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Nano-iron oxide coated on sand as a new sorbent for removal of arsenic from drinking water

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ABSTRACT

This study describes the removal of arsenic species including As(III) and As(V) from drinking water using magnetic iron oxide nanoparticles coated on sand (MIONCS). MIONCS was prepared by the batch method and characterized by a transmission electron microscope. The effects of different factors, pH on the synthesis of the adsorbent, contact time, amount of adsorbent, water sample volume, and adsorption capacity of MIONCS for arsenic removal were investigated. Under the optimum conditions, a 100% arsenic removal was achieved at pH value of 7 from a real drinking water with initial concentration of 87.0 $\mu g\ L^{-1}$ by MIONCS within 25 min. The Langmuir and Freundlich adsorption isotherms were investigated for a range of arsenic initial concentrations of 10–90 $\mu g\ L^{-1}$. Results suggest that the Langmuir isotherm is more adequate than the Freundlich isotherm in simulating the adsorption isotherm of arsenic. The adsorption capacity of the synthesized sorbent for arsenic calculated from Langmuir adsorption isotherms in batch experiments was 0.284 $mg\ g^{-1}$ ($R^2 = 0.999$), and the MIONCS adsorption rate constant is 502.2 $L\ mg^{-1}$. These findings indicate that the adsorption property of MIONCS has a great potential for arsenic removal from drinking water.

Keywords: Arsenic removal; Drinking waters; Magnetic iron oxide nanoparticles

1. Introduction

Arsenic has been known for centuries as a toxic element occurring naturally in the earth's crust and found throughout the environment [1]. High arsenic concentrations in natural waters and food samples are a worldwide problem. Millions of people are at serious risk of arsenic poisoning in many countries around the world, especially in West Bengal, India, and Bangladesh due to their exposure to high arsenic drinking water sources [2–4]. Drinking arsenic-rich water over a long period can result in various adverse health effects including cancers of the skin, bladder,

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kidneys and lungs, diseases of the blood vessels of the legs and feet and possibly also diabetes, high blood pressure, and reproductive disorders [5]. Therefore, it is really necessary to remove arsenic from water to make sure that our environment is safe. The detrimental health effects of arsenic have prompted many authorities, e.g. World Health Organization [6], European Commission [7], and United States Environmental Protection Agency [8], to reduce the maximum limit of arsenic in drinking water from 50 to 10 μg L^{-1}, due to its carcinogenic nature and other dermal effects. Arsenic is generally found as a contaminant in soil and water systems due to various anthropogenic sources, such as mining activity, discharges of industrial wastes, and agricultural application such as pesticides and herbicides, as well as from geochemical reactions [9]. It can occur in the environment in several oxidation states, but in natural waters, it is mostly found in inorganic form as oxyanions of trivalent arsenite or pentavalent arsenate. Organic arsenic forms may be produced by biological activity, mostly in surface waters, but they are rarely quantitatively important [10]. As is well known, the most commonly existing forms of arsenic species in aqueous environments are arsenate (as $H_2AsO_4^-$ and $HAsO_4^{2-}$) in well-oxidized waters and arsenite (as $H_3AsO_3^0$ and $H_2AsO_3^-$) in reduced environments [11]. Arsenite is 25–60 times more toxic than arsenate and more mobile in the environment [12]. The speciations of arsenic are dependent on pH, redox potential, and microbial activity [13]. The predominant As(III) compound is neutral in charge, while the As(V) species are negatively charged in the pH range of 4–10 [14]. Therefore, As(III) is less efficiently removed than As(V) from aqueous solutions by almost all of the arsenic removal technologies and pre-oxidation of As(III) to As(V) is necessary for better removal using some oxidizing chemical agents like chlorine and potassium permanganate. Under oxidizing conditions, $HASO_4^{2-}$ is dominant at higher pH while H_3AsO_4 predominates at extremely acidic conditions. The trivalent arsenic species are thermodynamically stable and dominant under reducing anaerobic conditions, while the pentavalent arsenic species are stable and predominant in the oxygen-rich aerobic conditions [15]. Many groups are working on remediation technologies and determination techniques of total arsenic and/or arsenic species. In parallel, developing cost-effective technologies such as ion exchange [16], coagulation (coprecipitation) [17], reverse osmosis [18], bioremediation [19], and adsorption [20] to remove arsenic from water has drawn great attention in the last 20 years. Adsorption is a common practice for arsenic removal from drinking water due to technological and cost advantages. Most of these technologies utilize the high adsorption and even higher retention of arsenic on iron oxide, iron hydroxides, and oxy (iron) hydroxide. Recently, it has been reported that Fe_3O_4-based materials are very effective in the removal of arsenic (arsenate and arsenite) due to their strong adsorption activities and the properties of being easily separated, collected, and reused by an external magnetic field [21–23]. The removal of adsorbents from solution with the use of magnetic field is more selective and efficient than centrifugation or filtration. In the recent years, Fe_3O_4 nanoparticles have attracted increasing attention in the field of environment protection and remediation because of the properties described above [24–26]. One inexpensive and yet effective method is to coat sand with nano-iron oxide and use this modified sand to remove arsenic [27]. However, the removal capacity of sand material was found to be insignificant for various heavy metal toxic ions [28,29]. Impregnation of sand with manganese or iron enables it to remove heavy metal toxic ions from aqueous solutions [30,31]. In this work, we describe a method for the removal of arsenic from drinking water based on a simple and economic preparation of the magnetic iron oxide nanoparticles coated on sand shell (MIONCS). At the end of the article, we examine the adsorption isotherms and maximum adsorption capacity.

