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Removal of Nitrate from Wastewater by Adsorption on Date Palm Waste (DPW)

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Abstract

The adsorption phenomenon is considered one of the most effective methods that can be used to eliminate the wastewater pollution that can have negative effects on the environment and human health due to the huge daily production of wastewater. Nitrate elimination is considered one of the most problems that we facing in the wastewater. In the present study, in order to efficiently remove nitrate, date palm waste (DPW) with high stability and good environmental adaptation was used as an adsorbent. A series of experiments were carried out in order to study the influence of date palm fiber on the adsorption capacity of nitrate, such as the contact time, the mass of adsorbent, the adsorbent particle size, and pH. From the studied parameters, we chose the optimum parameters to establish the kinetics and the isotherms of adsorption. The results obtained indicated that date palm fiber has good nitrate absorption capacity more than the ripes and the petioles.

Keywords: Adsorption, Date Palm Waste, Isotherm Adsorption, Kinetic Adsorption, Nitrate

1 Introduction

The amount of pollution spread in the world is directly related to the demographic explosion and technological development that the world knows. Environmental pollution can occur in many forms; air [1], soil, and water pollution [2], and it can be physical, chemical, or biological [3]. Water pollution is one of the most harmful pollution problems due to the great importance of water in our daily lives. Water is an important element in many domestic, industrial, and agricultural activities.

Nitrate is the stable ion form of nitrogen that is naturally occurring in the nitrogen cycle. Nitrate can be reduced by microbial action into nitrite (NO_2^-) or other forms, and nitrite can be oxidized to NO_3^- by biological and chemical processes [4]. Nitrate is a serious threat to drinking water supplies due to its high water solubility [5] and high concentrations [6]. The rise and fall of nitrate concentration in water compared to its natural ratio can pose a threat to our health and disturb our environment. In agricultural and industrial areas, water pollution by nitrates is an unavoidable problem. Drinking water or eating plants high in NO_3^- can lead to acute poisoning [7]. It may also be responsible for causing diabetes [8] and diverse kinds of cancers in humans [9]. Sewage and intensive use of fertilizers in agriculture can lead to the higher NO_3^- contamination of ground and surface water sources [10]. There are several physical, chemical, and biological methods to treat nitrate, such as coagulation, flocculation [11], chemical oxidation, ion exchange [12], electrochemical technologies [13], biodegradation, membrane filtration, and adsorption [14].

Among the previously mentioned techniques, the adsorption technique by solid adsorbents is considered one of the most efficient and preferential techniques for the treatment and removal of nitrate contaminants in water treatment. The adsorption phenomenon is characterized by simple design and low cost compared to other methods [15]. Adsorption is the surface phenomenon that removes organic and inorganic pollutants. It is carried out by the contact between a solution containing an absorbable solute and a solid with a highly porous surface structure. Due to the affinity between the adsorbate and the adsorbent, the attraction intermolecular forces cause some atoms, ions, or molecules of adsorbate to be deposited on the solid surface of the adsorbent [16]. The binding to the solid surface can be via physical bonds (physisorption) or chemical bonds (chemisorption [17].

In Adrar, an Algerian city located in the southwest, groundwater serves as the sole source of drinking water in urban areas and rural communities. Industrial and agricultural discharges can be a contaminant of water with a low ratio. It is therefore possible that they may remain undetected, thereby increasing the possible health risks. Therefore, it is necessary to limit this risk as much as possible by implementing an adequate treatment. The use of natural agricultural waste in adsorption

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without recourse to fabricated materials is considered a great challenge and is recommended for sustainable development in order to preserve the environment. The aim of this work is the elimination of nitrates present in water using date palm fiber as a local and available adsorbent.

2 Experimental

2.1 Raw substance characteristics

The raw substance, date palm wastes, used for the study of nitrate adsorption was obtained from the research unit in renewable energy in the Saharan medium, Adrar (figure 1).

Figure 1: Date palm wastes (DPW)

The raw substance was harvested from the same type of date palm variety called *Hmira*.

