



Chemical adsorption of iron ions from drinking water using Jordanian zeolitic tuff

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ABSTRACT

Jordanian natural zeolitic tuff is distributed in the northeast, central and southern parts of Jordan. Zeolitic tuff used in this work was brought from Al Hala Volcano piles located in the southern part of Jordan to investigate its capability to remove iron ions from drinking water. The main zeolitic minerals identified in Al Hala are phillipsite and chabazite. The effects of iron concentration, pH values, contact time, zeolite grain size and amounts of used zeolitic tuff were examined in the removal process using batch experiments. Two zeolitic tuff grain sizes with two different amounts were used and designated as HZ1 and HZ2, two iron concentrations from two different water wells were investigated and designated as W1 and W2 were examined for different contact time periods. Batch tests were performed to determine the ability of zeolite to remove Fe³⁺ (iron III) from drinking water. In static regime experiments, the use of the HZ2 type shows a higher percentage of Fe³⁺ removal as compared to the HZ1 type for the same time and concentration. The results indicate that iron ions can be removed from drinking water with approximately 100% efficiency at the beginning of the contact time regardless the concentration of iron ions in the water. Al Hala zeolitic tuff used in iron removal showed a high capability with complete Fe³⁺ removal from drinking water. The HZ2 is more efficient in iron ions removal than HZ1, the pH value is not significantly affected.

Keywords: Zeolitic tuff; Al Hala; Drinking water; Phillipsite; Iron ions

1. Introduction

The iron ion is a chemical contaminant in drinking water, causes many health problems. Several sources of drinking water suffer from high iron ion content in Jordan. That is why it is important to remove the iron ions from drinking water by cheap and available methods. McPeak and Aronovitch [1] mentioned four methods for iron removal from drinking water that are: oxidation followed by filtration, clarification followed by filtration, ion exchange, and iron removal by manganese greensand. One of the most important methods used to remove iron ion from water is by ion exchange capacity and cation selectivity.

There are some adsorbents, that are applied into the treatment of dye wastewater, such as activated carbon, bio-char, nanomaterials, and zeolite. Zeolites can be defined as microporous crystalline hydrated aluminosilicates, which can be used in various applications because of their unique physicochemical characteristics, such as ion exchange and adsorption-desorption properties (Ghasemi et al. [2]). Zeolite minerals are highly selective for cations and anions (Colella [3]). Zeolites can be used in radioactive wastewater treatment due to their high ion exchange selectivity and resistance to degradation from radiation (Ames [4]). Natural zeolite was applied in removing heavy metals from water (Pansini [5]). Natural zeolites have been utilized to

remove ammonium ions from wastewater (Mercer et al. [6]). Elboughdiri et al. [7] applied iron coating on the Saudi Arabia volcanic tuff for enhancing mercury adsorption from synthetic wastewater. The applications of natural zeolite include pollutants removal, both gas and wastewater, construction, catalyst, medical uses and food and agriculture (Hardi et al. [8]).

Natural zeolites in Jordan are available in huge quantities. One of the most relevant studies was carried out by Al Dwairi [9], who investigated the associated zeolites with pyroclastic in basaltic eruptions in Jordan and classified them into three areas: Northeast, Central, and South parts of Jordan. Many research works dealt with zeolitic tuff focusing on the geneses, geology, mineralogy, and petrology. Also, many research works rendered special care to the use of natural zeolite as ion exchangeable material in water and wastewater treatment (Blanchard et al. [10]; Zamzow et al. [11]; Wingenfelder et al. [12]; Shaheen et al. [13]; Zhang et al. [14]; Abd El-Azim and Mourad [15]; Omar et al. [16]; Borowski et al. [17]; Belova [18]; Elboughdiri [19]; Al-Abbad and Al Dwairi [20]).

Almjadleh et al. [21] utilized the natural and modified Jordanian zeolitic tuff for removal of cadmium (II+) from aqueous solutions. Manolov et al. [22] used the Jordanian zeolitic tuff as a raw material for the preparation of substrates used for plant growth. They concluded that the Jordanian zeolitic tuff has specific properties, such as high ion exchange capacity, high content of macro and microelements, which makes them among the good alternatives to the traditional potting media. Also, Aljbour et al. [23] removed phosphate from aqueous solutions by using natural Jordanian zeolitic tuff. Moreover, Al-Makhadmeh and Batiha [24] used Jordanian kaolin and zeolitic tuff for the removal of iron and copper from aqueous solutions using adsorption methods.

