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Removal of diazinon from aqueous solutions by the adsorption process of activated carbon produced from rice husk and enhanced with ultrasonic waves

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Abstract

Background: The World Health Organization (WHO) has classified diazinon as a dangerous chemical for human health and the environment. This research aimed to remove diazinon from aqueous solutions using the enhanced adsorption process with ultrasonic waves.

Methods: The adsorbent was prepared from rice husk after acid washing, and was placed in a furnace under nitrogen gas at a temperature of 700 °C for 2 hours. The effects of pH, initial diazinon concentration, adsorbent dose, contact time, and the combined effect of ultrasonic waves were investigated. The concentration of diazinon was measured by ultraviolet spectrophotometry at a wavelength of 247.5 nm.

Results: In this study, the pH_{pzc} of the adsorbent was determined to be 9. The highest removal efficiency (RE) was obtained at a pH of 8, adsorbent dosage of 2 g/L, and diazinon concentration of 20 mg/L. With the increase of the contact time, the RE increased, and maximum RE was obtained up to 78% at 60 minutes. The RE of diazinon using a combination of adsorption with ultrasonic waves increased up to 90%. In this study, with four recovery stages of the spent adsorbent, approximately 4% of the RE decreased. The equilibrium data were studied using Langmuir, Freundlich, and Temkin isotherms. Moreover, kinetic studies were evaluated by pseudo-first-order (PFO) and pseudo-second-order (PSO) models. The results indicated that the Langmuir isotherm and PSO model had the most agreement with the experimental data.

Conclusion: Application of this adsorbent with ultrasonic waves can be effective in removing diazinon from agricultural and pharmaceutical effluents containing diazinon.

Keywords: Diazinon, Adsorption, Ultrasonics, Spectrophotometry, Ultraviolet

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Introduction

In most parts of the world, increasing the concentration of pesticides in surface and groundwater sources is known as a threat to water resources (1,2). Despite the dangers of these pollutants, the number and amount of pesticide production are still increasing. Therefore, it endangers the life of aquatic cultures and affects the quality of water (3,4). Diazinon is one of the most widely used organophosphate pesticides that was produced in 1952 and has been widely used as an insecticide, acaricide, and nematicide (5,6). The World Health Organization (WHO) has classified diazinon as a hazardous chemical for aquatic animals, mammals, and humans (7,8). According to the recommendation of the European Union, the maximum permissible concentration of diazinon in drinking water guidelines is reported as 0.1 µg/L (9). This substance has a half-life of 2 to 4 weeks in the soil, and the contact threshold for humans is 0.025 mg/kg per day (10). The Chemical structure of diazinon was shown in Figure.1.

Diazinon removal methods include photocatalytic purification, advanced oxidation, biological treatment, membrane filters, ultrasonic, ultraviolet, ion exchange, and adsorption processes. Due to the high consumption of chemicals, high purification cost, incomplete removal, and time-consuming nature of most of the aforementioned methods, their use is not economical and applicable (11,12).

The adsorption process is one of the most effective physical methods for removing pollutants from the environment due to its simple design, low cost, ease of operation, proper maintenance, no need for final treatment, and cost-effectiveness (13,14).

There has been much interest in biosorbents over

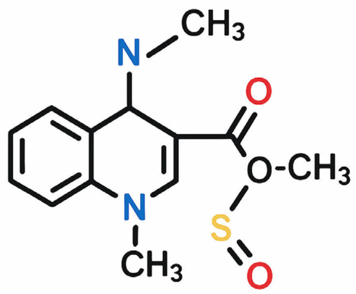

Figure 1. Chemical structure of diazinon

the decades, and they continue to be investigated due to different biomass yielding different results. Some impressive examples with high adsorption capacities are knotgrass, brewer's spent grain, fungi, berry seeds, and soybean hulls (15,16). Today, many studies are being performed on the use of agricultural waste in the adsorption process of various pollutants (17,18). Adsorbent derived from rice husk is a form of carbon that increases its porosity and internal surface due to the processing operation. This compound is a unique material due to its significant internal area, porous structure, high adsorption capacity, surface reactivation capability, and relatively low price compared to inorganic adsorbents such as zeolite (17,19).

