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The Effect of Wood Wool on Absorbing Oil Spills in Water Solutions

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Abstract

Background and Purpose: The oil Spill in water pollution has happened in Iran since the beginning of oil extraction, and their gradual accumulation in the environment is a threat to water resources; therefore, it is necessary to develop new technologies to remove and clean the oil spill. There are different methods to remove oil spills and their derivatives. In the present research, wood wool waste as a cheap adsorbent was used to remove oil spills from wastewater. Materials and Methods: First, the chemical compound was determined using XRF and XRD analysis, next, absorption tests discontinuously were carried out using the laboratory solutions containing oil and the optimal absorption conditions were investigated by changing the factors affecting absorption, which included pH, initial concentration of the pollutant, contact time and amount of adsorbent on the amount of absorption at different levels, and at last, the use of adsorbent for laboratory effluent was studied. Findings: The effect of adsorbent during the equilibrium time to absorb oil using wood wool as an adsorbent was higher at 15 min than at other contact times with a significant difference (84% absorption rate: $P<0.05$), and the lowest absorption was shown at 3 min with a significant difference (60% absorption rate: $P<0.05$). The highest absorption rate of wood wool was observed at pH=3, which showed a significant difference with other pHs (75% absorption rate: $P<0.05$). There was no significant difference between the effect of the amount of wood wool as an absorbent at 5 min and 10 min (70.83% and 70% absorption, respectively: $P<0.05$), and the lowest percentage of absorption with a significant difference was observed at pH=7 (56% absorption rate: $P<0.05$). The highest percentage of absorption with a significant difference compared to other values was observed in 2gr (82% absorption rate: $P<0.05$). The effect of the absorbent values had no significant difference in 0.5 and 0.25gr (75% and 60% absorption, respectively: $P<0.05$); but the least percentage of absorption with a significant difference compared to other values were observed in 0.25gr (60% absorption rate: $P<0.05$). The fit of the adsorption isotherms showed that the absorption of oil by wood wool as an absorbent corresponds to the Freundlich adsorption isotherm (R^2=0.9822).

Keywords: Wood wool, Oil spills, Factors absorption, Water Solutions

INTRODUCTION

Water is the most numerous substance in nature and exists in solid, liquid, and gas shapes. The cells of the human body are not able to survive without this substance. The first human attitude towards water was to drink and satisfy individual needs. Moreover, also, water is the best chemical solvent, in which many gases and minerals dissolve easily. Water is considered as the national capital of any country, which will be as valuable as oil shortly. In Iran as well as in the world, competition has increased for the use of water in urban, industrial, environmental and recreational uses (Taghvaii pour, 2016; Dehghani *et al.*, 2015).

Today, one of the most dangerous environmental pollution is oil spill (Durand, Bébouléne and Ducrozet, 1995). Crude oil consists of countless chemicals from a simple molecule of methane to sizable hydrocarbon molecules (more than 16 carbon atoms). Crude oil is composed mainly of aliphatic, alicyclic, and aromatic hydrocarbons. In oil-rich countries like Iran, operations such as exploration, extraction and exploitation of oil resources have caused many environmental problems despite many benefits. Contamination of water sources with petroleum compounds is one of the most important problems (Chen *et al.*, 2016; Fox *et al.*, 2016). The spill of oil into freshwater happens through accidents and wastes containing oil Oil-contaminated water is a major environmental concern caused by different industries and production processes. It presents a significant threat to the environment and various life forms due to its toxic nature (Sulyman *et al*., 2018; Zamri *et al*., 2021). Therefore, treating oily water and removing oil before discharge is vital to prevent environmental harm and protect ecosystems, wildlife, and human health (Al-Najar *et al.*, 2023; Cisterna-Osorio and Arancibia-Avila, 2019).

Exposure to these pollutants can cause the death of living aquatic organisms in water and have harmful effects such as reduced reproduction, inappropriate growth, problems in the feeding mechanism, and reduced defense against diseases (Bícego *et al.*, 2006). The intrusion of pollution into the waters of rivers, lakes, seas and oceans has left destructive effects; In such a way that not only aquatic life is endangered, but it also threatens the life of other land creatures that depend on providing their food via the sea (Yuan *et al.*, 2010).