2. Materials and methods

2.1. Reagents and instrumentation

Sand (particle size lower from 0.841 mm) was used as the supporting material for nano-iron oxide. Natural sand was collected from local river at the Ginekan site, Kerman, Iran. The standard solution of arsenic (1,000 μg mL^{-1}) was purchased from Merck (Darmstadt, Germany). In the calibration studies, working solutions were freshly prepared from the stock arsenic solution for each experimental run. All parameters were optimized in a real drinking water containing total arsenic concentration of 87.0 μg L^{-1} that collected from Negar city (Negar, Kerman). Analytical reagent grade chemicals were used for the preparation of all solutions. Similarly, $FeCl_3 \cdot 6H_2O$ and $FeCl_2 \cdot 4H_2O$, nitric acid, hydrochloric acid, and sodium hydroxide were purchased from Merck hydrochloric acid (0.1 mol L^{-1}) and sodium hydroxide (0.1 mol L^{-1}) solutions were prepared for pH adjustment. All solutions were prepared with deionized water. All bottles and glassware were acid washed and rinsed with deionized water before using.

A Varian AA220 model atomic absorption spectrometer (Australia) equipped with graphite furnace

(GTA-110 series), a deuterium-lamp background corrector, arsenic hallow cathode lamp (Varian) and argon (as purge gas) were used for the determination of arsenic. The optimum operating parameters for GFAAS are given in Tables 1 and 2. All pH measurements were made with a metrohm 827 model pH meter (Switzerland). A temperature-controlled shaker (Nüve ST-402, Ankara, Turkey), an ultrasonic bath (Bandelin electronic RK255H, Berlin, Germany) and oven GCA Precision Scientific model 28 (America) were used.

The morphology and size of nano-iron oxide were surveyed on a Hitachi-6,000 transmission electron microscope (TEM, Japan). Zeta potential was carried out on Zetasizer Nano Series (ZS90).

2.2. Preparation of MIONCS

Sand was soaked in 0.2 mol L^{-1} HNO_3 solution overnight and then rinsed 3–4 times with deionized water and dried in an oven at 100˚C for 24 h. Thus, sand with acidic treated with acid was used for coating. For the preparation of MIONCS, solutions of Fe (III) and Fe(II) with proper molar ratio 2:1 were prepared, respectively, by dissolving reagent grade $FeCl_3{\cdot}6H_2O$ and $FeCl_2{\cdot}4H_2O$ in deionized water and poured into a beaker. This beaker placed in a temperature-controlled shaker at 70–80˚C at 30 rpm and 0.1 M NaOH solution was added to it until pH reached 10. The Fe solution was mixed for another 20 min. A black precipitate was obtained confirming the synthesis of Fe_3O_4 [32]. Subsequently, the mixed solution was poured into 100 g sand and then put in a shaker at 30 rpm for 1 h and dried in an oven at 90˚C until only 10% of the water remained. The sample was then stirred at 30 rpm for 20 min to allow the stabilization of the coating process and abruptly dried at 90˚C in a drying oven. The resulting sample was heated at 150˚C for 8 h for complete drying and perhaps better coating stability of nano-iron on the sand surface. The small-sized particles of nano-iron oxide were aggregated on the sand surface. It was then cooled naturally to room temperature, and to remove traces of uncoated nano-iron on the sand, the dried sand was rinsed several times with deionized water, dried at 80˚C and stored in a stoppered polyethylene bottle. This dried MIONCS was used in our experimental.

