Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2019.24480

163 (2019) 198–205
September

Efficiency of an iron matrix-based filter in adsorption of arsenic from water

Seiran Shahmohammadi[a], Behzad Shahmoradi[b,*], Afshin Maleki[b], Jae-Kyu Yang[c], Seung-Mok Lee[d,*]

[a]*Department of Environmental Health Engineering, Faculty of Health, Kurdistan University of Medical Sciences, Sanandaj, Iran*
[b]*Environmental Health Research Center, Research Institute for Health Development, Kurdistan University of Medical Sciences, Sanandaj, Iran, emails: bshahmoradi@muk.ac.ir/bshahmorady@gmail.com (B. Shahmoradi)*
[c]*Department of Environmental Engineering, Kwangwoon University, Seoul, Korea*
[d]*Department of Environmental Engineering, Catholic Kwandong University, Ganeung 25601, Korea, email: leesm@cku.ac.kr*

Received 3 January 2019; Accepted 29 May 2019

ABSTRACT

Pollution of water resources, specifically ground water, with heavy metals and metalloids has turned into one of the greatest worldwide concerns. Among several inorganic pollutants, arsenic and pollution of natural water resources with arsenic have gained much attention due to the high toxicity of this metalloid. Therefore, this study was aimed at investigating adsorption of arsenate from water by a filter containing an iron matrix. The removal efficiency of all filter layers and the effect of the operational conditions including pH (6–9), flow rate (1–20 L/h), arsenate concentration (200–1,000 μg/L), turbidity (0–20 NTU) and chloride concentration (0–0.9 mg/L) were evaluated to optimize the process. Finally, the filter was applied for 10 h to adsorb As(V) from a water system contaminated with 510 μg/L As(V). The Pearson correlation coefficient was calculated using SPSS software. Accordingly, turbidity and flow rate variables with the values of –0.545 and –0.517, respectively, had the highest correlation with the adsorption efficiency of arsenic. The observed trends of removal efficiency at different operational conditions suggested 500 μg/L initial As(V) concentration, 2 L/h flow rate, pH 7, 0 NTU turbidity and 0 mg/L calcium hypochlorite concentration as the optimal adsorption conditions. The optimal extent of removal efficiency was found to be 98.7%. According to the findings of this study, the proposed filter has a high efficiency of adsorptive removal of As(V) from water.

Keywords: As(V); Filter; Adsorption; Iron filings; Activated carbon

1. Introduction

A global concern in natural water resources is the high concentration of arsenic (As) [1]. While As exists in several organic and inorganic forms, for example, arsenate As(V), arsenite As(III), dimethyl arsenic acid and monomethyl arsenic acid, it is commonly found as inorganic forms such as As(V) or As(III) in natural environments [2]. This element can diffuse into surface and ground waters through erosion of As-rich soils and stones, chemical and biological degradation, geochemical processes as well as the mine extraction, industrial and agricultural human activities [3]. The extent of As discharge in water resources is so noticeable that its presence in some regions of the world has turned it into an important and challenging issue. These regions include Argentina, Bangladesh, India, Chile, Mexico, Nepal, Taiwan, Turkey and Vietnam [4]. This environmental challenge should not be simply ignored since permanent exposure to As is harmful and leads to the increased risk

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* Corresponding authors.

of cardiovascular diseases [5]. In addition, long-term consumption of As-contaminated water in childhood increases the risk of diabetes for youngsters [2]. Since drinking water is one of the main ways of As uptake by humans, the US Environmental Protection Agency (USEPA) and World Health Organization (WHO) have reported the permissible level of As in water as 10 μg/L [6].

Different technologies have been developed to remove As from water, such as aeration/coagulation and flocculation, reverse osmosis, ion exchange, membrane filters, electrocoagulation and adsorption. While application of all these methods is accompanied with some disadvantages [7,8], the method of adsorption has been recognized as an effective and economic approach due to its simplicity, reliability and safety [9]. One of the most important adsorbents is zero-valent iron (Fe(0)). This adsorbent first attracted attention in the mid of 1990 s, when iron filings and scraps were applied to remove some metals and the byproducts of water chlorination [10]. In general, Fe(0) can act as a reactive agent for filtration of drinking water. The principal advantage of Fe-based filters is that they can be easily fabricated with locally accessible materials such as sands and iron particles used in welding, casting or iron nails [11]. The other advantage of this adsorbent includes continuous formation of new adsorption sites without adding any chemicals and the possibility of reaction/interaction between Fe(0) and the adsorbed pollutant as well as leading to adsorption, precipitation and oxidation of the pollutant [10]. Thus, using iron filters for As removal in rural areas where there is no access to potable and safe water to drink, is an encouraging approach [12]. With respect to the advantages of water treatment by the Fe(0) adsorbent, in this study, an iron matrix-based filter was designed and developed and its efficiency in removal of As from aqueous solutions was investigated at different operational parameters.

