Arsenate removal from water by an alumina-modified zeolite recovered from fly ash

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Abstract

A cancrinite-type zeolite was synthesized from Class C fly ash by molten-salt method. The product (ZFA) was used as the adsorbent for the arsenate removal from water. The adsorption equilibriums of arsenate are investigated on various adsorbents. ZFA showed a higher adsorption capacity ($5.1 \, mg \, g^{-1}$) than activated carbon ($4.0 \, mg \, g^{-1}$), silica gel ($0.46 \, mg \, g^{-1}$), zeolite NaY ($1.4 \, mg \, g^{-1}$), and zeolite 5A ($4.1 \, mg \, g^{-1}$). The relatively higher adsorption capacity of ZFA than zeolite NaY and 5A was attributed to the low Si/Al ratio and the mesoporous secondary pore structure of ZFA. However, it was found that the adsorption capacity of zeolites were generally lower than activated alumina ($16.6 \, mg \, g^{-1}$), which is ascribed to the small pores in zeolite frameworks.

The adsorption capacity of ZFA was significantly improved after loaded by alumina via a wet-impregnation method. The modified ZFA (ZFA-Al_{50}) with the optimum alumina loading showed an adsorption capacity of $34.5 \, mg \, g^{-1}$, which was 2.1 times higher than activated alumina. The Toxicity Characteristic Leaching Procedure (TCLP) leachability tests indicated that the spent ZFA and alumina-modified ZFA complied with the EPA regulations for safe disposal.
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1. Introduction

Arsenic is a recognized carcinogen for human and other living organisms. The most common valence states of arsenic in water are arsenate (As(V)) and arsenite (As(III)), while arsenate is more prevalent in surface water [1]. In January of 2001, the U.S. Environmental Protection Agency lowered the maximum concentration level (MCL) of arsenic in water system from 50 to 10 ppb [2]. Consequently, the removal of arsenic from water systems becomes urgent in order to comply with the new legislation.

A number of methods have been studied for arsenic removal from water, such as coagulation–precipitation, adsorption, ultra-filtration, reverse osmosis, and membrane separation [3–5]. Coagulation–precipitation using lime, alum, or ferric salts is the conventional method for arsenic removal in water systems [6]. However, the handling and disposal of produced bulky con-

taminated sludge requires further expenses and safe operations [3]. Moreover, the effectiveness of this method is significantly diminished when treating large amounts of water with low concentrations of arsenic [7]. As a competitive process, the adsorption process overcomes the disadvantages of the conventional coagulation–precipitation method as it only produces minimal amounts of sludge after treatment and it could efficiently treat the water with low concentrations of arsenic [6].

Natural adsorbents such as limestone, hematile, feldspar, and sandy soils have been studied for arsenic removal in water. Natural adsorbents were favorable for their low-cost and abundant sources, but they are generally not quite efficient for arsenic removal. Other synthetic adsorbents, including activated carbon [8], titania [9], activated alumina [10], iron oxides [11], and lanthanum compounds [12–14], were also used. These synthetic adsorbents generally showed much higher adsorption capacities; however, the higher costs of these materials hindered the use of these adsorbents. As a result, developing adsorbents with a high adsorption capacity and relatively low expenses has become a top priority to facilitate the use of adsorption process for arsenate removal.

Nomenclature

C_{eq} concentration of arsenate at equilibrium
        (mmol l^{-1})

q_{eq} amount of cations exchanged at equilibrium
        (mmol g^{-1})

Zeolite has been well recognized as excellent adsorbents, and it has been widely studies as adsorbents for arsenic removal [7,15–17]. In this study, we synthesized a cancrinite-type zeolite (ZFA) from Class C fly ash. This material was used as the adsorbent to remove arsenate from water. The adsorption behavior of arsenate was studied on different adsorbents. The adsorption capacity of ZFA was compared with other adsorbents including activated carbon, silica gel beads, and synthetic zeolites. To further improve the adsorption capacity, we applied a wet-impregnation method and modified the ZFA by loading with alumina. The adsorption behavior of arsenate on the modified ZFA was investigated and compared with the commonly used activated alumina.