Table 1
Optimum instrumental conditions for arsenic determination

Wavelength (nm)	193.7
Spectral bandwidth (nm)	0.5
Lamp current (mA)	10
Lamp type	HCl
Measurement mode	Peak height

2.3. Batch experiments of arsenic adsorption

Batch adsorption of arsenic was carried out in a beaker containing 3 g MIONCS adsorbent in the 500 mL of solution. The initial arsenic solution concentration of 87.0 μg L^{-1} arsenic was used, and the pH value was 7. The mixture was shaken for 25 min (equilibrium time) with 30 rpm at 25˚C, and then, an aliquot of supernatant was sampled at regular intervals and filtered through 0.45-μm membrane filter (Millipore) for aqueous arsenic analysis. The concentrations of arsenic in the water samples were determined.

The adsorption capacity (q_e, mg g^{-1}) of MIONCS was calculated by Eq. (1):

$$q_e = \frac{(C_0 - C_e)V}{W} \tag{1}$$

where C_0 is the initial arsenic concentration (mg L^{-1}) and C_e is the equilibrium arsenic concentration (mg L^{-1}), V is the volume of the sample solution (L), and W is the weight of the adsorbent (g).

3. Results and discussion

3.1. Characterization of adsorbents

The TEM sample and zeta potential were prepared by dispersing a small amount of dry powder in ethanol. Then, one drop of the suspension was dropped on 300-mesh copper TEM grids covered with thin amorphous carbon film. In Fig. 1, a TEM picture of the iron oxide nanoparticles is shown. The great majority of the particles have a size between 20 and 30 nm. In Fig. 2, a trimodal size distribution is shown and obtained with maxima at mean sizes of about 23.05, 25.8, and 29.4 nm.

3.2. Effect of pH on arsenic removal

The recovery of the arsenic was determined by applying the general procedure (Section 2.3) by varying the pH of the model solution in the range of 5–12. pH of the model solutions was adjusted to desired values with diluted hydrochloric acid (0.1 M) and/or sodium hydroxide solution (0.1 M). The variation in removal of arsenic with pH is shown in Fig. 3. These

Table 2
Optimum temperature program of GFAAS for determination of arsenic

Steps	Temperature (°C)	Time (s)	Argon flow rate (L min^{-1})
Drying	120	10	3.0
Ashing	1,400	5	3.0
Gas stopping	900	15	3.0
Atomization	2,600	2	0.0
Cleaning	2,650	2	3.0

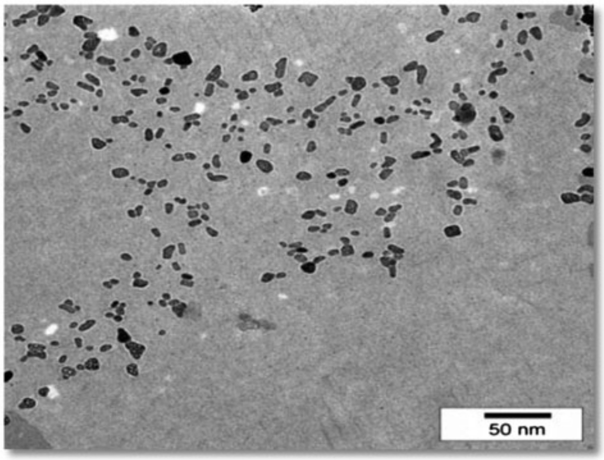

Fig. 1. Transmission electron picture of the Fe_3O_4 nanoparticles.