The propagation of ultrasonic waves in an elastic medium causes periodic initiation of expansion and contraction cycles that repeat one after the other. Contraction cycles increase the pressure and decrease the molecular distance, while in the expansion cycles, the distance between the molecules increases (20). This type of contraction and expansion mechanism on the molecules of pollutants such as diazinon can cause their destruction and dissolution in aqueous environments (21).

Considering the high consumption of diazinon in agricultural activities in the northern regions of Iran and the environmental pollution caused by it, and considering that the preparation of adsorbent from agricultural waste is available, abundant, and economical, this study aimed to remove Diazinon by an adsorbent produced from rice husk using ultrasonic waves. Also, in this study, the possibility of recovering the adsorbent used in the removal of Diazinon and determining its efficiency was investigated. So far, many studies have been conducted regarding the removal of environmental pollutants by agricultural wastes, but the removal of Diazinon with an adsorbent produced from rice husk using ultrasonic waves has not been addressed in other studies.

Materials and Methods

Preparation of the adsorbent from rice husk

First, rice husk was collected from the agricultural lands of northern Iran and washed with distilled water to remove any dust and impurities. Next, the desired sample (10 g) was mixed with 3% hydrochloric acid, 10% sulfuric acid, and washed completely with distilled water, and dried in an oven for 24 hours. Finally, the sample was placed inside a furnace under nitrogen gas at a temperature of 700 °C for 2 hours to prepare the adsorbent (22). The results of elemental analysis of CHNS for activated carbon derived from rice husk were shown in Table S1.

Preparation of diazinon solution samples

Diazinon with 99% purity and methanol solvent were used to prepare diazinon stock solution with a concentration of 1000 mg/L.

Determination of pH_{pzc} of the adsorbent

The pH_{pzc} of the adsorbent was determined based on the following method. In such a way that 50 mL of 0.1 M NaCl solution was poured into 150 ml Erlenmeyer flasks, and the pH of the samples was adjusted by sodium hydroxide and total hydrochloric acid in the range of 2 to 12. Then, 0.05 g of the adsorbent was added to the solutions and stirred for 48 hours by an electric stirrer, and the final pH was measured, and its curve was plotted against the initial pH. The point of the curve that intersected the bisector was determined as the adsorbent pH_{pzc} (23).

Designing adsorption experiments

To investigate the effective factors on the diazinon removal by the adsorbent produced from rice husk, to determine the optimal conditions of pH, equilibrium time, pollutant concentration, adsorbent dose, and temperature, 50 mL samples were prepared with specific concentrations of diazinon and the pH. A certain amount of adsorbent was poured into an Erlenmeyer flask. Then, the solution was stirred with a shaker at an average speed of 250 rpm for one hour and at ambient temperature. The samples were filtered, and the residual concentration of diazinon was measured by T80+UV-Visible Spectrometer from PG instruments Ltd at a wavelength of 247.5 nm (24). To calculate the removal efficiency (RE) and adsorption capacity in the adsorption system, equations 1 and 2 have been used (25).

$$\mathrm{R}\,(\%) = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

$$q_e = \frac{C_0 - C_e}{m} \times \mathrm{v} \quad (2)$$

Where R is diazinon removal percentage (%), q_e is adsorbent capacity in equilibrium, conditions (mg/g), C_0 is initial concentration of diazinon (mg/L), C_e is equilibrium diazinon concentration (mg/L), m is mass of the adsorbent (g), and V is volume of Diazinon solution (L).

Experiments related to the effect of ultrasonic waves on the removal of diazinon

To investigate the factors affecting the removal process of diazinon and ultrasonic waves, samples of 50 mL diazinon solution were prepared with optimal concentration and pH. The optimal amount of adsorbent was poured into an Erlenmeyer flask and placed in an ultrasonic chamber at a temperature of 20 °C for 15 minutes. Then, the samples were centrifuged for 7 minutes at a speed of 600 rpm and the residual diazinon was measured by UV spectrophotometry (25).

Results

Determination of the pH_{pzc} in the adsorbent

The pH_{pzc} of an adsorbent is the pH at which the electrostatic charge on the adsorbent surface is zero. This parameter is very important for the removal of contaminants in the adsorption process. As shown in Figure 2, the pH of the desired adsorbent is determined to be 9.

The effect of the initial pH of the solution on the removal of diazinon

The adsorption of diazinon by the adsorbent strongly depends on the pH of the solution. In this study, the RE of diazinon increased with increasing pH up to 8 and then decreased (Figure 3).