Al Dwairi [9] investigated the characterization of Jordanian zeolitic tuff and its potential use in wastewater treatment. Ibrahim and Jbara [25] have employed natural phillipsite-faujasite tuff from Jordan to remove paraquat from synthetic wastewater. They found that natural zeolites are suitable to remove paraquat. Al Dwairi et al. [26] made further tests and investigations on natural zeolite from south Jordan (Al Hala) to evaluate its adsorption capacity using breakthrough curves and by applying the Thomas and Yoon and Nelson models. The Thomas model analysis of the measured breakthrough curves revealed that the adsorbent HZ2 has a higher adsorption capacity to Cr(VI) ions (56.3 mg/g) than HZ1 (35.5 mg/g). Al-Abbad and Al Dwairi [20] have evaluated the effect of Jordanian zeolitic tuff from Al Hala Mountain in south Jordan for the removal of nickel(II) ions from water by Jordan natural zeolite as sorbent material. It was concluded that Al Hala zeolitic tuffs have shown acceptable efficiency in the removal of pollutants from the water.

Extending to the above studies, Al Hala zeolitic tuff (HZ) possess a positive impact on purification of water from sodium, cadmium, copper, and nickel ions. The capability of zeolitic tuff on removing iron(III) ions from drinking water need to be investigated. In this work, the zeolitic tuff from Al Hala Volcano in Jordan will be used to remove iron ions from the contaminated water. Jordanian zeolitic

tuff is plenty and available in most of the Jordanian areas with low cost. The implementation of Al Hala zeolitic tuff is expected to reduce the water purification cost and improve drinking water quality. Experiments for two zeolitic tuff grain sizes with two different amounts, two different water iron ion concentrations with different contact time will be performed; aiming to find the most suitable zeolitic tuff grain size, amount, and contact time for the selected water samples.

2. Materials and methods

The zeolitic tuff (ZT), which is used in this work was collected as channel samples from Al Hala Volcano located in southern part of Jordan as shown in Fig. 1. Al Hala zeolitic tuff is characterized by unique physicochemical properties, high ion-exchange capacity, excellent adsorption-desorption properties, low cost and widely available in Jordan. Tables 1 and 2 show the chemical composition (wt.%) and the physical characteristics of volcanic tuff from Al Hala Volcano. The mineral is crushed using a jaw crusher with an aperture of 5 cm (Fig. 2), then it was sieved into 0.350–0.750 mm sizes and designated as (HZ1) as shown in Fig. 3a, which is characterized by a high content of zeolitic minerals reaching to 85% of the total volume. Some of the sieved mineral (HZ1) was milled producing powder using a ball mill and designated as HZ2 (Fig. 3b). No other modifications or chemical treatments were carried out on all samples that are used in this work.

Scanning electron image shows that the main zeolitic mineral is phillipsite (Fig. 4). All physical processing were



Fig. 1. Location map of the southern Jordan basaltic tuff showing the Al Hala Volcano (Google Maps).

Table 1
Chemical composition (wt.%) for volcanic tuff from Al Hala Volcano (Al Dwairi [9])

Sample #	SiO ₂	Na ₂ O	Fe ₂ O ₃	MgO	Al ₂ O ₃	K ₂ O	CaO	MnO	TiO ₂	P ₂ O ₃	CO ₂	Sum
1	41.70	0.521	15.50	6.67	15.60	0.94	7.62	0.199	3.28	0.70	6.80	99.53
2	40.80	0.670	15.80	7.23	16.90	0.80	6.70	0.210	3.17	0.90	6.10	99.28
3	39.60	0.340	16.01	6.89	16.17	0.82	8.10	0.220	3.50	0.81	6.90	99.36
Average	40.70	0.510	15.77	6.93	16.223	0.853	7.473	0.210	3.32	0.803	6.60	99.40

Table 2
Physical characteristics of Al Hala zeolite

Density	1,950–2,350 kg/m ³
Bulk density	0.700–0.770 kg/m ³
Porosity	60.5%
Specific surface area	13.85–14.75 m ² /g
Permeability	0.001
Pore size	0.00035–0.00067 μ
pH	7.7–8.0
Color	White
Solubility	Insoluble
Melting point	>1,600°C
Boiling point	335°C

carried out in Natural Resources and Chemical Engineering Department Laboratories at Tafila Technical University.

