

Original Article

Fluoride Removal from Aqueous Solutions Using Bauxite Activated Mines in Yazd Province (Case Study: Kuhbanan Water)

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Abstract

Introduction: A high concentration of fluoride causes health problems in man. How to remove excess fluoride from water is the objective of this project. The study focuses on an evaluation of the performance of activated bauxite mines in Yazd province in order to remove fluoride from drinking water.

Materials and Methods: This study was an experimental one conducted on synthetic samples and water from May to November of 2013 in Kuhbanan region at the Environmental Health Engineering Research Center. Bauxite was taken from Sadr Abad and Shomal-e-Yazd mines. Bauxite analysis was performed using X-Ray Fraction and X-Ray Diffraction. The effects of pH, contact time, adsorbent dose and different concentrations of fluoride on defluoridation were determined. The optimal conditions were performed on real samples. In order to better understand the processes of absorption, synthetic and adsorption isotherms were studied. Fluoride content was determined by the SPADNS method and data analysis was performed by Excel.

Results: The maximum removal efficiency for synthetic solution was obtained by 25g/L bauxite of Sadr Abad mine and 30g/L bauxite of Shomal-e-Yazd mine at pH 7, with the contact time of 180 minutes as 75.31% and 59.90%, respectively and the removal efficiency of actual samples in optimum conditions was 51.21% and 36.68%. Freundlich isotherm with a correlation coefficient greater than 99% showed better results for both types of bauxite.

Conclusion: bauxite from Sadr Abad mine is more efficient than that from Shomal-e-Yazd mine.

Keywords: Adsorption isotherm, Fluoride, Shomal-e-Yazd Bauxite, Sadr Abad Bauxite

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Introduction

Water is an essential element for survival. So the most important factor in determining the usage of water in household, industrial and agricultural sectors is the chemical composition of water that is not suitable for drinking^[1]. The presence of fluoride in drinking water, according to concentration level^[2] and duration of continuous absorption^[3] can be helpful or harmful to humans. One of the main sources of human contact with fluoride is groundwater^[3, 4]. In the periodic table of elements, fluorine is the most electronegative and reactive element^[5]. Hydroxide ions are replaced with Fluoride in hydroxyapatite ($\text{Ca}_5(\text{PO}_4)_3\text{OH}$) structure, which is the main mineral in bones and teeth and form the Fluorapatite ($\text{Ca}_5(\text{PO})_3\text{F}$). Fluorapatite is more soluble than hydroxyapatite and causes tooth loosening and drooping (Dental fluorosis). In more severe cases dental fluorosis can result in skeletal fluorosis^[6]. According to the guidelines of the World Health Organization (WHO), the maximum acceptable concentration of fluoride in drinking water for warm and cold weather is 1 and 1.2mg/L, respectively. This difference is due to the use of more water in hot weather^[7]. According to available reports in many regions of the world, the fluoride concentrations in groundwater exceed the WHO guideline limits^[5] and it is estimated that more than 200 million people worldwide use drinking water with concentrations above set standards^[3]. Excess fluoride is a cause of major concern in the public health sector^[8]. Thus, it is necessary

to develop effective and economic methods for the removal of excess fluoride from drinking water. Technologies such as coagulation and chemical precipitation, surface absorption, membranous and electrochemical processes and ion exchange water are used to remove fluoride from water^[9]. Many of these techniques are not useful in developing countries and low-income areas due to the high cost and complexity of application^[10]. Thus, a growing tendency is to use local materials and inexpensive ways to remove fluoride from water^[11]. Absorption as an affordable and common method is favored in developing countries as compared to various other techniques^[12, 13]. Metal oxides, particularly iron and aluminum are good adsorbents for the removal of anions such as fluoride^[12]. One of the available local resources is bauxite ore. Bauxite is a mineral which mainly consists of oxides alumina, minor and variable amounts of silica, iron and titania^[7]. Das et al. in India in 2005, Peter et al. in Tanzania in 2009 and Sujana et al. in India in 2011 used activated titanium rich bauxite and bauxite-rich soil for the removal of fluoride^[14-16]. Lavecchia et al. in Italy in 2012, Sajidu et al. in Malawi in 2012 (An area with prevalence of fluorosis caused by high fluoride concentration) and Sujana et al. in India in 2011 used bauxite to remove fluoride^[7, 11, 16]. However, the efficiency of bauxite mines in fluoride removal in Yazd province had not been studied till now. Therefore, the aim of the study is to evaluate the efficiency of activated bauxite

mines in Yazd province in removing fluoride from aqueous solutions.