The effect of adsorbent dosage on diazinon removal

The effect of adsorbent dosage on the adsorption of diazinon from aqueous solutions was investigated, and the results are illustrated in Figure 4. As can be seen, the removal percentage of diazinon increases gradually when the dose of adsorbent is increased, and the adsorption capacity decreases with increasing adsorbent dosage from 0.5 to 2 g.

The effect of contact time on the diazinon removal

The effect of contact time on the RE of diazinon was shown in Figure 5. According to this Figure, in the first hour of the adsorption process, the removal percentages are significantly high. However, over time, the percentage of diazinon removal has gradually decreased.

The effect of diazinon concentration on the removal efficiency in the adsorption process

The effect of the initial concentration of diazinon on the adsorption process of the pollutant in different concentrations for 120 minutes was investigated (Figure 6). According to this Figure, as the concentration of diazinon increases, the RE of diazinon decreases.

Determination of the adsorbent recovery

In this study, the recovery and reuse of the produced adsorbent were investigated (Figure 7). It was shown that after four recovery stages, the RE of the adsorbent decreased by 3.9% due to the change in the structure of the adsorbent during the washing process.

Effects of ultrasonic waves on the removal of diazinon

In this study, the effect of the use of ultrasonic waves and ultrasonic waves with rice husk absorbent on the diazinon

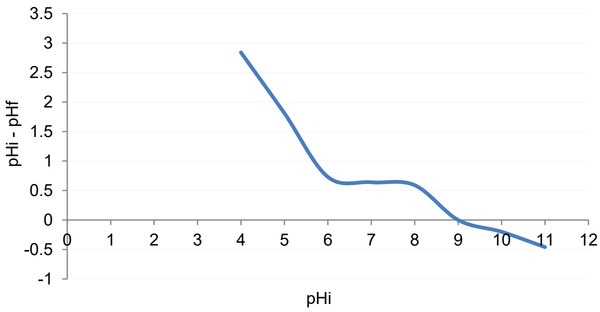

Figure 2. Determining the pH_{pzc} in the studied adsorbent

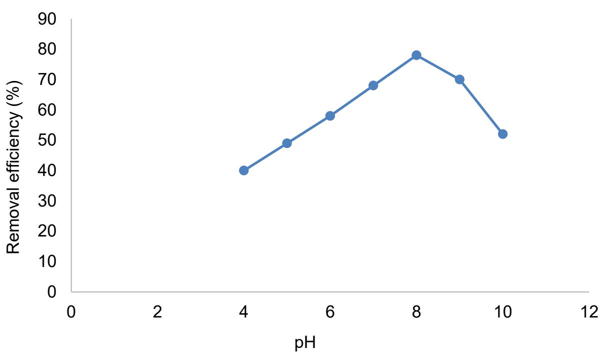

Figure 3. The effect of the initial pH on diazinon removal efficiency (Diazinon concentration: 20 mg/L; adsorbent dose: 2 g; temperature: 25°C; contact time: 60 min)

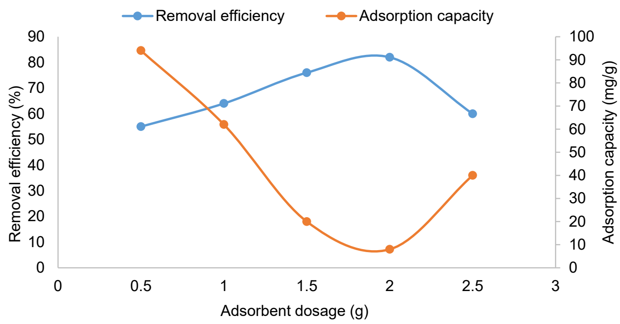

Figure 4. The effect of adsorbent dose on diazinon removal efficiency (Diazinon concentration: 20 mg/L; contact time: 60 min; temperature: 25°C; pH: 8)

