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COD reduction of potato processing wastewater using natural and commercial adsorbents

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ABSTRACT

Adsorption kinetics and equilibrium of chemical oxygen demand (COD) reduction from a potato processing wastewater using activated carbon as a commercial and clinoptilolite zeolite as a natural adsorbent was investigated in a batch system. The effects of different parameters such as COD initial concentration, contact time, and adsorbent dose in COD removal from wastewater was studied. The results showed that the COD removal percentage increased with increasing contact time and adsorbent dose. Maximum COD removal was 79% and 70% for COD initial concentration of 2,081 mg/L for activated carbon and clinoptilolite zeolite, respectively. According to study results, the pseudo-second-order and Freundlich were the most suitable models for describing the kinetic and equilibrium experimental data, respectively. This research demonstrated that clinoptilolite is a cheap, plentiful and high adsorptive capacity adsorbent that could be used as an alternative to commercial adsorbents such as activated carbon for COD reduction. The study results confirmed that natural zeolite has high adsorption capacity as same as activated carbon.

Keywords: Activated carbon; Clinoptilolite zeolite; Adsorption capacity; Pseudo-second-order model; Freundlich model

1. Introduction

In recent years, an increase in population, industrialization and commercial growth in developing countries such as Iran have increased not only water demand but also water pollution using the various pollutants. Among different industries, the food processing industry leads to the generation of large quantities of wastewater [1]. The potato processing plants use high volumes of water during different operation processes such as washing, peeling, blanching, slicing, and shredding of potato [2,3]. The potato processing industry consumes vast quantities of water and produces large quantities of wastewater consists high amounts of biodegradable compounds such as carbohydrates, starches, proteins, vitamins, amino acids, and sugars [4]. This complex wastewater has high chemical oxygen demand (COD), biochemical oxygen demand (BOD) and suspended solids [5–9]. The untreated effluents, when discharged into water bodies, cause an increase in water pollution. Therefore, it is necessary to treat these wastewaters before discharging into the environment. Various treatment technologies such as filtration, ion exchange, coagulation/flocculation, reverse osmosis, membrane bioreactor, electrocoagulation and electrodialysis have been used for COD reduction of wastewater [10–14]. However, it is mandatory to develop high-efficiency and low-cost techniques in wastewater treatment processes. Adsorption method due to its low operational costs and high availability is an effective and versatile method for the removal of various pollutants from wastewater [15–18]. In recent years, several porous materials such as activated carbon, mesoporous silica, zeolite, and biochar have experimentally been studied in wastewater treatment processes [19–23].

Among the different porous materials, activated carbon and natural zeolite are favorable for a wide range of

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applications because of their high removal ability of both organic and inorganic pollutants from wastewater [23–27]. Clinoptilolite zeolite, a mineral comprising silica and alumina tetrahedral microporous complex which naturally found in volcanic ashes, has significant adsorption properties [9]. Furthermore, the microporous structure, special surface reactivity and high surface adsorption capacity of natural zeolite make it a potential adsorbent for a wide range of pollutants [10,28]. Several studies have performed on the use of natural zeolites and activated carbon in water and wastewater treatments. Some researchers compared the adsorption efficiency of pecan shell-based granular activated carbon and commercial carbon for COD removal of municipal wastewater [19]. The performance of date-pit activated carbon for eliminating COD from a petroleum refinery wastewater in batch reactors was studied by some researchers too [10,12]. Omer et al. [22] evaluated the adsorption efficiency of clinoptilolite for COD and BOD reduction of leather tannery effluent. The results showed that the COD and BOD removal percentages were 65% and 75%, respectively.

Several studies were conducted on properties of activated carbon in COD removal but in this research, the main goal is the comparison of the ability of natural Iranian zeolite as a cheap adsorbent as compared to activated carbon in COD removal of potato wastewater due its high starch content has high treatment.

Therefore, the effects of operational parameters including initial COD concentration, contact time, and adsorbents doses of two adsorbents was investigated. Moreover, Freundlich, Langmuir, and Temkin isotherm models and pseudo-first-order, pseudo-second-order and Elovich kinetics equations were applied to describe COD removal ability of natural Iranian zeolite as compared to activated carbon.

2. Material and methods

2.1. Wastewater characteristics

The raw wastewater was obtained from a potato processing plant in Isfahan province, Iran. Some characteristics of the raw wastewater are shown in Table 1. During experiments, samples of wastewater were maintained at a temperature below 4°C to avoid any change in the physicochemical characteristics. Initial wastewater had COD of 4,000 mg/L. With a combination of this wastewater with deionized water, three concentrations of 510; 1,080; and 2,081 mg/L were supplied.