Materials and Methods

This study was an experimental one conducted on synthetic samples and water from May to November of 2013 in Kuhbanan region at the Environmental Health Engineering Research Center. An actual sample of drinking water was collected from Kuhbanan drinking water, located in the northwest of Kerman Province.

According to previous studies, the fluoride concentration in the water resources of this area was about 2.3-5.4 ^[17]. Bauxite was prepared from Sadr Abad and Shomal-e-Yazd activated mines located in Yazd province. Deionized water was used to prepare all solutions and analyses. The pH value of samples was set to 0.1N by H₂SO₄ and NaOH. PH measurements were performed by a digital pH meter (Model: Hanna211).

The chemical quality of actual water samples was determined initially. At first, the stones of both mines used in the present study were crushed, ground and sieved to obtain <250 μm size. The segregated materials were then heat treated at 200°C in an oven for 2 h. Then they were kept in cooled room and stored in desiccators for further use. X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD) were prepared in the Iranian Mineral Processing Research Center (model Philips X'PERT).

(XRF) and (XRD) were used to determine the chemical analysis and determination of phases in samples of bauxite powder.

In order to prepare stock solution, 0.221 mg of sodium fluoride, NaF, was dissolved in 1000 mL of deionized water (1ml=100 μgF⁻) and the experimental solutions of required concentrations were prepared from this solution. Fluoride analysis was measured according to SPADNS method and by using spectrophotometer at a wavelength of 570 nm. The SPADNS colorimetric method is based on the reaction between fluoride and zirconium – dye lake. Fluoride reacts with the dye lake, dissociating a portion of it into a colorless complex anion (ZrF₆²⁻); and the dye. As the amount of fluoride increases, the produced color becomes progressively lighter. Standard curves were constructed for concentrations of 0 – 1.40 mgF L⁻¹. All analysis methods were taken from “Standard methods for examination of water and wastewater” of the twentieth editing ^[18]. Adsorption tests were used to determine the equilibrium time and also sorption kinetics, optimal conditions (pH, contact time, adsorbent amount and the concentration of fluoride) and the study of sorption isotherms models. Removal efficiency was measured at pH (3-10), the initial concentration of fluoride (2, 4, 6mg/L), bauxite dose (2, 5, 10,20,25,30 g/L) and contact time (5-240 min).

The amount of fluoride adsorbed (q_e in mg/g) was determined according to equation 1.

$$q_e = \frac{(C_i - C_e)V}{m}$$

(1)

Where C_i and C_e are the initial and final concentrations of fluoride in solution (mg/L), V is the volume of solution (L) and m is mass of the adsorbent (g).

Langmuir and Freundlich isotherms models were investigated in order to study sorption

isotherms. Tests with real samples were also performed in optimal conditions and the removal efficiency was calculated. Data analysis was performed by Excel software.

Results

The results of the chemical quality of the drinking water of Kuhbanan region are shown in Table 1.

Table 1: Chemical quality of the drinking water of Kuhbanan

Parameter	Unit of measurement	value
pH	-	6.9-7.66
Temperature	(°C)	25-27
Sulfate (SO ₄ ²⁻)	mg/L	139
Chloride (Cl ⁻)	mg/L	126
Total solids (TS)	mg/L	711
Nitrate (NO ₃ ⁻)	mg/L	5.6
Calcium	mg/L	83
Potassium	mg/L	1.9
Magnesium	mg/L	49
Sodium	mg/L	95
Total dissolved solids (TDS)	mg/L	656
Residual chlorine	mg/L	0.5
Fluoride (F ⁻)	mg/L	2.3-2.7
Iron (Fe)	mg/L	<0.05
Aluminum(Al)	mg/L	<0.01

XRF and XRD analysis results of bauxite of Sadrabad and Shomal-e-Yazd mines are

presented in Table 2 and Figures 1 and 2, respectively.