2. Experimental

2.1. Preparation of ZFA and modified ZFA

Fly ashes (N.B. Power Co., Canada) were used as received. A weight ratio of 1:1.2 of fly ash (6 g) and NaOH pellets (7.2 g, Reagent Grade, Fisher) were mixed together and ground into fine powder in a mortar. The well-mixed powder was calcined at 600 °C for 2 h with a temperature increase rate of 20 °C min^{-1}. After the calcination, the powder became a loose lump with a color change from brown to dark green. The lump was ground into powder again and added into 80 ml of deionized water. The obtained mixture was heated to 95 °C in an oil bath for 72 h with stirring (400 rpm) and heat refluxing. Then, the mixture was transferred to an autoclave and kept in the oven at 140 °C for 3 days. The solids were recovered from autoclave, washed with deionized water, and dried in oven at 100 °C. The obtained ZFA solids were stored in a vacuum desiccator for future use. The synthesis conditions of ZFA have been optimized based on our previous work [18].

The synthesized ZFA was further modified by loading it with various amounts of alumina via a wet-impregnation method. A calculated amount of aluminum chloride hexahydrate (99%, Sigma–Aldrich) was mixed with 0.5 g of ZFA in 200 ml deionized water. The solution was stirred at 300 rpm for 2 h and then fast dried in furnace by calcining at 400 °C for 4 h with a temperature increase rate of 20 °C min^{-1}. The prepared alumina-modified ZFA samples are designated as ZFA-Al$_i$, where i is determined by the following equation:

$$i = \frac{\text{mass of alumina impregnated on ZFA}}{\text{mass of ZFA}} \qquad (1)$$

In this study, alumina-modified ZFA samples were prepared with four different loadings, i.e., i = 10, 30, 50, and 80.

2.2. Characterization

The Si/Al ratio of ZFA was determined by dissolving ZFA solids in HF/HNO$_3$ (v/v = 1/1, regent grade) and analyzing Si and Al levels by inductively coupled plasma-Atomic Emission Spectrometer (ICP-AES, Spectro Ciros) equipped with Circular Optics system in the 22 CCD detectors (Spectro). The specific surface area of ZFA and modified ZFA was determined by nitrogen adsorption–desorption isotherms measured at the liquid nitrogen temperature (77 K) on a Quantachrome Autosorb-1 Surface Area and Pore Size Analyzer. Samples were degassed at 200 °C for 12 h in vacuum before measurement. The BET specific surface area was determined by fitting the linear portion of the BET plot to BET equation. Pore size distribution was calculated based on the desorption plot of N$_2$ adsorption–desorption isotherm using the Barrett–Joyner–Halenda (BJH) method. The data analysis was performed on the built-in computer program Autosorb 1® provided by the analyzer.

2.3. Adsorption isotherm and kinetics

The adsorption of arsenate on various adsorbents was studied in batch experiments at 25.0 ± 2.0 °C in an air-conditioned room as monitored by a digital wall-mounted thermometer (Cole-Parmer). The adsorption capacity of ZFA were studied and compared with activated carbon (Fisher), silica gel (Sigma–Aldrich), zeolite 5A (Sigma–Aldrich), and zeolite NaY (Zeolyst). The alumina-modified ZFA, i.e., ZFA-Al$_{10}$, ZFA-Al$_{30}$, ZFA-Al$_{50}$, and ZFA-Al$_{80}$, were investigated for their adsorption capacities of arsenate. Activated alumina (Fisher) was also used as a reference adsorbent. Stock solution of 10 mg l^{-1} sodium arsenate pentahydrate (NaH$_2$AsO$_4$·7H$_2$O, 99%, Sigma–Aldrich) was prepared. Varied amounts of adsorbents were added into polyethylene bottles containing 250 ml arsenate stock solution to obtain the adsorbent loading as 0.2, 0.5, 1, 2, and 5 g l^{-1}. For adsorbents with very high or low adsorption capacity, adsorbent loadings as 0.1 and 10 g l^{-1} were also used respectively to get more readable results. The bottles were mounted on a rotary shaker (KS-130, IKA) and shaken for 24 h at 480 rpm. The pH of the solutions was re-adjusted to 6.0 ± 0.2 by measuring the pH of the solution at the set time intervals (t = 2, 4, 10 h) and adding trivial amounts of HNO$_3$ or NaOH solutions by aliquotor (100–1000 μl, Nichipet EX). After shaking for 24 h, the pH of solutions was checked to ensure the final pH was below 8.0. The solution was filtrated though a 0.45-μm nylon syringe filter (Cole-Parmer). The concentration of arsenate in the filtrated solutions was determined by ICP-AES.

