APPLICATION OF IRON IMPREGNATED ACTIVATED CARBON FOR REMOVAL OF ARSENIC FROM WATER

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ABSTRACT

The presence of arsenic in drinking water is one of the greatest threats to public health. The aim of this experimental investigation was to study the removal efficiency of As(v) from water by application of iron-impregnated activated carbon(Fe-AC). Coating of activated carbon with iron salt was carried out by impregnation method and thermo-chemical reactions with using of 100 mM $FeCl_3$ solution. The Fe-AC was pulverized using ASTM standard sieves with the range of 16-20 mesh. The solid structure and surface characteristics of Fe-AC were determined using conventional techniques. Batch adsorption experiments were carried out with 300 and 600µg/L arsenate. Langmuir, Freundlich and Dubinin-Radushkevich models were used to describe the isotherm and energy of adsorption. The results of this study showed that 2.5 h contact time was enough to achieve the equilibrium. The adsorption of As(v) had a good compliance with Langmuir model ($R^2 = 0.995$) and the maximum adsorption capacity was obtained as 0.024 mg/g. The results of kinetic studies showed that As(v) adsorption on Fe-AC may be limited by film diffusion step (m=0.26). The mean free energy of adsorption (E) calculated from Dubinin-Radushkevich isotherm was found to be 1.52 kj/mol which implies that the adsorption of As(v) on Fe-AC is a physical adsorption. The results indicated that Fe-AC is one of the suitable adsorbents which can be used for the treatment of arsenic contaminated waters.

Key words: Arsenic, Adsorption, Water, Isotherm models, Activated carbon, Iron impregnated

INTRODUCTION

Arsenic is a ubiquitous element in the environment that can be found in trace amounts in rocks, water, soils and even in air. Nowadays, pollution of water with arsenic is one of the most important understudy topics of the public health organizations (Ahsan *et al*., 2007).

The problems related to As are distributed among millions of people in several countries such as Bangladesh, India, Nepal (Matschullat, 2000) Pakistan, Mexico, Mongolia, Germany, Thailand, China, Iran, Chile, USA, Canada and Myanmar

(Mesdaghinia *et al.,* 2005; Mondal *et al*., 2006*)*. In these countries, As-contaminated water and endemic exposure with this pollutant emerged as a single catastrophe affecting the people in these regions. Survey of groundwater quality in Iran shows that drinking water in some rural areas in Kurdestan province, located in the west of Iran, has high concentrations of arsenic (Mesdaghinia *et al.,* 2005).
Survey of environmental geology in Iran shows that deposits and sediments in some parts of central zone of eastern Alborz, located at $54°$ $33'$ – $55°$ $15'$ E and $36°$ $15'$ - $36°$ $48'$ N in north

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east of Iran, have as high as 140ppm of arsenic. Analysis of water quality shows that in this area, surface and groundwater has been affected by the high concentration of As and its contaminations (Shariati and Aghanabati , 2004).

Considering the geochemical data in North West of Iran indicates that Zarshuran As-Au deposits located at 36° 43′ 21″ N and 47° 8′ 25″ E, 42 km north of the town of Takab in the West Azarbaijan province has a high concentration of As. Analysis of chemical constituents of water in this region shows that some of samples have high concentration of As (Modaberi and Moore , 2004).

Arsenic occurs in the environment in variable oxidation states (-3, 0, +3, +5) (Modaberi and Moore , 2004). Arsenic can not be destroyed but can be converted from one state to other states (Mondal *et al.*, 2006). Arsenic is present in natural waters in both organic and inorganic forms (Katsoyiannis and Zoubolis, 2002 ; Nakajima *et al.*, 2005) The most common form of inorganic As in aqueous solutions is arsenate [As (v)] and arsenite [As (III)] (Cebrian *et al.*, 1983 ; WHO, 2001; Mesdaghinia *et al.,* 2005).

Survey of origins of contaminants shows that water is one of the most important media through which arsenic enters into the human body worldwide (Frumkin *et al.*, 2001; Mondal *et al.*, 2006). Based on WHO reports, absorption of As through the skin is minimal and thus hand-washing, bathing and laundry with water containing As do not pose human health risks, but drinking of As contaminated water pose the greatest threat to public health (WHO, 2001 ; Ahsan *et al.*, 2006).

