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Adsorption of hexavalent chromium from aqueous solutions by wheat bran

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ABSTRACT: In this research, adsorption of chromium (VI) ions on wheat bran has been studied through using batch adsorption techniques. The main objectives of this study are to 1) investigate the chromium adsorption from aqueous solution by wheat bran, 2) study the influence of contact time, pH, adsorbent dose and initial chromium concentration on adsorption process performance and 3) determine appropriate adsorption isotherm and kinetics parameters of chromium (VI) adsorption on wheat bran. The results of this study showed that adsorption of chromium by wheat bran reached to equilibrium after 60 min and after that a little change of chromium removal efficiency was observed. Higher chromium adsorption was observed at lower pHs, and maximum chromium removal (87.8 %) obtained at pH of 2. The adsorption of chromium by wheat bran decreased at the higher initial chromium concentration and lower adsorbent doses. The obtained results showed that the adsorption of chromium (VI) by wheat bran follows Langmuir isotherm equation with a correlation coefficient equal to 0.997. In addition, the kinetics of the adsorption process follows the pseudo second-order kinetics model with a rate constant value of 0.131 g/mg.min The results indicate that wheat bran can be employed as a low cost alternative to commercial adsorbents in the removal of chromium (VI) from water and wastewater.

Key words: *Heavy metals, natural adsorbents, isotherm, kinetics*

INTRODUCTION

One of the heavy metals that has been a major focus in water and wastewater treatment is chromium and the hexavalent form of it has been considered to be more hazardous due to its carcinogenic properties (Karthikeyan *et al.*, 2005). Chromium has been considered as one of the top $16^{th.}$ toxic pollutants and because of its carcinogenic and teratogenic characteristics on the public, it has become a serious health concern (Torresdey *et al.*, 2000).Chromium can be released to the environment through a large number of industrial operations, including metal finishing industry, iron and steel industries and inorganic chemicals production (Gao *et al.*, 2007). Extensive use of chromium results in large quantities of chromium containing effluents which need an exigent treatment. The permissible limit of chromium for drinking water is 0.1 mg/L (as total chromium) in EPA standard (EPA, 2007). In addition, National Iranian standard for Cr(VI) concentration in drinking water is 0.05 mg/L (ISIRI number 1053, 1991). There are various methods to remove Cr(VI) including chemical precipitation, membrane process, ion exchange, liquid extraction and electrodialysis (Verma *et al.*, 2006). These methods are non-economical and have many disadvantages such as incomplete metal removal, high reagent and energy requirements, generation of toxic sludge or other waste products that require disposal or treatment. In contrast, the adsorption technique is one of the preferred methods for removal of heavy metals because of its efficiency and low cost (Li *et al.*, 2007). For this purpose in recent years, investigations have been carried out for the effective removal of various heavy metals from solution using natural adsorbents which are economically viable such as agricultural wastes including sunflower stalks (Sun and Shi, 1998), Eucalyptus bark (Sarin and Pant, 2006), maize bran (Singh *et al.*, 2006), coconut shell, waste tea, rice straw, tree leaves, peanut and walnut husks (Karthikeyan *et al.*, 2005). The bran of wheat is the shell of the wheat seed and contains most nutrients of wheat. This bran is usually removed in the processing

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of wheat into flour. Recently a few studies have been done on removing heavy metals such as pb(II) (Bulut and Baysal, 2006), Cu(II) and Cd(II) (Farajzadeh and Boviery Monji, 2004) by wheat bran. In this study, wheat bran had been used for Cr(VI) removal from aqueous solution. The aims of this study are to 1) investigate the chromium adsorption from aqueous solution by wheat bran 2) study the effect of different parameters such as contact time, pH, adsorbent dose and initial chromium concentration on adsorption process and 3) find optimum adsorption isotherm as well as the rate of adsorption kinetics.

MATERIAL AND METHODS

This study was accomplished in environmental engineering laboratory, department of civil and environmental engineering, Amirkabir University of technology in 2007. The details of materials and methods of the study are discussed as below:

Preparation of adsorbent

The wheat bran was ground and particle sizes between 297 and 595 μm were obtained by passing the milled material through standard steel sieves. Then, they used for experiments without washing or any other physical or chemical treatments.