ZFA-Al$_{50}$ was selected as the adsorbent for the adsorption kinetics study for its optimized loading of alumina, which showed the highest adsorption capacity among the alumina-modified ZFA samples. Activated alumina (Fisher) was used as a reference adsorbent to compare the adsorption kinetics with ZFA-Al$_{50}$. Aqueous solutions of 100 mg l^{-1} arsenate were prepared by dissolving the sodium arsenate pentahydrate in deionized water. The initial pH was adjusted to 6.0 ± 0.2. Then, the solution containing adsorbents was stirred for 5 h at 400 rpm on a magnetic stirrer. Samples were taken at the set time intervals

and immediately filtered through 0.45-μm nylon syringe filters. The concentration of arsenate in the filtration was determined by ICP-AES.

2.4. Effect of pH on arsenate adsorption

ZFA-Al$_{50}$ was used to investigate the pH effect on the arsenate adsorption. The adsorption experiment was repeated for five stock solutions (10 mg l^{-1}) with different initial pH levels varying from 2.0 to 12.0. The adsorbent loading was the same (1.0 or 0.5 g l^{-1}) for ZFA-Al$_{50}$ and activated alumina. After shaking for 24 h, the pH of sample solution was measured as the equilibrium pH. Concentrations of arsenic and aluminum in the solution were determined with ICP-AES.

2.5. Leachability test

The leachability of the spent ZFA and ZFA-Al$_{50}$ adsorbents were tested by following the Toxicity Characteristic Leaching Procedure (TCLP) which is recommended by EPA [19]. The spent adsorbents were transferred into a polyethylene bottle containing 0.1N acetic acid at pH 5. The bottle was mounted in the rotary shaker and shaking for 18 h. Then, 20 ml sample was collected from the suspension and filtered through a 0.45-μm nylon syringe filter. The final pH of the extracted solution was 5.4.

3. Result and discussion

3.1. Properties of fly ash, ZFA, modified ZFA, and other adsorbents

The composition of fly ash was determined as: SiO$_2$: 31.6%; Al$_2$O$_3$: 27.8%; Na$_2$O: 27.7%; CaO: 6.4%; Fe$_2$O$_3$: 2.1%; P$_2$O$_5$: 1.5%; MgO: 1.4%; SO$_3$: 0.9%; other: 0.7% [18]. It is categorized as Class C fly ash in terms of the specification of ASTM C618-99. The crystallographic structure of ZFA has been shown that the dominant crystal phase of ZFA is carbonate cancrinite Na$_6$Ca$_2$Al$_6$Si$_6$O$_{24}$(CO$_3$)$_2$·2H$_2$O [18].

Fig. 1 shows the pore size distributions of ZFA and ZFA-Al$_i$, which are determined from their nitrogen adsorption–desorption isotherms at the temperature of liquid nitrogen (77 K). It is shown that the pore size of ZFA centered at 20–500 Å (1 Å = 0.1 nm). The mesoporous secondary pore structure may be favourable for the adsorption of arsenate anions because arsenate anions are below 20 Å [20]. The BET specific surface area of ZFA is 265.81 m^2 g^{-1}, which is larger than most natural zeolites and also among the highest specific surface areas in previous studies on the conversion of fly ash to zeolites [21–23]. The rel-
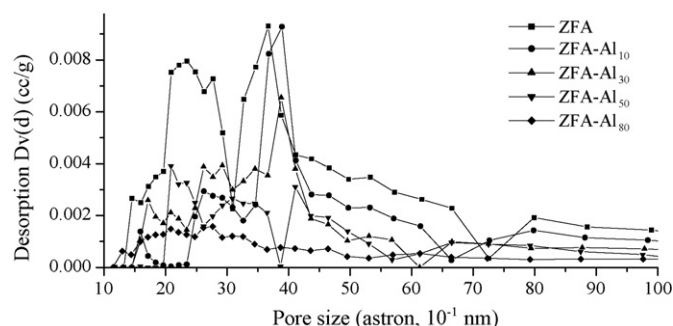

Fig. 1. The pore size distribution of: (a) ZFA; (b) ZFA-Al$_{10}$; (c) ZFA-Al$_{30}$; (d) ZFA-Al$_{50}$; (e) ZFA-Al$_{80}$.