2.2 Preparation of adsorbent

The date palm waste (ripe, petiole, and fiber) washed good by tap water to remove the dirt stuck on the waste, then soaked in distilled water for two hours to dissolve as much as possible of salts contained in the samples. The samples dried in an oven at a temperature of 105° C for two hours, grinding by a grinder with low rotation speeds to avoid the heating of samples, then sieved through the sieve pore size. We obtained from each sample three sub-samples with pore size of 0.2, 1, and 2mm.

2.3 Adsorption parameters

A stock solution of nitrate of concentration 1000 mg/L was prepared by dissolving 1.37 g of $NaNO_3$ in 1000 ml of distilled water. Dilute nitrate stock solution from 1000 mg/L to 20, 30, 40 and 50 mg/L was carried out by using distilled water under dilution law.

The study of the adsorption parameters allows us to follow the adsorption phenomenon and its mechanism.

2.3.1The adsorbent nature effect

In this parameter, we studied the effect of date palm waste nature (ripe, petiole, and fiber) on nitrate adsorption. We prepared three beakers, each of them contains 1 g of adsorbent (ripe, petiole, and fiber) and 40 ml of nitrate solution with a 1000 mg/L concentration. After 2 hours of magnetic stirring, at room temperature, the three adsorbents (palm, diet and fiber) are filtered and analyzed by a UV-Visible adsorption spectrophotometer at λ_{max} 520 nm.

2.3.2 Contact time effect

The contact time was obtained by mixing 1 g of DPF with 40 ml of nitrate solution (1000 mg/L) under magnetic stirring for 1, 2, 3, and 4 hours. The mixture (DPF-Nitrate) was filtered and analyzed by a UV-Visible at λ_{max} 520 nm.

2.3.3 Mass effect

Four masses 0.1, 0.25, 0.50, 1, 1.5, 2 and 2.5 g of date palm fiber are used. Each mass was mixed with 40 ml of nitrate solution (1000 mg/L). The contact time was fixed at 4 hours according to the results obtained from the contact time effect on the nitrate adsorption by DPF.

2.3.4 Particle size effect

We weighed 1.5 g for each fiber size (0.2, 1 and 2 mm). Each mass was mixed with 40 ml of nitrate solution (1000 mg/L) and stirred for 4 hours.

2.3.5 pH effect

To study the influence of pH on nitrate adsorption, we adjusted the pH of solutions to the following pH values; 2, 5.77, 11 and 12. Under magnetic stirring; 1.5 g of fibers particle size 0.2 mm was mixed with 40 ml of nitrate solution (1000 mg/L) for 4 hours.

2.4 Adsorption isotherm

Four nitrate solutions were prepared with initial concentrations of 20, 30, 40, and 50 mg/L. In a beaker, 40 mL of each solution was mixed with 1.5 g of date palm fibers (0.2 mm). After 4 hours of stirring, the suspensions were separated by filtration and the supernatant concentrations were analyzed by a spectrophotometer at λ_{max} = 520 nm.

2.5 Adsorption kinetics

The nitrate adsorption kinetics on date palm fiber was carried out by mixing 1.5 g of date palm fibers (0.2 mm) with 40 ml of nitrate solution (20 mg/L). The mixture was placed under magnetic stirring for 4 hours. During the stirring process, samples are analyzed every 30 min by using a UV-vis spectrophotometer at 520 nm.

3 Results and discussion

3.1 Adsorption parameters

3.1.1 The adsorbent nature effect

The influence of date palm waste (ripe, petiole, and fiber) nature on the nitrate adsorption was studied to determine the best adsorbent among these wastes. From the results showed in the figure 2, we note that the fibers showed the highest adsorption ratio compared to the ripes and the petioles. The recorded adsorption capacities were; 0.266 for ripes, 3.733 for petioles, and 4.266 for fibers. These differences in adsorption capacity can be explained by the presence of more adsorption sites in the fibers than the ripes and petioles.