Batch sorption experiments

The sorption studies were carried out at 25±1°C. Solution pH was adjusted with H_2SO_4 or NaOH. A known amount of adsorbent was added to samples and was agitated by jar test at 250 rpm agitation speed, allowing sufficient time for adsorption equilibrium. Then, the mixtures were filtered through filter paper, and the Cr(VI) ions concentration were determined in the filtrate using DR/4000U spectrophotometer by colorimetric techniques according to the standard method No. 3500-Cr B (standard methods, 1992). The effects of various parameters on the rate of adsorption process were observed by varying contact time, t (5, 10 , 15, 20 , 30, 45, 60, 120 and 240 min), initial concentration of chromium ion, C_o (2.5, 5, 7.5, 10, 12.5 and 15 mg/L), adsorbent concentration, W (1, 2, 3, 4, 5 and 6 g/100 mL) and initial pH of solution (2, 3, 5, 7, 9 and 11). The solution volume (V) was kept constant (200 mL). The chromium removal (%) at any instant of time was determined by the following equation:

$$Chromium\ removal\ (\%) = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

where, C_o and C_t are the concentration of chromium at initial condition and at any instant of time, respectively. To increase the accuracy of the data, each experiment was repeated 3 times. Adsorption isotherm studies were carried out with different adsorbent doses ranging from 1 to 6 g/100 mL while maintaining the initial chromium concentration at 5 mg/L.

RESULTS AND DISCUSSION

Effect of contact time on chromium adsorption

Contact time is one of the effective factors in batch adsorption process. In this stage, all of the parameters except contact time, including temperature (25 °C), adsorbent dose (2 g/100 mL), pH (3), initial chromium concentration (5 mg/L) and agitation speed (250 rpm), were kept constant. The effect of contact time on chromium adsorption efficiency showed in Fig. 1. As it is shown, adsorption rate initially increased rapidly, and the optimal removal efficiency was reached within about 1 h to 87.6 %. There was no significant change in equilibrium concentration after 1 h up to 4 h and after 1 h, the adsorption phase reached to equilibrium.

Effect of pH on chromium adsorption

The pH of the aqueous solution is clearly an important parameter that controlled the adsorption process. The experiments of this stage were done under the conditions of constant temperature (25 °C), agitation speed (250 rpm), contact time (1 h), adsorbent dose (2 g/100 mL) and initial chromium concentration (5 mg/L). pH of solution was changed and the chromium removal was investigated. The experimental results of this stage are presented in Fig. 2. As it is shown, the optimum pH of solution was observed at pH of 2 and by increasing pH, a drastic decrease in adsorption percentage was observed. This might be due to the weakening of electrostatic force of attraction between the oppositely charged adsorbate and adsorbent that ultimately lead to the reduction in sorption capacity (Baral *et al.*, 2006). Adsorption of hexavalent chromium varies as a function of pH with H_2CrO_4, $HCrO_4^-$, $Cr_2O_7^{2-}$ and CrO_4^{2-} ions appear as dominant species (Gaballah and Kilbertus, 1998). At pH of 2, $HCrO_4^-$ is the dominant species. The surface charge of wheat bran is positive at low pH, and this may promote the binding of the negatively charged $HCrO_4^-$ ions. The $HCrO_4^-$ species are most easily exchanged with OH^- ions at active surfaces of adsorbent under acidic conditions as shown in Eq. 2 (Ar is adsorbent surface) (Argun *et al.*, 2006):

