Batch study of arsenate (V) adsorption using Akadama mud: Effect of water mineralization

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Abstract

Akadama mud, consisting mainly of different forms of iron and aluminum oxide minerals, was used for arsenate (V) adsorption from aqueous solutions. The adsorption process fitted the first-order kinetic equation and the Langmuir monolayer model well. The adsorption capacity, estimated by the Langmuir isotherm model, was 5.30mg/g at $20 \pm 0.5°C$. The effects of the solution properties (initial concentration of As (V), pH, temperature, and mineralization degree) on As (V) removal were investigated. Various mineralization degrees in underground water were simulated by adjusting the ionic strength of the solution or adding coexisting ions to the contaminated solution. It was found that mineralization of the water significantly influenced the arsenic adsorption. The existence of multivalent metallic cations significantly enhanced the As (V) adsorption ability, whereas competing anions such as fluoride and phosphate greatly decreased the As (V) adsorption. This result suggests that Akadama mud is more suitable for arsenic adsorption in low-level phosphate and fluoride solutions. The loaded Akadama mud could be desorbed at polar pH conditions, especially in acidic conditions, and more than 65% As (V)

1 sorption has been achieved at pH 1.

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1. Introduction

10 In recent years, one of the top 20 most hazardous substances [1], inorganic

11 arsenic such as As (V) and As (III), has become one of the most serious

12 water-pollution problems in many parts of the world. Natural arsenic pollution of

13 drinking water supplies has been reported from over 70 countries, and about 150

14 million people are threatened by this serious health hazard [2]. Among them, more

15 than 70% live in Asia, especially in South and Southeast Asia, encompassing

16 Bangladesh, China, and India [3, 4]. Many research studies have indicated that high

17 arsenic concentrations in drinking water might cause many chronic and cancer-related

18 public health problems, such as cancer and skin diseases [5-9]. In 2002, the US

19 Environmental Protection Agency (USEPA) adopted a new maximum limit for arsenic

20 in drinking water of 10μg /L, replacing the old limit of 50μg /L [10]. However, the old

21 standard is still in use in China, India, and some other developing countries where a

22 high risk of arsenic poisoning exists [2].

1   It has been reported that there are several methods available for arsenic removal

2   from aqueous medium, including adsorption, ion exchange, lime softening, reverse

3   osmosis, coagulation, and precipitation [11 -13]. Considering its advantages and

4   limitations, adsorption has been recognized as a favorable technology with the merits

5   of convenient processing, high removal efficiency, potential regeneration, and

6   freedom from sludge. Many earlier studies focused on synthetic adsorbents such as

7   hydrous ferric oxide, activated alumina, activated bauxite, and biomass waste-derived

8   activated carbon [14-17]. Although these materials are regarded as effective

9   adsorbents due to their high surface area and positive charge, there are several

10   problems (high reagent cost, energy requirements, and second contamination caused

11   by residual chemicals) associated with their use [18]. A more suitable adsorbent

12   should be selected by evaluating the following essential factors: low cost, high

13   efficiency, and easy access [19, 20]. Therefore, Akadama mud, which is common and

14   inexpensive in Japan, was used in this study. Akadama mud is a deposit of volcanic

15   ash, a quality soil medium, which is widely utilized as a cultivating mud for plants

16   due to its osmotic properties for both dissolved oxygen (DO) and water. Bang et al.

17   [21] reported that the presence of DO in aqueous medium increased the rate of

18   iron-oxide corrosion and arsenic removal. Therefore, the high DO content of

19   Akadama mud may be helpful for efficiently removing arsenic.

20   Besides some commonly researched factors, such as the adsorbent conditions

21   (dosage and particle size) and solution properties (initial concentration, pH, and

22   temperature), water mineralization also greatly influences arsenic adsorption. In

general, phosphate impedes arsenate adsorption due to competitive effects [22-31].

However, the effects of some other anions, like nitrate and sulfate, are not consistent

across different studies [22-25]. Guo et al. [22] reported that the presence of nitrate

(10mg/L N) does not significantly affect arsenate removal with siderite, while Sun et

al. [23] demonstrated that the presence of high-level nitrate anions (100mg/L N) can

accelerate arsenate removal on zero-valent iron. Zhang et al. [24] determined that the

presence of sulfate (above 10mg/L) is favorable for As(V) sorption in natural iron

ores. In contrast, Partey et al. [25] reported that arsenic adsorption using laterite iron

concretions exhibits no distinct shifts with the addition of sulfate (10mg/L S). These

conflicting reports imply that arsenic removal differs significantly with various

adsorbents and mineralized water. Therefore, given the fact that the degree of water

mineralization varies in most contaminated aqueous systems, it is important to

investigate the effects of water mineralization on arsenic removal by a particular

absorbent.

The present study sought to investigate Akadama mud as an alternate arsenic

adsorbent. The effects of the initial arsenic concentration, pH, temperature, and degree

of mineralization (ion competition and ionic strength) on the arsenic removal

efficiency were researched. Kinetic and equilibrium studies concerning arsenic

removal by Akadama mud were also discussed.