Considering the lethal impact of arsenic on human health, WHO in 1993 and National Health and Medical Research Committee, Australia, in 1996 had recommended maximum contaminant level (MCL) of arsenic in drinking water as 10 and 7µg/L, respectively. The European commission in 2003 has also reduced the MCL of As from 50 to 10µg/L (Katsoyiannis and Zouboulis , 2004). Japan and Canada have reduced the MCL for arsenic in drinking-water to 10 and 25 ppb, respectively. In February 2002, the USEPA lowered the MCL of arsenic in drinking water system from 50 to 10µg/L and set a required compliance date of January 23rd , 2006 (Vaughan

and Reed , 2005).

Based on several studies, it is concluded that contamination of surface and groundwater resources in Iran is possible and should be recognized as an important problem; thus, substitution of contaminated drinking-water resources or elimination of this pollutant from water and provision of healthy drinking water is necessary.

Several processes are available for removing arsenic from drinking-water resources, including enhanced coagulation, membrane systems (Vaughan and Reed, 2005), ion exchange, adsorption onto activated alumina and other oxyhydroxides (Rau *et al*., 2003), lime softening, electrodialysis (Kim and Nriagu, 2000), adsorption by metal oxides, zero valent iron (Vaughan and *et al*., 2005)and photocatalyst adsorbent system (Nakajima *et al*., 2005). Although coagulation is an efficient process, but based on enhancing effects of phosphate on the mobility of As(v) it may be told that presence of elevated concentration of phosphate and silicate may dramatically decrease the arsenic removal by coagulation (Meng *et al*., 2000).

Many types of adsorbents have been used for the removal of arsenic from water. Several promising technologies like coating of granular materials with metallic compounds are being used for the removal of As from water. The pretreatment of activated carbon with Cu(II) lead to 30% increase in arsenic removal (Sun *et al*., 2006). Iron compounds are the preferred group of substances for arsenic adsorption, being hematite, goethite iron oxide-coated materials and granular ferric hydroxide (GFH) (Rau *et al*., 2003). Because As has high affinity for iron compounds and this leads to low leaching of adsorbed arsenic from exhausted adsorbent (Meng *et al*., 2001; Zhang and Itoh, 2006;), the aim of this research was focused on removal of As(v) from water with using of iron-impregnated activated carbon as an adsorbent material.

MATERIALS AND METHODS
Reagents / solutions and instruments
All of the chemicals used for the solutions were reagent grade and were used without further purification. The water used in solutions was

purified with a MQ-UV water device (Millipore). The electrical conductivity (EC) of the distilled water used for the experiments was lower than 3 μmohs/cm. The stock solution of 1000 mg/L As(v) was prepared by dissolving the $Na_2HAsO_4.7H_2O$ in tap water. As(v) intermediate solutions (100mg/L) were prepared by diluting the stock solutions with deionized water. Finally, 300 and 600 μg/L As(v) spiked water were prepared from intermediate solution, from which solutions of various concentrations according to experimental needs were prepared. The pH was measured using Hach pH meter (Hach Co., USA).

The iron salt solution used for impregnation/coating of AC was prepared by dissolving ferric chloride nonohydrated $FeCl_3$ (Merck Co.) in deionized water. Quantitative determination of arsenic was conducted with UV-visible spectrophotometer system (PU8700 Philips) at a wavelength of 535nm, by the silver diethyldithiocarbamate method (minimum detectable quantity=1μg As) commonly known as SDDC method, as mentioned in the standard methods for the examination of water and wastewater (APHA, 2005).

Preparation and characterization of Fe-AC
The fresh activated carbon was rinsed three times in deionized water and then dried at 110°C over night and cooled in a desiccator. The AC was then pulverized using ASTM standard sieves in the range of 10-16 mesh (1.18-2 mm) (ASTM, 2007). Coating of AC with iron was carried out by impregnation method and thermo-chemical reactions with using of 100 mM $FeCl_3$ solution. In this process, pH of suspension was adjusted to ~8.5 by addition of NaOH and HCl (0.1N) solution. Because the pH_{ZPC} of fresh activated carbon was 4.5, increasing the slurry pH to higher than pH_{ZPC} leads to negative charge abundance on AC surface and enhancing of impregnation process (Mondal *et al*., 2007).

The AC granules were stirred thoroughly in iron salt solution to obtain a uniform mixture. The solid to liquid ratio was 1:10 and the suspension temperature was controlled on 70°C. After 24 h the suspension was filtered and the solid phase was dried at 120°C over night. The product of this process was cooled to room temperature and washed thoroughly with deionized water until a clear supernatant was obtained. The washed granules were then dried at room temperature to be used as iron- impregnated activated carbon (Zhang *et al*., 2007).

The solid structures of Fe-AC were analyzed using scanning electronic microscopy (SEM) and energy dispersive X-Ray (EDX) techniques (XL30 Philips model). The spectrums of EDX technique approved the presence of iron on AC surface and adequate efficacy of applied impregnation process.