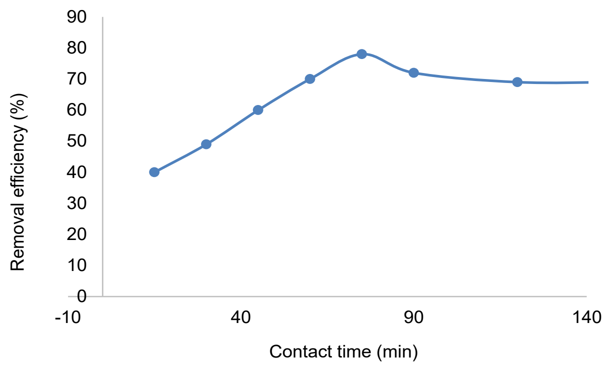

Figure 5. The effect of contact time on the removal efficiency of diazinon (Diazinon concentration: 20 mg/L; adsorbent dose: 2 g; pH: 8; temperature: 25 °C)

RE was investigated, and the results are shown in Figure 8.

Adsorption isotherms

The isotherm is considered a fundamental factor in determining the capacity of an adsorbent and optimizing the consumption of the adsorbent. Usually, Langmuir, Freundlich, and Temkin models are used to determine the appropriate isotherm in the adsorption process, and the corresponding equations are shown in relations 3 to 5, respectively.

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{C_e K_L q_m} \qquad (3)$$

Where q_e and q_m (mg/g) are the equilibrium and maximum adsorption capacities, respectively; K_L is the Langmuir constant and C_e (mg/L) is the equilibrium concentration of the adsorbate (24).

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

Where K_f and n are related to the adsorption capacity of an adsorbent and sorption intensity, respectively (24,25).

$$q_e = B_T \ln K_T + B_T \ln C_e \qquad (5)$$

Where B_T is the Temkin constant, which is related to the heat of sorption, K_T is the Temkin isotherm constant, and T is the absolute temperature (25).

In this study, the adsorption isotherms of Langmuir, Freundlich, and Temkin were investigated, and the relevant results are shown in Table 1 and Figures S1 and S2.

In this study, the R^2 coefficient related to the Langmuir isotherm ($R^2 = 0.991$) was greater than the R^2 coefficient related to the Freundlich ($R^2 = 0.940$) and Temkin ($R^2 = 0.983$) isotherms.

Determination of kinetic models in the adsorption process of diazinon

Adsorption kinetics was investigated to better understand the dynamics of diazinon adsorption on the adsorbent and to prepare a predictive model that allows estimating the amount of adsorbed ions during the process. Usually, pseudo-first-order (PFO) and second-order (PSO) are used to determine the kinetics of surface adsorption processes.

Equations 3 and 4 are used to determine PFO and PSO, respectively (24,25):

$$\frac{{}^{d}q_t}{dt} = k_1 (q_e - q_t) \qquad (6)$$

$$\frac{t}{q_t} = \frac{1}{n} + \frac{t}{q_e} \qquad (7)$$

The kinetic parameters of diazinon adsorption on the

Table 1. Parameters resulting from fitting equilibrium data with different isotherms (24).

Model	Formula	Parameters	Values
Langmuier	$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{C_e K_L q_m}$	n	0.31
		K_f (mg $^{-1 (1/n)}$. L $^{1/n}$. g^{-1})	1.34
		R^2	0.940
Freundlich	$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e$	q_{max} (mg.g^{-1})	18.71
		K_l (L.mg^{-1})	0.200
		R^2	0.991
Temkin	$q_e = B_T \ln K_T + B_T \ln C_e$	B_T (L. mg^{-1})	19.902
		A_T (L/g)	0.230
		R^2	0.983

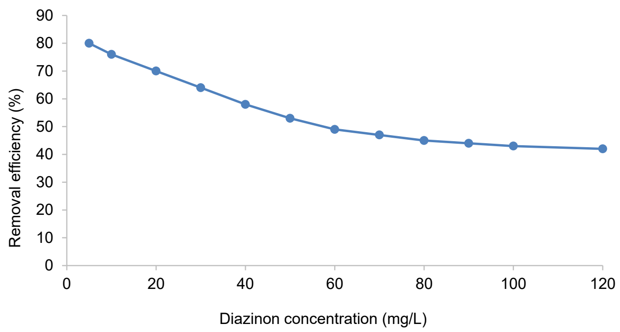

Figure 6. Diazinon concentration on diazinon removal efficiency (Adsorbent dosage: 2 g; contact time: 60 min; temperature: 25 °C; pH: 8)