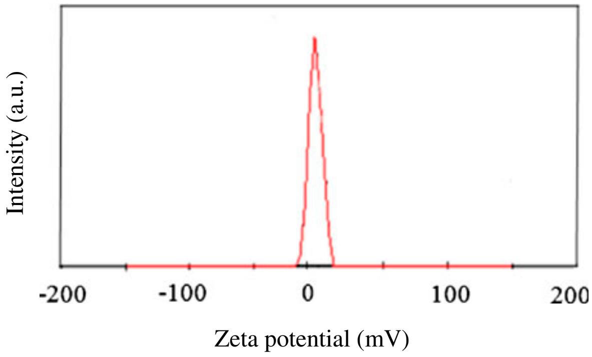

Fig. 2. Zeta potential of Fe_3O_4 nanoparticles.

results reveal that at pH 7, 100% removal was achieved for arsenic. The percent removal decreases to 96.7% at pH 8, 92.5% at pH 9, 87.9% at pH 10, 73.7% at pH 11, and 61.5% at pH 12. This effect can be attributed to the electrostatic repulsion between arsenic and the adsorbent. The results suggest that the MIONCS has a high efficiency for removing arsenic from drinking water, and it is a potential adsorbent for solving environmental problems. pH 7 was selected as an optimum pH for solid-phase extraction of the arsenic for subsequent experiments.

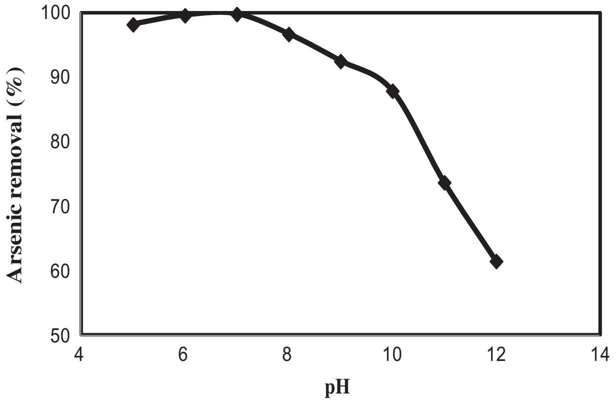

Fig. 3. Effect of pH on removal of arsenic by MIONCS. Initial [As]: 87.0 μg L^{-1}; amount of adsorbent, 3 g; sample volume, 500 mL; equilibration time, 25 min.

3.3. Effect of adsorbent amount

The influence of adsorbent amount on arsenic removal at a fixed initial arsenic concentration of 87.0 μg L^{-1} and neutral pH is shown in Fig. 4. The

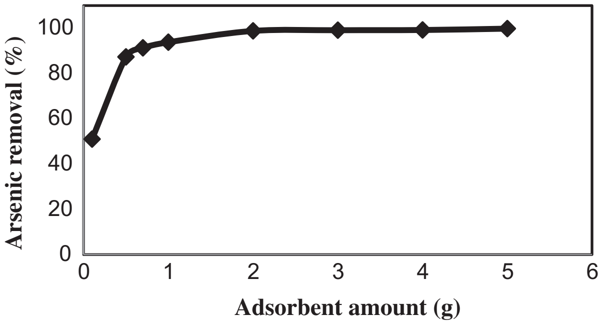

Fig. 4. Effect of adsorbent amount on removal of arsenic by MIONCS. Initial [As]: 87.0 μg L^{-1}; initial pH 7; sample volume, 500 mL; equilibration time, 25 min.

removal of arsenic increased from 51 to ~100% with an increase in adsorbent amount from 0.1 to 5.0 g, respectively, and this is due to the higher active site/As ions ratio. However, it was noticed that after a dosage of 3 g, there was no significant change in the percentage removal of arsenic. The amount of 3 g has been selected as it was sufficient to bring down the arsenic level to below 0.01 mg L^{-1} (10 ppb) as per WHO guidelines.

3.4. Effect of contact time

Fig. 5 presents the adsorption of arsenic on the MIONCS adsorbent surfaces for 87.0 μg L^{-1} of initial arsenic concentration at 25℃, which reveals that the percentage of adsorption of arsenic increases with increasing time. During the initial 15 min, percentage removal of arsenic increases dramatically, which we attributed to the fact that there were plenty of available adsorption sites. With the adsorption process proceeding, the adsorption sites were occupied gradually and percentage removal increased slowly. After 25 min, the percentage removal of arsenic did not exhibited visible change, which we ascribed to the fact that the equilibrium distribution of arsenic in the solution and on the surface of the adsorbent was reached. Thus, the equilibration time of 25 min was applied in the determination of adsorption isotherms (Fig. 5).