The Fe-AC was characterized by pH_{zpc}, Brunauer-Emmett-Teller (BET) and iodine adsorption from solution (Lowell *et al*., 2004). The pH_{zpc} of the Fe-AC was determined using the batch equilibrium technique with 1:100 solid to liquid ratios in 0.01 M $NaNO_3$ solution as an inert electrolyte (data not shown). Briefly, 0.5 g of washed and dried activated carbon was added to 50 mL of 0.01 M $NaNO_3$. The initial pH of the electrolyte solution was adjusted in the range of 2 to 12 by adding 0.1N HNO_3 and NaOH (Karthikeyen and Siva Ilango, 2007). This solution pH_s was considered as initial pH. The solutions were allowed to equilibrate for 24 h in an isothermal shaker (Model GFL 3017) with 120 rpm at 25±1°C. The suspensions were filtered through filter papers, and the pH values were measured again, using an ion pH meter as final pH. A blank test without solid phase was also made in order to eliminate the influence of interferences (Smiciklas *et al*., 2009).

The iodine number of Fe-AC was determined by using a 0.1N standardized iodine solution; titration was carried out by 0.1N sodium thiosulfate (ASTM, 2007). The BET specific surface area and Barrett-Joyner-Halenda (BJH) specific surface area were determined by nitrogen adsorption-desorption measurement and application of the linear BET equation (data not shown). Also, the porosity of Fe-AC was determined with the conventional adsorption of N_2 gas at 77 °K and 91.43 KP pressure (Lowell *et al*., 2004). Analysis of data for calculation of BET and BJH surface area (m²/g), pore volume and porosity was carried out by Belsorb software (Ver.5).

Batch studies of As adsorption with Fe-AC
Batch adsorption experiments were carried out in room temperature (25 ± 1°C) with the shaker (GFL 3017) set at 150 rpm. Starting with 100 mL

flasks, containing various amount of adsorbent (0, 5, 10, 13, 15, 20, 25 and 30 g/L) and 50-75 mL of As spiked water of known concentration (300 and 600 µg/L Arsenate) were added. The flasks were then shaken for 24 h to reach equilibrium and the supernatant was filtered through a 0.45 µm filter membrane and analyzed to find As concentration. From preliminary kinetic tests, it was concluded that 2.5 h was long enough to achieve the equilibrium. The arsenic removal efficiency and amount of arsenic adsorbed on the adsorbent were calculated by Eqs.1 and 2, respectively.

$$E = \frac{C_o - C_e}{C_o} \times 100 \tag{1}$$

$$q_e = \frac{C_o - C_e}{W_m} \times V \tag{2}$$

where E is the removal efficiency, q_e is the adsorption amount (mg/g), C_0 and C_e are the concentrations of As in the initial solution and supernatant after adsorption; V, W_m are the volume of solution (L) and adsorbent mass(g), respectively (Rezaee *et al.*, 2009; MWH 2005).

The Langmuir and Freundlich equations were used to describe the equilibrium between adsorbate and adsorbent which can be represented as:

$$\frac{C_e}{q_e} = \frac{1}{q_{max} \cdot b} + \frac{C_e}{q_{max}} \tag{3}$$

$$Log\ q_e = Log\ k + \frac{1}{n} Log\ C_e \tag{4}$$

Mechanism and energy of adsorption
Since Langmuer and Freundlich isotherms do not give information about mechanisms and energy of adsorption, this information were determined with using of Dubinin – Radushkevich isotherm which can expressed as:

$$Ln\ q_e = Ln\ q_{max} - k\ \varepsilon^2 \tag{5}$$

where ε (Polanyi potential) is [RT Ln(1+(1/C_e)], q_{max} is the adsorption capacity(mg/g), k is a constant related to adsorption energy R and T are the gas constant ant temperature (°K), respectively (Kundu and Gupta, 2007). The mean energy of adsorption (*E*) may be calculated from k value using the following equation:

$$E = - (2k)^{-0.5} \tag{6}$$

The numerical value of energy is in the range of 1-8 and 9-16 kj/mol for physical and chemical adsorption, respectively (Kundu and Gupta, 2007).
In order to predict the adsorption efficiency of the process and affinity between As(v) and Fe-AC adsorbent, the dimensionless separation factor R_L can be used by the following equation:

$$R_L = \frac{1}{1 + b\ C_o} \tag{9}$$

where C_0 is the initial concentration of As (v) (mg/L) and b is the Langmuir isotherm constant. According to Kundu and Gupta (2007) and other studies (Singh and Pant , 2004) it has been shown that in using mathematical calculations, R_L indicates the shape of the isotherm. Value of $R_L < 1$ represents the favorable adsorption but $R_L > 1$ describes unfavorable adsorption (Samarghandi *et al.*, 2009).