2.2. Adsorbents

Powder activated carbon termed as 'H carbon' was purchased from Merck (Germany). The pH_{zpc} of the activated carbon was found to be 8.2 and exhibit positively charged surface when the solution pH is below 8.2 and negatively charged surface while pH is over 8.2. The Iranian clinoptilolite zeolite applied in this study was obtained from the Firoozkooh mine, northeast of Firoozkooh city. Table 2 illustrates the chemical composition of the Iranian clinoptilolite zeolite applied in this research.

The structural characterization of activated carbon and Iranian clinoptilolite zeolite was investigated using different analysis methods such as specific surface area Brunauer–Emmett–Teller (BET) and scanning electron microscopy with energy dispersive X-ray (SEM-EDX).

Fig. 1 indicates the surface area and pore size distribution of activated carbon and clinoptilolite zeolite analyzed using the N_2 adsorption–desorption method. The BET specific surface areas obtained for the adsorbents are reported in Table 3. The lowest and the highest mean pore diameter

Table 1
Some characteristics of the potato processing plant wastewater

COD[a] (mg/L)	4,000
BOD[b] (mg/L)	1,865
pH	6.5
EC[c] (dS/m)	0.40

[a]COD: chemical oxygen demand
[b]BOD: biochemical oxygen demand
[c]EC: electrical conductivity

Table 2
Chemical composition of Iranian clinoptilolite zeolite

Chemical composition	Percentage (%)
SiO_2	66.85
Al_2O_3	10.87
Fe_2O_3	0.61
TiO_2	0.39
CaO	2.98
MgO	1.28
Na_2O	1.54
K_2O	1.52
Loss on ignition	14.3
Total	100

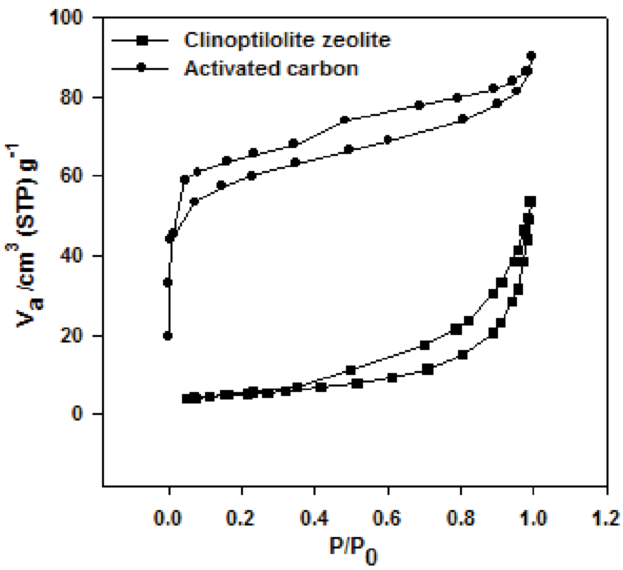


Fig. 1. Nitrogen adsorption–desorption isotherms of activated carbon and clinoptilolite zeolite.

Table 3
Structural properties of the activated carbon and clinoptilolite zeolite

Sorbent	BET-plot			
	P/P_0 (cm^3 g^{-1})	V_m (cm^3 g^{-1})	d_{pore} (nm)	S_a (m^2 g^{-1})
Clinoptilolite zeolite	8.17	4.15	18.09	18.06
Activated carbon	0.13	49.35	2.53	214.82

P/P_0: total pore volume
V_m: monolayer volume
d_{pore}: mean pore diameter
S_a: Brunauer–Emmett–Teller (BET) specific surface area

were observed to be 2.53 and 18.09 nm for the activated carbon and clinoptilolite zeolite, respectively.

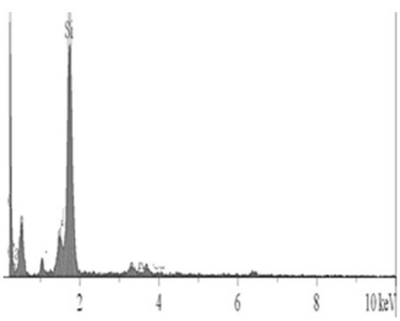
Fig. 2 shows surface morphology and elemental composition of activated carbon and clinoptilolite zeolite obtained using SEM equipped with EDX. The surface of clinoptilolite zeolite is coarse with the composing of the agglutinative blocks (Fig. 2a). While the surface of activated carbon is filled with hollow holes and very fine particles in agglomeration at the surface (Fig. 2b). The outcome of the EDX analysis is quantitative elemental in each adsorbent which indicates the inset Tables in the EDX spectra (Fig. 2). The Si and Al value were found to be 33 and 6 wt.%, respectively, for clinoptilolite zeolite and the C of activated carbon was calculated as 94 wt.%. In fact, these elements are the main building of activated carbon and clinoptilolite zeolite structure.