2. Methods and materials

This research was a pilot-scale study performed in three steps of iron matrix fabrication, filer construction and efficiency assessment.

2.1. Preparation of the iron matrix

First, to prepare the iron matrix, Fe(0) (iron filings) were obtained from welding wastes recovered from a local welding workshop. Then, the iron filings were washed, immersed in water for 72 h and dried under natural sunlight conditions (T = 20°C ± 5°C). Subsequently, the filings were inserted in acetic acid (filings to acid ratio = 10–20 wt.%) for 24 h, dried and again immersed in the acid. This acid treatment was performed three times. Thereafter, a stream of water with flow rate of 55 L/h was passed through the iron fillings for 4–6 d to produce the required pores of the final product, which was later dried under natural sunlight conditions. The final product formed was iron matrix. These steps helped to enhance adsorption efficiency of the main contributor and the formation of iron hydroxide ($Fe(OH)_2$) [13]. In addition to applying the iron matrix, the activated carbon adsorbent was used to enhance adsorption of As and to eliminate the color and taste resulted from the iron filings.

2.2. Filter fabrication

To fabricate the filter, a cylindrical reactor made of Plexiglas with 25 cm height, 11 cm inner diameter and 12 cm outer diameter was designed and fabricated. The reactor was filled from down to top by 600 g fine-grained sands with 3 mm diameter (h = 4 cm), 200 g activated carbon with 0.2–2 mm diameter (h = 5 cm), 500 g iron matrix with 0.2 mm diameter (h = 5 cm), and 600 g coarse-grained sands with 6 mm diameter (h = 4 cm), respectively. Two water reservoirs with 22 cm dimension were placed at the top and bottom of the reactor to save the polluted and treated water. The fabricated iron matrix-based filter is schematically depicted in Fig. S1.

2.3. Adsorbent characterization

To determine the surface morphology of the utilized activated carbon and iron matrix particles, scanning electron microscopy (SEM; TESCAN VEGA II) was used. The specific surface area and pore distribution of the iron matrix was analyzed by the BET (BELSORP Mini II) method. Moreover, the elemental composition of the iron matrix was determined using X-ray fluorescence (XRF) spectroscopy by a PW 2404 spectrometer.

2.4. Efficiency evaluation

Adsorption efficiency of the filter was evaluated by operating the reactor in a continuous flow mode. The As(V) solutions with 200–1,000 μg/L concentration were prepared by dissolving Na_2HAsO_4 in deionized water. The chemicals were purchased from the Merck company (Germany). To measure the concentration of As, a graphite furnace atomic absorption instrument was used. pH level of the solutions was adjusted using 0.01 N HCl and NaOH and measured by a Wagtech pH meter. Turbidity of the solutions was determined by a Merck turbidity meter. The As removal efficiency was optimized by varying the solution pH, flow rate, initial As(V) concentration, solution turbidity and chloride concentration. After analysis of the synthetic solutions, a real As-contaminated well was treated by the fabricated filter to evaluate its removal efficiency. The Pearson correlation coefficient was calculated using SPSS software (Version 17).

3. Results and discussion

3.1. Filter properties

Fig. 1 shows the SEM images of the utilized activated carbon and iron matrix particles. According to the SEM image of the activated carbon sample (Fig. 1a), its cellular structure has resulted in the formation of a porous texture. In fact, the presence of these pores facilitates diffusion of pollutants toward the internal parts of the raw material, enhances the quality of the adsorption process and enables achievement of a high-quality product [14]. According to the SEM image of the iron matrix sample (Fig. 1b), particles of iron were irregular, dense and spherical shape with a diameter of 4–100 nm. On the other hand, the smaller diameter of the adsorbent particles and the higher surface area to volume