However, there are many applications for absorbents because it is possible to collect and completely remove oil and its derivatives from contaminated areas by these materials in a short time. Absorbents simplify the ability to convert liquid oil into semi-solid and solid forms (Lim and Huang, 2007; Ibrahim *et al*., 2010).

Accessibility, low price, high capacity and speed of absorption of petroleum compounds, hydrophobicity, long retention time, simplicity of oil recovery from the adsorbent, reusability, high buoyancy, biodegradability, and environmentally friendliness are some effective factors in the selection of adsorbents for the removal of petroleum compounds. Hydrophobicity and lipophilicity of adsorbents, however, are some primary determining parameters for selecting adsorbents to remove organic compounds. Oil absorbents are classified into three main groups: synthetic organic materials, inorganic compounds, and plant organic materials (Lim and Huang, 2007; Lee *et al*., 1999; Teas *et al.*, 2001). The limitations of inorganic compounds and organic materials created motivation to find new absorbents like natural organic materials with biodegradability (Teas *et al.*, 2001; Adebajo *et al.*, 2003).

Among the important and effective factors in the selection of adsorbents to remove petroleum compounds can mention to the accessibility, low price, high absorption capacity and speed, long retention time, ease of oil recovery from the adsorbent, reusability, high buoyancy, and biodegradability, high physical and chemical resistance against shape change, optical decomposition and environmental-friendliness.

Natural absorbents are cheap, available, and have acceptable absorption intensity. Many natural absorbents have been processed and are available as special absorbents for oil stains. This increases the efficiency of the adsorbent to absorb oil. Nowadays, the application of natural materials and absorbents is increasing to a great extent. Corn, cotton, straw, rice hulls, wood particles, and even waste materials such as feathers and wool have attracted the attention of researchers for reasons like biodegradability, renewability, low price, and less effect on the ecosystem (Rezanezhad and Sharifi, 2020).

Wood wool is short and long fibers made of the wood of silver poplar, willow, and poplar. Its cellulosic and porous structure makes it able to absorb large amounts of oil in its pores(Johansson, 1994).

Therefore, the purpose of this study was to the effectiveness of wood wool in absorbing oil from water, oil and oil-water mixture, as well as the effect of various factors such as contact time, initial pH, the concentration of oil spill in the effluent and the amount of adsorbent on the amount of absorption.

INSTRUMENTS AND METHODOLOGY

In the present study, first, wood wool was chosen as the adsorbent and crude oil as the adsorbent to determine the efficiency of different adsorbents for absorbing the oil spill from the water surface. The properties of the absorbent were scrutinized through FTIR and XRF analytical techniques. Adsorbent performance in water and oil absorption was tested separately. The effect of different factors, then, including contact time, initial pH, the concentration of oil spill in wastewater, and amount of adsorbent, was studied on the amount of absorption by the adsorbent. Considering 4 treatments, i.e., pH-dependent adsorption, wastewater-initial-concentration-dependent adsorption, contact-time-dependent adsorption, and adsorbent-dependent adsorption, the research was conducted as follows.

Determination of absorbent properties

Infrared radiometry was carried out using the Fourier[1](FT-IR) Transform to determine the absorption spectrum and types of functional groups of wood wool. The FT-IR beam of the absorbent provides information on the chemical structure and the surface functional groups of the absorbent (polar or non-polar) (Genieva *et al.*, 2008). To determine the chemical compound of the sample, a fluorescence spectrometer (XRF) manufactured by Philips, model 1480PW was used.

[1] FT-IR: Fourier transform infrared spectroscopy

Preparation of absorbents

Wood wool as an absorbent sample was provided as an absorbent. After preparation, absorbent samples were washed several times with distilled water to remove dust and dried in a furnace at 105°C for 14 hours. Next, the absorbent samples were crushed using a Moulinex machine and granulated with a sieve smaller than 2 mm (Ahmad *et al*., 2005).