2.3. COD removal experiments

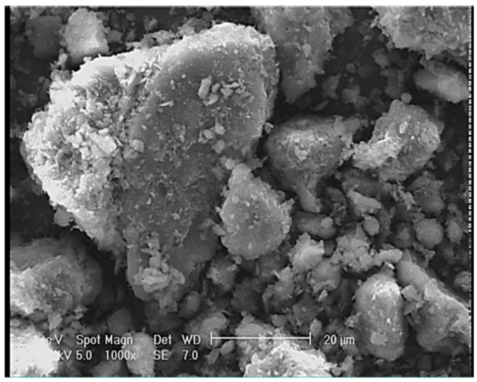
All batch experiments were performed in a glass conical flask and combined wastewater of potato processing with different doses of activated carbon and clinoptilolite zeolite, stirred well and kept in contact for 12 h at 25°C temperature then filtered. The COD concentration, pH and electrical conductivity (EC) in wastewater samples were measured twice (before and after adsorption) using UV-Vis spectrophotometer (model UV/Vis 2100) at the wavelength 600 nm, pH-meter and EC meter, respectively.

The effects of different parameters such as contact time (0.5, 1, 2, 5, 12, 18, and 24 h), initial concentration (510; 1,080; and 2,081 mg/L) and adsorbent dose (0.2, 0.5, 1, 1.5, and 2 g in 40 ml) on the adsorption process was investigated.

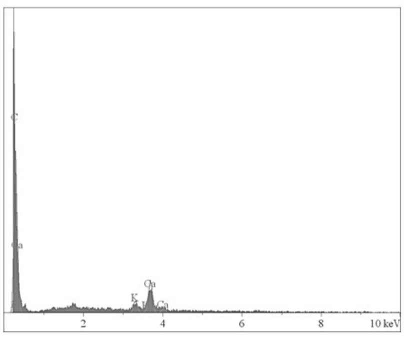
(a)



Element	Intensity (c/s)	Atomic wt.%
O	36.35	53.19
Na	10.89	3.76
Al	32.54	6.16
Si	181.71	33.32
K	11.29	1.86
Ca	10.24	1.72
		100.00



(b)



Element	Intensity (c/s)	Atomic wt.%
C	132.73	94.09
K	13.69	1.43
Ca	37.34	4.47
		100.00

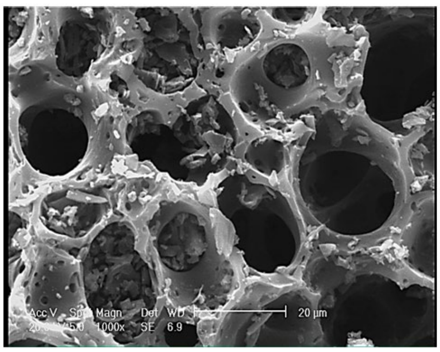


Fig. 2. SEM images with EDX spectrum results of activated carbon (a) and clinoptilolite zeolite (b).

Kinetics of COD adsorption by adsorbents was also studied at the selected time intervals. In this experiment, 2 g samples of each adsorbent were added into a conical glass flask containing wastewater COD concentration of 510 or 2,040 mg/L.

Adsorption isotherm study was performed using potato processing wastewater containing different initial concentrations of COD from 510 to 2,040 mg/L. All the experiments were performed in triplicates. The COD removal percentage was defined as:

$$\% \text{ Adsorption} = \frac{\left(\text{COD}_0 - \text{COD}_t\right)}{\text{COD}_0} \times 100 \quad (1)$$

where COD_0 (mg/L) and COD_t (mg/L) are the initial and final concentrations of COD in wastewater, respectively.

2.4. Adsorption kinetic experiments

Time-dependent COD adsorption data onto the activated carbon and clinoptilolite zeolite was described using the pseudo-first-order, pseudo-second-order, and Elovich kinetic models. The kinetic models are given in Table 4, with different linear and non-linear [29,30].

Pseudo-first-order model is based on the fact that the rate of occupation of adsorption sites which was proposed by Lagergren in 1898 for adsorption of oxalic and malonic acid onto charcoal. The pseudo-second-order model is based on adsorption rate for available surface sites (Table 4). where q_e is the equilibrium adsorption capacity (mg/g); q_t is the value of adsorbed COD (mg/g) at the time t (min); K_1 is the constant adsorption rate in the pseudo-first-order model (min^{-1}); K_2 is the constant adsorption rate in the pseudo-second-order model (min^{-1}).