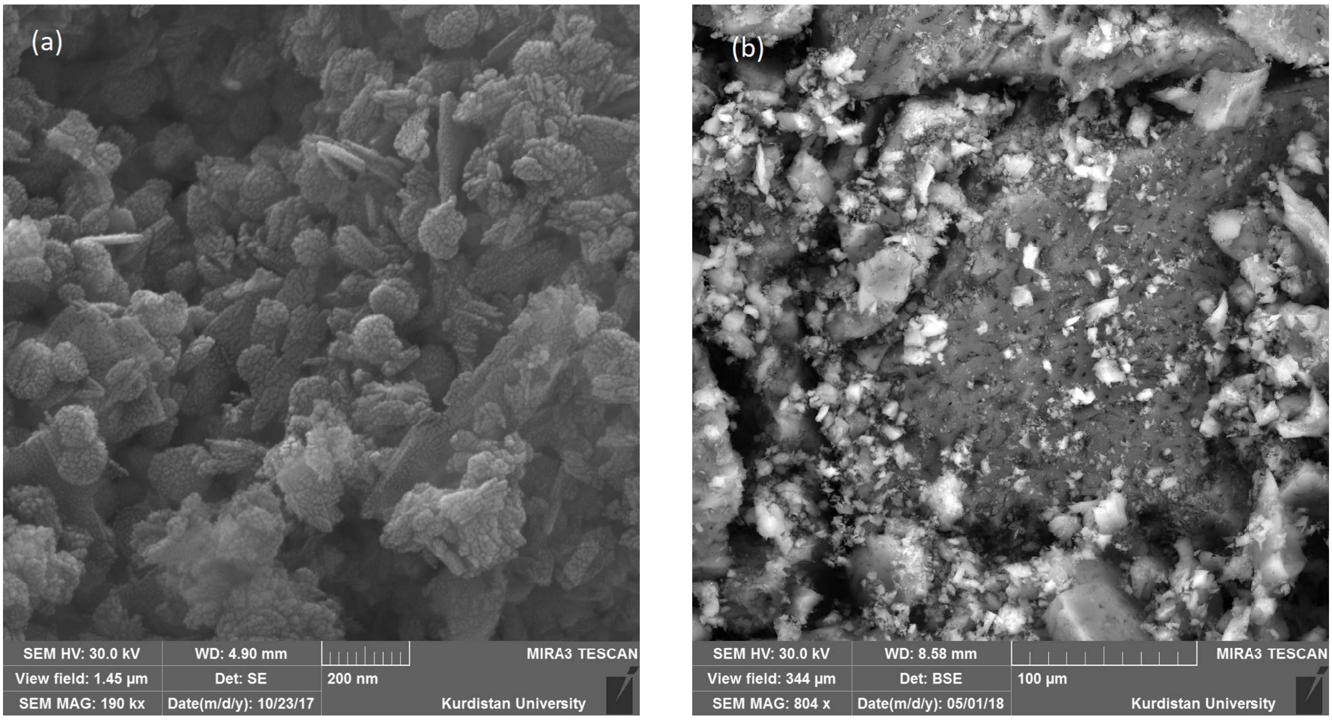

Fig. 1. SEM images of (a) iron matrix and (b) activated carbon.

ratio resulted in enhanced adsorption efficiency. This result was similar to that by Casentini et al. [10].

The BET analysis was carried out for the iron matrix. This analysis showed that the average pore size of the iron matrix equals to 9.272 nm. Moreover, the total volume of the pores is 0.055 cm^3/g, and the effective surface area of the adsorbent is 23.098 m^2/g. The density of adsorbent was measured as 4,545 kg/m^3. The BET results of this study are consistent with those of Kanel et al. [15]. They reported 25 m^2/g specific surface area and 20 nm pore. In addition to the SEM and BET analyses, the iron matrix was analyzed by XRF spectroscopy. The obtained results show that the iron matrix is composed of Fe_2O_3 (98.684%) and trivial amounts of other compounds ($MnO = 0.375\%$, $Al_2O_3 = 0.191\%$, $CaO = 0.084\%$, $SiO_2 = 0.078\%$ and $P_2O_5 = 0.033\%$).