RESULTS
Composition and characteristics of Fe-AC
Physical and chemical analysis of prepared adsorbent were carried out with conventional methods and other available analytical techniques. Figs. 1a and 1b show the SEM (500X magnification) and EDX analysis of Fe-AC and Table 1 shows the important physical parameters and major chemical constituents of the Fe-AC adsorbent. The EDX analysis provides valuable information on the distribution of active metal in the structure based on carbon and shows the

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presence of Fe as high as 79.75 wt% on surfaces of activated carbon; so, these evidences confirm that the whole procedure is a suitable method for coating of iron on surface of activated carbon.

In SEM analyses for better reflection of the irradiated electrons from the surface of target material and preparing of obvious and brightness picture, these surfaces have been covered by Au metal and the brightness points is a good indicator for presence of metalloid elements in the surface of materials that led to reflection of electrons from the surface of adsorbent and detected with microscope detector. As shown in Fig.1a, the SEM of Fe-AC has several brightness points which is an indication of the probable presence

of Fe in these points. As mentioned above, this phenomenon was confirmed by EDX analysis.

The results obtained from N_2 equilibrium adsorption isotherm at 77°C and other surface characteristics of Fe-AC are summarized in Table 1. Comparison of the BET surface area of virgin activated carbon (data not shown) and Fe-AC showed that impregnation of adsorbent surface with iron leads to decreasing of BET surface area. This phenomenon can be related to filling of several parts of surface area, porosity and pore volume of activated carbon with iron compounds with surface area relatively smaller than activated carbon.
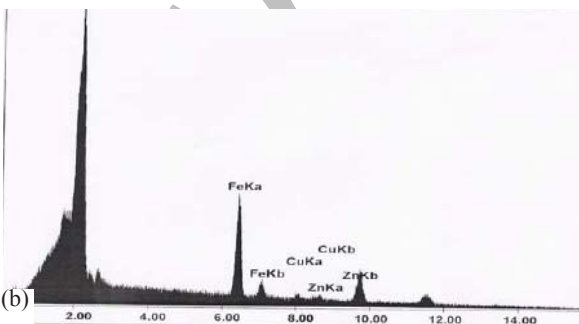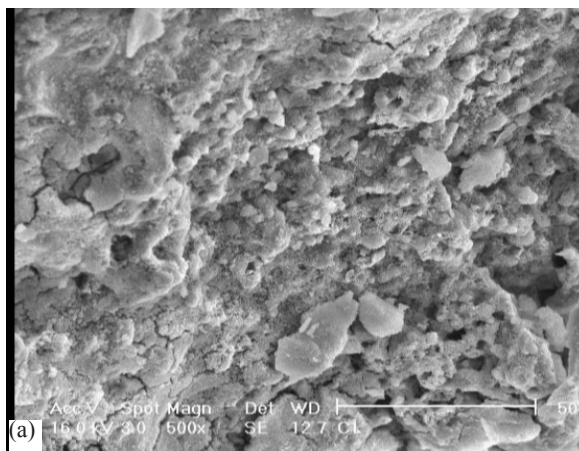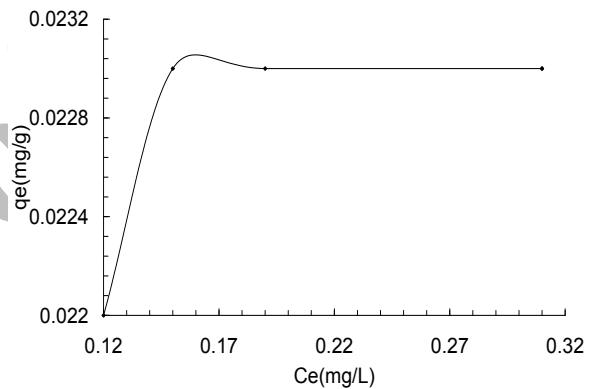

Fig. 2 : Experimental and theoretical qe versus Ce plots for the As(v) on Fe-AC (Equilibrium time=2.5 h)