Another kinetic model has seen the considerable applications in adsorption processes of pollutants from aqueous solutions is the Elovich's model which proposed by Roginsky and Zeldovich in 1934 to describe the adsorption of CO on manganese dioxide [31]. This model had the considerable applications used for liquid–solid systems and is suitable where desorption does not occur because of low surface coverage [10].
where in Elovich's model x is the initial adsorption rate (mg/g min); $1/y$ is the parameter related to the number of sites available for adsorption (mg/g).

2.5. Adsorption isotherm experiments

The adsorption isotherms for the COD removal from potato processing wastewater were analyzed with two well-known adsorption isotherm models including Langmuir and Freundlich isotherms. The basic assumption of Langmuir's theory is maximum adsorption happens at specific homogeneous sites within the adsorbent [32]. The Langmuir adsorption isotherm is shown in the following equation:

$$\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{C_e}{q_m} \quad (2)$$

where q_e is the capacity of adsorbent for adsorbate (mg/g); C_e is the concentration of adsorbate in solution at equilibrium (mg/L); q_m is the maximum adsorption capacity for adsorbate (mg/g).

In the Langmuir isotherm model, the amount of q_m and K_L can be calculated from the linear plot of C_e/q_e vs. C_e.

The Freundlich isotherm provides an empirical isotherm, which is commonly used to describe the adsorption characteristics for the heterogeneous surfaces and has a non-uniform distribution of adsorption heat. Nonlinearity form the Freundlich isotherm model expresses the following equation [25].

$$q_e = K_F C_e^{\frac{1}{n}} \quad (3)$$

where C_e is the concentration of adsorbate in solution at equilibrium (mg/L); K_F is the Freundlich adsorption constant (mg/g); n is the dimensionless Freundlich parameter.

In the Freundlich isotherm model, the amount of K_F and $1/n$ calculates by plotting $\log q_e$ vs. $\log C_e$. Linear form Freundlich isotherm model is indicated by the following equation:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (4)$$

Temkin isotherm is based on the effects of indirect adsorbent–adsorbate interactions, which increased interactions between adsorbate and adsorbent leading to decrease linearly in the heat of adsorption [30]. This model is indicated by the following equation:

$$q_e = B \,\text{Ln}\left(A C_e\right) \quad (5)$$

Table 4
Kinetic models and their different forms of linear and non-linear

Kinetic models	Non-linear forms	Linear forms	Plots for linear forms
Pseudo-first-order	$\frac{dq}{dt} = k_1\left(q_e - q_t\right)$	$\text{Ln}\left(q_e - q_t\right) = \text{Ln } q_e - k_1$	$\log\left(q_e - q_t\right) \text{vs. } t$
Pseudo-second-order	$\frac{dq}{dt} = k_1\left(q_e - q_t\right)^2$	$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} - \frac{1}{q_e} t$	$\frac{t}{q_t} \text{vs. } t$
Elovich	$q_t = \frac{1}{y}\text{Ln}\left(1 + xyt\right)$	$q_t = \frac{1}{y}\text{Ln}\left(xy\right) + \frac{1}{y}\text{Ln}$	$q_t \text{vs. Ln } t$

where B is the constant related to the heat of adsorption; A is the equilibrium binding constant (Lm/g).

In the Temkin isotherm model with a linear plot between q_e and $\text{Ln}C_e$ determines the values of both constants A and B. The linear form of Temkin isotherm is given by Eq. (6):

$$q_e = B\,\text{Ln}\,A + B\,\text{Ln}C_e \tag{6}$$

3. Results and discussion

3.1. Effect of contact time

The effect of contact time on removal percent of COD from potato processing wastewater using activated carbon and clinoptilolite zeolite is shown in Fig. 3. The COD removal percent increased initially with increasing contact time and reached a constant amount after 12 h. Accordingly, 12 h contact time was suitable for the removal of COD using activated carbon and clinoptilolite zeolite in two different initial concentrations of COD (510 and 2,040 mg L^{-1}). The maximum COD removal from wastewater in COD initial concentrations of 510 mg L^{-1} using 2 g in 40 mL dose of activated carbon and clinoptilolite zeolite was obtained 72.1% and 62.2%, respectively. For a COD concentration equal to 2,040 mg/L, COD removal percentages were 79.7% and 70.8%, respectively. Similar results were obtained by other studies concerning the effects of contact time on adsorption efficiency of wastewater [33].

