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Application of cane bagasse adsorption on nitrate removal from groundwater sources: adsorption isotherm and reaction kinetics

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ABSTRACT

Removal or reduction of nitrogen compounds in natural waters resulted from discharge of industrial and agricultural activities, which has environmental and human health problems, is necessary. In this study, the effect of initial nitrate concentration (15.5, 50, 80, 120 and 200 mg/L), pH (1–7) and contact time (4.5–36 h) on the adsorption efficiency of cane bagasse modified by $ZnCl_2$ was investigated to remove nitrate from groundwater sources in Izeh city, Khuzestan province of Iran. Adsorption performance was evaluated using Freundlich, Langmuir and Bet isotherms and the first-order and pseudo-second order feed removal models. Results showed that for modified bagasse, the system reached equilibrium after 32 h and the maximum adsorption occurred at pH = 4. Also, the removal efficiency increased by decreasing the concentration of nitrate. The adsorption process followed the Langmuir isotherm. The results of the study showed that the modified bagasse fiber absorbent has a great potential for nitrate removal. Because of the inexpensiveness of bagasse fiber and especially bagasse abundance in Khuzestan province, it is suggested to remove nitrate from groundwater of Izeh city via cane sugar (bagasse) modified by zinc chloride.

Keywords: Cane bagasse; Nitrate; Groundwater; Isotherm; Kinetics

1. Introduction

The increasingly growing industrial and agricultural activities have led to the introduction of various organic and mineral contaminants into aquatic ecosystems. So, the protection of the environment, consequently and public health have been a major challenge facing human societies. Nitrate can be mentioned as the main form of nitrogen, which is among the pollutants that its presence in excessive amounts in the environment has a negative impact on the ecological status of the waters, the life of aquatic organisms, and public health [1]. Nitrogen is considered as an essen-

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tial nutrient for plants and living organisms. However, the excessive entrance of nitrogen compounds through urban, industrial and agricultural wastewater is one of the most important sources which threatens water quality [2]. Due to its structural properties and negative charge, nitrate is easily leached out via irrigation water and reaches to the groundwater sources. The accumulation of nutrients (specifically nitrogen compounds) in surface and groundwater results from sewage discharges, could cause many problems for aquatic ecosystems, consequently human and animal health [3]. Some of the adverse impacts include eutrophication, destruction of water quality, increasing treatment costs, decrease in dissolved oxygen concentration from surface waters and complications on human health, and in some cases even death among newborns under the six months age [4–10]. Therefore, it is very necessary to eliminate or reduce nitrogen pollutants in the receptor environments. Conventional wastewater treatment could not always provide the required quality in terms of discharge standards for wastewater nitrogen compounds. This is especially true when entering considerable amounts of wastewater into water resources which considered as a major problem. Various procedures are used to remove nitrate from contaminated water, including denitrification [11], adsorption [12], ion exchange [13], distillation [14], reverse osmosis [15], etc. Nowadays investigations are aimed at finding simple and inexpensive methods for application in treatment of contaminated water, especially in developing countries, which are unable to use costly methods due to economic problems. The adsorption process gained much attention in comparison with other treatment techniques in terms of simplicity and flexibility in design, easy operation, low initial cost, reuse of wastewater, etc. [16].

The process does not influence by target pollutant toxicity and does not require any hazardous chemicals. In addition, the absorbed contaminant can be recycled, which is useful in some cases [17]. In recent years, the use of natural adsorbents has been developed to eliminate all kinds of contaminants from organic and inorganic compounds, good results have been achieved [18–23]. One of the natural absorbents is bagasse. Bagasse or sugar cane is a by-product of cane extraction that contains chemical compounds, including cellulose, hemicellulose, lignin, sulfur, potassium, etc., which has the ability to absorb contaminants with associated sites. According to studies, 99% of cane sugar in Iran, which is over 100 ton/ha, is produced in Khuzestan province, Iran [24]. Many studies have emphasized the use of bagasse in the removal of pollutants [19,25,26].