Table 2: The Percentage of Metal oxides measured in bauxite of Sadr Abad and Shomal-e-Yazd mines by XRF method

Constituent %	bauxite Shomal-e-Yazd	bauxite Sadr Abad
Al₂O₃	34.38	45.77
Fe₂O₃	19.93	24.77
SiO₂	17.27	10.20
TiO₂	4.74	5.87
CaO	6.62	0.52
MgO	3.35	2.35
K₂O	1	0.01
P₂O₅	0.09	0.10

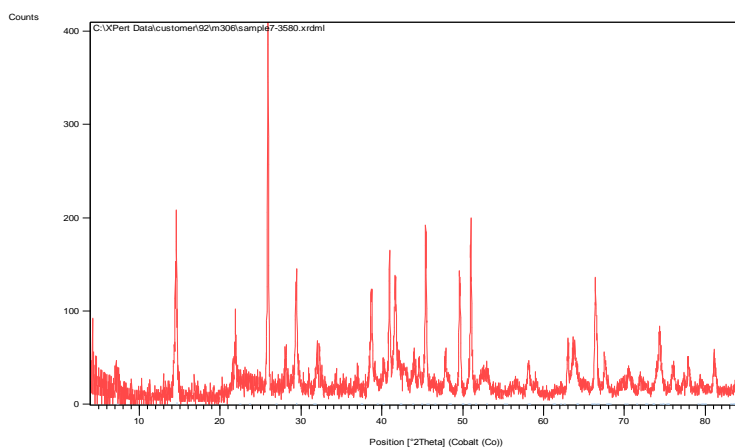


Figure 1: XRD pattern of Sadr Abad mine

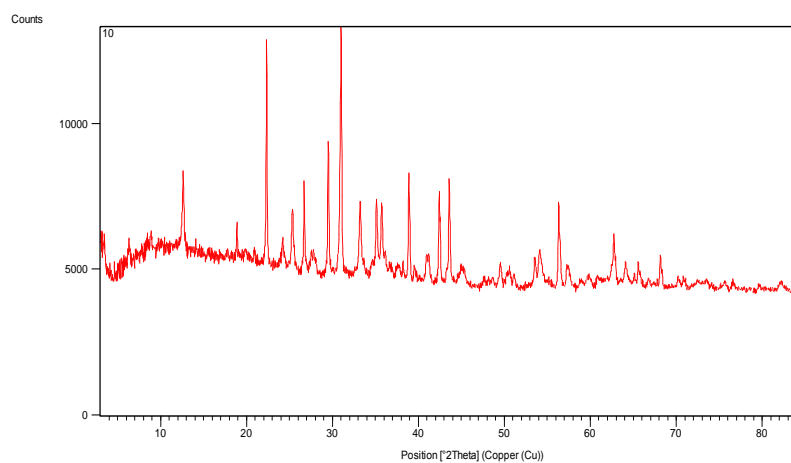


Figure 2: XRD pattern of Shomal-e-Yazd mine

The results of the effect of contact time on fluoride absorption capacity by bauxite of Sadr

Abad and Shomal-e-Yazd are shown in Figure 3.