However, one of the reasons for the increase in removal efficiency of COD with an increase in contact time might be due to that the adsorbate molecules have enough time to be captured by the adsorbents and as a consequence, the increase in contact time can enhance the removal efficiency.

3.2. Effect of adsorbent dose

Adsorbent dose is an important factor in the removal of pollution from the wastewater. Therefore, the effects of activated carbon and clinoptilolite zeolite different doses on COD removal efficiency from wastewater were investigated by varying the adsorbent dose from 0.2–2 g in 40 ml, of potato wastewater in COD of 1,080 mg/L. The results are presented in Fig. 4 showed that higher percentage removal is achieved for higher adsorbent dose, and in comparison with clinoptilolite zeolite, the COD removal of activated carbon is higher at identical dosages. The results of this case are in corresponding with a higher specific surface area of activated carbon as compared to natural zeolite. Because activated carbon has a specific surface area about 11.9 times of natural zeolite. As illustrated, higher COD removal percent can be attributed to the increase in the number of available adsorption sites. However, some of the studies reported that increase in adsorbent dose can be leading to higher COD removal percent from wastewater (4).

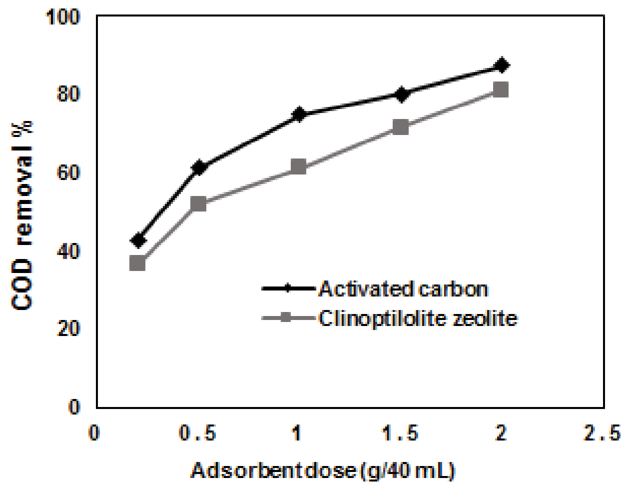


Fig. 4. Influence of the adsorbent dose on COD removal efficiency from wastewater. (Conditions: COD concentration, 1,080 mg/L; run time 12 h, pH: 5.23).

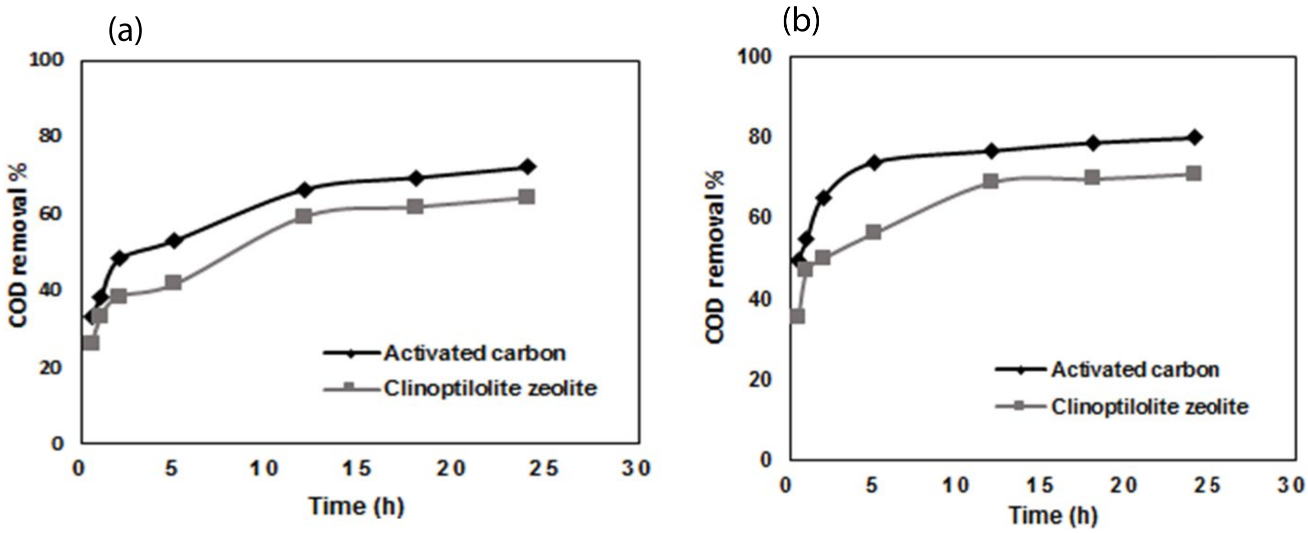


Fig. 3. The efficiency of COD removal from wastewater at various times by using activated carbon and clinoptilolite zeolite. (a) Initial COD concentration 510 mg/L and (b) initial COD concentration 2,040 mg/L.