3.5. Evaluation of equilibrium adsorption isotherm

Equilibrium adsorption isotherm, which represents the relationship between the equilibrium concentration of adsorbate in solution and temperature, assesses the adsorption capacity of adsorbent and facilitates the exploration of the adsorption mechanism. The Langmuir [33] and Freundlich [34] models were the frequently used isotherm models. The Langmuir theory was valid for monolayer adsorption onto a surface containing a finite number of identical adsorption sites and when there is not important interaction between the adsorbate molecules adsorbed on the adjacent adsorption sites of the adsorbent. The Freundlich isotherm model is an empirical theory available for a heterogeneous surface possessing sites with different sorption energy, which suggested that

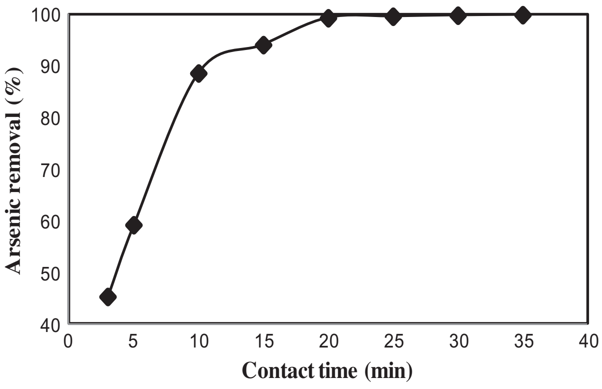

Fig. 5. Effect of contact time on removal of arsenic by MIONCS. Initial [As]: 87.0 μg L^{-1}; initial pH 7; amount of adsorbent, 3 g; sample volume, 500 mL.

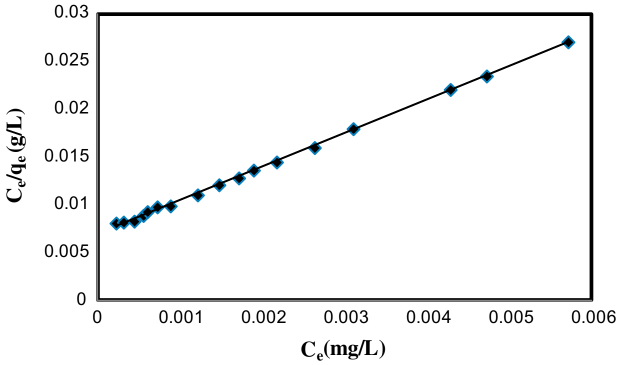

Fig. 6. Adsorption isotherm obtained for arsenic sorption by MIONCS using Langmuir equation (linearized form).

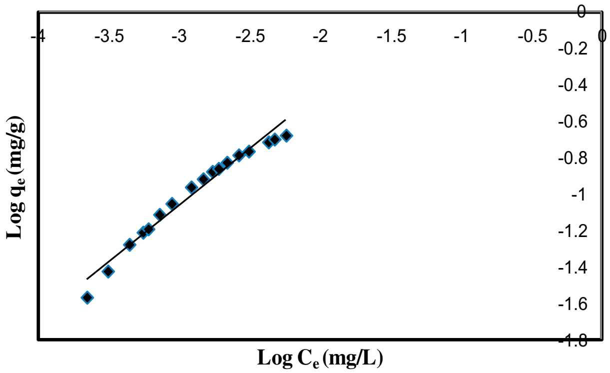

Fig. 7. Adsorption isotherm obtained for arsenic sorption by MIONCS using Freundlich equation.

Table 3
The related parameters of Langmuir and Freundlich isotherms for arsenic adsorbed on MIONCS at pH 7, temperature 25℃, equilibration time 25 min

Langmuir model			Freundlich model		
q_m (mg g^{-1})	b (L mg^{-1})	R^2	K_F (L mg^{-1})	$1/n$	R^2
0.285	502.5	0.999	6.1	0.61	0.968

Table 4
Comparison of magnetic iron oxide nanoparticles coated on sand shell and other adsorbents for arsenic removal

