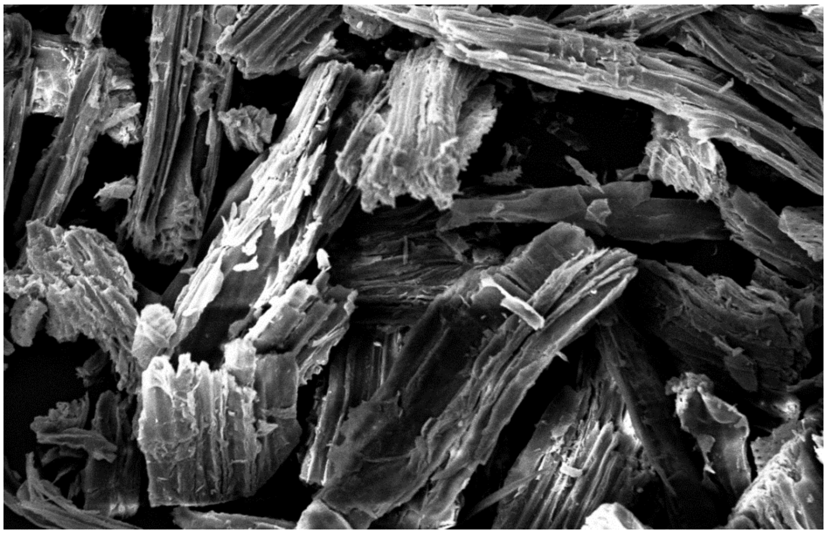

Figure 1. FE-SEM image of pistachio tree sawdust.

Effect of solution pH

To determine the effect of pH on the rate of RR-11 dye removal by pistachio sawdust, pHs of 4, 6, 7.5, 9 and 11 at an initial dye concentration of 40 mg/L and an adsorbent of 0.25 g/L were investigated and analyzed. The results were calculated in terms of removal efficiency. Figure 2 shows the effect of solution pH on dye removal efficiency.

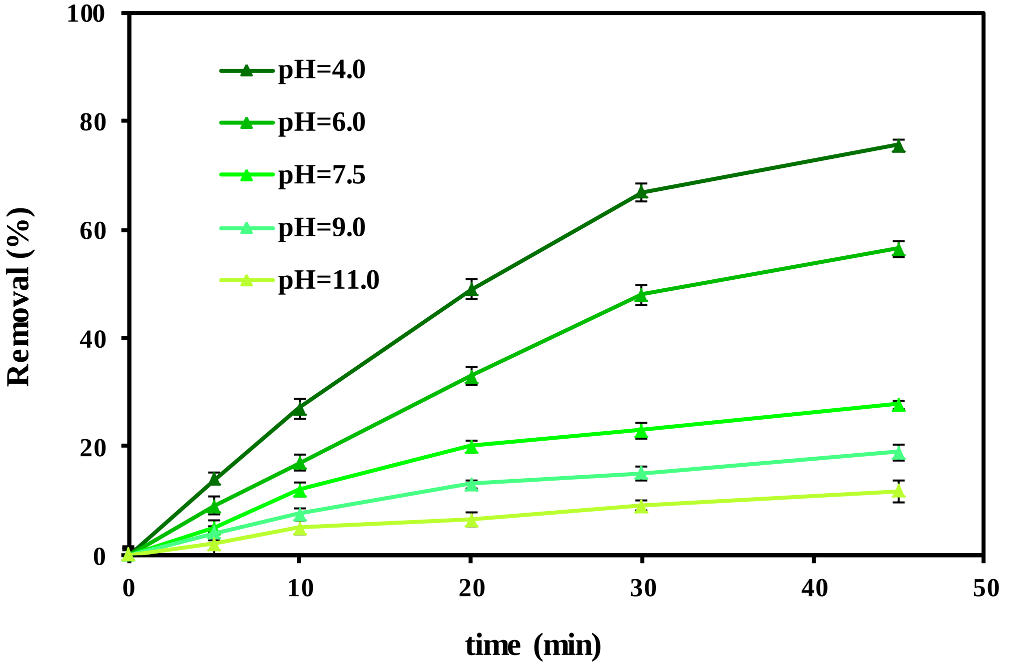

Figure 2. Effect of solution pH on RR-11 removal (initial dye concentration 40 mg/L and adsorbent amount of 0.25 g/L)