3.2. Effect of the operational parameters

3.2.1. As adsorption efficiency of the filter layers

Fig. 2 displays the removal efficiency for As(V) in each layer of the filter. The iron matrix exhibits the highest removal efficiency for As(V) and the activated carbon layer adsorption shows about 30%. While sands have no impact on As(V) removal. Evaluation of the adsorption efficiency of all filter materials demonstrated that the iron matrix can remove 71%, 68%, 49% and 48% of the As(V) ions from the stream of water containing 500 μg/L As(V) at the 1 l/h flow rate within 1, 2, 3 and 4 h, respectively. Under the same conditions, the activated carbon layer adsorbs 30%, 18%, 10% and 4% of the As(V) ions within 1, 2, 3 and 4 h operation time, respectively. While the coarse-and fine-grained sand particles adsorb no As(V) ions. Therefore, the applied iron matrix has the greatest removal efficiency for As(V) among all adopted filter materials. The high efficiency of this adsorbent can be attributed to the fact that the Fe(0) particles are converted into iron hydroxide particles as an outcome of corrosion in the presence of oxygen. Some of the produced iron hydroxide particles stick to the surface of the matrix while some other iron hydroxides suspense in the solution and act as a strong As(V) adsorbent. Under such conditions, adsorption can be considered as the main process of As(V) removal [16]. The mechanism for removing As(V) by Fe(0) involves adsorption, reduction, surface precipitation and coprecipitation with corrosion products including ferric and ferro hydroxides [17] Meantime, the activated carbon granules do not show a high extent in As(V) adsorption. The reason for this result may be

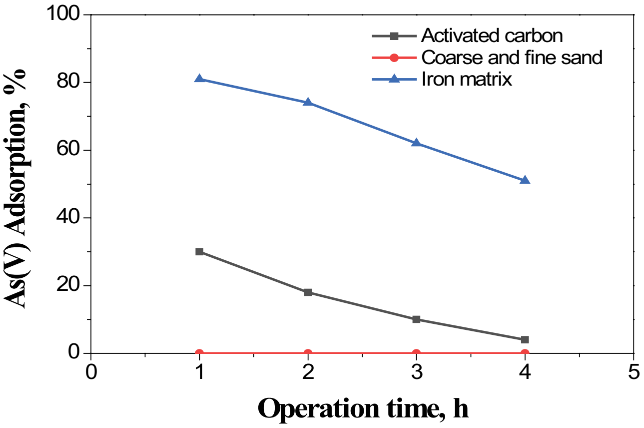

Fig. 2. As(V) adsorption efficiency of the filter layers.

regarded as the adsorption of As(V) onto activated carbon is a physisorption process [18].

3.2.2. Effect of pH

Fig. 3 exhibits the efficiency of the filter for adsorptive removal of 500 µg/L As(V) at various pH levels (6, 7, 8 and 9). With respect to Fig. 3, increasing pH from 6 to 9 declines the adsorption efficiency of the filter. So that, the adsorption process is pH dependent and the maximal As(V) adsorption is observed at pH 6. To be more specific, changing the pH value of the starting solution from 6 to 7, 8 and 9 gives the removal percentages of 99.3%, 98.7%, 96.9% and 94.1%, respectively. These values indicate that the best adsorption performance of the filter can be obtained in acidic and neutral conditions.

Effect of pH on adsorption of pollutants depends on the type of the pollutant and the zero-point charge (zpc) pH of the adsorbent. According to Figs. S2 and S3, the pH_{zpc} values of the iron matrix and activated carbon are 7.5 and 7.3, respectively. At pH levels lower than these pH_{zpc} values, the surface of the two adsorbents would be positively charged due to the increase of H^+ and the reduction of the OH^- ions. Consequently, electrostatic attraction between the positively charged particles and the As(V) anions would promote the adsorption reaction [19]. On the other hand, at pH values above the pH_{zpc}, both As(V) and the adsorbents have negative charges, causing little adsorption due to electrostatic repulsion [20]. Furthermore, at higher pH levels, hydroxyl ions can compete with the As(V) anions for adsorption onto the active sites of the adsorbents. In addition, under alkaline conditions, $Fe(OH)_2$ can be precipitated on the surface of adsorbent, causing occupy of the active adsorption sites and inhibit progress of the adsorption reaction [21]. Similarly, Kanel et al. [15] reported 100% removal of the As(V) by Fe(0) from pH 3 to 7 and Yao et al. [3] observed the highest efficiency of As removal (95.27%) at pH 6. Whereas, Vieira et al. [22] reported the maximum adsorption capacity of As(V) and As(III) as 7.3 and 4.2 mg/L at pH 7 and 20°C, respectively.