Fig.1: Scanning electron micrograph of Fe-AC (a) and EDX spectrum (b)
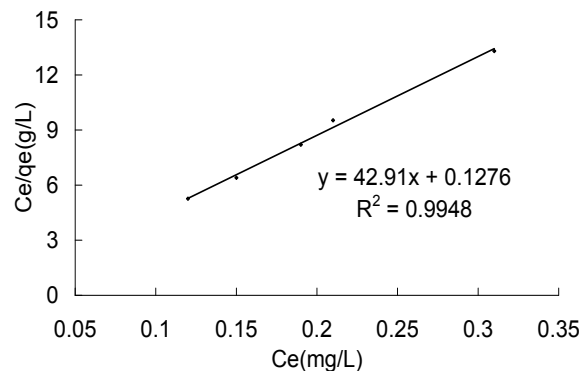
Fig. 3: Langmuir isotherm for As(v) adsorption
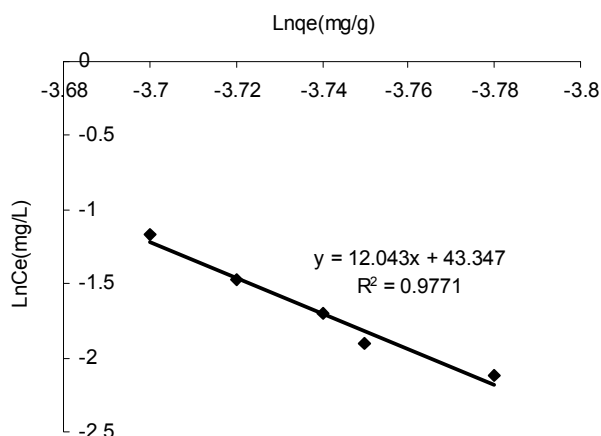
Fig. 4: Freundlich isotherm for As(v) adsorption
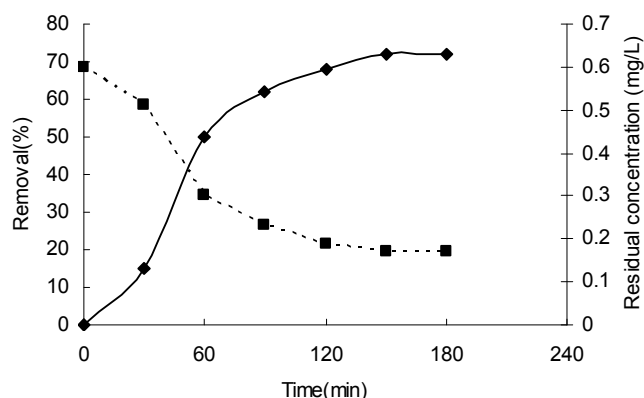
$y = 12.043x + 43.347$
$R^2 = 0.9771$

Fig.5: Removal efficiency of As(v), C_0 = 600 µg/L, Fe-AC dose=13 g/L

The sorption isotherm

The results of experimental isotherms carried out at ambient temperature are presented in Fig. 2. It can be seen that adsorption isotherm exhibits an L shape, which corresponds to the classification of Giles. In this classification, type L assumes monolayer formation in the active sites of the surface and all the adsorption sites are supposed to be equivalent.

Figs. 3 and 4 show the observed (symbols) and simulated (lines) isotherms for the Fe-Ac adsorbent. Comparisons between R^2 in these models show that the adsorption of As(v) has

a good compliancy with Langmuir adsorption model ($R^2 = 0.99$ versus 0.97). Confirmation of the experimental data into Langmuir isotherm model indicates monolayer coverage of As(v) and supports prior results shown in Fig. 2. Based on Eq. 3 and equation curve on Fig. 3, the q_{max} and b related to the maximum capacity of adsorbent and energy of adsorbent were calculated as 0.024 mg/g, and 320L/mg , respectively.

Adsorption kinetics and effect of contact time

Fig. 5 demonstrates the effect of contact time (0-180 min) on the adsorption of As(V).
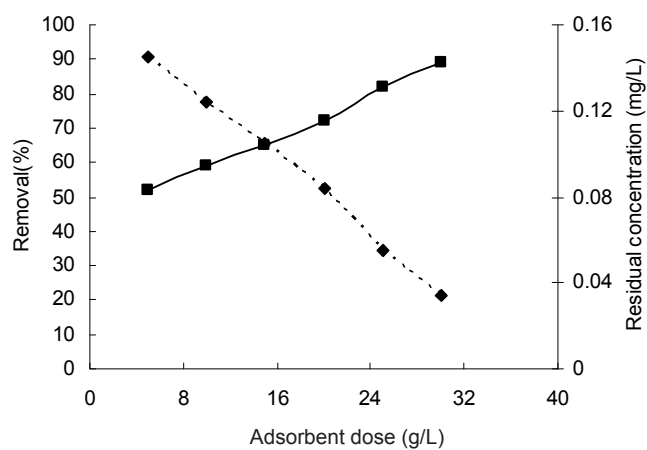
150