Absorption of oil from water test

Simulated wastewater was used in this experimental study. To prepare simulated wastewater, 150 milliliters of oil were mixed with one liter of distilled water. Then, it was shaken by a shaker for 10 minutes to mix water and oil.

Effect of different parameters on the absorption process

The effect of various factors affecting absorption was investigated and calculated to achieve the maximum amount of absorption. For this purpose, the mixture of petroleum and absorbent, first, was poured into a 100-ml Erlenmeyer in a ratio of 1:1 and the rest was added to drinking water. Second, it was stirred by a shaker at 150 rpm and passed through filter paper. Next, it is placed in an oven for 24 hours at a temperature of 40-50°C to evaporate its water. Finally, the amount of absorbed oil was obtained using the Gravimetric method based on equation (1):

Equation (1):

$$\text{Absorption percentage} = \frac{\text{Secondary amount of oil} - \text{Primary amout of oil}}{\text{Total amount}} \times 100$$

Among the items that effects were studied on the absorption of petroleum compounds were: pH, the amount of adsorbent, contact time, and oil spill concentration, which the amounts of different variables were pH: (7, 5, 3), wood wool: (0.25, 0.5, 1.5, and 2 grams), contact time: (intervals of 3, 5, 10, and 15 minutes), and oil spill: (350, 250, 150, and 450 milliliters).

Data Analysis

The findings were analyzed by Excel and SAS9.4 software. Excel was used to draw graphs of the effect of pH, temperature, amount of adsorbent, concentration, and contact time on the absorption of grease and oil from simulated wastewater, and SAS software was used to process the data. In addition, the Mean comparison was performed between the parameters by Duncan's test at the 0.05 level (Abolhasani and Pirestani, 2019).

Findings

X-ray diffraction was used to determine the characteristics of the tested absorbent (Figure1).

(Table 1) shows the state of the major phase of wood wool waste, which was in the shape of amorphous or non-crystalline.

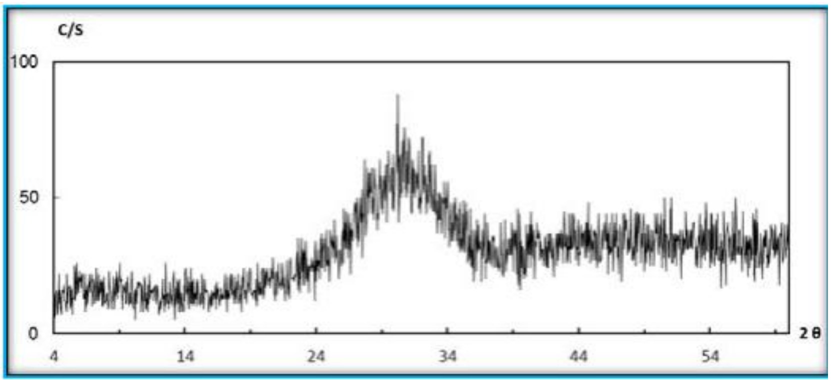

Figure 1. X-ray diffraction diagram

Table 1. Determining the mineral properties of wood wool waste

Major Phase(s)	Minor Phase(s)	Trace phase(s)
Amorphous	----	-----

In (Table 2), the results of variance analysis during the experiment were shown. Based on the findings of this test, the different measured levels had an important effect ($p<0.01$).

Table 2**.** Variance analysis

Indicator	Degree of freedom	F	Significant level
pH	2	283.00	**
Detention time	3	324.67	**
Adsorbent amount	3	286.00	**
Adsorbate concentration	3	762.33	**

** Indicates a significant difference in values at the 0.01 level.

Effect of absorbent amount on oil absorption

According to (Table 3), the amount of oil in 2 grams was more than other amounts of this absorbent (82%). The lowest amount was seen at 60% with a significant difference of 0.25 grams, which is not significantly different from the amount of 0.5 grams ($p<0.01$).

Table 3. Mean comparison of absorption percentage of absorbent in different amounts (gr)

Absorbent(gr)	0.25	0.5	1.5	2
	75 ±1[c]	75 ±1[c]	79± 1[b]	82±1[a]

The letters indicate significant differences in each column at the α=0.05 level.