The water used in all batch experiments was obtained from two wells; the first sample (W1) was obtained from the effluent from Sad Attannour well located in Wadi Elhesa



Fig. 2. Zeolitic tuff from Al Hala Volcano.



a



b

Fig. 3. Processed zeolitic tuff used in experiments: (a) HZ1 with 0.35–0.75 mm and (b) HZ2 powdered zeolitic tuff.

area, northern part of Tafila, while the second water sample (W2) was obtained from Al-Damkhie area, eastern part of Karak Governorate (Table 3). Water samples were filtered using filter paper to get rid of suspended solids.

The ions exchange mechanism is performed by adsorption process. Zeolites contain calcium ions (Ca^{2+}) attached

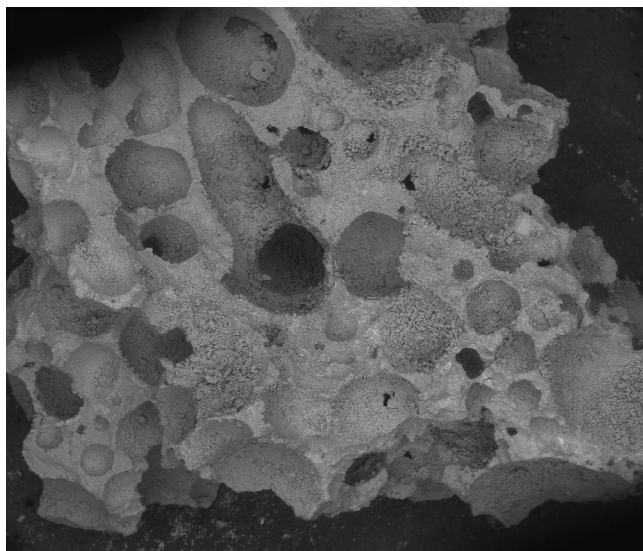


Fig. 4. Scanning electron image for zeolitic mineral from Al Hala area.

to its structure by ionic bond, when the iron ions (Fe^{3+}) contact the zeolite, the Fe^{3+} has more attractive forces than Ca^{2+} (because the charge of Fe^{3+} is more than Ca^{2+}) so that, the Fe^{3+} replaces Ca^{2+} and the Fe^{3+} are removed from water. The released calcium causes an increase in its concentration in the water as demonstrated in Fig. 5. Thus, the pH value is not significantly affected during this process. After adsorption process, no changes in the zeolite structure or surface characterization just the change occur on the type of anion exchanged.

2.1. Experimental procedure

Batch experiments were conducted to investigate HZ capacity to remove Fe^{3+} ions from drinking water. The effects of iron concentration, contact time, zeolite amounts, and zeolite grain size were examined in this work. Two types of HZ were used: HZ1 and HZ2 (Table 4), and the removal experiments were carried out by using glass flasks containing a specific volume of 100 ml of drinking water. Two fixed amounts of zeolitic tuff (5 and 10 g), which contains almost 60% from zeolitic mineral were added to 100 mL drinking water samples. The 5 and 10 g used mass of HZ zeolitic tuff is suitable for the adsorption process as it has been noted from the performed experiments. The containers were stirred for different time periods (2, 6, 8, 10, 12, and 24 h). The treated wastewater was filtered and analyzed for Fe amounts. The percentage removal of Fe ions from solution was determined using the following equation:

Table 3

Chemical composition and specifications of W1 and W2 samples and their comparison with the specifications of drinking water in Jordan

Test type		W1	W2	Jordanian drinking water specifications [27]
Chloride Cl^{-} (mg/L)		0.0	162	<500
Turbidity (NTU)		24.9	28	<5.0
Total dissolved solids (mg/L)		312	645	<1,000
Specific conductivity ($\mu\text{S}/\text{cm}$)		624	1,140	<400
pH		7.58	7.44	6.5–8.5
NH_4^{1+} (mg/L)		0.01	0.15	<0.2
Fe^{3+} (mg/L)		3.06	1.70	<1
Total hardness (mg/L)	CaCO_3	100	116	120–180
	Mg^{2+}	40	41.1	20–50
	Ca^{2+}	60	89.3	1.0
Total coliform		<1	<1	<1
<i>Escherichia coli</i> (<i>E. coli</i>)		<1	<1	<1