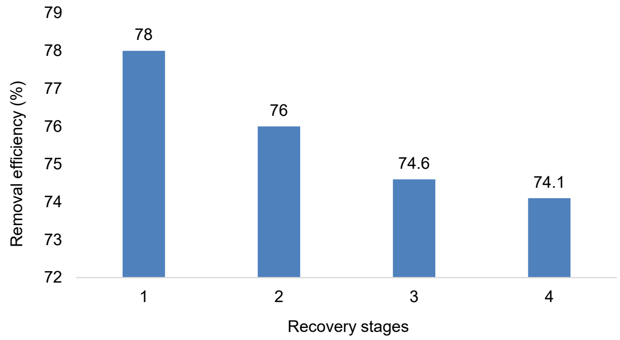

Figure 7. Diazinon removal in each different stage of the adsorbent recovery (Diazinon concentration: 20 mg/L; dose adsorbent: 2 g; contact time: 60 min.; temperature: 25 °C; pH: 8)

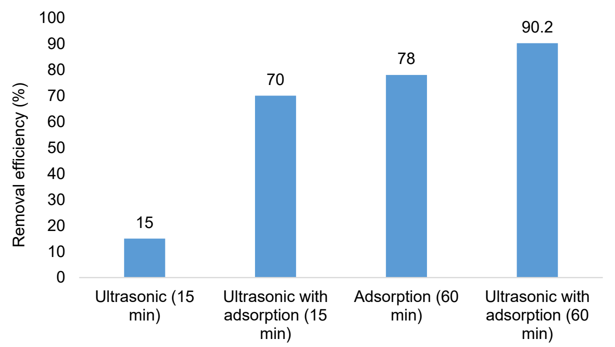

Figure 8. Effect of ultrasonic waves on the removal of diazinon (Diazinon concentration: 20 mg/L; adsorbent dose: 2 g; temperature: 25 °C; pH: 8)

studied adsorbent are shown in Table 2. As shown in this table, the R^2 of the PSO model was greater than the R^2 of the PFO model.

Table 2. Kinetic constant values in the diazinon adsorption process by the studied adsorbent (24,25)

Model	Formula	Kinetic constants		
PFO	$\frac{dq_t}{dt} = k_1(q_e - q_t)$	K_1(min^{-1})	q_e (mg.g^{-1})	R^2
		0.785	71.58	0.931
PSO	$\frac{t}{q_t} = \frac{1}{n} + \frac{t}{q_e}$	K_2(min^{-1})	q_e (mg.g^{-1})	R^2
		0.040	9.72	0.984

Identifying the physical structure of the biosorbent using different analyses

Field emission scanning electron microscopy (FE-SEM) analysis

The internal structure of the adsorbent produced from rice husk before and after the diazinon adsorption process is shown in Figure S3. As shown in the figure, the rough surfaces inside the adsorbent and the high porosity, which is a good characteristic for an adsorbent in the adsorption process, are visible.

Fourier Transform Infrared (FT-IR) analysis

Using FT-IR spectra, the different functional groups of the adsorbent produced from rice husk at wavelengths of 400 to 4000 cm^{-1} are shown in Figure S4.

Energy dispersive X-ray spectroscopy (EDX) analysis

The results of the analysis of the various elements present in the structure of the studied adsorbent using the EDX experiment are shown in Figure S5**.** In this Figure, each peak in the spectrum is specific to a type of element, and peaks with higher heights indicate a higher concentration of each element in the adsorbent.

X-ray diffraction (XRD) analysis

The patterns of XRD analysis related to before and after adsorption of the biosorbent show the production and removal of diazinon in the range of 2 Theta = 4-70° (Figure S6). According to the XRD analysis, in the structure before adsorption, the largest spatial distance of 464 nm corresponds to the angle of 26.408 Theta°. However, after the adsorption process, the spatial distance obtained at 2 Theta° = 26.408 decreased and reached 315 nm.

Brunauer- Emmett- Teller analysis

According to Table S2, the total pore volume, specific surface area, and mean pore diameter in the adsorbent produced from rice husk by the Brunauer- Emmett- Teller (BET) analysis were determined to be 0.284 cm^3/g, 460.96 m^2/g, and 2.47 nm, respectively.