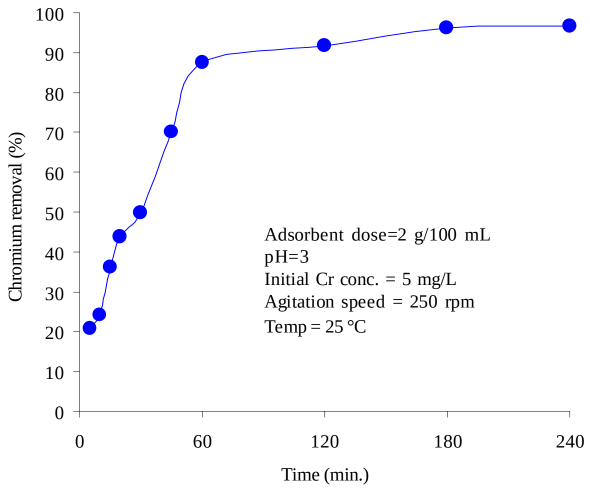

Fig. 1: Effect of contact time on adsorption process efficiency

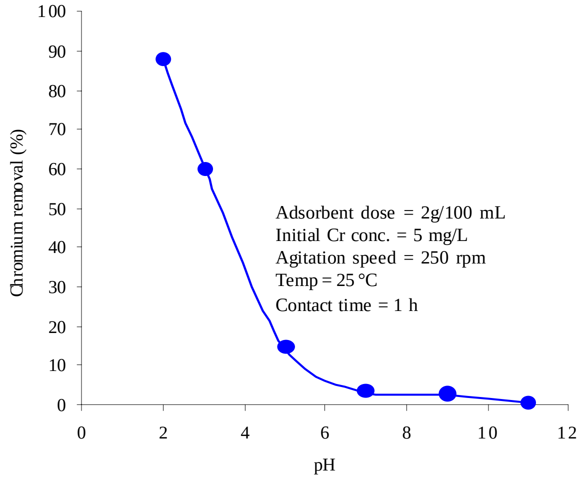

Fig. 2: Effect of pH on Cr (VI) removal

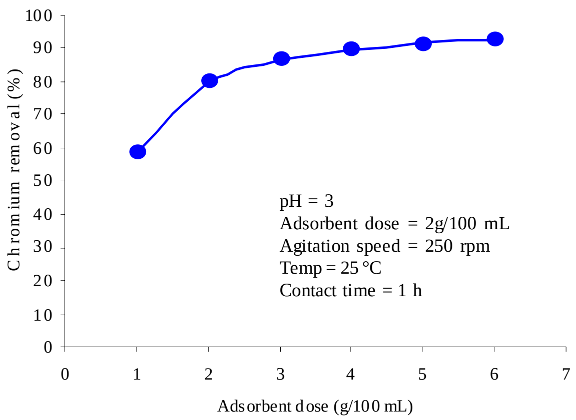

Fig. 3: Effect of adsorbent dose on Cr (VI) removal

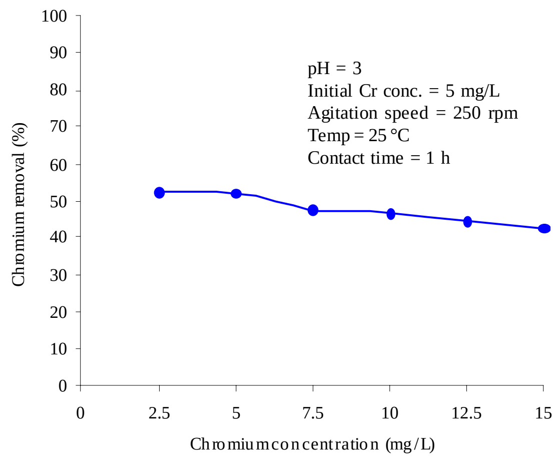

Fig. 4: Effect of chromium concentration on Cr(VI) removal

$$ArOH + HCrO_4^- + H^+ \longrightarrow ArHCrO_4 + H_2O \quad (2)$$

Effect of adsorbent dose on chromium adsorption

At this stage, the experiments were done under the conditions described at previous stage with constant pH of 3 and variable adsorbent dose (1, 2, 3, 4, 5 and 6 g/100 mL). The effect of adsorbent dose on the adsorption of chromium by wheat bran was presented in Fig. 3. As illustrated in Fig. 3, chromium removal efficiency increased with increase in adsorbent dose, since contact surface of adsorbent particles increased and it would be more probable for $HCrO_4^-$ and$Cr_2O_7^-$ ions to be adsorbed on adsorption sites and thus adsorption efficiency increased (Morshedzadeh *et al.*, 2007).

Effect of initial chromium concentration on adsorption process

Initial concentration is one of the effective factors on adsorption efficiency. The experiments were done with variable initial chromium concentration (2.5, 5, 7.5, 10, 12.5 and 15 mg/L) and constant temperature (25 °C), pH (3), agitation speed (250 rpm), contact time (1 h) and 2 g of adsorbent dose (2 g/100 mL). The experimental results of the effect of initial chromium concentration on removal efficiency were presented in Fig. 4. As Fig. 4 is shown , chromium removal efficiency decreased with the increase in initial chromium concentration. In case of low chromium concentrations, the ration of the initial number of moles of chromium ions to the available surface area of adsorbent is large