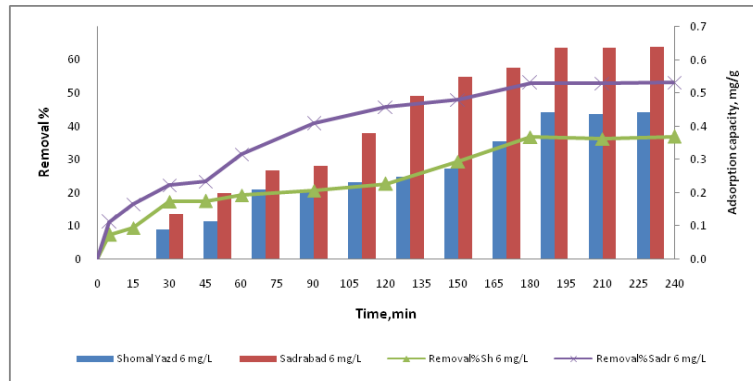


Figure 3: The effect of contact time on the absorption capacity of fluoride (1 g of bauxite, pH 7 ± 0.1 , the fluoride concentration 6mg/L, shaking in 300 rpm and room temperature)

The results showed that the removal efficiency of fluoride at different pHs by both ore bauxites (Sadr Abad and Shomal-e-Yazd mines) had a similar outcome. Removal efficiency was low in acid and alkaline pH. In a situation involving constant concentration of 6mg/L and constant value of 5g/L of bauxite of Sadr Abad mine and 180 minutes for contact time with increasing pH from 3 to 7, fluoride removal efficiency increased from 8.80% to 53.11% and at pH 10 it decreased to 11.34%.

The removal efficiency in similar conditions by the bauxite of Shomal-e-Yazd mine changed from 8.12% to 36.82%. For both bauxites, the maximum absorption was obtained at pH 7. The study of the effect of initial concentration of fluoride on removal efficiency showed the following: with increasing initial concentrations of fluoride at concentrations of 2, 4 and 6mg/L at 180 min, removal efficiency by 5 g/L of Sadr

Abad mine's bauxite decreased to 74.67, 60.35 and 53.11 % and by bauxite of Shomal-e-Yazd mine it decreased to 54.06, 41.24 and 36.82%, respectively. With mass increasing Sadr Abad mine bauxite from 2 to 25gr/L and fluoride concentration of 6mg/L at 180 minutes; removal efficiency changed from 46.58 to 75.31%. In similar conditions, Bauxite of Shomal-e-Yazd mine reached equilibrium of 35gr/L and the removal efficiency of 55.13% was obtained.

In order to study the fluoride adsorption kinetics by bauxite of Sadr Abad and Shomal-e-Yazd mines, Pseudo first order and Pseudo second order kinetics were studied. Fluoride removal by the Sadr Abad bauxite followed Pseudo first order kinetic ($R^2=0.987$) and it followed Pseudo second order kinetic ($R^2=0.989$) by Shomal-e-Yazd bauxite. Pseudo first order and Pseudo second order kinetics results are shown in Figures 4 and 5.

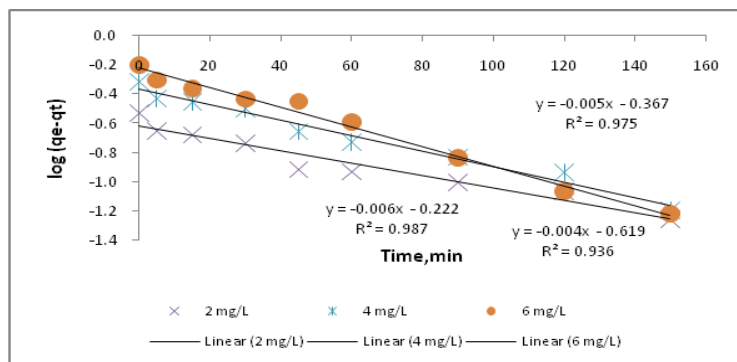


Figure 4: Pseudo first order kinetic (Initial concentrations of fluoride 2, 4, 6 mg/L, 5g/L bauxite of Sadr Abad mine shaking in 300 rpm and room temperature)