atively high BET surface area was considered to be favourable the adsorption of arsenate on ZFA. As seen from the pore size distributions, the loading of alumina did not remarkably change the mesoporous secondary pore structure of ZFA though mesoporous pores were gradually filled by alumina crystals as the loading amount increased. In ZFA-Al$_{10}$, the pores ranged from 20 to 30 nm in ZFA were significantly diminished after loading with alumina. These pores, however, were well preserved in ZFA-Al$_{30}$ and ZFA-Al$_{50}$. In stead, a continuous decrease was observed for pores with sized ranged from 30 to 40 nm as the alumina loading increased from ZFA-Al$_{10}$ to ZFA-Al$_{30}$ and ZFA-Al$_{50}$. From these observations, it is implied that the impregnated alumina formed crystals with the size between 20 and 30 nm at low alumina loading. These alumina crystals filled in the cavities (secondary pores) in the ZFA structure. With an increasing loading of alumina, larger sizes of crystals (30–40 nm) were formed during the impregnation and calcination processes and became the dominant size of alumina crystals. Smaller pores of 20–30 nm were preserved, which still provide easy accesses for the approaching arsenate anions in the solution. The volume of pores of 20–100 nm in ZFA-Al$_{80}$ is the lowest; however, the high loading of alumina in ZFA was not necessarily favourable for the adsorption as the inner surface area may not be able to perform as active adsorption sites for arsenate anions due to the blockage of pores.

Meanwhile, decreasing BET specific surface areas are observed as ZFA was impregnated with increasing amounts of alumina: ZFA-Al$_{10}$ (111.6 m^2 g^{-1}) > ZFA-Al$_{30}$ (98.1 m^2 g^{-1}) > ZFA-Al$_{50}$ (65.7 m^2 g^{-1}) > ZFA-Al$_{80}$ (61.1 m^2 g^{-1}). The magnitude order of BET surface area also suggests that the inner cavities of ZFA were gradually filled by alumina crystals with an increasing amount of alumina loading.

The data of the properties of zeolite 5A, NaY, and activated alumina are from the suppliers. Properties such as the

Table 1
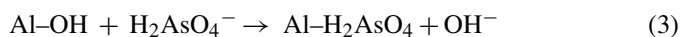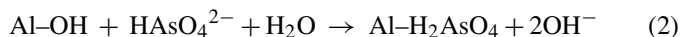Properties of zeolite 5A, zeolite NaY, and activated alumina

	Si/Al (mol/mol)	Cation	Surface area (m^2 g^{-1})	Pore size (nm)	Supplier
Zeolite 5A	~1.0	Calcium	n.a.	0.5	Linde
NaY	2.55	Sodium	900	0.74	Zeolyst
Activated alumina	n.a.[a]	n.a.	150	11	Sasol

[a] Al$_2$O$_3$% is 72 wt.%, Na$_2$O% is 0.002%, and the other part is moisture. Data is from the supplier. The adsorption isotherm data has been adjusted as 100% Al$_2$O$_3$.

Si/Al ratio, specific surface area, and pore size are shown in Table 1.

3.2. Adsorption mechanism

The prevalent forms of arsenate ion at pH 6.0–8.0 are $HAsO_4^{2-}$ and $H_2AsO_4^-$ [17]. The adsorption of arsenate on zeolites and alumina can be described by the ligand exchange reactions [7,17]:

$$Al–OH + HAsO_4^{2-} + H_2O \rightarrow Al–H_2AsO_4 + 2OH^- \quad (2)$$

$$Al–OH + H_2AsO_4^- \rightarrow Al–H_2AsO_4 + OH^- \quad (3)$$

The Al–OH shown in Eqs. (2) and (3) is the terminal OH group present on zeolites and activated alumina. This mechanism was well supported by the observations that the pH of solutions significantly increased during the entire adsorption process. During the experiment, it was necessary to adjust the pH to 6.0 ± 0.2 at the set time intervals to control the pH under 8.0 to avoid potential structure collapse of ZFA (cancrinite). It is also concluded from Eqs. (2) and (3) that the aluminol groups present on ZFA and activated alumina are reactive sites for arsenate adsorption.