3.2.3. Effect of initial As(V) concentration

The adsorption test was performed by changing the initial As(V) concentrations (200–1,000 µg/L) at 2 L/h flow rate and at pH 7. As shown in Fig. 4, as the initial concentration of As(V) increased from 198 to 955 µg/L, the removal efficiency decreased from 99.4% to 95.5%. Eventually, application of 200, 400, 500, 600, 800 and 1,000 µg/L As(V) leads to 99.4%, 98.92%, 97.98%, 96.4% and 95.5% removal efficiency, respectively. It means that the highest adsorption efficiency is obtained at 200 µg/L As(V) concentration. This observation could be attributed to the fact that the numerical ratio of adsorbate molecules to accessible adsorption sites is low and the extent of adsorption is independent on the initial As(V) concentration at lower concentrations. Moreover, at low concentrations, adsorption of the As(V) anions onto the surface may occur through mono-layer adsorption behavior. As an outcome, the rate of As(V) adsorption on the iron matrix and the activated carbon particles increases. However, with the increase of As(V) concentration, the positively charged active surface of the two adsorbents decreases, accessibility of the As(V) anions to active adsorption sites reduces and adsorption decreases due to the reduced rate of mass transport. In other words, since the numerical ratio of pollutant molecules to active adsorption sites increases at higher As(V) concentrations, competition for adsorption onto the adsorption sites increases, resulting in decrease of the adsorption percentages [23,24].

3.2.4. Effect of flowrate

Fig. 5 depicts the effect of different flow rates (1–20 L/h) on adsorption of 500 µg/L As(V) by the filter at pH 7. The highest efficiency was observed at 1 L/h and the removal efficiency was decreased as the flow rate further the increased. Even though the greatest removal efficiency was observed at the flow rate of 1 L/h, 2 L/h was selected as the optimal flow rate since the As(V) concentration in the effluent by applying this flow rate (7 µg/L) was lower than the permissible limit. The decrease of adsorption efficiency with the increase

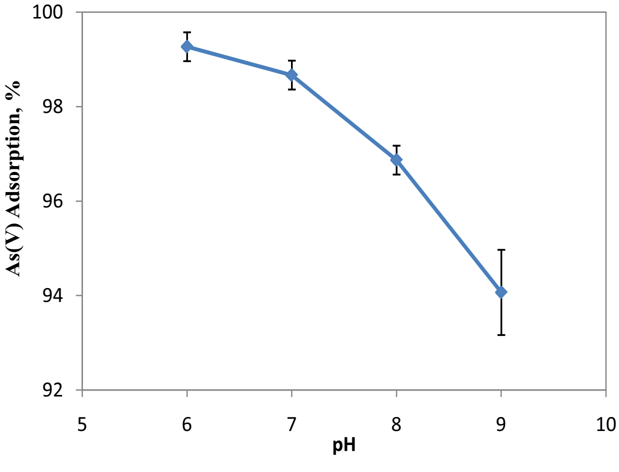

Fig. 3. Effect of solution pH on the adsorption As(V): initial As(V) concentration = 500 µg/L, flow rate = 2 L/h.

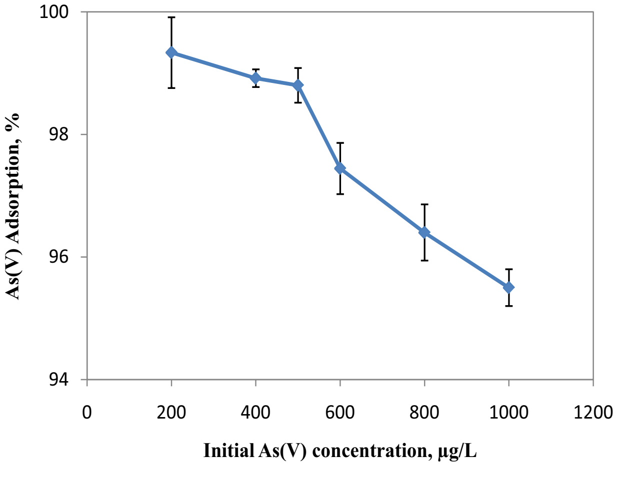

Fig. 4. Effect of initial As(V) concentration on the adsorption efficiency at flow rate 2 L/h and pH = 7.

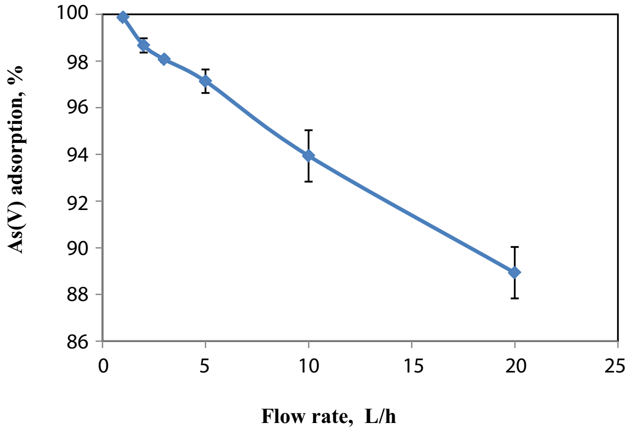

Fig. 5. Effect of flow rate on the adsorption As(V): initial As(V), concentration 500 μg/L, pH = 7.