Modification of bagasse with various chemical agents via increasing the functional groups and the diffusion of internal particles (increased porosity) improves its efficiency compared to the unmodified form. According to the above, this study investigated the application of modified bagasse under different operating conditions, including equilibrium time, pH and concentration of nitrate inlet on nitrate adsorption and in parallel, the kinetics of adsorption and adherence to adsorption parameters through adsorbent isotherm with the aim of removing nitrate from groundwater sources of Izeh city. The aim of the present study was the determination of cane bagasse adsorption efficiency in nitrate removal from groundwater of Izeh city, Khuzestan province of Iran.

2. Materials and methods

2.1. Type of study

In this experimental study, the adsorption potential of zinc chloride- modified bagasse in nitrate-nitrogen removal from groundwater sources in a laboratory scale and batch was studied.

2.2. Area studied

The city of Izeh has a population of 140,000 that at present, drinking water at an average of 29500 m^3/d is provided by through the number of 13 wells. Due to the fact that the main problem of increasing nitrate in the wells of Izeh is reported, therefore, samples were taken from Jamoshi well.

2.3. Regent and chemical

All chemicals used were of analytical reagent grade and were purchased from Merck Company, Germany. Synthetic solutions were prepared with deionized water.

Instruments

- Dry Heat, UN55 model, Memmert Co, Germany
- pH meter, 60 model, Jenway Co, England
- Elemental analysis O/CHNS, Vario EL III, Elementar Co, Germany
- Digital Balance, GF=2000, AND Co, Japan
- Spectrophotometry, UV2100 model, unique Co, America

2.4. Operation

Baggase was prepared from Amirkabir sugar cane development fields located at Ahwaz-Abadan Road. In order to increase the adsorption sites and product performance, bagasse was rinsed well with distilled water and then washed at 105°C for 6 h. Chemical activation was carried out using zinc chloride ($ZnCl_2$) for 8 h at room temperature. In the next step, it was placed in an oven at 115°C for 24 h in order to dry completely. It was then washed with a 1.2 molar $CaCl_2$ solution. It was washed three times with distilled water to reach a constant pH and dried in an oven for 24 h at 115°C (18). Finally, it was kept in glass containers in the package for the experiments. It should be noted that all stages of sampling, sample storage and testing were carried out according to standard methods [27].

2.5. Experimental method

It should be noted that nitrogen, carbon, and hydrogen in modified and un-modified bagasse absorbent were determined by elemental O/CHNS analysis (Table 1). Determination of the anionic iodine value of produced adsorbent conducted using the standard method ASTM D4607-94 [28]. The methylene blue adsorption test on modified bagasse adsorbent was also carried out using a discontinuous process [29]. Water solubility of modified and inactive bagasse absorbent was determined using ASTM D5029-98 standard [30].

Table 1
Elemental parsing of baggase in active and inactive mode

Adsorbent	C, %	H, %	N, %
Inactive	29.43	6.1	0.85
Modified	38.25	7.8	4.65

First, we carefully weighed a certain amount of each absorbent (50 g) with a digital scale and mixed it with 500 ml of water and boiled for 20 min. The cooled solution was filtered and 100 ml of the filtered sample was poured into a pre-weighed cleaned container and the container having the mixture was placed in an oven at 150°C for 60 min in order to evaporating the solution and to completely drying the residual. After drying, we re-weighed the container and the residual. The weight difference of the container was determined in the un-contaminated state and after drying, which is equal to the dry residue. Then, from Eq. (1), the solubility of water in each of the adsorbents was determined.

$$Solubility\,in\,water = \frac{(B-A)\times(D)}{(C)\times(E)}\times 100 \quad (1)$$

where A: initial weight of the container, g; B: The initial weight of the container and dry residue, g; C: Amount of consumed water, ml; D: adsorbent dose, g; E: amount of filtered sample taken, ml.

To determine the density of the adsorbent, first weigh out a specific volume of each adsorbents and then put the samples in an oven for 24 h at a temperature of 110°C to drying. After drying we reweighed the dried specimens. The weight difference of the samples is equal to dry weight before and after drying. Finally, the density of adsorbents was determined, using Eq. (2) [31].

$$\rho = \frac{m_s}{V_t} \quad (2)$$

where m_s Dry sample weight, g; V_t = total volume, ml; ρ = density, g/ml.