3.3. Adsorption results

Fig. 2 shows the adsorption results of arsenate on ZFA and other adsorbents including activated carbon, silica gel, synthetic zeolites, and alumina-modified ZFA. q_{eq} is the adsorbed sorbate (mg g^{-1}) at equilibrium, while C_{eq} is the concentration (mg l^{-1}) of sorbate at equilibrium. At the investigated condition, it is shown that activated carbon shows an adsorption capacity of 4.0 mg g^{-1}, while the cancrinite-type ZFA (before loading with alumina) shows a slightly higher adsorption capacity (5.1 mg g^{-1}). Since terminal Al–OH groups were the reactive sites, the adsorption capacity of ZFA is mainly determined by the amount of terminal Al–OH groups accessible for the approaching arsenate anions. Silica gel beads are also used in this study

to test the sole effect of surface silanol groups on the adsorption of arsenate anions. The silica gel beads used in this study has been characterized in our previous study which indicated that it had sufficient silanol groups on the surface [24]. It is shown in Fig. 2 that the amount of adsorbed arsenate on silica gel is very limited. This result agrees well with the previous reported results that the adsorption capacity of surface silanol groups is substantially low [12,25]. It also confirms that it is the aluminol groups rather than silanol groups that act as active adsorption sites in the adsorption process. The low adsorption capacity of sinanol can also be implied from the pH observations that during the adsorption process no significant pH increase was observed when silica gel beads were used as adsorbents.

A comparison between ZFA and synthetic zeolites shows that ZFA had a higher adsorption capacity than zeolite NaY and 5A. It can be seen from Table 1 that the zeolite NaY has a much higher specific areas than ZFA, which should enable a favorable diffusion of arsenate anions into the inner structure of ZFA. However, the high Si/Al ratio leads to a less amount of available Al–OH groups in NaY. With a low Si/Al ratio ($\sim$1.0), ZFA exhibits a much higher adsorption capacity for arsenate anions. Shevade and Ford [17] also reported that the low Si/Al ratio was a governing factor for the performance of zeolite for arsenic adsorption.

In addition, despite that zeolites have been extensively studied for using as the adsorption, a direct comparison on the adsorption capacity of commercial alumina and zeolite is not available. One of the important notes from this study is that, by comparing the performance of synthetic zeolites and alumina, it is found that zeolites showed significantly lower adsorption capacities than activated alumina. The adsorption capacity of zeolite is greatly limited since zeolites are microporous materials. Synthetic zeolites, such as zeolite 5A and zeolite NaY, had quite large surface areas but they generally had a pore size of 5 and 7.4 Å, which is very close to $HAsO_4^{2-}$ and AsO_4^{3-} but smaller than $H_2AsO_4^-$ [20]. Another reason that should be taken into account is the composition of zeolites. As we know, zeolites are generally crystalline aluminum-silicates. Since silanol groups have substantially low adsorption capacity for arsenate anions, the presence of silicate reduces the surface aluminol density on the surfaces which eventually leads to a reduced adsorption capacity. This conclusion can be proved by the relationship between the Si/Al ratio and the adsorption capacity of ZFA, zeolite 5A, and zeolite NaY. This finding may be very important since synthetic zeolites are generally more costly than activated alumina. Although zeolites may have fast kinetics due to their large surface areas, activated alumina is preferable for practical use from an engineering point of view when the adsorption capacity is more important.

Fig. 2 shows that the adsorption capacity of ZFA was remarkably increased by two to three times, depending on the loading amount. An increasing adsorption capacity was obtained as the loading amount of alumina increased from 10% to 50%. The higher adsorption capacity was attributed to the alumina crystals dispersed in the mesoporous secondary pores in the inner structure of ZFA. These impregnated alumina crystals can provide more adsorption sites (Al–OH) to the sorbate anions,
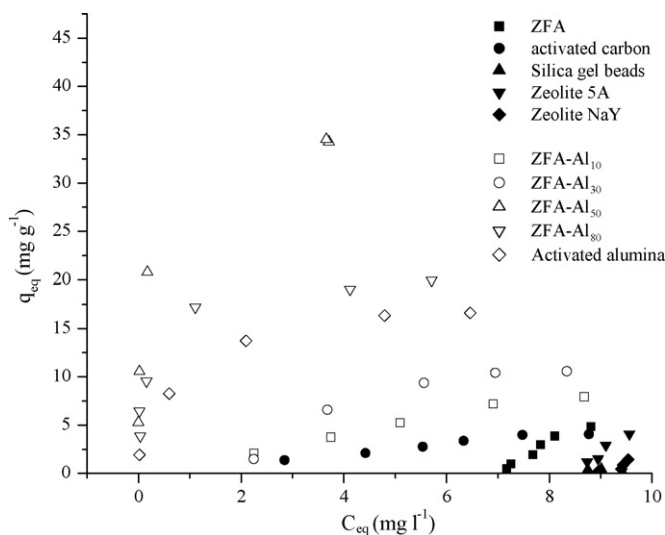

Fig. 2. Adsorption results of arsenate on different adsorbents.