of flow rate can be associated with contact time. Increasing flow rate increases the velocity and volume of the inlet solution. It causes decrease in contact time between As(V) and the adsorbents. As an outcome, the adsorption efficiency of the filter decreases with the increase of flow rate [25]. In accordance with this finding, Mehta and Chaudhari [1] stated that As(V) removal through filtration was enhanced with decreasing flow rate to 0.75 L/h. Nevertheless, Genç-Fuhrman et al. [26] concluded that the performance of filtration columns can be promoted by decreasing flow rate (prolonging contact time) and decreasing the initial As(V) concentration. Furthermore, Smith et al. [25] observed that the adsorption efficiency for As(V) decreased with the reduction of contact time between unoccupied adsorbent beds and As(V) at higher flow rate. When the flow rate ranged from 40 to 60 L/h, the output increased due to the reduced contact time between water and iron adsorbent; hence, the flowrate of 20–30 L/h was proposed [12]. Calo et al. [27] found that the concentration of As(V) reduced from 92.4 to 35.3 μg/L within 0.5 h contact time, with undetectable concentration within 24.2 h contact time.

3.2.5. *Effect of residual chloride*

To evaluate the effect of residual chloride on adsorption of As(V) by the filter, 0 to 0.9 mg/L calcium hypochlorite was added to a 500 μg/L As(V) solution at pH 7 and 2 L/h flow rate. Fig. 6 shows that the presence of 0.0, 0.3, 0.6 and 0.9 mg/L hypochlorite leads to the 98.7%, 98.93%, 99.3% and 99.7% removal, respectively. It means that chloride ion can increase the efficiency of the filter. The underlying reason is that calcium hypochlorite can react with water to dissociate into the hypochlorite and calcium ions. Calcium can covalently bind to As(V) and produce $CaAsO_4$ precipitate [28]. In confirmation of this statement, Robins et al. [29] noted that the most common technique of As(V) removal from aqueous solution is its precipitation as arsenite sulfide, calcium arsenate and ferric arsenate. In addition, Pokhrel and Viraraghavan [30] observed 86.5%, 95.4% and 65.8% As(V) removal by adding 200 mg/L Ca^{2+}, Fe^{2+} and Mg^{2+}, respectively, to the adsorption system with iron oxide-coated biomasses.

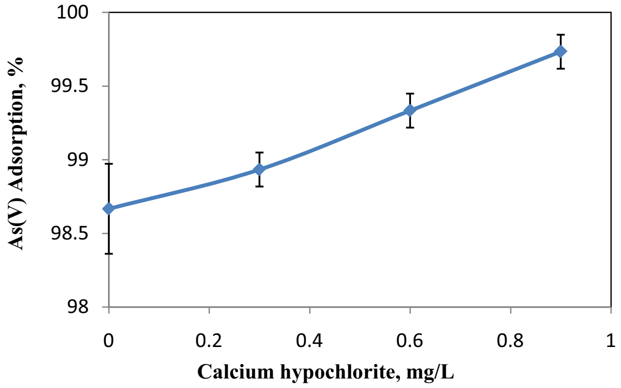

Fig. 6. Effect of calcium hypochlorite on the adsorption of As(V): initial As(V) concentration 500 μg/L, pH = 7.

3.2.6. *Effect of turbidity*

The effect of solution turbidity (0–20 NTU) on adsorption of 500 μg/L As(V) at pH 7 was studied. Fig. 7 shows that the As(V) removal increases as the turbidity decreases. So that, the greatest percentage of As(V) removal (98.6%) is observed at 0 NTU and increasing turbidity to 20 NTU reduces the removal percentage to 83%. A similar observation is reported by Niu et al. [31], who assessed the performance of a very fast and porous filter in treating turbid solutions. They observed the greatest removal percentage (about 77%) at 25 to 50 NTU and the lowest efficiency (70%) at 100 NTU. They also reported increased coagulant consumption with the increase of turbidity. In addition, Zhang et al. [32] reported that the best turbidity reduction (96.75%) was related to 1 NTU turbidity by a filter containing anthracite, microelectrolyzer and sand.