The method used in the D2867-99ASTM [30] standard was utilized to determine the mass moisture content of the adsorbents. In order to determine the moisture content of each adsorbent, certain amount of absorbent weighed and placed in an oven at 105°C for 24 h to dry the specimen. After cooling the specimen, the container was weighed again. Eq. (3) was used to determine the moisture content of the adsorbent mass.

$$\text{Mass moisture, } \% = \frac{(\text{C}-\text{D})}{(\text{C}-\text{B})}\times 100 \quad (3)$$

where B: The initial weight of the container, g; C: The weight of the container and initial sample, g; D: The weight of the container and dried sample, g.

Adsorption of nitrate from equilibrium solution by bagasse-modified fibers under adsorbent mass and initial concentration of nitrate (5–120 mg/l), pH and equilibrium time conditions were investigated. The samples were centrifuged for 5 min at 5000 rpm and passed through filter and analyzed by the spectrophotometer device [25].

2.6. Adsorption isotherm

Langmuir and Freundlich models are shown in Eqs. (4) and (5) [18].

$$\frac{C_e}{q_e} = \left(\frac{1}{k_1 q_m}\right) + \left(\frac{1}{q_m}\right) C_e \quad (4)$$

where Q_e: amount of the component absorbed per unit of mass, mg/g; C_e: equilibrium concentration of absorbent in solution after adsorption, mg/g; Q_m: absorption capacity; K: Langmuir constant.

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

where C_e: Concentration balance, mg/l; q_e: Capacity at the time of equilibrium, mg/g; n and K_f: Freundlich constant

2.7. Kinetic adsorption

Adsorption kinetics are one of the important characteristics in the evaluation of adsorption efficiency. Eqs. (6) and (7) show the relations between two first-order and pseudo-second-order models, respectively. In order to obtain the kinetics of adsorption, the adsorption charts of proposed models were plotted and kinetic model was obtained based on the data overlaying [19].

$$\ln(q_e - q_t) = \ln(q_e) - K_f \cdot t \quad (6)$$

$$\frac{t}{q_t} = \frac{1}{\left(K_s\, qe^2\right) + \frac{t}{q_e}} \quad (7)$$

where q_t and q_e: The rate of nitrate absorption was in time t and balance time (mg/g).

3. Results

3.1. Adsorption characteristics

Results of elemental analysis showed that 29.43% of the bagasse consisted of inactive carbon, while this ratio increased in modified bagasse, which accounted for 38.25% (Table 1). Determination of the bagasse water solubility showed that bagasse is not soluble in water, so that no solubility is observed. Also, the density and moisture content (w/w) of modified bagasse was determined to be 180 kg/m^3 and 12% respectively. Significant increase in nitrogen content in modified bagasse shows that the reaction between the adsorbent with produced ammonium compound is efficiently carried out and ammonium groups are formed rapidly in modified bagasse. Similar results were obtained in Gao et al. study for preparation of anionic adsorbent from rice stalk, pure alkaline, lignin and sugar cane bagasse, in which the nitrogen content was in the range of 5.8–0.36% [32]. Hafshejani et al. [20] obtained carbon, nitrogen, sulfur, and oxygen content in modified cationic bagasse as 56.2, 4.1, 0.8 and 21.1%, respectively. The higher the carbon content, as compared to other elements, reflects the presence of carbon in functional groups such as carboxylic. According to Mohan et al. [33], and considering to carbon content, zinc chloride-modified bagasse was in the 3rd carbon class, which is less than Hafshejani et al. [20] and Kanawade et al. [34] findings.