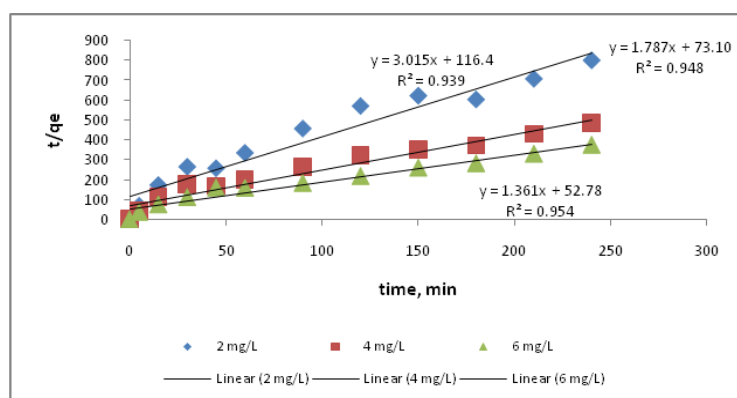


Figure 5: Pseudo second order kinetic (Initial concentrations of fluoride 2, 4, 6 mg/L, 5g/L bauxite of Shomal-e-Yazd mine, shaking in 300 rpm and room temperature)

Constants of Pseudo second order kinetic for initial fluoride concentrations of 2, 4, 6 mg/L are listed in Table 3.

Table 3: The constants of Pseudo first order and Pseudo second order kinetics of initial fluoride concentrations (2, 4, 6 mg/L)

absorption kinetic	Parameter	Bauxite of Shomal-e-Yazd F ⁻ , mg/L			Bauxite of Sadr Abad F ⁻ , mg/L		
		2	4	6	2	4	6
Pseudo first order	k_1	0,0046	0,0046	0,0046	0.009	0.011	0.013
	q_e	0,511	0,644	0,744	0.538	0.692	0.800
	R^2	0,780	0,784	0,888	0.936	0.975	0.987
Pseudo second order	k_2	0,146	0,154	0,042	0.078	0.043	0.035
	q_e	0,233	0,352	0,494	0.331	0.559	0.734
	R^2	0.949	0.989	0.891	0.939	0.948	0.954

The study of the data of Langmuir and Freundlich adsorption isotherm models showed that fluoride removal of Sadr Abad and Shomal-e-Yazd bauxite mines is better explained by

Freundlich isotherm model. Freundlich adsorption isotherm model for both mines is shown in Figures 6 and 7.

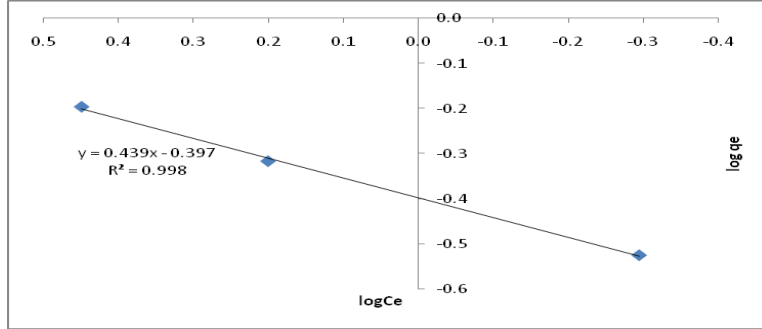


Figure 6: Freundlich adsorption isotherm (pH 7, the amount of bauxite of Sadr Abad Yazd mine 5g/L, F⁻ concentration 2, 4, 6mg/L, shaking in 300 rpm and room temperature)

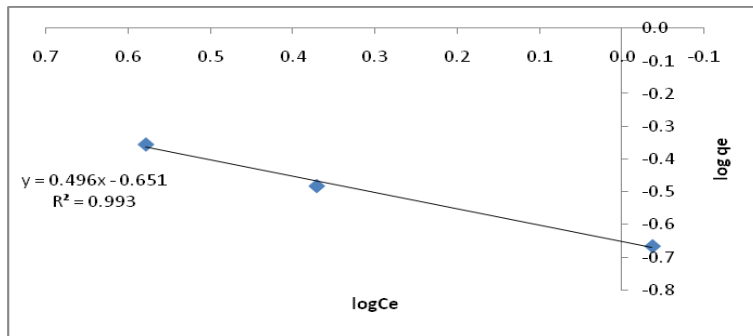


Figure 7: Freundlich adsorption isotherm (pH 7, the amount of bauxite of Shomal-e-Yazd mine 5g/L, F⁻ concentration 2, 4, 6mg/L, shaking in 300 rpm and room temperature)

The parameters of adsorption isotherm models by Sadr Abad and Shomal-e-Yazd bauxite mines are given in Table 4.