Fig. 6: Effect of adsorbent dose for As (v) removal, contact time=150min, C_0= 300 μg/L

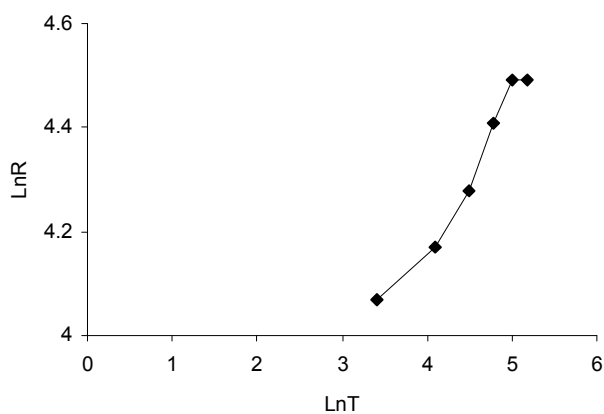Fig. 7: Ln % removal vers. Ln contact time
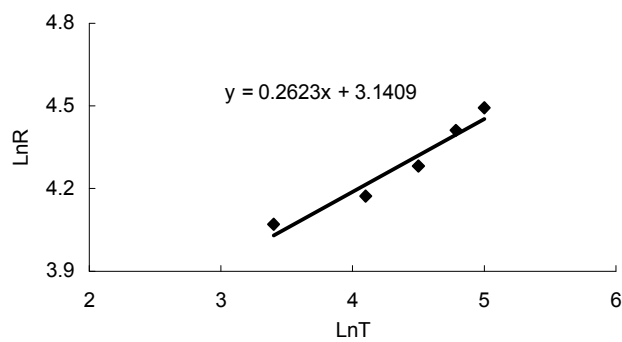
$y = 0.2623x + 3.1409$

Fig. 8: Determining of m coefficient for As adsorption kinetic

Adsorption efficiency increased with increase in contact time upto 150 min and a maximum removal efficiency of 75 % was achieved and remained relatively constant (180 min). The result of this study showed that most of the removal (62%) occurred in the contact time of 80 min. Fig. 5 shows that the adsorption of As with these conditions can not meet the Iranian Standard Institute guideline values, thus decreasing of As concentration or increasing the adsorbent dosage is necessary. As shown in Fig. 6, decreasing of As concentration to 300 µg/L and increase of Fe-AC upto 25 g/L led to meet the Iranian Standard Institute guideline values (ISIRI., 1999). Based on the relationship between arsenic removal and contact time, the adsorption kinetics may be evaluated and expressed as:

$$R = K_t t^m \tag{7}$$

Eq. 5 can be expressed in linearized form as:

$$Ln\ R = Ln\ k_t + m\ Ln\ t \tag{8}$$

where R is the As (v) removal efficiency (%), t = contact time (min) and k_t and m are the constants.

A plot of Ln R versus Ln t (Figs. 7 and 8) shows that the sorption kinetics of As with Fe-AC can be divided into two linear phases: (I) the first phase, corresponds to rapid sorption phase and (II) the second phase, in which the removal becomes constant, indicates equilibrium condition.

Applying the Eq. 8 shows the values of m and Ln k_t as 0.26 and 3.1409, respectively. For pore diffusion to be the rate limiting step, the value of m should be 0.5, and the rate of adsorption may be varied with the square root of time ($t^{0.5}$). However, in this research, the rate of adsorption varies with $t^{0.26}$ which implies that the pore diffusion is not the rate limiting step. Furthermore, as shown in Fig. 8 (that the plot do not pass through the origin), it is confirmed that pore diffusion was not the rate-limiting step (Kundu and Gupta , 2007). Therefore, the rate limiting step may be film diffusion.

According to Eq. 9 and C_0 , the value of this parameter for adsorption of As(v) on the Fe-AC was found to be 0.01 (within 0 and 1) which indicates favorable adsorption in this experimental study. Fig. 9 shows the plot of Lnqe versus ε^2. The value of ε was calculated directly from related equation and k can be calculated from the intercept of this curve. In this research, the value of E was calculated as 1.52 kj/mol which implies that adsorption of As(v) on Fe-AC is of physical type.
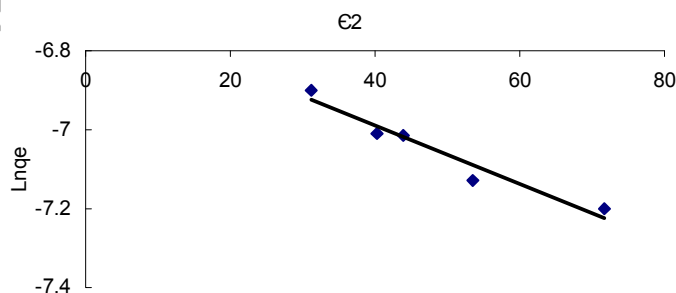

Fig. 9: D-R isotherm for adsorption of As(v) on Fe-AC

DISCUSSION

In this research, the impregnation feasibility of activated carbon with iron salt and the potential of this adsorbent for the removal of As(v) from tap water have been investigated.