3.2. pH effect

The soluble acidity is among the factors affecting adsorption. The adsorption process is influenced by the pH due to the change in adsorption surface charge. Fig. 1 shows the variations in the amount of absorbed nitrate as a function of the primary pH. As can be seen, for $ZnCl_2$-modified bagasse adsorbent, the highest removal rate at pH = 4 was 77%, and the removal efficiency decreased by increasing pH value. In this case, absorbent had positive charge. The results showed that the efficiency of nitrate removal decreased with increasing pH. Also, the increase in removal efficiency by decreasing pH is due to positive charge in absorbent surface in lower pH From pHzpc, and given the fact that nitrate is in the anionic compounds group, it causes electrostatic gravity of the adsorbent and the desired pollutant, thus increasing the efficiency [35]. On the other hand, at pH greater than pHzpc, the negative charge of the adsorbent surface increases and creates a repulsive force between the adsorbent and the anionic contaminant and consequently resulted to decrease in removal efficiency. Tahir et al. [19] studied the surface adsorption of bagasse for removal of color and reported similar results. As the pH increased, the adsorption rate decreased and the highest adsorption was achieved in alkaline medium. Saad et al. [36] reported optimal pH for the adsorption of methyl bromide with bagasse in the range of 3–6, although the process less affected by pH.

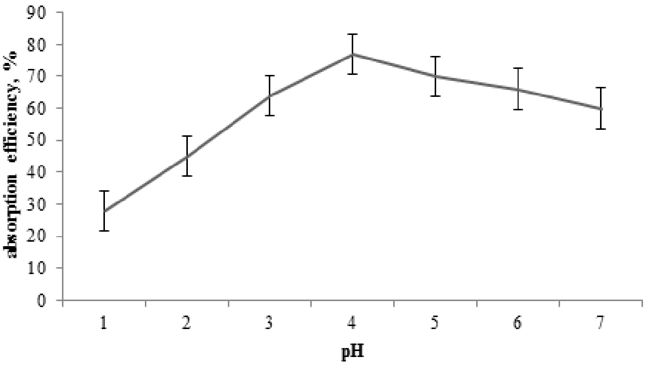

Fig. 1. pH effect on nitrate adsorption by modified bagasse.

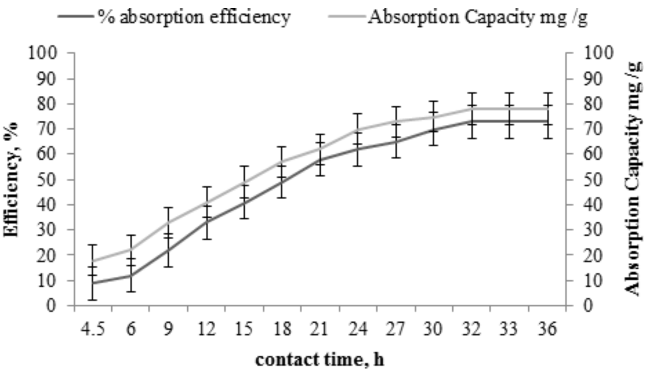

Fig. 2. Changes in efficiency and absorption capacity based on the time of equilibrium for modified bagasse.

3.3. Contact time

After determining the optimal pH, the effect of the equilibrium time on nitrate adsorption by modified bagasse adsorbent was studied. At this stage, the mass of 3 g and concentration of 20 mg/L and pH = 4 were considered as constant parameters and the contact time was changed from 4.5 to 36 h. Fig. 2 shows the changes in nitrate adsorption efficiency with time variations by bagasse adsorbent. The nitrate removal percentage is directly related to contact time, means that increasing the contact time from 3 h to 32 h, removal efficiency increased due to the abundance of adsorption sites and the high difference between the concentration of the absorbed substance in the solution and its amount in absorbent surface. In most studies that have been carried out on the adsorption process, the process efficiency is directly related to contact time [37,38]. It can also be stated that by increasing the time, the opportunity for effective collisions and random movements of nitrate molecules increased, which subsequently increased the chance of collisions with active positions and increased adsorption [39]. On the other hand, van der Waals forces are strengthened between nitrate and bagasse. But over time, the adsorption process becomes linearly equation due to the formation of a contaminant layer at the adsorbent surface. So that the time remains ineffective in the adsorption efficiency. Over time, it is difficult to occupy the remaining empty surface area, since a repellent is produced between adsorbed molecules and molecules that are in the soluble phase [20]. According to the results of the experiments, absorbance amount does not change for absorbent after 32 h. In this case, the amount of nitrate-nitrogen absorbed on the adsorbent surface will be equal to the amount of nitrate-nitrogen present in the solution phase. So the contact time of 32 h obtained as the equilibrium time. Samiksha and Mane [39] studied the removal of chemical oxygen demand via bagasse and concluded that, with increasing contact time, the oxygen demand was reduced and obtained the equilibrium time as 150 min. The study of Soleimani et al. on the removal of groundwater nitrate with mineral adsorbents had equilibrium time of 24 h [40]. Hafshejani et al. [20] also investigated the adsorption of nitrate ions by bagasse, and concluded that the removal efficiency was reached to equilibrium after 60 min. The equilibrium time in this study is less than recent studies. The difference in the findings is related to the structural variation of the utilized adsorbents and their chemical compositions. One of the reasons for the higher equilibrium time in the present study than other studies can be due to the type of contaminant or absorbent structure. Generally, the larger the size of the adsorbent molecule, adsorption is increased due to the higher charge density.