Effect of pH on oil absorption

According to (Table 4), the highest (75%) and lowest (56%) amounts of oil absorption were seen at pH=3 and pH =7, respectively, which have a significant difference with pH=5 ($p>0.01$).

Table 4. Mean comparison of absorption percentage of wood wool at different pHs

pH	3	5	7
	75 ± 1[a]	62 ± 1[b]	56 ± 1[c]

The letters indicate significant differences in each column at the α=0.05 level.

Effect of contact time on oil absorption

According to (Table 5), the highest amount of absorption (84%) occurred in 15 minutes. There was no significant difference in the amount of oil absorption between 5 and 10 minutes, and the lowest amount of absorption (60%) was observed in 3 minutes ($p>0.01$).

Table 5. The mean comparison table of the absorption percentage of wood wool at different times (minutes)

Time (min)	3	5	10	15
	60±1[c]	70 ± 1[b]	70.83±0.763[b]	84±1[a]

The letters indicate significant differences in each column at the α=0.05 level.

Effect of initial concentration of oil spill on absorption efficiency

According to (Table 6), the amount of oil absorption in 450 ml was 88.33% with a significant difference more than other concentrations, and the lowest amount was seen in 150 ml with a significant difference of 60% ($p<0.01$).

Table 6. Mean comparison of the percentage of wood wool in different concentrations (ml)

Pollutant concentration (milliliter)	150	250	350	450
	60 ± 1[d]	71 ± 1[c]	84.66±0. 577[b]	88.33±0. 577[a]

The letters indicate significant differences in each column at the α=0.05 level.

To have a proper fitting adsorption model on adsorption isotherms, it should be mentioned that these isotherms describe the adsorption of soluble materials on solid materials based on quantitative units at stable temperatures. To do this, the fitting of the achieved results with Langmuir and Freundlich adsorption models for the wood wool as an absorbent is shown in (Figures 2 and 3). The parameters of the models are also presented in (Table 4) for the wood wool.