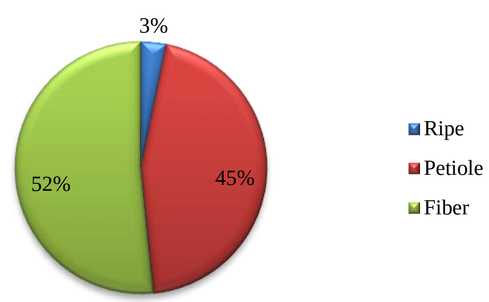

Figure 2: The effect of the adsorbent nature

3.1.2 Contact time effect

The contact time is a parameter that influences the adsorption yield and the adsorption equilibrium (adsorbate-adsorbent). The time effect allows us to determine the equilibrium time, which corresponds to the saturation of the active site.

As shown in figure 3, there is a correspondence between the contact time and the amount of nitrate absorbed. Based on the obtained results, the curve showed in the first hour of contact a rapid rise in the absorption capacity due to the large numbers of easily accessible active sites on the adsorbent surface. Because the kinetics of fixation are limited by the low concentration of adsorbate, this first step is an important part of the adsorption phenomenon [16]. In the phase limited between 1 and 4 hours, we saw a gradual rise until reaching the equilibrium state at 4 hours, with an absorption capacity (Qe) equal to 10 mg/g which remained constant after 4 hours of contact. In this phase, the occupation of the adsorption sites requires a diffusion of the adsorbate inside the adsorbent micropores [18] (deep sites or less accessible sites), which leads to a slow rise in the adsorption capacity before reaching the equilibrium state after the saturation of the adsorbent specific surface [19].

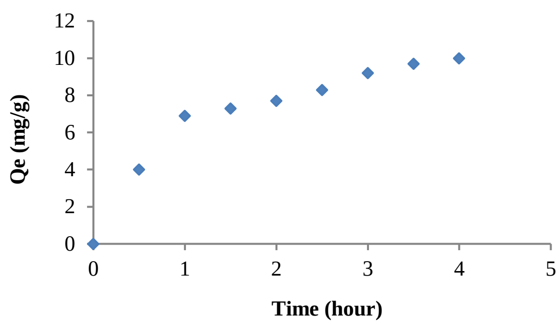

Figure 3: The effect of contact time on nitrate adsorption

3.1.2 Mass effect

The figure 4 shows the nitrate adsorption yield by different masses of date palm fibers at room temperature.

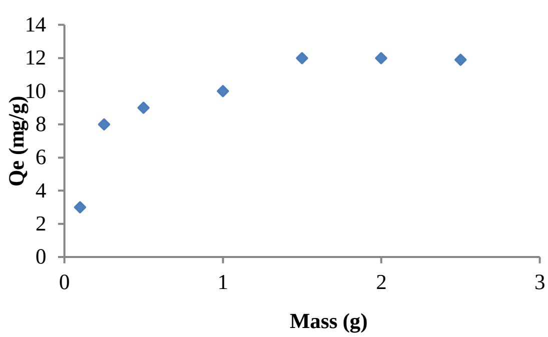

Figure 4: Mass effect of date palm fibers on the nitrate adsorption

The curve shows that the sorption capacity (Qe) of adsorption increases with the adsorbent mass up to 1.5 g, where the percentage remains constant. This behavior can be explained by the increase of adsorption sites due to the adsorbent mass increasing, which leads to the elimination of the nitrate solution.

3.1.3 Particle size effect

The particle size analysis of the date palm fibers was carried out by sieving in the range of 0.2 – 2 mm.

As shown in figure 5, the capacity of nitrate adsorbed is affected by the particle size of the adsorbent, where the adsorbent particles adsorb the adsorbate up to a stable value, after which the adsorption capacity remains constant (saturation). The external particle surface area decreases as the particle sizes increase, where the maximum adsorption capacity was recorded at a particle size of 0.2 mm. This is due to the increase in available surface area due to the narrowing of the intermolecular spaces and therefore gives the best adsorption capacity.

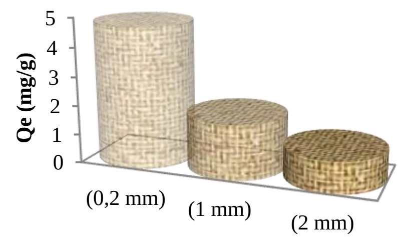

Figure 5: Particle size effect on nitrate adsorption

3.1.4 pH effect

The pH parameter is considered as a critical factor [20] during the adsorption process [21] [22]. The aqueous solution pH at pH: 2, 5.7, 11, and 12 was tested to evaluate the effect of pH conditions on the nitrate removal by DPF. The surface area charge is positive for solutions with a pH less than 5.7 and negative for solutions with a pH greater than 5.7.