3.2.7. *As(V) adsorption efficiency of the filter with real sample*

In order to investigate the filter efficiency in treating real samples, it was applied to a real sample (V = 20 L and flow rate = 2 L/h) taken from a well located in Gilaku (latitude: 35°27′16.36″N and longitude 47°43′50.91″E), Qorveh, Kurdistan, West of Iran. The characteristics of the examined sample was EC = 875 μs/cm, pH = 6.89, total hardness = 675 mg/L, calcium = 220 mg/L, magnesium = 30.4 mg/L, methyl orange alkalinity = 284 mg/L, sulfate = 45 mg/L, nitrate = 33.3 mg/L, and As = 510 μg/L. According to Fig. 8, the efficiency of the filter in removing As(V) from the sample was almost constant within the first 3 h (99.6%, 99.4% and 99.2%). After that, the efficiency reduced to 98.2% and 98% after 4 and 5 h of the reaction time, respectively. The decreasing removal trend was continued after 5 h, demonstrating a significant decrease in As(V) removal, showing 86.1% removal after 10 h. This value indicates 20% efficiency loss within 10 h. It should be noted that the filter pores were blocked after this period due to the permanent corrosion of the iron matrix, and the water stream was no longer able to pass through the filter. The high efficiency of As(V) removal by the filter can be ascribed to the existence of metallic cations in water and the pH level of the sample. The existence of the metal

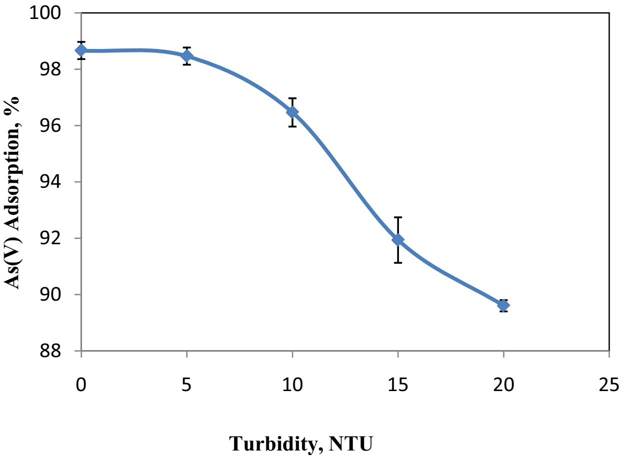

Fig. 7. Effect of turbidity on the adsorption As(V): flow rate 20 L/h, initial As(V), concentration 500 µg/L, pH = 7.

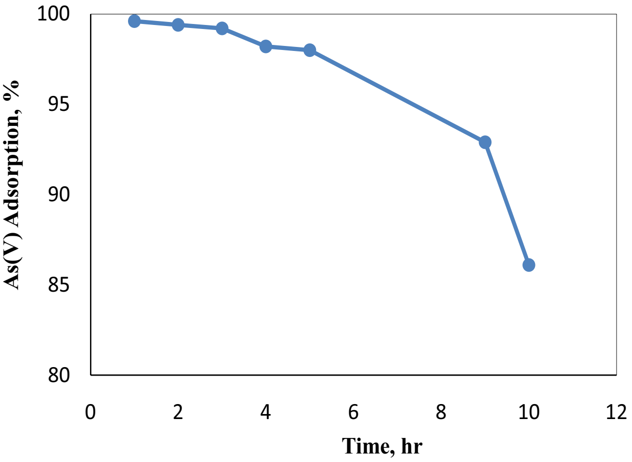

Fig. 8. As adsorption efficiency of the filter with real sample: flow rate 2 L/h, initial As(V) concentration = 510 µg/L and pH = 7.

Table 1
Pearson correlation analysis

Removal As	pH	*Q* (flow rate)	As concentration	$HClO^-$	Turbidity
Pearson correlation	–0.223	–0.517**	–0.158	0.283	–0.545**
Sig. (2-tailed)	0.038	0.000	0.045	0.015	0.000

**Correlation is significant at the 0.01 level (2-tailed).
*Correlation is significant at the 0.05 level (2-tailed).

cations increases the positive charge on the surface of the adsorbents and improves their capacity of anion adsorption. In fact, divalent cations such as Mg^{2+}, Mn^{2+} and Ca^{2+} exhibit the same behavior of iron for As(V) adsorption. Moreover, the presence of calcium bicarbonate enhances As(V) removal by providing an appropriate bed or nucleus for the growth of iron hydroxyl [30,33]. In Table 1, values of high significance level are highlighted using two stars; the turbidity and flow rate have the highest correlation with the efficiency of As(V) removal. Table 1 indicates that the Pearson correlation coefficient for As(V) concentration, turbidity, residual chlorine, flow rate and pH is –0.158, –0.545, 0.283, –0.517 and 0.223, respectively. Since the calculated standard deviation for all variables is less than 0.05, the correlation is significant, that is, the variables considered are effective in As(V) removal efficiency.