Table 4

Used zeolitic tuff grain size and Fe concentration of W1 and W2

Al Hala zeolitic tuff		Drinking water	
Sample	Grain size	Location	Fe^{3+} (mg/L)
HZ1	(0.350–0.750) mm	Sad Attannour well (W1)	3.06
HZ2	Grounded to powder	Al-Damkhie well (W2)	1.70

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

where C_0 and C_t are the initial and residual concentration at time t in (mg/L).

2.2. Methods of chemical analysis

Chemical analysis of original and treated water samples were conducted according to the standard methods of the water and wastewater examination. The determination of iron metal concentration was carried out using DR 6000™ UV-VIS Spectrophotometer at wavelength of 510 nm with an accuracy of ± 1 nm.

3. Results and discussion

Batch experiments were carried out using two types of Al Hala zeolitic tuff (HZ1 and HZ2) on two types of

drinking water wells (W1 and W2) containing different concentrations of Fe ion (Table 3). The results obtained from these experiments are listed in Tables 5–8 and illustrated in Figs. 6–9. It is important to mention that the insets indicate the enlarged regions of the main curves to exhibit the error percentages in a clear manner.

It is noticeable that HZ2 showed a faster removal and higher efficiency of iron removal from water compared to HZ1.

The adsorbed iron was trapped in the HZ structure, so the rich iron zeolite can be used as slow-release fertilizer. As shown in Tables 5–8, the Fe^{3+} concentration is decreased with stirring time.

To verify the achieved results in this work, the removal percentage has been compared with those reported by Lima et al. [28], who used zeolite to remove iron from irrigation water, they achieved 98% removal percentage which is well compared to the results achieved in the present work.

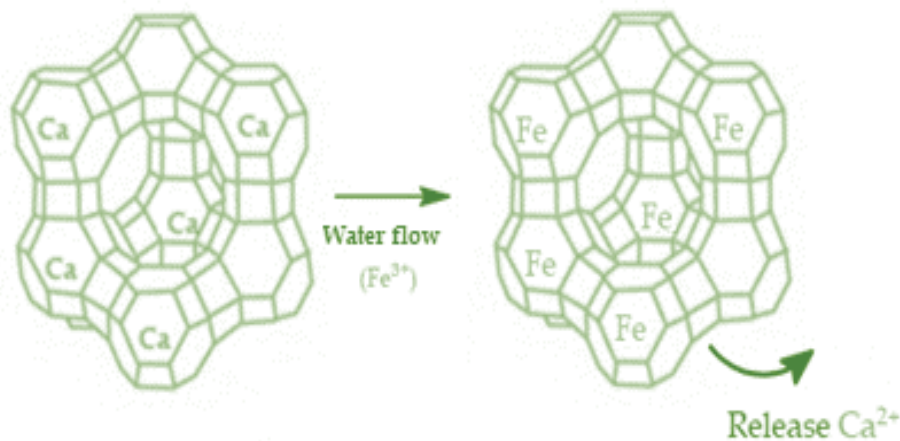


Fig. 5. Mechanism of ion-exchange between Ca^{2+} and Fe^{3+} .

Table 5
Percentage removal obtained from batch experiments for Fe^{3+} from W1 using 5 g of HZ

Al Hala zeolitic tuff	Initial well concentration C_0 (mg/L)	Stirring time (h)	Treated water concentration C_t (mg/L)	Percentage removal (%)
HZ1	3.06	2	0.30	87.2
		6	0.25	91.8
		8	0.20	93.4
		10	0.15	95.0
		12	0.10	96.7
		24	0.10	96.7
		24	0.20	93.4
HZ2	3.06	6	0.15	95.0
		8	0.10	96.7
		10	0.05	98.3
		12	0.03	99.0
		24	0.01	99.6
		24	0.01	99.6