The results of this study shows that the impregnation of activated carbon with $FeCl_3$ leads to increase the iron content on activated carbon surface. Comparison of the results of this research with other publications shows that the procedures used in our study are more efficient. Zhang *et al* (2007) reported that coating of activated carbon surface with iron leads to the increase of iron content upto 8% wt, but this study showed that the constituents may cover up to 79.8% wt that is higher than that reported by Zhang *et al*.(2007).

Several studies have shown that impregnation of adsorbents surfaces with salt metals leading to decrease of Iodine number and BET surface area. Zhang *et al*.(2007) reported that after impregnation of activated carbon with iron salt the BET surface area and Iodine number decreased from 1000 to 800mg/g , and from 1026m²/g to 807m²/g , respectively. Similar results were found in our study and Iodine number and BET surface area decreased from 215 mg/g to 145 mg/g, and from 630 m²/g to 478 m²/g, respecting .Since the adsorption capacity of adsorbent materials is related to the surface specification of these materials, therefore results showed that the surface characteristics of virgin and modified activated carbon used by Zhang *et al*., (2007) were better than what was used in this study.

Another surface characteristic of activated carbon is the total pore volume. In this study, the parameter of Fe-AC was 0.23 cm³/g which was lower than that used by Zhang *et al*. (2007) reported as high as 0.606 cm³/g. These differences in surface characteristics may be related to iron settled on pore volumes and surfaces of activated carbon having lower surface area than activated carbon. These results can be confirmed by the comparison of iron contents of activated carbons which were used by Zhang *et al*., and the present study in which decreasing of BET surface area in the prior study was lower (21%) than the late study (24%).

Adsorption of arsenic on Fe-AC as shown in Fig. 5 is relatively a fast reaction and increasing of contact time upto 180 min leads to increase of adsorption efficiency. Also, Fig. 5 shows that most of the removal (62%) occurs in a contact time of 80 min and maximum uptake of pollutant (75%) was achieved on 150 min. Gupta *et al*.,(2005) reported similar results but their results showed the efficiency of iron oxide coated cement (IOCC) for the removal of arsenic higher than that of Fe-AC which was used in this study. Also, their results showed that adsorption of As on IOCC was markedly faster than that achieved in this study, so the results of Gupta *et al*. (2005) show that most of the arsenic adsorption (65%) occurs in a contact time of 40 min (Gupta *et al*., 2005) but, in our study the rate of this phenomenon was slower. These differences may be related to the surface characteristics of adsorbents.

Manju *et al*.,(1998) used the copper coated coconut husk carbon for the removal of As and reported that increasing of contact time can lead to increase of As adsorption and saturation of adsorbent or equilibrium was reached after 4 h. These results show that adsorption of As by this adsorbent is slower than the Fe-Ac which was used in our study. Similar result was reported by Reazaee *et al*. (2009) in adsorption of bacterial endotoxin with bone char. They reported that the adsorption efficiency of endotoxin was elevated with laps time and equilibrium was achieved after 6h contact time and the adsorption efficiency did not change afterward (Rezaee *et al*., 2009). These findings show that the contact time of solute and adsorbents for the removal of different pollutants with adsorption process depends on the adsorbents and adsorbates characteristics and determination of the minimum contact time is critical parameter in adsorption studies.

As shown in Fig. 6, This figure shows a positive and direct correlation between the adsorption efficiency of As and Fe-AC dosage exists and indicates the availability of a large number of active sites on adsorbent surface. This phenomenon shows that the adsorption process takes place at specific sites correlated with adsorption efficiency. These results confirmed those of Gupta *et al*.(2005) which reported increasing of virgin and iron oxide coated cement dosage lead to elevation of As(v) adsorption .Karthikeyan *et al*.(2007) also reported that

increase of adsorbent dosage upto 30 g/50 mL leads to elevation of fluoride adsorption from water and adsorption did not change beyond this dosage. Although, elevation of adsorbent dosage leads to increase of active sites for adsorption, but this phenomenon may not lead to high adsorption capacity of adsorbent.