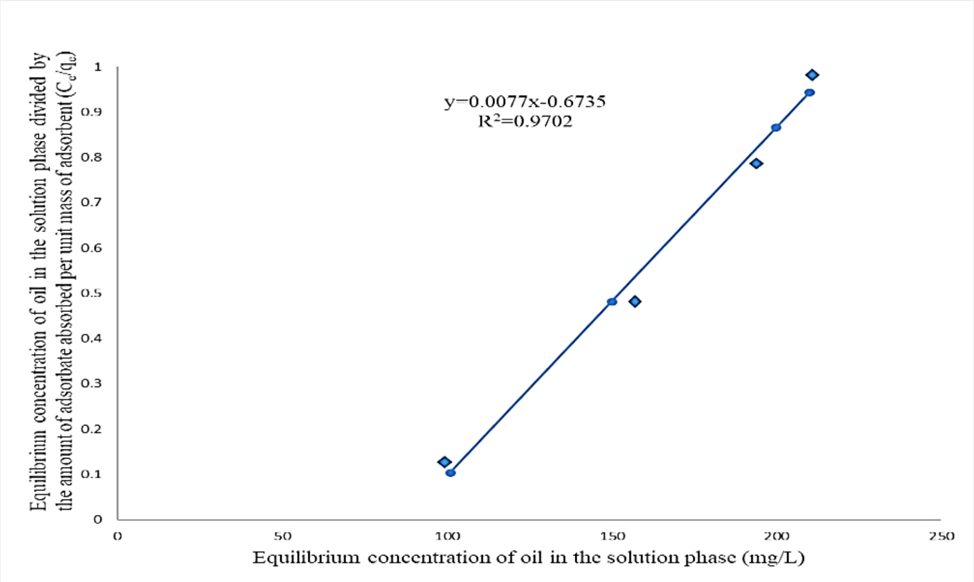

Figure 2. Langmuir adsorption isotherm for oil absorption by wood wool

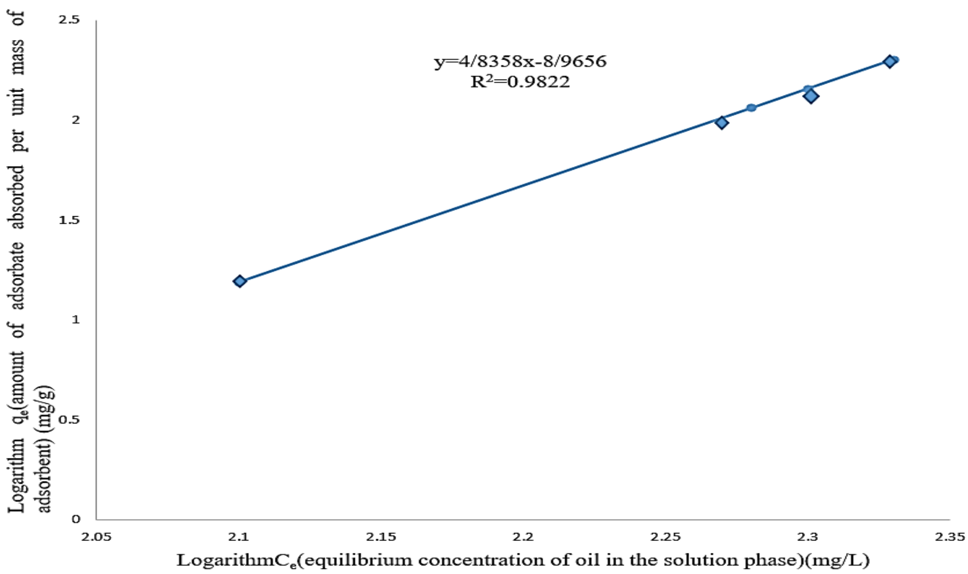

Figure 3. Freundlich adsorption isotherm for oil absorption by wood wool

Oil absorption by wood wool follows the Freundlich model because of a higher R^2 (0.9822). The K_f coefficient, which is a measure of absorption power, was 10^{λ}g/L. Freundlich model's n/1 coefficient, which is a measure of absorption intensity, was 8358.4 for oil using wood wool. In the Langmuir model, the wood wool as an adsorbent, the maximum oil absorption capacity (b) was 129.87 mg of oil per g of wood wool, and its absorption tendency constant (K_L) was 0.01 mg/L and the calculated R_L values for were 0.05 oil absorption. This value is between 0 and 1. The obtained R_L indicated the optimal absorption by the adsorbent.

Table7. Langmuir and Freundlich adsorption isothermal constants for the wood wool as an adsorbent

Coefficient	Model	Amount
k_f(L/g)	10^{λ}	
1/n	4/8358	Freundlich $n\frac{logCe}{1} + logqe = logK_f$
R^2	0/9822	
K_L(L/mg)	0/01	
b(µg/g)	129/87	Langmuir $Kb + \frac{\frac{c}{b}}{1} = \frac{c}{x}/m$
R^2	0/9702	
R_L	0/05	

DISCUSSION

Using absorbents is currently considered the most important and widely used method (Ong *et al*., 2007). In the present study, the maximum absorption of oil using wood wool occurred at an acidic pH of 3 (75%). As the pH increased, the amount of absorption decreased; So, the lowest amount of absorption occurred at pH=5 with a significant difference (56%). pH was one of the important factors that affected the absorption process through the effect on the structure of the pollutant and the surface charge of the absorbent (Banat *et al.*, 2000). The reason for the decrease in absorption percentage with increasing pH can be related to the anion structure of the adsorbate. Studies showed that at high pH, the dominant surface electric charge on the surface of the adsorbents was negative. Therefore, with the increase in pH, the number of negative charges increased, and regarding the anionic nature of the adsorbate in these conditions, the electrostatic attraction between the adsorbent and the pollutant, and absorption efficiency decreased (Varghese *et al*., 2004; Sayyahzadeh *et al*., 2015). The findings are consistent with the study of Banat *et al*. (2000). In their study, the percentage of phenol adsorption on bentonite decreased with increasing pH (Banat *et al.*, 2000).