Discussion

Investigating the effect of initial pH on diazinon removal

pH is one of the most important parameters affecting the efficiency of the adsorption process in water and wastewater treatment. This parameter plays an important role in the changes in electrical charge between the type of solution and the surface of the adsorbent. Therefore, determination of the optimum pH can have a significant effect on increasing the efficiency of the adsorption process (26,27). According to Figure 3, the pH_{pzc} of the adsorbent is 9, and it is clear that at pH 9, the adsorbent is electrically neutral. In this study, the highest percentage of diazinon removal (78%) was obtained at pH 8. In explaining the cause of these results, it should be said that at a solution pH of less than 9, H^+ ions increase on the surface of the adsorbent and due to the creation of van der Waals attraction force between the positive ions on the surface of the adsorbent and the negative charges in the diazinon solution, the RE in the adsorption process increases (28,29). At pH above 9, the adsorbent donates its positive charges (H^+ ions) to the solution and the adsorbent surface becomes negatively charged. In this case, the negative functional groups in the solution dominate, and the electrostatic repulsion between the negative charges of the adsorbent surface and the negative functional groups in the diazinon solution reduces the efficiency of diazinon removal in the adsorption process (30,31). In a study by Baharum et al on the removal of diazinon from aqueous solutions using an adsorbent produced from modified coconut shells, the best adsorption conditions (98% RE of diazinon) were obtained at pH 7 and a dosage of 10 g/L (32). In a study by Sean et al on the removal of diazinon from aqueous solutions using modified adsorbents produced from agricultural wastes, the adsorption capacity range was 6.8 to 15.8 mg/g at pH 5 (33).

Investigating the effect of adsorbent dosage on the diazinon adsorption process

The adsorbent dose is also an effective factor in the adsorption process of a pollutant (34). As can be seen in Figure 5, increasing the adsorbent dose increased the RE of diazinon and decreased the adsorption capacity of this adsorbent. In this study, the maximum RE of diazinon was achieved at a dosage of 2 g. In addition, the maximum adsorption capacity of the adsorbent was obtained at 80 mg/g at a dosage of 0.5 g. In justification of this phenomenon, it can be mentioned that with increasing the adsorbent dose, the amount of active surfaces and the number of adsorption sites for removal of the pollutant increase (35). However, according to the adsorption capacity relationship, increasing the adsorbent dosage increases the denominator of the relationship and decreases the adsorption capacity (36). In the study by Akbarlou et al, to investigate the adsorption of diazinon by commercial activated carbon, the most suitable adsorption conditions were determined at pH 5, a dose of 10 g/L, and a contact time of 30 minutes (37).

Investigating the effect of contact time and diazinon concentration on the adsorption process

Other factors affecting the adsorption rate of a pollutant in the adsorption process include contact time and pollutant concentration (23). According to Figure 6, in the early stages of the surface adsorption process, due to the presence of a large number of active sites on the adsorbent surface and the presence of a concentration gradient in the two phases of solution and solid, the adsorption rate is significantly high. In this study, the maximum RE of diazinon was at a contact time of 60 minutes, a dosage of 2 g, a concentration of 20 mg/L of diazinon, and pH 8. However, as time passed, the efficiency of diazinon removal gradually decreased. In this study, the pollutant RE decreased with increasing diazinon concentration. The lack of active sites at high concentrations of a pollutant reduces the surface adsorption capacity of an adsorbent, and since each adsorbent has certain adsorption sites, with increasing concentration of a pollutant, these active surfaces are filled and the adsorption capacity of the adsorbent increases. Eventually, increasing the concentration increases the adsorption capacity until the active sites of an adsorbent are occupied by the contaminant (26). In the study of Moussavi et al, in the removal of diazinon from polluted waters using activated carbon modified with ammonium chloride, a maximum removal of 97.5% was achieved at a concentration of 20 mg/L of diazinon and an adsorbent dose of 0.3 g, at neutral pH and a contact time of 30 min (38).

Recovery and reuse of adsorbent

The recovery of the adsorbent after the adsorption process is one of the most important issues from an economic and environmental point of view. In this study, the recovery and reuse of the adsorbent for the RE of diazinon was investigated. The recovery of the adsorbent was carried out by distilled water, sulfuric acid (50 mL, 0.1 M), and sodium hydroxide (50 mL, 0.1 M) at the optimum pH and room temperature (25 ± 2°C). The recovery of the studied adsorbent after four uses reduced the RE of the adsorbent by only 4%. In a study by Seyedi et al on the recovery from adsorbents used in the removal of pentachlorophenol, after the third adsorption-desorption cycle, the removal percentage of PCP was reduced by only about 2.36% (21). In a study by Taghavi et al on the recovery of adsorbents used in the removal of trichlorophenol pollutants, the RE of this contaminant after four stages of adsorption and desorption decreased about 4% (25).