Table 6
Percentage removal obtained from batch experiments for Fe³⁺ from W1 using 10 g of HZ

Al Hala zeolitic tuff	Initial well concentration C_0 (mg/L)	Stirring time (h)	Treated water concentration C_t (mg/L)	Percentage removal (%)
HZ1	3.06	2	0.25	91.8
		6	0.15	95.0
		8	0.10	96.7
		10	0.08	97.3
		12	0.05	98.3
		24	0.03	99.0
		2	0.20	93.4
HZ2	3.06	6	0.10	96.7
		8	0.08	97.3
		10	0.03	99.0
		12	0.01	99.6
		24	0.01	99.6

Table 7
Percentage removal obtained from batch experiments for Fe³⁺ from W2 using 5 g of HZ

Al Hala zeolitic tuff	Initial well concentration C_0 (mg/L)	Stirring time (h)	Treated water concentration C_t (mg/L)	Percentage removal (%)
HZ1	1.07	2	0.22	87.0
		6	0.20	88.2
		8	0.15	91.1
		10	0.10	94.1
		12	0.09	94.7
		24	0.05	97.0
		2	0.15	91.1
HZ2	1.07	6	0.11	93.5
		8	0.09	94.7
		10	0.04	97.6
		12	0.02	98.8
		24	0.01	99.4

Table 8
Percentage removal obtained from batch experiments for Fe³⁺ from W2 using 10 g of HZ

Al Hala zeolitic tuff	Initial well concentration C_0 (mg/L)	Stirring time (h)	Treated water concentration C_t (mg/L)	Percentage removal (%)
HZ1	1.70	2	0.20	88.2
		6	0.18	89.4
		8	0.13	92.3
		10	0.10	94.1
		12	0.07	95.8
		24	0.03	98.2
		2	0.10	94.1
HZ2	1.70	6	0.04	97.6
		8	0.01	99.4
		10	0.00	100
		12	0.00	100
		24	0.00	100

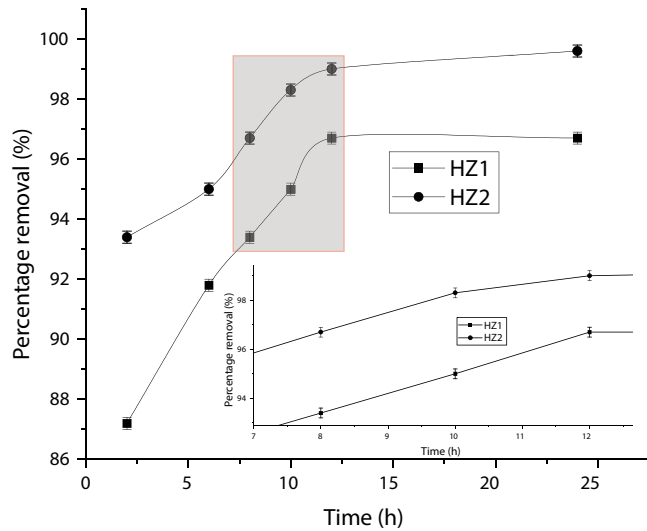


Fig. 6. Percentage removal obtained from batch experiments for Fe^{3+} from W1 using 5 g of HZ. The inset indicates the enlarged region of the main curves to exhibit the error percentages in a clear manner.

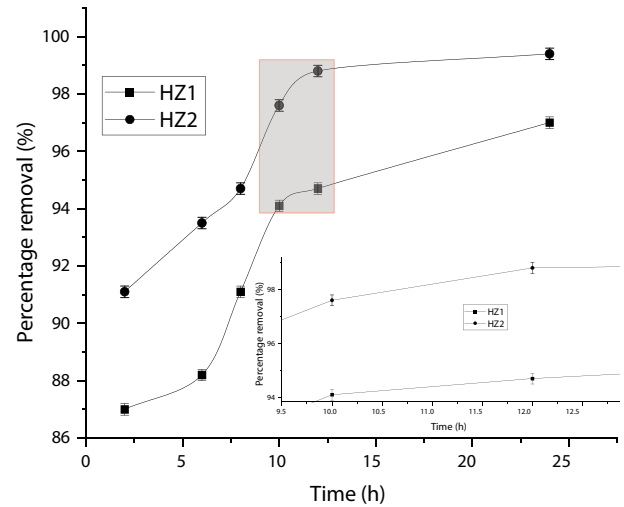


Fig. 8. Percentage removal obtained from batch experiments for Fe^{3+} from W2 using 5 g of HZ. The inset indicates the enlarged region of the main curves to exhibit the error percentages in a clear manner.