The same results were reported by Varghese *et al*, (2004). As they argued, the effect of pH in increasing pollutant absorption on the absorbent surface is related to the relationship between pH and the electric charge of the absorbent surface. These researchers reported that at a pH lower than pHzpc, the absorbent surface is surrounded by proton-containing carboxylic agents, and this, in turn, increases the positive charge on the absorbent surface. In their study, considering the anionic nature of the adsorbents, the electrostatic attraction between the adsorbate and the pollutant, and so the removal efficiency grew. This condition was the same for the adsorbent used in this study (Varghese *et al.*, 2004).

In Ahmad *et al.*'s (2005) study, which was conducted on the removal of residual oil in wastewater using natural adsorbents, chitosan, bentonite, and activated carbon, the adsorption of residual oil on bentonite and activated carbon was significant under the conditions of acidic. The pH of the solutions affected the surface of charge of these adsorbents and their degree of ionization made them perform better as oil adsorbents. In the current study, the effect of changing the contact time on the process of oil absorption by the adsorbent implied that the absorption efficiency increased while the increase in contact time of the wood wool with the oil-containing simulated wastewater. The highest amount of oil absorption by wood wool was observed at 15 min with a significant difference compared to 3 and 5 mins (84%), and the lowest one was discovered at 3 min with a significant difference (60%). Sayyahzadeh *et al*. (2015) conducted research on the removal of petroleum hydrocarbons from oil refinery wastewater using natural adsorbents such as almond and walnut shells. They pointed out that as time went up to 30 min, the absorption increased, but with more growth in time, absorption remained unchanged (Sayyahzadeh *et al.*, 2015).

Salehi *et al*. (2014) showed an increase in contact time, the absorption rose because many unoccupied active sites on the absorbent surface were occupied as time elapsed (Salehi Rad, Haghighat Kish and Hosseini Verkhani, 2014). In the study of Razavi *et al*. (2014), which was conducted on the use of raw rice hulls in removing crude oil from aquatic environments, the percentage of crude oil absorption using rice hulls increased as the contact time grew, and it, then, remained stable (Razavi and Mirghfari, 2014). Hosseini *et al*. (2017) argued that in the removal of hydrocarbons from the sludge of diesel fuel tanks using zeolite in Semnan, increasing the time due to the growth in the number of collisions between the pollutant and the adsorbent caused the absorption percentage to go up (Majid Hosseini and Shenavaei Zare, 2017).

In this study, the effect of changing the amount of adsorbent on the oil absorption process from wastewater showed that with the increase in the amount of adsorbent, the percentage of oil absorption by the adsorbent increased; however, higher amounts of adsorbent did not change the absorption percentage. The highest amount of absorption by wood wool was in the 2 g/L of absorbent (82%), and there was a significant change in the absorption percentage with the increase in the amount of absorbent. Moreover, the lowest amount of absorption with a significant difference compared to other values was observed at 0.25 g/L (60%).

In the study of Banat *et al*. (2000), as the adsorbent grew, the absorption increased resulting from the rise in the active and effective surface area in absorption, but finally, adding more amounts of adsorbent did not affect on increasing absorption. There was an equilibrium between the adsorbent and the adsorbate in the solution, and after that no absorption took place. At first, increasing the amount of adsorbent, much adsorption space was available to the adsorbent at the same concentration, and then, increased the absorption percentage (Banat *et al.*, 2000). However, the adsorbent was able to accumulate only a certain amount of the pollutant and became saturated, so adding more amount of adsorbent did not affect on increasing the absorption percentage.