Investigating the effect of ultrasonic waves on the efficiency of diazinon removal in the adsorption process

Ultrasonic technology is one of the newest technologies available in water and wastewater treatment. Ultrasonic waves are a type of sound waves in which the wave motion is in the longitudinal direction. At frequencies above 20 kHz, the transmitted waves can cause changes in pollutants (20). Through ultrasound, the pore structure of adsorption materials can be fully utilized, and the mass transfer between adsorbents and adsorbates becomes more rapid (21). In this study, the effect of ultrasonic waves on increasing the efficiency of diazinon removal in the adsorption process was investigated. According to Figure 8, the RE of diazinon by ultrasonic waves alone was 15%, while the application of ultrasonic waves with the adsorbent for 15 and 60 minutes increased up to 70% and 90%, respectively. In a study by Hu et al on the removal of Acenaphthene by Palm shell activated carbon enhanced with ultrasonic waves at a frequency of 50 kHz and a power of 600 watts, a removal efficiency of more than 90% was achieved (39).

Study of adsorption isotherms

Various mathematical models such as Langmuir, Freundlich, Temkin, Elovich, Redlich-Peterson, and Langmuir-Freundlich are used to study isotherms and describe equilibrium reactions. (25). The Langmuir isotherm model is suitable for monolayer adsorption on an adsorbent surface with limited and homogeneous adsorption sites. The Langmuir isotherm is defined by the following equation:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{C_e K_L q_m} \tag{3}$$

Where q_e and q_m (mg/g) are the equilibrium and maximum adsorption capacities, respectively; K_L is the Langmuir constant and C_e (mg/L) is the equilibrium concentration of the adsorbate. The separation factor expressed by R_L contains the essential characteristics of the Langmuir isotherm and is calculated as follows:

$$R_L = \frac{1}{(1 + K_L C_0)} \tag{8}$$

Where C_0 (mg/L) is the initial concentration of the adsorbate. The R_L determines the nature of the adsorption process and is an indicator to determine the efficiency of this process. The adsorption is categorized as follows: $R_L > 1$, the adsorption process is undesirable; $R_L = 1$, the process is linear; $0 > R_L < 1$, the process is appropriate, and $R_L = 0$, the adsorption process is irreversible (24).

The Freundlich isotherm is an empirical equation suitable for the adsorption process on heterogeneous and multilayered surfaces. The linear form of this model is as follows:

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \tag{4}$$

Where K_f and n are related to the adsorption capacity of an adsorbent and sorption intensity, respectively (24,25).

The Tamkin isotherm model determines the effects of indirect adsorbent/adsorbate interactions on the adsorption process. According to this model, the heat of adsorption of all molecules in the layer decreases linearly due to the interaction of the adsorbent and the adsorbate (25). The linear relationship of the Tamkin isotherm model is presented as follows:

$$q_e = B_T \ln K_T + B_T \ln C_e \quad (5)$$

where B_T is the Temkin constant, which is related to the heat of sorption, K_T is the Temkin isotherm constant, and T is the absolute temperature (25).

Due to the higher R^2 correlation coefficient of the Langmuir isotherm compared to other investigated isotherms, the experimental and laboratory data of this study follow the Langmuir isotherm.

Study of adsorption kinetics

To better understand the mechanisms controlling the adsorption process and evaluate the performance of the rice husk biosorbent, PFO and PSO kinetic models were used in this study. Comparison of the experimental and theoretical data from the mentioned models was performed using the correlation coefficient (R^2). The equation of the PFO model discovered by Loggren is as follows:

$$\frac{{}^{d}q_t}{dt} = k_1(q_e - q_t) \quad (6)$$

In the above equation, K_1 is the rate constant (min^{-1}) and the parameters q_t and q are the adsorption capacity at each time t and at the equilibrium time (mg.g^{-1}). The PSO kinetic model, based on the proportionality between pollutant adsorption on active sites on the adsorbent surface, is expressed by the following equation (25).

$$\frac{t}{q_t} = \frac{1}{n} + \frac{t}{q_e} \quad (7)$$

Table 2 shows that due to the higher correlation coefficient ($R^2 = 0.984$) of PSO compared to PFO ($R^2 = 0.931$), the kinetic model of diazinon adsorption by the adsorbent used follows the PSO model.