In addition, Fig. 5 indicates that increasing the dose of adsorbent can be leading to a decrease in the adsorption capacity. This means that low adsorbent dosage has a higher adsorbent capacity as compared to high adsorbent dosage.

3.3. Effect of initial COD concentration with different adsorbent dose

The COD removal percent for two adsorbents activated carbon and clinoptilolite zeolite in three COD concentrations are presented in Fig. 6. The results showed that adsorbent dose of 2 g/40 ml, and by increasing COD concentration, COD removal decreases two adsorbents. In this case, the ratio between the amount of COD and the adsorbent dose is so important. With an adsorbent dose of 0.2, using COD concentrations of COD concentration of 510; 1,080; and 2,040 mg/L were 2,550; 5,400; and 10,200 times, respectively. Therefore, by increasing the ratio of COD concentration to adsorbent dose COD removal percent is reduced. However, in two doses of 1 and 2 g in 40 ml, this process was reversed. These results were similar to the results of other studies [34]. As a result of these changes, it can be seen that adsorbent dose is a key factor in COD removal.

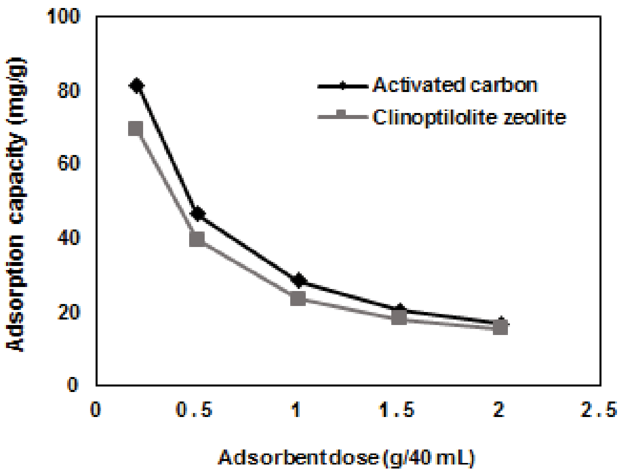


Fig. 5. Influence of the adsorbent dose on COD adsorption capacity. (Conditions: COD concentration, 1,080 mg/L; run time 12 h, pH: 5.23).

3.4. Adsorption isotherms

The equilibrium adsorption isotherms of COD were determined at different doses of activated carbon and clinoptilolite zeolite. The different isotherm models parameters, along with the regression coefficients, are presented in Table 5. The results indicated that the experimental data better describes the equilibrium adsorption with the Freundlich model with higher R^2 values than others. According to the Freundlich isotherm model if $1 < n_f < 10$, the adsorption is favorable. Therefore, as consequence adsorption mechanisms of COD from wastewater by using of different dose of activated carbon and clinoptilolite zeolite is suitable.

3.5. Adsorption kinetics

COD removal kinetic experiments result from wastewater by using activated carbon and clinoptilolite zeolite indicated in Fig. 7. It can be observed from Fig. 7 that COD removal kinetic data showed a better fit with the pseudo-second-order kinetic model and provided the best correlation to the data compared to pseudo-first-order and Elovich. The different adsorption kinetic model parameters are presented in Table 6. Based on the pseudo-second-order kinetic model, this suggests that adsorption mechanisms for removal COD from wastewater by using activated carbon and clinoptilolite zeolite is based on rapid adsorption.