As shown in Figure 2, as the pH of the solution increases, the removal efficiency decreases so that the maximum removal efficiencies at pHs 4, 6, 7.5, 9 and 11 are 75.6 and 56.4, 27.8, 18.9 and 11.7%, respectively. Given the results, the maximum removal efficiency was obtained at pH being equal to 4; this pH is introduced as the optimal pH, and in the rest of the study, pH value was considered to be constant i.e. 4.

Effect of initial dye concentration

To determine the effect of initial RR-11 dye concentration on removal efficiency by pistachio sawdust, concentrations 20-100 mg/L of the dye were investigated. The pH of the samples was constant, being equal to the value optimized in the previous phase (pH = 4), and the specified amount of adsorbent was equal to 25 g/L. Sampling was conducted at specified intervals and in the range of 5 to 60 minutes, and finally the removal efficiency was calculated, the data for which are presented in Figure 3.

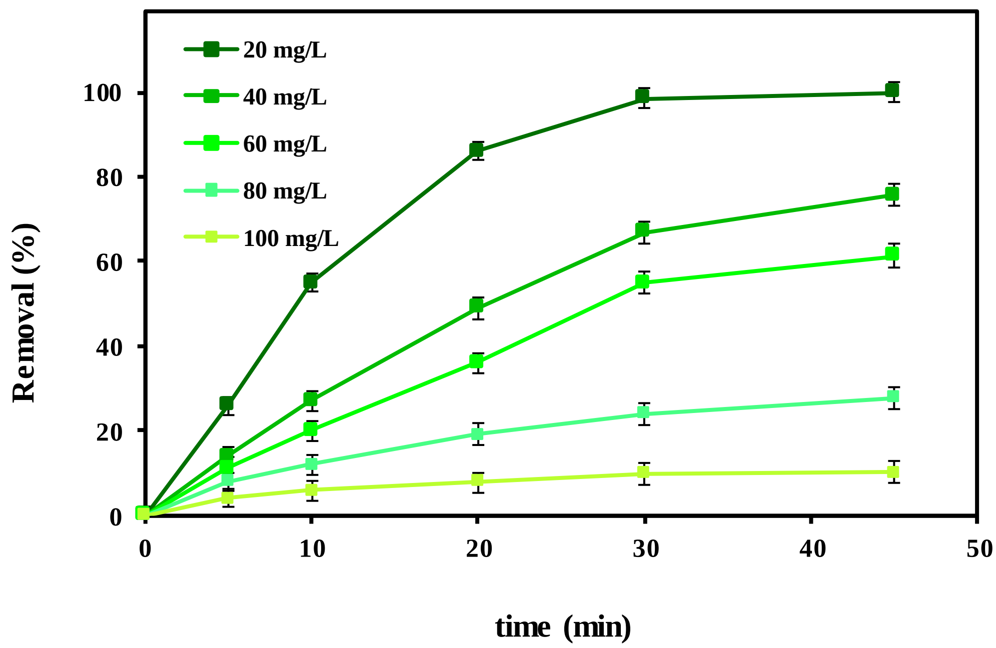

Figure 3. Effect of initial concentration on RR-11 removal (pH of 4 and adsorbent of 0.25 g/L)

As Figure 3 shows, with increasing the initial concentration of RR-11 dye, the removal efficiency decreases so that at concentrations of 20, 40, 60, 80 and 100 mg/L, the removal efficiency is 100%, 75.6%, 61.3%, 27.8% and 10.2%, respectively. In other words, the removal efficiency decreases as the RR-11 dye concentration increases. Given the results, the maximum removal efficiency was obtained at a concentration of 20 mg/L, which is equal to 100%. Thus, to determine the effect of other parameters, a concentration of 40 mg/L was introduced as the optimal concentration.