Investigation of structural characteristics of the adsorbent produced from rice husk

FE-SEM analysis

Figure S3 shows the internal structure of the adsorbent produced from rice husk before and after the diazinon adsorption process. Considering these figures, the rough surfaces inside the adsorbent are visible and have high porosity, which is a good characteristic for an adsorbent in the adsorption process.

FT-IR analysis

In this study, using FT-IR spectra, different functional groups in the used adsorbent were investigated at wavelengths of 400 to 4000 cm^{-1} (Figure S4). The $^{-}$OH group is at the peak of 3418 cm^{-1}, which leads to the formation of weak hydrogen bonds. The peaks at 470 and 1098 cm^{-1} are related to Si-O compounds of silica in the adsorbent produced from rice husk (22). The observed peak at 1600 cm^{-1} is related to C = C bending vibration, which decreased with increasing temperature (25).

EDX analysis

In this study, the EDX test was used to determine the elements and chemical compounds present in the adsorbent. Therefore, using EDX, the types of elements and atoms present in the adsorbent produced from rice husk were identified. In Figure S5, the size and type of the spectra of different elements in the adsorbent are reported by EDX. In this method, each peak in the spectrum corresponds to a type of atom, and peaks with higher heights indicate a higher concentration of an element (25). According to this Figure, the highest concentration of the element in the adsorbent produced from rice husk is related to carbon.

BET analysis

Specific surface area is an important factor for the determination of the adsorption capacity of adsorbents. According to the IUPAC classification, diameters less than 2 nm are microporous, between 2 and 50 nm are mesoporous, and greater than 50 nm are macroporous (23). Based on Table S2, the total pore volume and the specific surface area in the adsorbent produced from rice husk by BET were determined as 0.2844 cm^3/g and 460.96 m^2/g, respectively.

In Table 3, the performance of different adsorbents in the process of diazinon adsorption is presented.

Conclusion

This research aimed to remove diazinon from aqueous environments using an ultrasound-enhanced adsorption process. In this study, under conditions of pH = 8, diazinon concentration of 20 mg/L, adsorbent dose of 2 g, contact time of 60 min, and temperature of 25 °C, an efficiency of 78% was achieved. In this study, under conditions of pH 8, diazinon concentration of 20 mg/L, adsorbent dose of 2 g, contact time of 60 minutes, and temperature of 25°C, an efficiency of 78% was achieved, which was increased to 90% using ultrasonic waves. According to the results obtained in the present study, the adsorbent produced from rice husk waste has a high ability to remove diazinon. In this study, the RE of diazinon by the adsorbent produced from rice husk did not decrease significantly with repeated use in the adsorption process. The results displayed that the Langmuir isotherm and PSO model were consistent with the experimental data. Studies of the structural parameters of this adsorbent, such as BET, FE-SEM, FTIR, EDX, and

Table 3. Comparison of different adsorbents in terms of removal performance

Adsorbent	pH	t(min)	dose(g/L)	Concentration (mg/L)	q_e (mg/g)	Efficiency (%)	References
Watermelon	6	30	1.0	0.00017	-	95.1	(40)
Activated carbon	5	30	10	20	71.4	95.2	(37)
NH4Cl-induced activated carbon	7	30	0.2	20	100	97.5	(38)
MK-BZK	3	15	10	10	2.25	97	(41)
Activated carbon (walnut shells)	7	10	0.4	60	-	55	(42)
Rice Husk- induced US	8	60	2	20	10	78	This study

XRD, confirm these results. Therefore, it can be proposed as a cheap, efficient, and available adsorbent for removing diazinon from wastewater containing diazinon pesticide, such as pharmaceutical industry effluent and agricultural wastewater.

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Competing interests
The authors declare that they have no conflict of interests.

Ethical issues
The Ethical Committee of the Babol University of Medical Sciences approved this study (Ethical code: IR.MUBABOL.REC.1402.136).

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Supplementary files
Supplementary file 1 contains Tables S1 and S2 and Figures S1-S6.

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