Table 4: Langmuir and Freundlich isotherm parameters for adsorption of fluoride on Sadr Abad and Shomal-e-Yazd bauxite mines

Isotherm model	Bauxite of Shomal-e-Yazd			Bauxite of Sadr Abad		
	q _m	k _L	R ²	q _m	k _L	R ²
Langmuir	0.663	0.485	0.964	2.132	0.967	0.982
Freundlich	n	k _f	R ²	n	k _f	R ²
	2.016	0.521	0.993	2.277	1	0.998

The study of the results of fluoride removal from real water samples by bauxite of Sadr Abad and Shomal-e-Yazd mines showed that when 25gr/L of bauxite of Sadr Abad mine was added to Kuhbanan water samples, the fluoride concentrations were equal to 2.74mg/L at 180 minutes and neutral pH (7-7.66); Fluoride concentration decreased to 1.28mg/L (Removal efficiency of 53.21%) and in the same conditions by addition of 35mg/L bauxite of Shomal-e-Yazd mine, the fluoride concentration decreased to 1.74 (Removal efficiency of 36.82%).

Discussion

XRF analysis showed that oxides alumina, silica, iron and titania are four main elements of bauxite ore in Sadr Abad and Shomal-e-Yazd mines which form 86.65 and 76.32% of their chemical compounds, respectively. This combination is similar to the reported elements of bauxite in the studies of 2011 in India by Sujana and the study of 2012 in Italy by Lavecchia et al. [7, 16], but the percentage of composition was different. Titanium and silica percentage in Sadr Abad and Shomal-e-Yazd mines showed high values compared to the results of these studies. Gomoro et al. in Ethiopia in 2012 attributed bauxite combinations a different percentage in various parts to laterite stage of soil that occurs in the last stages of bauxite formation [19]. XRD patterns of bauxite from Sadr Abad mine showed that diaspore, hematite, chlorite and anatase form the main phase and rutile, Lizardyt, chlorite and corundum are the minor phases. XRD patterns of bauxite from Shomal-

e-Yazd mine showed that diaspore, hematite, dolomite, quartz and anatase were the main phases and calcite, rutile, clinocllore, muscovite were the secondary phases.

The results showed that at a constant mass of 5gr/L of bauxite from Sadr Abad and Shomal-e-Yazd mines, fluoride concentration of 6mg/L, increased with time from 5 to 180 minutes; absorption capacity increased from 0.13 to 0.63mg/g and 0.08 to 0.44mg/g. During the initial period, many active sites were present on the adsorbent surface. Therefore, the removal efficiency increased with time. Sujana et al. in India in 2011 used bauxite to remove fluoride from contaminated groundwater and a similar behavior was observed with the passage of time [16].

PH is one of the factors that have significant impact on the adsorption process. According to the results, fluoride removal efficiency in both bauxites at neutral pH was the highest removal percentage and at acidic and alkaline pH was the lowest removal percentage. Due to formation of weak hydrofluoric acid (HF) efficiency was negligible at pHs less than 5. Due to the competition of hydroxide ions (OH^-) and fluoride ions (F^-) for adsorption position on bauxite, a significant absorption was not observed at PHS over 10. The obtained results corresponded with the results of the studies of Sajana et al. in India in 2011, Sajidu et al. in Malawi in 2012 and Mohapatra et al. in India in 2004 [11, 16, 23]. The initial concentration of fluoride is the most effective factor for transformation of fluoride ion from the liquid phase to adsorbent solid phase [20]. The results

showed that in constant contact time (180 min) and constant amount of bauxite (5g/L), with increasing concentrations of fluoride from 2 to 6 mg/L by bauxite of Sadr Abad and Shomal-e-Yazd the removal efficiency reduced from 74.67 to 53.11% and 54.06 to 36.82%, respectively. Gomoro et al. conducted a survey in Ethiopia in 2012 and reached a similar conclusion. They expressed the saturation of absorption active sites as a reason for this reduction ^[19].