4. Conclusion

Among different layers of the fabricated filter, the iron matrix exhibits the highest removal efficiency for As(V) and the activated carbon granules show a lower extent of As(V) adsorption. The removal efficiency of the filter showed an inverse relationship with solution pH, flow rate, initial As(V) concentration and turbidity but showed a direct relationship with the concentration of calcium hypochlorite. The optimal processing conditions were determined in 98.7% As(V) removal. Application of the filter to a well sample containing 510 µg/L As(V) led to 99.6% removal within 1 h. In general, the findings of this study declared that the fabricated filter can provide a high efficiency of As(V) adsorption, in addition to the advantages of low-cost precursors, feasible establishment and operation. The only limitation of the filter observed was the blockage of its pores after 10 h operation. Since replacement of the filter does not require any particular technology, this limitation can be ignored and the filter can be adopted to treat arsenic-contaminated aqueous solutions.

Acknowledgments

This research was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (NRF-2019R1I1A3A01062424).

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Supplementary Information

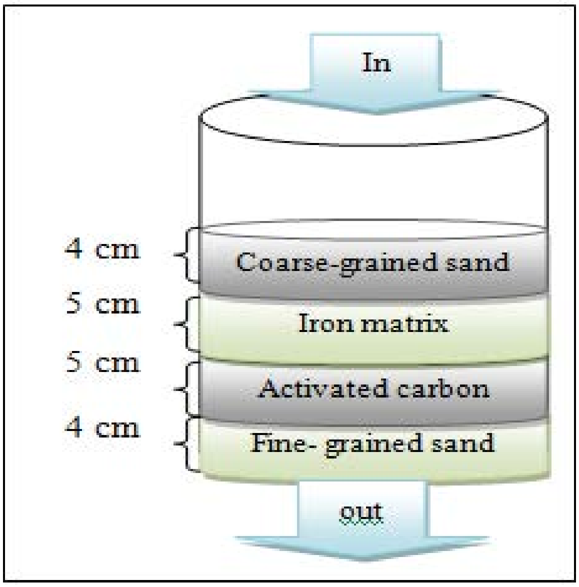

Fig. S1. Schematic of the iron matrix-based filter.

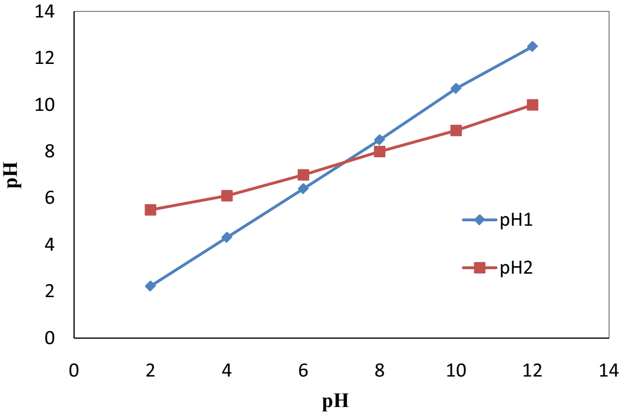

Fig. S2. Characteristic determination of the pH_{zpc} of iron matrix.

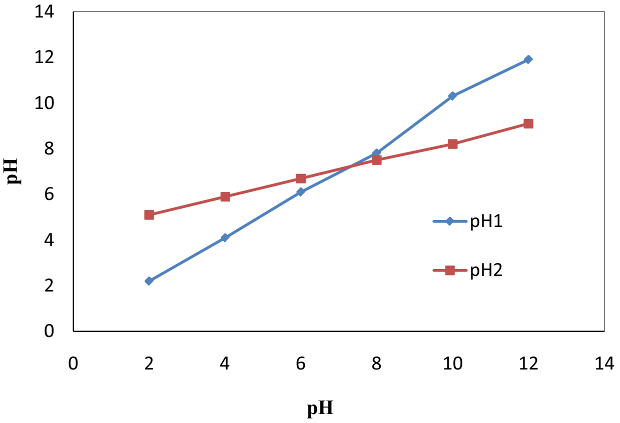

Fig. S3. Characteristic determination of the pH_{zpc} of activated carbon.