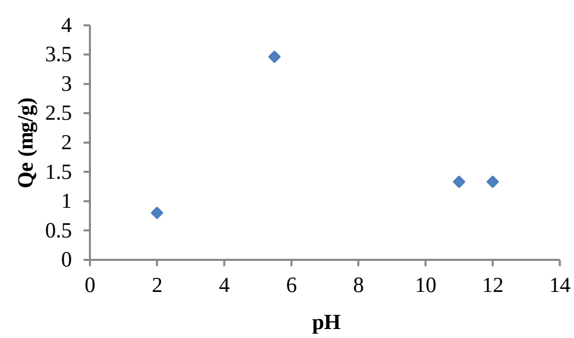

Figure 6: Effect of pH on nitrate adsorption

As shown in figure 6, the increase in pH increases the amount of nitrate adsorbed, where the higher adsorption was at pH 5.77, which corresponds to the pH of date palm waste [23]. At pH 5, a decrease in efficiency was observed due to the negative charge of the surface area and the competition between the hydroxyl and nitrate ions [24]. The electric charge of the absorbent is affected by the pH of the medium due to the ionisation of the surface functional groups [25].

3.2 Adsorption isotherm

Langmuir, Freundlich and Temkin adsorption isotherms are commonly used to describe the adsorption data of the adsorbate onto the adsorbent [26] [27]. Langmuir, Freundlich and Temkin adsorption isotherms were obtained from tracing the values of

C_e/Q_e versus C_e, $\log Q_e$ versus $\log C_e$ and Q_e versus $\ln C_e$ respectively (Figure 7).

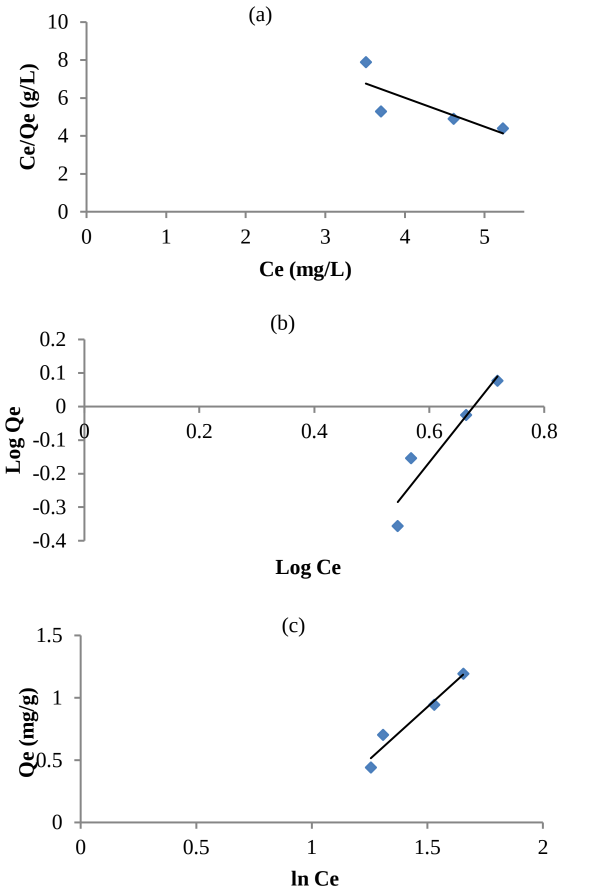

Figure 7: Adsorption isotherm models for nitrate solutions (20, 30, 40 and 50 mg/L) adsorption on DPF. "(a)" Langmuir, "(b)" Freundlich, and "(c)" Temkin