3.4. Initial concentration nitrate

The effect of initial concentration of nitrate on adsorption of nitrate ions by adsorbent, was investigated through

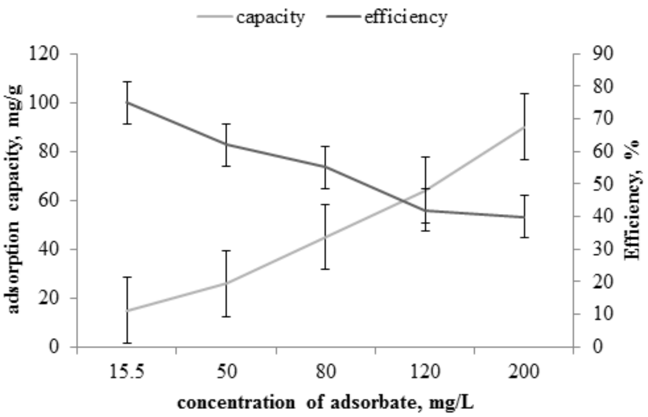

Fig. 3. Efficiency and adsorption capacity based on the initial concentration of nitrate for modified bagasse.

variations in nitrate concentrations (15.5, 50, 80, 120 and 200 mg/l), and the optimum mass of each adsorbents at 32 h equilibrium time for modified bagasse and in optimum pH. Considering Fig. 3 for modified bagasse, it was also found that by increasing the concentration from 15.5 to 200 mg/L, the efficiency decreased from 75% to 40%, while in this case the adsorption capacity showed increasing trend. This indicates the fact that adsorption is strongly a function of the initial concentration of nitrate.

The reason for this is that increasing the adsorbates on absorbent, available adsorption sites on the adsorbent surface will be reduced, which will consequently reduce the efficiency of the removal. Also, due to the possibility of a collision between the adsorbent and the absorbate, the adsorption capacity of the adsorbent will also increase so that some of the contaminants are not absorbed from and will out of the process. Also, in the study of Hafshejani et al. [20], with increasing initial concentration, the removal efficiency is reduced rapidly. The study by Alavi et al. [18] confirms the results of the present study so that the adsorption efficiency increases with decreasing initial concentration and decreasing absorbate volume. In the study of Soleimani et al., with increasing initial concentration of fluoride, removal efficiency with bagasse adsorbent decreased and adsorption capacity increased [40].

3.5. Adsorption isotherm

Regression coefficients in nitrate adsorption were determined using active bagasse absorbent for Langmuir, Freundlich and Bet isotherm models (Table 2). The observed values of the regression coefficient show that the active bagasse absorbent follow the Freundlich and Bet isotherms ($R^2 = 0.96$) and Langmuir ($R^2 = 0.93$). The study found that Langmuir's data matching was less than Freundlich, which contradicts Aloma's results [41]. The results of the Tahir study are in agreement with the present study, so that the adsorption does not follow Freundlich and Langmuir models [19]. When the desired isotherm is obtained, it means the process of mass transfer from the solid phase to the liquid phase. According to results, the higher adsorption of Freundlich isotherm shows that the adsorption process is carried out in a multilayer and homogeneous surface. The *n* value represents the spatial distribution of energy sites, which *n* value more than 1 representing high energy sites [35]. In this study, the *n* value was close to 1.