Table 2
Adsorption capacities of various adsorbents

Adsorbent	Adsorption capacity ($mg\,g^{-1}$)
ZFA	5.1
Activated carbon	4.0
Silica gel beads	0.46 (negligible)
Zeolite 5A	4.1
Zeolite NaY	1.4
ZFA-Al$_{10}$	7.9
ZFA-Al$_{30}$	10.6
ZFA-Al$_{50}$	34.5
ZFA-Al$_{80}$	19.9
Activated alumina	16.6

while the inner structure of modified ZFA was still easy to be accessed by arsenate anions with a proper loading amount of alumina. The highest adsorption capacity ($34.5\,mg\,g^{-1}$) was obtained with ZFA-Al$_{50}$, which is 2.1 times higher than that of the activated alumina ($16.6\,mg\,g^{-1}$). The adsorption capacity decreased in ZFA-Al$_{80}$ ($19.9\,mg\,g^{-1}$), as shown in Table 2. The abrupt decrease of the adsorption capacity of ZFA-Al$_{80}$ is due to that the mesopores in ZFA were filled with alumina crystals and they might hinder the approaching sorbate anions although more adsorption sites (Al–OH) were provided. Moreover, compared with activated alumina which had the macropores (11 nm) and a specific area of $150\,m^2\,g^{-1}$, the higher adsorption capacity observed in ZFA-Al$_{50}$ could provide more terminal Al–OH groups are available for the sorption of arsenate anions due to the well-dispersion of the impregnated alumina crystals in the ZFA structure.

3.4. Effect of pH on arsenate adsorption

Fig. 3 shows the adsorption results on ZFA-Al$_{50}$ at different initial pH levels. The results indicate that more arsenate anions were adsorbed under pH 4.9–10.1. When the initial pH was 4.9 and 7.0, more than 90% fraction of the initial arsenate was
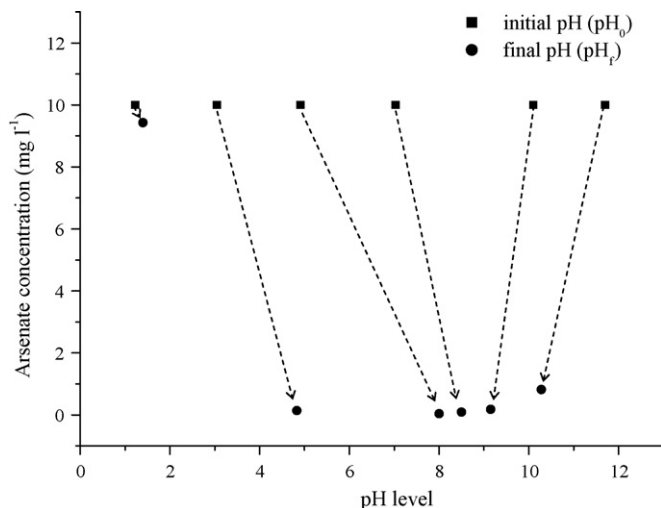
removed. A steep decrease in sorption occurred when the initial pH increased from 10.1 to 11.7. Similar effects of pH on As(V) adsorption on TiO$_2$ were reported by Dutta et al. [26]. It is suggested that the adsorption of strong acid anions by metal oxides and hydroxides typically decrease with increasing pH [27]. Meanwhile, the surface potential of metal oxides becomes more negative when pH is greater than the pH$_{pzc}$, which is not favourable for the adsorption of arsenate anions. Dutta et al. [26] reported that the adsorption rate of As(V) on TiO$_2$ decreased with increasing pH and attributed it to increased electrostatic repulsion between the negative charged surface and charge of anionic arsenate species. The adsorption of arsenate anions follows the surface ligand reaction shown in Eqs. (2) and (3), from which we can also imply that a higher proton concentration can facilitate the adsorption process. The decrease at initial pH 1.2 is due to the dissolution of alumina and the structural collapse of cancrinite.