Effect of adsorbent amount (pistachio sawdust)

The results of the effect of the amount of adsorbent are shown in Figure 4. As shown in Figure 4, with increasing the amount of pistachio sawdust as an adsorbent, the removal efficiency increases, so that by increasing the sawdust in different amounts including 0.10, 0.20, 0.35, 0.45, and 0.60 g/L, removal efficiency showed an ascending trend, being 52.1%, 70.6%, 68.59%, 83.5%, 95.8% and 99.1%. Thus, according to the obtained results, a concentration of 0.6 is introduced as the optimal adsorbent concentration.

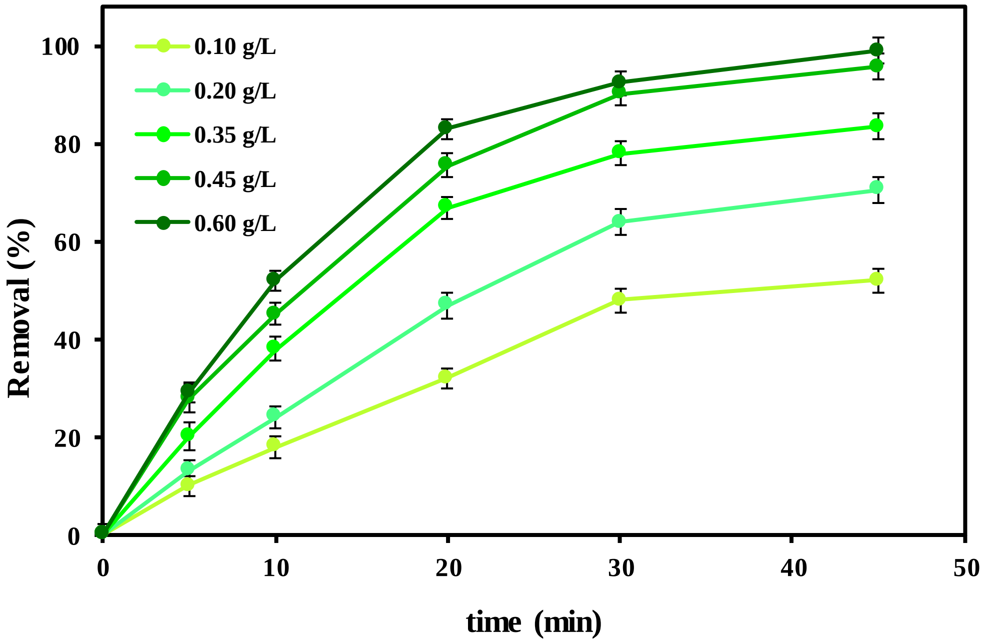

Figure 4. Determining the effect of adsorbent amount on RR-11 removal (pH 4 and initial dye concentration of 40 mg/L)

Effect of reaction time

The results of the reaction time effect are shown in Figure 5. As shown in Figure 5, with increasing reaction time, the removal efficiency increases so that at reaction times 5, 10, 20, 30, 45, 60 and 100 minutes, the removal efficiency adopts an ascending trend, being equal to 27.8%, 45.2%, 75.6%, 90.3%, 95.8%, 99.2% and 99.4%, respectively. This shows that pistachio sawdust has an efficiency of more than 99% and reaches equilibrium in 60 minutes. Thus, the equilibrium time was introduced to be 60 minutes.