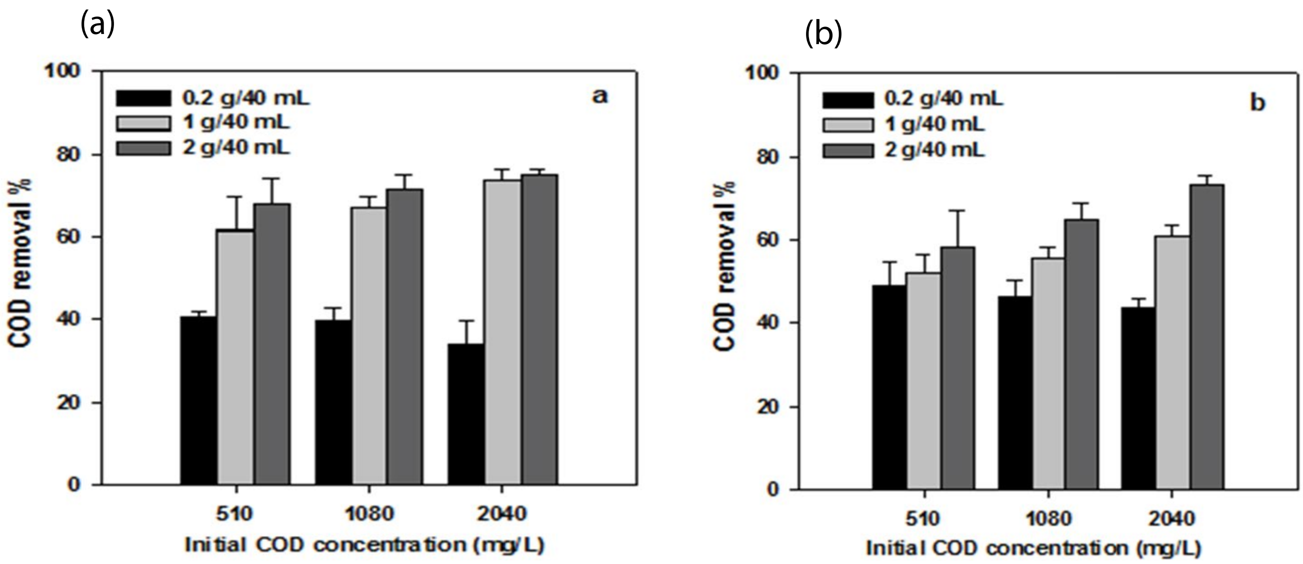


Fig. 6. Effects of different initial COD concentrations on the COD removal efficiency from wastewater (a) natural zeolite and (b) activated carbon. (Conditions: run time 12 h, pH: 5.23).

Table 5
Isotherm model parameters for the adsorption of COD on activated carbon and clinoptilolite zeolite

Adsorbent	Model	Parameters	Dose of adsorbent		
			0.2 g/40 mL	1 g/40 mL	2 g/40 mL
			Values	Values	Values
Activated carbon	Langmuir	q_m (mg/g)	416.66	47.39	49.01
		K_L (L/mg)	3.78×10^{-4}	10×10^{-4}	7.62×10^{-4}
		R_L	0.77	0.72	0.8
		R^2	0.958	0.995	0.953
	Freundlich	$1/n$	0.808	1.583	1.311
		K_F	1.44	12.65	7.79
		R^2	0.999	0.997	0.999
	Temkin	A (L/mg)	333.84	1,638.93	1,445.65
		B	0.015	0.002	0.003
		R^2	0.995	0.98	0.974
Clinoptilolite zeolite	Langmuir	q_m (mg/g)	714.28	81.3	19.49
		K_L (L/mg)	2.87×10^{-4}	4.7×10^{-4}	11×10^{-4}
		R_L	0.84	0.8	0.7
		R^2	0.991	0.987	0.997
	Freundlich	$1/n$	0.852	1.3	1.675
		K_F	1.42	7.91	22.57
		R^2	0.999	0.999	0.999
	Temkin	A (L/mg)	445.07	181.15	1,712
		B	0.011	0.029	0.002
		R^2	0.973	0.948	0.982