Ghanizadeh and Asgari (2009) reported that although the increasing of adsorbent dosage has positive and direct correlation with adsorption efficiency of Methylene Blue dye with bone char, but this condition attributed with decreasing of adsorbed solute on mass unit of adsorbent. These results may be related to the fact that with an increase in adsorbent dosage, several active sites remained unsaturated and in adsorption process determination of optimum dosage of adsorbent should be carried out specifically (Ghanizadeh and Asgari, 2009).

Survey of the adsorption isotherm of As(v) within Fe-AC (Figs. 3 and 4) show that based on equation curve and related R^2, As(v) adsorption could be described by Langmuir isotherm . Hsu *et al*., (2008) studied the adsorption of As with iron oxide-coated sand and reported that adsorption kinetic was fitted to the Langmuir isotherm, aswell. Similar results were reported by Anirudhan and Unithan (2007). These researchers surveyed the As(v) removal using anion exchanger and reported that adsorption isotherm in all conditions exhibits an L-shape which corresponds to Giles classification and implies monolayer adsorption in active sites of surfaces and supposing the equivalent for all sites which complies with Langmuir assumption. Based on these results, they concluded that the adsorption kinetic of As (v) complies with Langmuir isotherm (Anirudhan and Unnithan, 2007). Based on related equations and Figs 2, 3 and 4 , the reported results on As (v) adsorption kinetics confirmed the results of this experimental study.

In adsorption studies the dimensionless parameter of R_L was used for prediction of adsorption situations. Based on Eq. 9 the value of this parameter for this study was obtained as 0.01 and this level of R_L shows favorable adsorption. Manju *et al*.(1998) surveyed the adsorption of As by coconut husk carbon and reported that R_L values lies between 0 and 1. Similar result was reported by Rezaee *et al*. (2009) on adsorption of

bacterial endotoxin with bone char.

Survey of adsorption process shows this process might be controlled with film or pore diffusion. In this experimental study, determination of limiting step was carried out using Eqs.7 and 8. The results of these equations were shown on Figs. 7 and 8. As shown, the curve of Fig. 7 has 2 steps, which in the first step adsorption is fast, but in the second phase the system is equilibrated. As mentioned before; the coefficient of m could be related to adsorption conditions and shows the limiting phase of process. Plot of the curve for the second phase of Fig. 7 (shown in Fig. 8) shows that m = 0.26.

Several studies show that for pore diffusion to be the rate limiting step, the value of m should be equal 0.5 and the rate of adsorption should be varied with the square root of time ($t^{0.5}$). However, in this research, the rate of adsorption varied with $t^{0.26}$ which implies that the pore diffusion was not the rate limiting step. Furthermore, plotted curve on Fig.10 did not pass through the origin, which confirms the fact that pore diffusion is not the rate-limiting step. Therefore, the rate limiting step may be film diffusion. These results were confirmed by Kundu and Gupta (2007). In adsorption of As within iron oxide coated cement , m was reported to be 0.42 that implies the film diffusion process is a limiting phase (Kundu and Gupta, 2007) . Another study carried out by Gupta *et al*. (2005) showed similar results, so that the value of m for two types of adsorbents were determined to be 0.65 and 0.42, which implies the adsorption of As within virgin and iron oxide-coated sand limited by film diffusion step. Although in these researches the values of m were different, but the results of all of these studies show that the same process is a limiting step on As adsorption.

Since, the traditional isotherms do not give information in this field; hence the D-R isotherm was used for determination of adsorption type and the related energy. This task was carried out based on Eqs. 5 and 6. Data analysis plot of D-R isotherm in this experimental study showed that the adsorption energy As(v) on Fe-AC is equivalent to 1.52 kj/mol . The numerical value of energy in the range of 1-8 and 9-16 kJ/mol. implies the physical and chemical adsorption, respectively. Based on the value of E, the adsorption of As(v) on Fe-AC is physical. This result was confirmed in other studies. Kunda and Gupta (2007) reported

the adsorption energy of As on iron oxide-coated cement as 2.86 kj/mol which implies physical adsorption. Singh and Pant (2004) also reported the adsorption energy of As on activated alumina as 7.45 kj/mol, which confirm the results of this study and show that As(v) adsorption on Fe-AC is a physical process.

Based on the results of this study, it may be concluded that although, iron- impregnated activated carbon is one of the suitable adsorbents used for the removal of arsenic from water (As concentrations upto 300 µg/L), but improvement of the adsorption performance for the removal of higher concentrations of As and provision of the regulated standards may require the impregnation with higher concentrations of iron salt or other metals providing that their As adsorption affinities are higher than the iron.

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