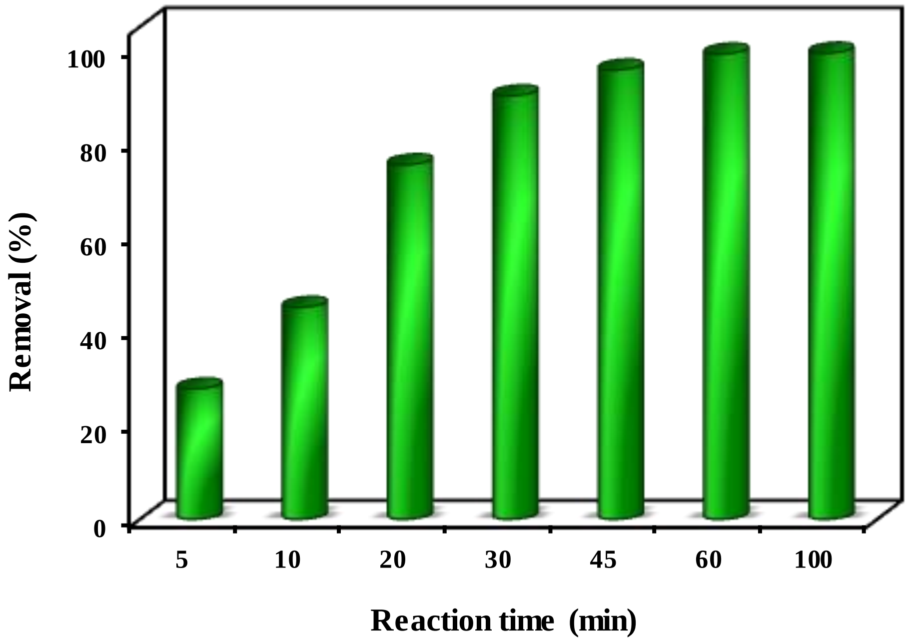

Figure 5. Effect of reaction time on RR-11 removal (pH of 4 and dye concentration of 40 mg/L, and adsorbent amount of 0.60 g/L)

Isotherm and adsorption kinetics

Adsorption isotherms are very important to establish the appropriate relationship for the equilibrium curve and to optimize the design of an adsorption system to remove contaminants. Absorption equilibrium occurs when the amount of contaminant adsorbed on the adsorbent surface is equal to the amount of contaminant absorbed. The distribution of the pollutant molecule between the liquid and adsorbent phases is a measure of the equilibrium position in the adsorption process and is generally expressed by one or more adsorption isotherm models. In this study, Freundlich and Langmuir linear isotherms were investigated. The results are shown in Table 1 [16, 19].

Table 1. The parameters and coefficients constant of Langmuir and Freundlich isotherms.

Langmuir isotherm				Freundlich isotherm		
$\frac{Ce}{qe}=\frac{1}{bqm}+\frac{Ce}{qm}$				$lnq_e = \ln K_f + \frac{1}{n} lnC_e$		
q_m (mg.g^{-1})	b (L.mg^{-1})	R_L	R^2	K_f	n	R^2
52.7	1.3	0.011	0.9906	28.4	2.3	0.9028

In order to study the adsorption kinetics and the rate of adsorption reactions, it is essential to evaluate the adsorption kinetics. Two models of first-order kinetics and second-order kinetics are extensively investigated for adsorption processes. First-order kinetics show that adsorption occurs by penetration of a layer, and second-order kinetics show that chemical adsorption is a step-slower and controls the adsorption process. The first-order kinetics is based on solid phase capacity, and the second-kinetics is based on solid phase adsorption. In this study, first and second-order kinetics were investigated. The results are shown in Table 2.

Table 2. The parameters and coefficients constant of first and second-order kinetics model.

First-order kinetic constants			Second-order kinetic constants		
$\log(q_e - q_t) = \log(q_e) - k_1 t/2.303$			$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$		
K_1 (min^{-1})	q_e (mg/g)	R^2	K_2 (g.mg.min)	q_e (mg/g)	R^2
0.019	43.5	0.9025	0.027	38.7	0.9934

4. Discussion

According to the results obtained (Figure 2), the maximum removal of RR-11 dye occurred at a pH of 4, which is due to the anionic structure of RR-11 dye and pH_{zpc} of sawdust. Given that the pH_{zpc} of sawdust from pistachio trees is 2.5, at a pH above pH_{zpc}, the predominant electrical charge at the surfaces of gravity is negative. Moreover, at a pH of less than pH_{zpc}, the predominant electric charge on the surfaces of gravity is cationic and positive. Due to the fact that the surface charge of RR-11 paint is negative and anionic, so with increasing pH, the number of negative charges increases and the electrostatic repulsion force between the adsorbent and the pollutant increases and finally the removal efficiency decreases; at a pH of less than 5.2, the adsorbent charge (pistachio sawdust) is positive but the molecule of RR-11 dye still has a negative and anionic charge, and electrostatic attraction occurs and the dye removal efficiency will thus increase.