Adsorbent	pH	Conc. range mg L^{-1}	Temp. (℃)	Capacity (mg g^{-1})	References
Iron oxide-coated quartz (IOCQ)	6.0	1.5	30	0.097	[28]
Iron oxide-coated sand	7.6	0.10	22 ± 2	0.043	[36]
Iron oxide-coated sand (IOCS)	–	0.325	22 ± 2	0.018	[37]
Activated alumina	7.6	1.0	25	0.18	[38]
MnO_2 (MOl)	7.9	<1.0	25	0.172	[39]
Iron oxide-uncoated sand	7.0	0.01–0.08	27 ± 2	0.006	[40]
Modified iron oxide-coated sand (MIOCS)	7.2	0.5–3.5	50	0.14	[42]
Magnetic iron oxide nanoparticles coated on sand (MIONCS)	7.0	0.087	25	0.285	This study

the adsorption capacity is related to the equilibrium concentration of the adsorbate. The linearized Langmuir and Freundlich isotherm model can be expressed by Eqs. (2) and (3), respectively:

$$\frac{C_e}{q_e} = \frac{1}{bq_m} + \frac{C_e}{q_m} \quad (2)$$

$$\log q_e = \log K_F + \frac{1}{n}\log C_e \quad (3)$$

where C_e (mg L^{-1}) is the equilibrium concentration of adsorbate in solution, q_e (mg g^{-1}) is the equilibrium adsorption capacity, q_m (mg g^{-1}) is the maximum adsorption capacity of MIONCS for monolayer coverage, b (L mg^{-1}) is a constant related to adsorption free energy, K_F ($mg^{1-(1/n)}$ $L^{1/n}$ g^{-1}) is a constant related to sorption capacity, and n is an empirical parameter related to sorption. Values of q_m and b can be calculated from the slope and intercept of linear plots of C_e/q_e vs. C_e for Eq. (2) and values of K_F and n can be obtained from Eq. (3). The Langmuir isotherm constants obtained from fitting the experimental data to linearized Langmuir isotherm (Fig. 6), and two Freundlich isotherm constants (i.e., K_F and $1/n$) obtained from fitting the experimental data to Freundlich equation isotherms (Fig. 7) are tabulated in Table 3. The sorption coefficient, K_F, was found to be 6.1 (mg g^{-1}) (L $mg^{-1})^{1/n}$, and the adsorption intensity $1/n = 0.61$ Numbers in the brackets are standard deviations. In the Freundlich isotherm model, K_F is a measure of the adsorption capacity (larger K_F indicates a larger overall capacity), whereas the parameter $1/n$ is a measure of the strength of adsorption, representing the concentration of arsenic in a saturated column. The absorbent capacity q_e was determined from Eq. (3) for the equilibrium concentration $C_e = 87.0$ μg L^{-1} and was found to be equal to 0.134 mg g^{-1}. Langmuir model is more suitable in simulating the adsorption isotherm of arsenic onto MIONCS. The correlation coefficients of Langmuir equation (0.999) are higher than that of the Freundlich, which suggests that the adsorption belongs to the monolayer adsorption [35]. The adsorption rate constant of the model is 502.5 L mg^{-1}, and the maximum adsorption capacity is 0.285 mg g^{-1}.

3.6. Comparison with other adsorbents

For comparison of MIONCS and previously reported adsorbents [27,36–42], the Langmuir adsorption capacity was considered. Results of another isotherm study with different adsorbents are also presented in Table 4. An analytical comparison shows that MIONCS is better than many other adsorbents in terms of adsorption capacity. The main advantages of MIONCS are the substantially low cost, availability of the materials, possibility to be prepared by sol–gel method, simple, rapid and effective arsenic removal ability, rapid and simple separation with external magnetic field and economic feasibility. This suggests that the adsorption property of MIONCS gives the material great potential for applications in arsenic removal from drinking water.

4. Conclusion

MIONCS adsorbent was prepared by loading nano-iron oxide onto sand, and its performance for arsenic removal from drinking water was investigated by batch adsorption experiments and characterized by TEM. The results show that arsenic ions are chemisorbed on the surface of MIONCS. The adsorption rate of arsenic is very fast, and equilibrium time is around

25 min. It is also observed that the experimental results of adsorption isotherms fit the Langmuir model well and the maximum adsorption capacity is 0.285 mg g^{-1} at pH 7, which is higher than most of the available arsenic adsorbents. The facile synthetic conditions, efficient and fast adsorption process, as well as simple and convenient magnetic separation process make a potential adsorbent for removal of arsenic from water at neutral pH. MIONCS adsorbent can decrease the arsenic concentration to less than 10.0 μg L^{-1} in drinking water with total arsenic concentration of about 90 μg L^{-1}.

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