Table 1: Isotherm equations (Bulut and Baysal, 2006; Argun *et al.*, 2006)

Isotherm name	Isotherm equation	Parameters
Langmuir	$q_e = \frac{\theta.b.C_e}{1+b.C_e}$	C_e: the equilibrium concentration(mg/L) q_e: the amount adsorbed per amount of adsorbent at the equilibrium(mg/g) θ: (mg/g) and b(L/mg): the Langmuir constants related to the maximum sorption capacity and energy of adsorption, respectively.
Freundlich	$q_e = KC_e^{\frac{1}{n}}$	K(mg/g) : an indicator of the adsorption capacity $\frac{1}{n}$ (mg/L): adsorption intensity
D-R	$q_e = X'_m \exp(-K'\varepsilon^2)$	ε (the Polanyi potential) = RT ln(1 + 1/C_e) q_e: the amount of metal ions adsorbed per unit weight of wheat bran(mg/g) X'_m: the adsorption capacity of the sorbent (mg/g) C_e: the equilibrium concentration of metal ions in solution(mg/L) K´: constant related to the adsorption energy (mol^2/KJ^2) R: gas constant (kJ/K.mol) T: temperature (K)

Table 2: Adsorption isotherms constants

Isotherm type	Isotherm constants	R^2
Langmuir	θ=0.942 (mg/g), b=0.251 (L/mg)	0.997
Freundlich	K=0.18, n=1.291	0.985
D-R	K' = 0.134 (mol^2/Kj^2), X'_m = 0.301 (mg/g)	0.975

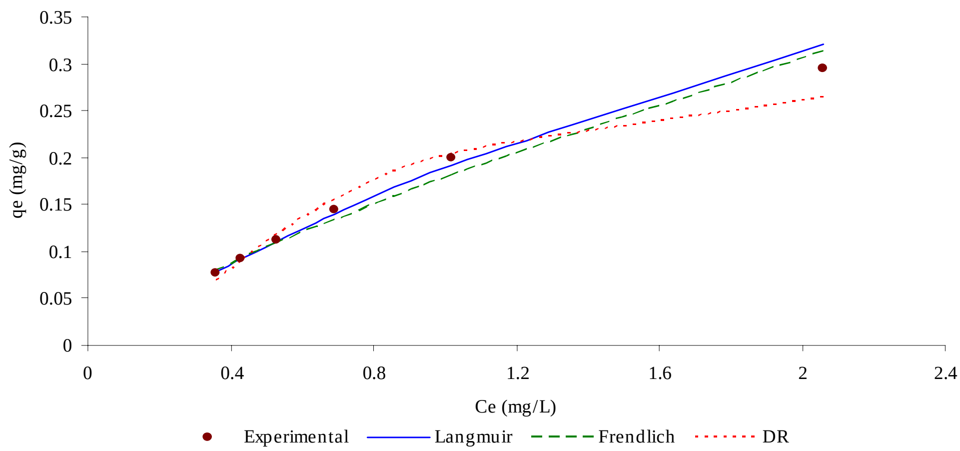

Fig. 5: Comparison of the experimental and predicted isotherms for the adsorption of Cr(VI) on wheat bran (pH= 3; initial Cr conc.= 5ppm; agitation speed= 250rpm; Temp= 25±1°C; contact time= 1 h)

and subsequently the fractional adsorption becomes independent of initial concentration. However, at higher concentrations, the available sites of adsorption become fewer, and hence the percentage removal of metal ions which depends upon the initial concentration, decreases (Yu *et al.*, 2003).

Adsorption isotherms

The distribution of metal ions between the liquid phase and the solid phase can be described by several isotherm models such as Langmuir, Freundlich and Dubinin–Radushkevich (D-R). The isotherm equations of these models are summarized in Table 1. The Langmuir model assumes that the uptake of metal ions occurs on a homogenous surface by monolayer adsorption without any interaction between adsorbed ions. However, the Freundlich model assumes that the uptake of metal ions occurs on a heterogeneous surface by monolayer adsorption (Bulut and Baysal, 2006) and Dubinin–Radushkevich (D-R) isotherm assumes a heterogeneous surface, too (Argun *et al.*, 2006). In order to find the most appropriate model for the