The alumina species was also detected in the solution after shaking for 24 h. Significant aluminium levels were found in the solutions at extreme initial pH levels, i.e., $249.8\,mg\,l^{-1}$ (initial pH 1.2), $124.2\,mg\,l^{-1}$ (initial pH 11.7). Low aluminium species were also detected in the solutions with milder pH levels: 14.85 (initial pH 3.1), 1.93 (initial pH 4.9), 3.68 (initial pH 7.0), 11.85 (initial pH 10.1). The aluminium leachment at initial pH 10.1 may be due to the structure collapse of ZFA. At pH 3.1, however, the aluminium leachment is mainly due to the dissolution of impregnated alumina at the acidic condition.

3.5. Adsorption kinetics and leachability test

It has been well established that the adsorption of arsenate is a diffusion-controlled process. As shown in Fig. 4, the kinetics results are well fitted with second-order kinetics model. The adsorption rate was determined by the diffusion rate of arsenate anions into ZFA. Terminal Al–OH groups at the outer surface of ZFA could readily react with the arsenate anions in the solution, while inner Al–OH sites can only be reactive once they can encounter the diffused arsenate anions. For the modified-ZFA, the mesoporous structure of ZFA enables a fast-
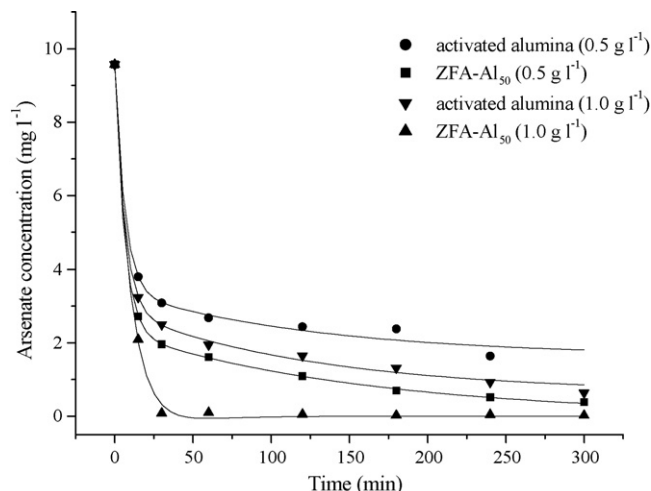

Fig. 3. pH effects on the adsorption of arsenate on ZFA-Al$_{50}$.

Fig. 4. Adsorption kinetics of arsenate on ZFA-Al$_{50}$ and activated alumina.

diffusion of arsenate anions and thus fast adsorption kinetics was obtained.

It was found that the arsenate concentrations in the TCLP extraction solutions were 0.05 and 0.13 mg l^{-1} for ZFA and ZFA-Al$_{50}$, respectively, which are lower than the EPA regulation (5.0 mg l^{-1}). According to the TCLP procedures, the spent ZFA and ZFA-Al$_{50}$ were safe to dispose.

4. Conclusions

A cancrinite-type zeolite (ZFA) was synthesized from fly ash and used as an adsorbent to remove arsenate from water. ZFA showed a higher adsorption capacity than active carbon, silica gel, and synthetic zeolites including zeolite NaY and zeolite 5A. The higher adsorption of ZFA was attributed to its low Si/Al ratio of ZFA, which provided sufficient terminal Al–OH groups as reactive sites for the adsorption of arsenate anions. It was also found that the zeolite-type material had a generally lower adsorption capacity for arsenate than the commonly used activated alumina.

The adsorption capacity of ZFA was significantly improved by loading with alumina via a wet-impregnation method. The ZFA with an optimum loading of alumina, ZFA-Al$_{50}$, showed a 2.1 times higher adsorption capacity than activated alumina. The pH effect investigation indicates that a pH range of 4.9–7.0 could optimize the performance for ZFA-Al$_{50}$. The adsorption kinetics study shows that the adsorption for 10 mg l^{-1} arsenate solution was completed within 1 h at the ambient condition using 1.0 g l^{-1} ZFA. Leachability tests showed that the spent ZFA and ZFA-Al$_{50}$ were safe to dispose.

Acknowledgements

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