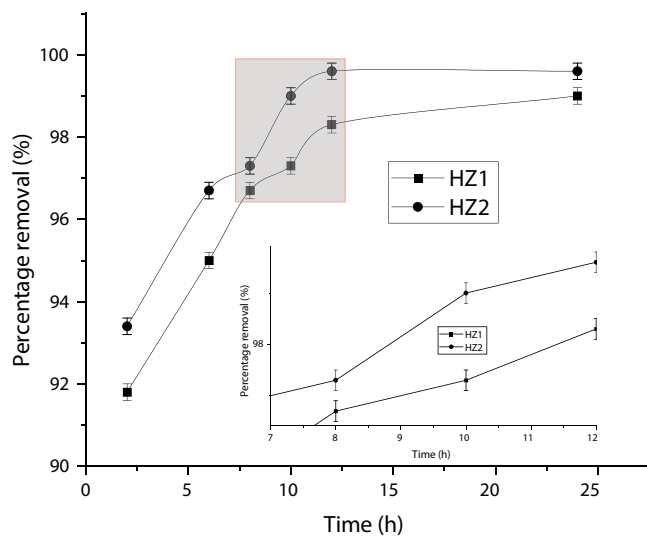


Fig. 7. Percentage removal obtained from batch experiments for Fe^{3+} from W1 using 10 g of HZ. The inset indicates the enlarged region of the main curves to exhibit the error percentages in a clear manner.

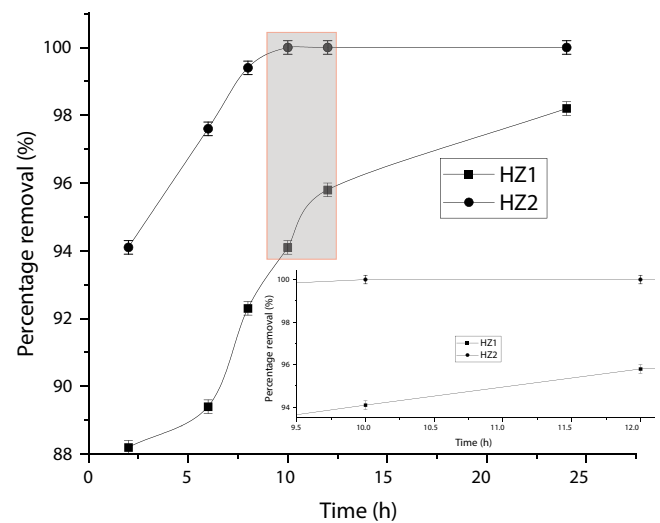


Fig. 9. Percentage removal obtained from batch experiments for Fe^{3+} from W2 using 10 g of HZ. The inset indicates the enlarged region of the main curves to exhibit the error percentages in a clear manner.

4. Conclusions

The use of zeolitic tuff from Al Hala volcanic region located in the southern part of Jordan (HZ1 and HZ2) proves to be an effective ion exchange material. Al Hala zeolitic tuff were able to remove efficiently the iron ions from drinking water. The performed experiments showed that HZ2 (zeolitic tuff powder) has more power in removing iron ions than HZ1. Even though both sizes could achieve drinking water within the standards, HZ2 (for W1 = 3.06 mg/L and 5 g HZ) removal percentage reached to 99.6% at 24 h stirring time, where HZ2 (for W1 = 3.06 mg/L

and 10 g HZ) were able to reach 99.6% at 12 h stirring time and stay there. Increasing the stirring time increases the iron ions removal. Zeolite tuff was able to reach 100% removal percentage at shorter time with lower water sample concentrations. The efficiency of the batch process is influenced by the grain size of HZ particles and the concentration of an iron ion in drinking water.

The pH value is not significantly affected due to the ion exchange process.

From the achieved results, utilization of zeolite can be a promising alternative in removing Fe^{3+} , this is an inexpensive process and an interesting issue for the next works.

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