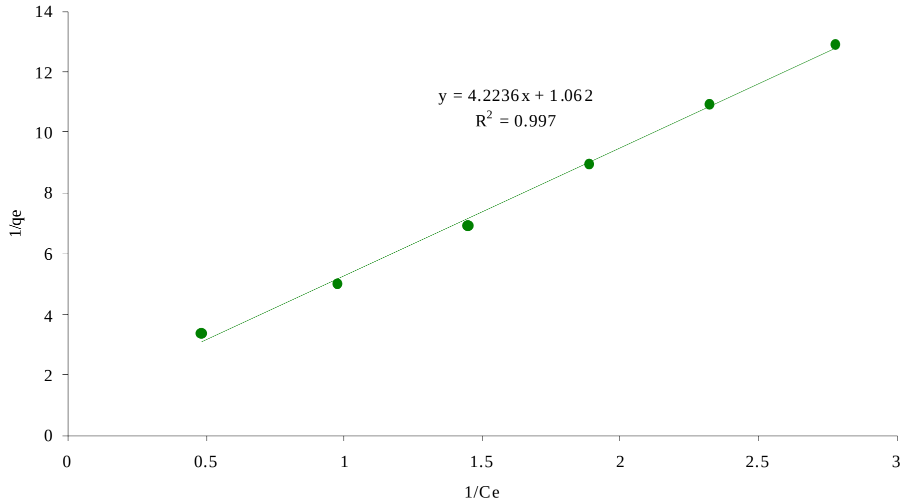

Fig. 6: Langmuir isotherm (pH=3, Temp=25 °C, C_0=5 mg/L)

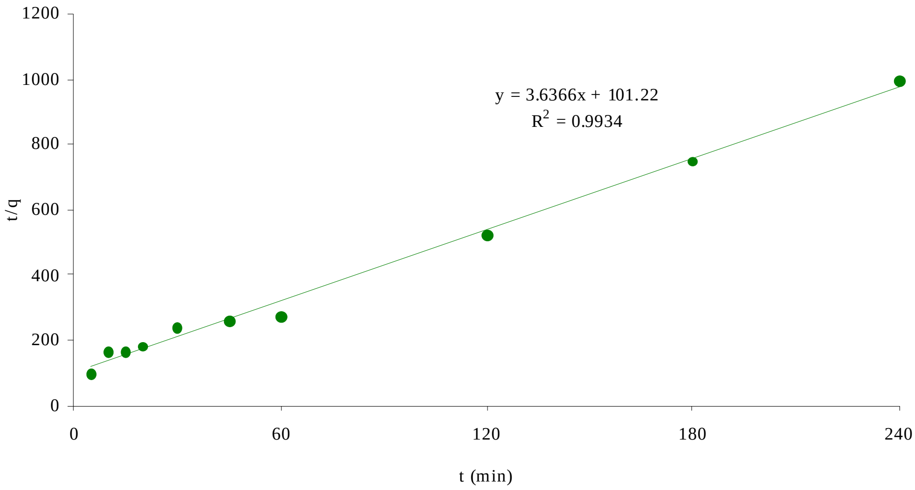

Fig. 7: Pseudo-second-order kinetics plot for adsorption of chromium on wheat bran

chromium adsorption, the data were fitted to each isotherm model. The obtained isotherm parameters and correlation coefficients (R^2) are presented in Table 2. The experimental and predicted isotherms for wheat bran at 25±1 °C are given in Fig. 5. The results showed that the Langmuir adsorption isotherm was the best model for the chromium adsorption on wheat bran with R^2 of 0.997 (Fig. 6). The essential features of Langmuir adsorption isotherm can be expressed in terms of a dimensionless constant called separation factor or equilibrium parameter (R_L), which is defined by the following relationship (Hall *et al.*, 1966; Malik, 2004):

$$R_L = \frac{1}{1+bC_0} \tag{3}$$

where, C_0 is the initial Cr(VI) concentration (mg/L). The R_L value indicates the shape of the isotherm to be ireversible ($R_L = 0$), favorable ($0 < R_L < 1$), linear ($R_L = 0$)

or unfavorable (R_L >1) (McKay *et al.*, 1982; Malik, 2004). Through the above-mentioned equation, R_L value for investigated Cr-adsorbent system is found to be 0.44. From the value of R_L, it is confirmed that wheat bran is desirable for adsorption of chromium from wastewater under the conditions used in this study. For Freundlich isotherm, as it was shown in Table 2, n is equal to 1.291. The situation $n > 1$ is most common and may be due to a distribution of surface sites or any factor that cause a decrease in adsorbent-adsorbate interaction with increasing surface density (Reed and Matsumoto, 1993) and the values of *n* within the range of 2-10 represent good adsorption (Mckay *et al.*, 1980; Ozer and Pirincci, 2006).