Table 2
Regression of Isotherm model

Isotherm model	Parameter	Modified bagasse
Langmuir	Q_0 (mg/g)	76.6
	b (L/ mg)	0.8
	R^2	0.93
	RMSE	1.15
Freundlich	K_f (L/ g)	73.33
	n	0.85
	R^2	0.96
	RMSE	0.02
Bet	R^2	0.96
	RMSE	1.85

3.6. Kinetic

The adsorption rate can be described by kinetic parameters to predict the adsorption mechanism. The dynamics of the nitrate adsorption process was described by modified bagasse with first-order and pseudo-second-order adsorption models. Table 3 shows the constants of kinetic models and regression coefficients for first-order and pseudo-second-order models for three concentrations of 100, 150 and 200 mg/L in bagasse, activated with $ZnCl_2$ with a ratio of 1–2. In order to investigate the correlation of the kinetic adsorption model, we need to consider two factors of regression coefficient (R^2) and the difference between the actual adsorption rates in terms of q_e (real) equilibrium and the concentration obtained by models q_e (CaL). In the present study, nitrate adsorption via modified bagasse fibers is at most below 16%, which confirms that the results of experiments are consistent with the pseudo-second-order adsorption model. The results of this study are consistent with the results of Arselen [42] and Zheng [43].

The results of the present study are in consistent with other studies (Table 4).

4. Conclusion

The results of current study showed that bagasse can be successfully used in groundwater nitrate treatment. According to the results, the efficacy of bagasse in nitrate removal is better in acidic pH and the adsorption reaction has been achieved in a 32-min interval. Also, removal percentage decreased with increasing initial concentration of ammonia nitrogen and decrease of absorbent dose. Maximum nitrate removal was obtained in pH: 4, contact time: 32 h and nitrate initial concentration: 5 mg/L. Isotremic studies and kinetics of the nitrate uptake process showed that bagasse adhered to Langmuir isotherm and kinetics of psudo first and second order kinetic. Regarding to many sources of sugar cane production in the country, especially in Khuzestan province, according to a report from the Agri-

Table 3
Constants and absorption kinetic regression coefficients for nitrate absorption by modified bagasse

Nitrate concentration mg/L	q (mg/g) (actual)	First-order kinetic model				Second-order kinetic model			
		$10^{-3} \times K_f$ (min^{-1})	CaLq (mg/g)	R^2	real (q_e) CaL (q_e)	$10^{-4} \times K_s$ (g·mg^1·mi)n^{-1}	CaLq (mg/)	R^2	real (q_e) CaL (q_e)
100	87.51	7.75	70.5	0.991	17.2%	1.45	85.5	0.982	2%
150	86.3	10.5	78.4	0.984	12.3	1.25	83.06	0.962	3.33%
200	86.2	14.3	115.5	0.973	29.3%	3.45	89.9	0.971	3.7%

Table 4
Literature references about use of sugar cane bagasse as an adsorbent

Absorbent	Adsorbate	Equilibrium time, min	PH optimum	Optimum adsorbate dosage, mg/L	Ref.
Sugar cane bagasse fly ash	Malachite green	20		8×10^6	[19]
Sugar cane bagasse	Oil-Polluted Water	180	3	200 mg/L	[44]
Modified sugar cane bagasse biochar	Nitrate	90	2–11	1–40	[20]
Modified bagasse with $NaHCO_3$	Froride	60	7	2	[45]
Sugar bagasse	Phenol	50	3	2	[41]

cultural Jihad Organization of Khuzestan province it currently has over 145,000 hectares of sugar beet cultivated in the province, and an annual production plan of 110,000 tons of cane sugar in the province and a large amount of cane sugar (bagasse), So using bagasse will be cost effective. Therefore, the bagasse is suggested order to remove the nitrate from water resources especially in Khuzestan province (Izeh city) due to its great resources.

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