One of the important and influential factors in the adsorption process is the initial concentration of the pollutant. In this study, the initial concentration of RR-11 dye was investigated in the range of 20 to 100 mg/L. The results showed that the initial concentration of RR-11 dye had a negative effect on the removal efficiency. In other words, increasing the initial concentration of RR-11 dye reduced the removal efficiency (Figure 3). The reason behind the decreasing trend in removal efficiency by increasing the initial concentration of RR-11 dye can be explained by the fact that at a constant amount of adsorbent, the active sites of adsorption are constant, but with increasing concentration of the contaminant, the number of RR-11 dye molecules in the reaction medium increases. Thus, the removal efficiency decreases (20, 21). Investigating the effect of adsorbent amount on adsorption processes is another issue that should be taken into account. The amount of adsorbent was applied in the range of 0.1 to 0.6 g/L, and its effect on RR-11 dye removal efficiency was measured. In this study, given the high adsorption sites and surface area, with increasing the amount of adsorbent, the removal efficiency has increased as well. It can also be stated that, at a constant concentration of the contaminant, by increasing the amount of adsorbent, the ratio of active sites present at the adsorbent surface in relation to RR-11 dye molecules is high; the removal efficiency will thus increase. While at low adsorbent amounts, the ratio of active sites to RR-11 dye molecules decreases, and as a result the adsorption decreases (22, 23).

Reaction time is an integral part of any technique, especially adsorption technique. In this study, reaction times of 5-100 minutes was investigated. The results have indicated that the RR-11 dye adsorption process has an increasing trend, and the adsorption process reaches equilibrium in 60 minutes, and shows a relatively stable trend. The effect of reaction time can be expressed as such that by increasing the reaction time, the collision likelihood of the contaminant molecule with the adsorbent increases, and as a result, the removal efficiency increases. In other words, the residual value decreases and the adsorption capacity increases (24-26).

As can be seen in Table 1, the value of R^2 for the Freundlich isotherm was 0.9028, and as for Langmuir isotherm, it was 0.9906, indicating that the adsorption process follows the Langmuir isotherm with a higher R^2 value. Moreover, b values were reported to be 1.3 and the maximum adsorption capacity was 52.7 mg of pollutant per gram of adsorbent. An isotherm is used that can be used to evaluate the suitability of sawdust for RR-11 dye adsorption. In Langmuir model, a dimensionless coefficient called the separation factor (R_L) is used to express the main feature and characteristic of the isotherm, which can be used to evaluate the suitability of sawdust in absorbing RR-11 dye. The status of the isotherm can be interpreted according to the value of the separation factor. If the calculated R_L value is more than 1, the adsorption is undesirable, if it is 1, the adsorption is linear, if it is zero, the adsorption is irreversible, and if it is between 0-1, the adsorption is optimal. The R_L value in this study was 0.011, being between 0-1. Thus, absorption is desirable. The results of adsorption kinetics (Table 2) showed that the adsorption process follows the second-order kinetics. The amount of R^2 in RR-11 dye removal was 0.9934. The coefficient K_2 was 0.027, and q_e value was 38.7 mg/g.

5. Conclusion

In this study, the removal efficiency of RR-11 dye was investigated using pistachio sawdust and the effect of important variables including initial dye concentration, pH, adsorbent amount and reaction time on RR-11 dye removal efficiency was investigated. According to the results obtained, the maximum removal efficiency (under optimal conditions) was 99.2%. Moreover, kinetics and adsorption isotherm studies indicated more compatibility with second-order kinetics and Langmuir isotherm. It can be concluded that pistachio sawdust can be a suitable and reliable adsorbent; it can replace expensive and costly adsorbents. It can be applied as an effective method to remove dye with high efficiency over a short time without producing toxic and harmful by-products to treat contaminated aquatic environments.

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