Adsorption kinetics

In order to define the adsorption kinetics of heavy metal ions, the kinetics parameters for the adsorption process were studied for contact times ranging from 1 to 240 min by monitoring the removal percentage of the Cr(VI). The data were then regressed against the Lagergren equation (Eq. 4), which represents a first-order kinetics equation (Namasivayam and Yamuna, 1995), and against a pseudo-second-order kinetics equation (Eq. 5) (Ho, 1995).

$$\log(q_e - q_t) = \log q_e - \frac{K_1}{2.303} t \quad (4)$$

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \quad (5)$$

where, q_t is the metal uptake per unit weight of adsorbent (mg/g) at time t, q_e is the metal uptake per unit weight of adsorbent (mg/g) at equilibrium, and k_1 (min^{-1}) and k_2 (g/mg.min) are the rate constants of the pseudo-first-order and pseudo-second-order kinetics equations, respectively (Argun *et al.*, 2006). The slopes and intercepts of these curves were used to determine the values of K_1 and K_2, as well as the equilibrium capacity (q_e). The first and second order kinetics constants are presented in Table 3. The results indicated that the adsorption process follows pseudo-second-order model. The plot of t/q_t versus $1/q_e$ gives a straight line as shown in Fig. 7.

Table 3: Kinetics constants

Parameter	Kinetics order	
	Pseudo-First order	Pseudo-Second order
q_t (mg/g)	0.232	0.275
Rate constant (K)	0.029 (g/mg.min)	0.131(1/min)
Correlation factor (R^2)	0.971	0.993

Comparison of adsorption capacity of wheat bran with other adsorbents

Direct comparison of wheat bran with other adsorbent materials is difficult, owing to the different applied experimental conditions. In the present study, wheat bran has been compared with other adsorbents based on their maximum adsorption capacity for Cr(VI) and shown in Table 4. It can be observed that the wheat bran compares well with the other adsorbents listed in Table 4. Activated rice husk carbon and modified oak sawdust are adsorbents that exhibited higher adsorption capacity. This could be primarily due to the initial carbon content, activation process as well as the pore development due to the basic morphology of the raw material (Garg *et al.*, 2004). Hence, wheat bran can be considered to be viable adsorbent for the removal of Cr(VI) from aqueous solutions.

In this study, the effect of wheat bran on Cr(VI) removal was examined. The results indicated that the adsorption process reached to equilibrium after 60 min and at this time, the chromium removal was 87.6 %. Among all of the selected parameters, pH of solution was the most effective on chromium removal. The results showed that the highest adsorption of chromium on wheat bran (87.8 %) was at pH of 2.

Table 4: Comparison of adsorption capacities of Cr(VI) with other adsorbents

Adsorbents	Adsorption capacity (mg/g)	pH	C_o (mg/L)	Reference
Activated rice husk carbon	0.8	2	10	Bishnoi *et al.*, 2004
Activated alumina	1.6	4	10	Bishnoi *et al.*, 2004
Sawdust	0.229	2	5	Morshedzadeh *et al.*, 2007
Pine leaves	0.277	2	5	Morshedzadeh *et al.*, 2007
Raw rice bran	0.07	2	5	Oliveira *et al.*, 2005
Modified oak sawdust	1.7	3	-	Argun *et al.*, 2006
CETYL-amended zeolite	0.65	-	-	Santiago *et al.*, 1992
EHDDMA-amended zeolite	0.42	-	-	Santiago *et al.*, 1992
Wheat bran	0.942	3	5	Present study

It was observed that the removal percentage increased at the lower initial chromium concentration and higher adsorbent doses. The results showed that the Langmuir adsorption isotherm was the best model for the chromium removal on wheat bran with a correlation coefficient (R^2) of 0.997. The kinetics analysis of the study showed that the adsorption of Cr(VI) ions onto wheat bran could be well described with the pseudo-second-order kinetics model and the rate constant for the process was found to be 0.131 g/mg.min at 25 °C. Based on the results of this research, wheat bran can be considered as an effective, available and natural adsorbent for removing chromium from aqueous solutions.

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