By increasing bauxite mass, removal efficiency increased, which could be due to increasing active sites in the higher absorption amount. Removal efficiency reached equilibrium with increasing mass absorbent of 25 and 35g/L for bauxites of Sadr Abad and Shomal-e-Yazd mines and the removal efficiency was roughly constant at higher values. In 2011 Malakootian et al. used industrial waste of vegetable oil, in 2009 Sujana et al. used minerals containing iron in India and in 2004 Mohapatra et al. used mineral oxides for removal of fluoride in India. They found that an increase in the amount of adsorbent did not have a significant effect on removal efficiency and this would explain that in presence of excess adsorption remains only a limited amount of free fluoride ions, which corresponds with the results of this study ^[21-23]. In amounts greater than 35gr/L of bauxite of Shomal-e-Yazd mine, fluoride concentrations were more than the WHO guidelines. It can also be said that the bauxite of Shomal-e-Yazd mine did not have the required performance to remove fluoride in concentrations greater than

6mg/L. Lavecchia, Sajidu and Sujana et al. have shown that Langmuir and Freundlich isotherm models can be used to describe the adsorption of fluoride on bauxite ^[7, 11, 16]. Therefore, the experimental data were analyzed by the two adsorption isotherms models. The outcomes showed that Freundlich adsorption isotherm provided better results than Langmuir isotherm. The correlation coefficient of Freundlich adsorption isotherm for bauxite of Sadr Abad mine is higher than 0.99 ($R^2=0.998$) and for bauxite of Shomal-e-Yazd mine is ($R^2=0.993$). The results showed that when 25gr/L of bauxite of Sadr Abad mine was added to Kuhbanan water samples, the fluoride concentration decreased from 2.74 to 1.28mg/L and residual fluoride concentrations descended below the guidelines of WHOM (< 1.5 mg/L). In the same conditions with the addition of 35 gr/L of bauxite from Shomal-e-Yazd mine the fluoride concentration decreased to 1.74 higher than the WHO guidelines. In 2012 Gomoro and his colleagues in Ethiopia found a significant relationship between the fluoride removal capacity and percentages of Al_2O_3 and Fe_2O_3 ^[19]. Percentages of Al_2O_3 and Fe_2O_3 in bauxite of Sadr Abad mine (70.24%) were more than those of Shomal-e-Yazd mine (54.31%). So it could be the reason for the high fluoride removal capacity of Sadr Abad Bauxite. Removal efficiency in real samples by bauxite from Sadr Abad and Shomal-e-Yazd mines were 53.21 and 36.37%, respectively and it was less than synthetic samples that could be due to the presence of interfering ions in real samples such as nitrate, chloride, and sulfate.

The presence of these ions with fluoride competed with the absorption of zinc, iron, and aluminum of bauxite.

Thus, the removal efficiency of the actual sample decreased. In 2011 Sujana et al. in India used bauxite to remove fluoride from ground water and expressed that the presence of nitrate, chloride and sulfate was the result of removal efficiency of fluoride reduction in ground water samples ^[16].

Conclusion

A comparison of the data associated with the study showed that the efficiency of Sadr Abad mine's bauxite was more than that of Shomal-e-Yazd in fluoride removal. However, both bauxite results showed that increasing contact time and adsorbent dosage in neutral pH raises removal efficiency. As an available and abundant matter in many parts of the country and also due to

having such features as easy application, low cost, effectiveness and not requiring sophisticated technology, Bauxite can be used as an adsorbent to remove fluoride from contaminated water. An assessment of the regenerative adsorbent and optimization process is suggested for future studies.

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