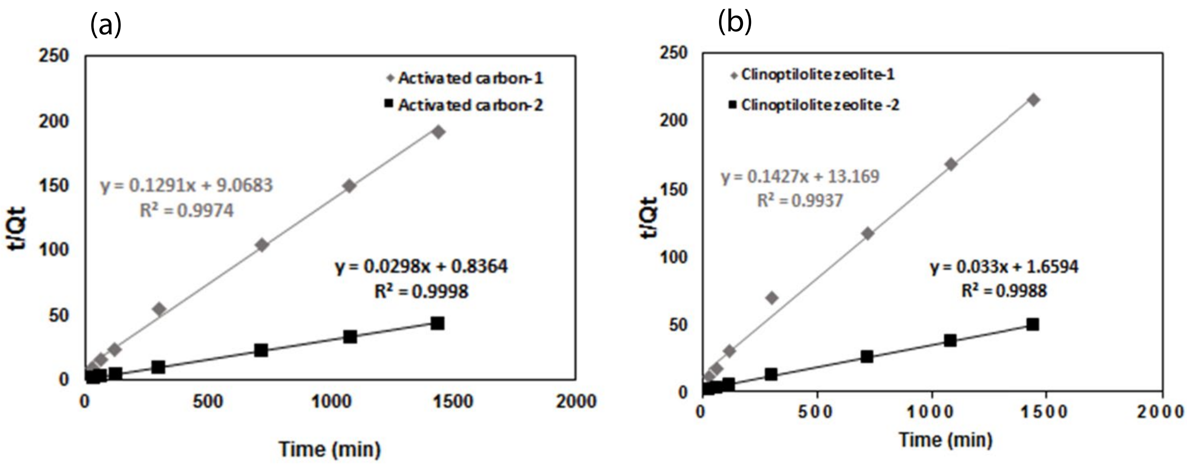


Fig. 7. Pseudo-second-order kinetic model to adsorption COD in the activated carbon (a) and clinoptilolite zeolite and (b) with different initial COD concentration (1) 520 mg/L and (2) 2,081 mg/L.

Similar results in other studies have been reported which for the adsorption of COD from wastewater the pseudo-second-order kinetic model act as a suitable kinetic model [10,35,36].

3.6. Reuse of treated wastewater to irrigate

The reuse ability of activated carbon and clinoptilolite zeolite for the elimination of COD from potato processing plant wastewater was also investigated (Fig. 8). The experimental results displayed that the activated carbon and clinoptilolite zeolite maintained their usability towards COD for initial concentrations of 2,081 mg/L even after five cycles. The results showed that the potato processing plant wastewater can be used for application in irrigation of green spaces after 2 cycles of treatment by activated carbon and 3 cycles of treatment by clinoptilolite zeolite. The residual COD of

Table 6
Parameters of pseudo-first-order, pseudo-second-order, and Elovich kinetic models

Adsorbents	Pseudo-first-order			Pseudo-second-order			Elovich		
	q_e (mg/g)	K_1 (min^{-1})	R^2	q_e (mg/g)	K_2 (g/mg min)	R^2	X (mg/g min)	Y (g/mg)	R^2
Activated carbon-1	5.07	0.0019	0.455	7.74	0.0018	0.997	1.27	1.07	0.989
Activated carbon-2	2.25	0.0018	0.536	33.55	0.001	0.999	2752.81	3.45	0.954
Clinoptilolite zeolite-1	2.07	0.002	0.368	7.07	0.0015	0.998	3.17	1.08	0.964
Clinoptilolite zeolite-2	8.11	0.0023	0.599	30.3	0.0006	0.993	2.359	3.85	0.974

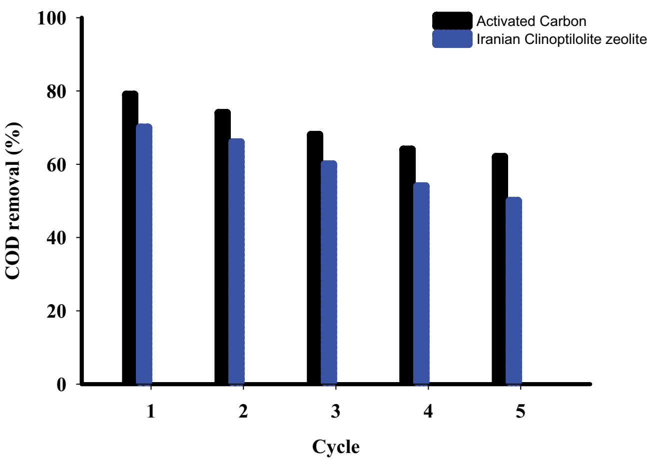


Fig. 8. The removal efficiency of COD using the activated carbon and clinoptilolite zeolite after five cycles.

treated wastewater is below 150 mg L^{-1}. The results were satisfactory according to Iranian standards.

4. Conclusion

The results obtained from this study indicate that adsorption is a versatile and highly efficient method with low-cost adsorbents to treat potato processing wastewater. The maximum COD removals were 79% and 70% in initial COD concentration of 2,040 mg/L for activated carbon and clinoptilolite zeolite, respectively. Findings showed that the COD removal percentage increased with increasing the contact time and adsorbent dose. Besides, kinetic and isotherm studies indicated that kinetic and isotherm data were best fitted to the pseudo-second-order model and Freundlich isotherm model. This study demonstrated that the Iranian clinoptilolite zeolite is a cheap, plentiful and high adsorptive capacity adsorbent that could be an alternative to the commercial adsorbents such as activated carbon for COD removal from wastewater. Therefore, clinoptilolite zeolite due to its low cost and high performance can be used in many factories for wastewater treatment instead of commercial adsorbents. Despite all benefits of adsorbents usage in the wastewater treatment industry, the main concern using adsorbents is sludge production resulting in environmental problems. Therefore, finding effective methods for adsorbents reuse is an important case should be examined in future studies.

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