All the adsorption isotherms parameters with the correlation coefficients are listed in Table 1. These isotherm parameters allow determination of the equilibrium ion exchange/adsorption capacities of nitrate. Usually, the Langmuir isotherm is used to describe the adsorption mechanism of molecules in monolayers without adsorbate-adsorbate interactions [25], but the negative values for the Langmuir isotherm constants indicate the inadequacy of this isotherm to explain the adsorption mechanism. The Freundlich and Temkin models would be applicable. The correlation coefficients R^2 obtained indicate that the adsorption of nitrate by date palm fibers may be successfully achieved by the Langmuir, Freundlich, and Temkin isotherms. Both the Freundlich and Temkin isotherms presented correlation coefficients close to unity. The constant related to the adsorption energy is low, indicating that the adsorption process is physical (physisorption).

As shown in figure 8, the adsorption equilibrium of nitrate onto DPF was reached in 120 min, which corresponds to the high rate of nitrate adsorption. Usually, the adsorption rate mainly depends on the contact of nitrate and the adsorbent's active sites. The particle sizes of DPF and dilute concentration of nitrate contributed significantly to nitrate contacting with active sites, so as to reach the adsorption equilibrium in short time.

3.3 Adsorption kinetic

Adsorption usually occurs between the functional groups on the surface of the adsorbent and the adsorbate [28]. As shown in table 2, the experimental Qe values do not agree with the calculated Qe obtained from the linear plots of the first order, while they agree with the calculated Qe obtained from the linear plots of the second-order kinetic model, which also explains why the second-order model totally agrees with the experimental data. The second-order nitrate adsorption reaction was based on physical sorption via the sharing or exchange of ions between adsorbent and adsorbate [31].

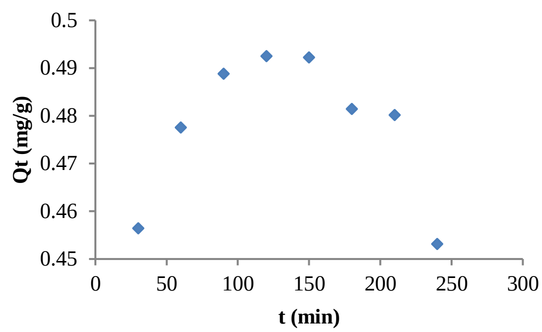

Figure 8: Contact time of nitrate solution (20 mg/L)

Table 1: Langmuir, Freundlich and Temkin parameters for adsorption of nitrate on DPF

Langmuir parameters			Freundlich parameters			Temkin parameters		
R^2	K_L (L/mg)	Q_m (mg/g)	R^2	K_F (mg/g)	n_F	R^2	A_t (L/mg)	b_t (KJ/mol)
0.619	- 18.488	- 0.655	0.886	0.034	0.463	0.948	0.387	1.476

'Q_e' is equilibrium adsorption capacity (mg/g or mmol/g). 'C_e' is equilibrium concentration in solution (mg/L or mmol/L). 'Q_m' is maximum adsorption capacity of the adsorbent (mg/g). 'K_l' is Langmuir adsorption isotherm constant (L/mg or L/mmol). 'K_f' is the adsorbent–adsorbate relative affinity in the adsorption process. 'A_t' is equilibrium binding constant (l/mg). 'b_t' is adsorption energy (J/mol).