Hussein *et al*. (2009) investigated that although the growth in adsorbent amount increased the removal efficiency of the pollutant (phenol), the amount of absorption per unit mass of the adsorbent decreased because of the unsaturation of some active sites on the adsorbent surface (Husseien *et al.*, 2009). Razavi *et al*. (2014) conducted a study on the use of raw rice hulls to remove crude oil from aquatic environments. According to their research, the percentage of absorption went up to the saturation level with the increase of the adsorbent amount from 1 to 10 g/L (Razavi and Mirghfari, 2014). Another study by Razavi *et al*. (2013), which was about using rice hulls in removing engine oil from water, showed a rise in the amount of adsorbent from 0.1 to 1 gram (Behran engine oil concentration=7 g/L) caused the percentage of the engine oil absorption to increase, but adding more amount of adsorbent showed no effect on increasing the absorption percentage (Razavi *et al.*, 2012).

In the present research, the absorption efficiency of wood wool began to increase as the oil in wastewater grew from 150 to 450 mL so that the highest percentage of absorption was at the concentration of 450 mL with a significant difference of 88.33%, and the lowest percentage, at the concentration of 150 mL, was 60%. In the study of Sayyahzadeh *et al*. (2015), which was conducted on the removal of petroleum hydrocarbons from oil refinery wastewater using natural adsorbents like almond and walnut shells, the adsorbent concentration was significantly increased from 0.2 to 1 g/L and the removal efficiency remained with no change, but as the concentration increased from 1 to 5 g/L, this rate gradually decreased (Sayyahzadeh *et al.*, 2015).

Razavi *et al*.'s (2014) study implied that the percentage of crude oil absorption using raw rice hulls increased initially with the increase of initial oil concentration from 1 to 7 g/L (45-25%) and then stabilized (around 51%). By increasing the initial concentration of the oil compound, the absorption rose, and then no change was observed in absorption reaching a certain concentration. Adsorbents have a specific absorption capacity, so the absorption process remained unchanged after reaching the maximum amount (Razavi and Mirghfari, 2014). Although, increasing the initial concentration of petroleum compounds, on the one hand, makes the adsorbent absorb less water (Liew *et al*., 1993), on the other hand, more molecules of the petroleum compound were available to the adsorbent, so the absorption space was quickly occupied by the adsorbent (Lim and Huang, 2007). As soon as the adsorbent was saturated, however, adding a higher concentration of the petroleum compound did not influence the absorption. In addition, despite the increase in the initial concentration of petroleum compounds because of the constant amount of absorbent, the absorption percentage experienced no change (Durand *et al.*, 1995).

Razavi *et al*. (2013) in another study on the use of rice hulls to remove engine oil from aquatic environments, in the case of Behran engine oil, argued that the absorption percentage initially increased as the initial oil concentration from 7 to 1 g/L grew (80%), and then decreased at the concentration of 100 g/L of oil (75%). At last, after comparing the coefficients of determination of linear isotherms in oil adsorption with each other, the Freundlich model with high R_2 was chosen to be absorbed by wood wool. The K_L coefficient for the absorbent, the b coefficient for the wood wool, and the R_L coefficient were 0.01, 129.87, and 0.05, respectively.

In the study by Sayyahzadeh *et al*. (2015) on the removal of petroleum hydrocarbons from oil refinery wastewater using natural adsorbents such as almond and walnut shells, the maximum adsorption capacity of a single layer of activated carbon of almond and walnut shells, respectively, was estimated about 83 and 59 mg/g (Sayyahzadeh *et al.*, 2015). In the study by Razavi *et al*. (2014) which was conducted on the use of rice hulls for removing crude oil from aquatic environments, the absorption of crude oil on rice hulls was a better fit using the Freundlich model, and the maximum absorption of crude oil in aquatic environments by rice hulls in optimal conditions was about 55%.

CONCLUSION

In our study, the absorption efficiency increased as the contact time went up. The absorption of oil by wood wool as an adsorbent was the highest in 15 minutes. Furthermore, the fall in pH caused the amount of absorption for oil to rise. In the study of the effect of changing the initial concentration of oil, increasing the initial concentration in wastewater increased the absorption. Moreover, regarding the amount of adsorbent, the amount of absorption increased with a growth in the amount of adsorbent. Drawing absorption isotherms disclosed that oil absorption by using wood wool followed the Freundlich model. Finally, it was concluded that wood wool has a high efficiency in absorbing oil from wastewater and it can be used in the treatment of industrial wastewater.

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