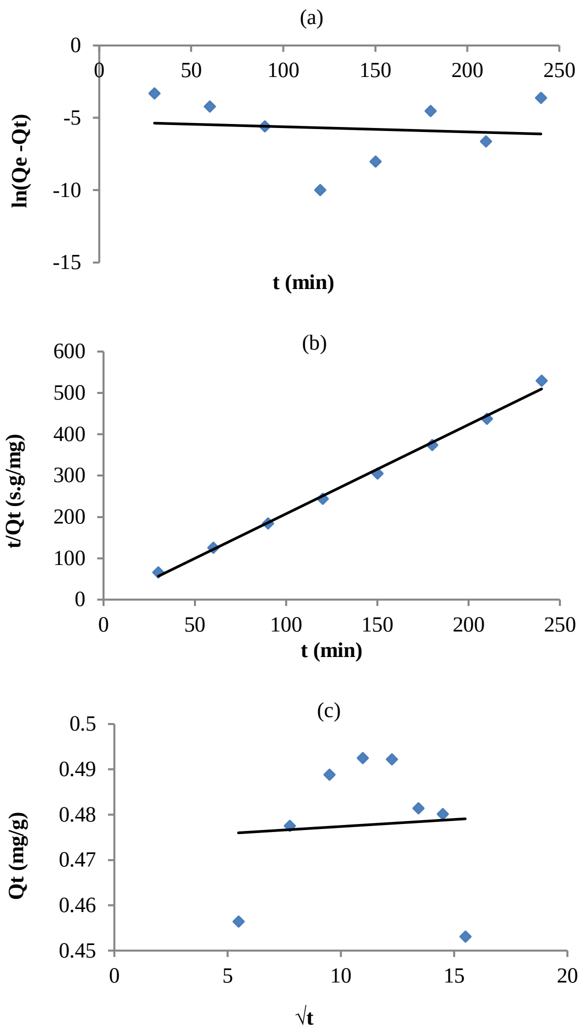

Figure 9: Adsorption kinetic for nitrate solution (20 mg/L) adsorption on DPF. “(a)” the first-order, “(b)” the second order, and “(c)” intraparticle diffusion

The reaction order is a very important parameter in determining reaction mechanisms. In order to follow the analysis of nitrate adsorption kinetics onto DPF, three kinetic models were studied; first order, second order, and the intraparticle diffusion model. Both first order and second order equations are used to describe the curve of adsorption kinetics, while intraparticle diffusion is used to control the adsorption rate [29] [30]. The adsorption kinetic of nitrate by DPF was checked by tracing the evolution of log (Q_e-Q_t) versus t, (t/Q_t) versus t, and Q_t versus $t^{1/2}$ respectively, with the correlation coefficients for each curve shown in Figure 9.

The first-order model and intraparticle diffusion model were not valid for the nitrate adsorption due to non-linear progression with low correlation coefficients of these models. The second order was valid for the nitrate adsorption due to the linear progression with a correlation coefficient greater than 0.995.

4 Conclusions

The aim of this work is to study the elimination of nitrate by using date palm fiber (DPF), a waste residue produced in large amounts during the pruning of palm trees in the season of cultivation, as an adsorbent in this work. Several parameters were studied, such as contact time, adsorbent mass, particle size, and pH solution. This study was also supported by the study of isotherm and kinetic adsorption. From the results obtained, we see that the date palm fibers showed a good adsorption capacity for nitrate, represented by an optimal contact time of 4 hours, an optimal mass of 1.5 g, and an optimal size of 0.2 mm. Adsorption of nitrate was favorable at pH 5.77. The adsorption isotherms modeling allowed us to say that the Freundlich and Temkin models perfectly simulate the physical adsorption of nitrate. The adsorption kinetics indicates that the second-order model gives the best description of nitrate adsorption onto the DPF surface.

Aknowledgment

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Ethical issue

Authors are aware of, and comply with, best practice in publication ethics specifically with regard to authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests and compliance with policies on research ethics. Authors adhere to publication requirements that submitted work is original and has not been published elsewhere in any language.

Competing interests

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

Authors’ contribution

All authors of this study have a complete contribution for data collection, data analyses and manuscript writing.

Table 2: Values of adsorption rate constants for nitrate (20 mg/l) adsorption on DPF

$t_{(contact.)}$ (min)	Q_e exp (mg/g)	First-order kinetic model $\ln(Q_e - Q_t) = \ln Q_e - k_1 t$			Second-order kinetic model $\frac{t}{Q_t} = \frac{t}{Q_e} + \frac{1}{k_2 Q_e^2}$			Intraparticle diffusion model $Q_t = K_{id} t^{1/2} + C$		
		k_1 (min^{-1})	Q_e (mg/g)	R^2	k_2 (g $mg^{-1}.min^{-1}$)	Q_e (mg/g)	R^2	k_{id} ($mg.g^{-1}.min^{-1/2}$)	C	R^2
120	0.440	0.003	0.005	0.012	- 0.572	0.463	0.995	0.0003	0.474	0.004

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