2. Materials and methods

2.1. Materials

Akadama mud, provided by the Makino Store, Kiyosu, Japan, was crushed and

sieved to obtain fractions of particles smaller than 150 mesh. The sample was then

washed with deionized water and dried at 105°C for 24h. The dry sample was sieved

again through a 150-mesh screen for future research.

As (V) stock solutions (1000mg/L) were prepared by dissolving sodium

hydrogen arsenate ($Na_2HAsO_4 \cdot 7H_2O$, Wako Pure Chemical Industries) in deionized

water (DI) with a pH of 6.9. Coexisting ions were provided by analytical-grade

chemicals (KCl, $CaCl_2$, $FeCl_3$, NaCl, NaF, $NaNO_3$, Na_2SO_4, and Na_3PO_4; Wako Pure

Chemical Industries), and the solution ionic strength was adjusted to 0.001, 0.01, and

0.1M with NaCl.

2.2. Methods

2.2.1. Characterization methods

The prepared Akadama mud was analyzed in order to determine its physical,

mineralogical, and chemical properties. The specific surface area and pore size

distributions were determined by a gravimetric nitrogen Brunauer–Emmett–Teller

(BET) specific surface analysis device (Coulter SA3100, US). Morphological features

of the Akadama mud before and after As (V) adsorption were analyzed by a scanning

electron microscope (SEM, JSM-6700F, JEOL, Japan). Chemical analysis was

conducted using SEM coupled with an Energy Dispersive X-ray (EDX) spectroscopic

detector. Akadama mud mineralogical phase characterization was carried out using

quantitative X-ray diffraction (XRD) (RINT2200, Rigaku, Japan). The As (V)

concentrations were measured by an inductively coupled plasma atomic emission

spectrometer (ICP-AES, Model ICPS-7500, Shimadzu, Japan). The detection limit of

this method varies, ranging from 0.5 to 1.0μg/L for arsenic.

2.2.2. Batch experiments

Batch equilibrium experiments were carried out in a water thermostat (20±0.5°C) without any agitation. 300 ml of synthetic As(V) solution (10mg/L) was loaded into a conical flask, with an adsorbent dosage of 10g/L. Samples were taken at certain intervals and filtered through a 0.45μm membrane filter to measure the residual As(V) concentration by ICP-AES. Isotherm experiments were conducted at 20, 35, 50, and 65°C with an agitation speed of 200rpm. 50 ml polyethylene centrifuge tubes filled with 40ml of As(V) solution (in concentrations ranging from 5 to 100mg/L) and 10g/L Akadama mud were used in the isotherm experiment. Blank tests demonstrated that no arsenic was adsorbed onto the tube walls during the reaction period.

The effect of pH on As(V) sorption was investigated by adjusting the pH value of the solution from 1 to 13 (by adding 1.0M HCl or 1.0M NaOH). The influence of the HCl or NaOH addition on ionic strength could be neglected (less than 1%). In order to study the relationship between the degree of water mineralization and As(V) removal, we adjusted the ionic strength of the solution or added other coexisting ions to simulate various mineralization degrees in underground water. The solution ionic strength was adjusted to 0.001, 0.01, and 0.1M by adding analytical grade NaCl. Coexisting -ion tests were performed using As(V) solutions of 50mg/L containing 5, 10, 25, and 50mg/L of cations, including K^+ (as KCl), Ca^{2+} (as $CaCl_2$), and Fe^{3+} (as $FeCl_3$), and anions, including NO_3^- (as $NaNO_3$), F^- (as NaF), SO_4^{2-} (as Na_2SO_4), and PO_4^{3-} (as Na_3PO_4). These ions represented univalent, bivalent, and trivalent ions,

1 respectively.

2 *2.2.3. Desorption of used sorbent*

3   In this study, HCl (1M) and NaOH (1M) solutions were prepared in order to

4 carry out extraction experiments [32]. The absorbed Akadama mud was separated

5 from the solution and re-suspended in an acid or base solution with 200rpm agitation

6 for 12h. After the reaction, the supernatant was separated by a 0.45μm membrane

7 filter. The As(V) concentration in the supernatant was then analyzed to evaluate the

8 desorption efficiency.

9 **3. Results and Discussion**

10 *3.1. Characterization of the treated Akadama mud*

11   The BET specific surface area of Akadama mud (<150 mesh) was 149.72sq.m/g,

12 and its pore volume was 0.1636ml/g. The BJH (Barrett–Joyner–Halenda) pore-size

13 distribution revealed that the observed pore sizes mostly varied between 2 and 50nm

14 (65.13%), according to the IUPAC classification, which is typical for mesoporous

15 materials. An SEM image of pristine Akadama mud (Fig. 1a) indicates that fragment

16 surfaces were covered with ruptured spherical particles. The random distribution of

17 small particles between flat and coarse areas implies the existence of amorphous

18 particles. Fig. 1b presents the Akadama mud used for adsorption in the initial As(V) of

19 100mg/L. It was found that acicular or prismatic partials appeared on the surface,

20 which elevated the adsorption site and hence the specific surface. The local chemical

21 composition of the sorbent was determined by SEM-EDX, together with XRD

22 patterns. The result is presented in Fig. 1c and Table 1. Pristine Akadama mud

consists mainly of salic mineral; the major metallic oxide content is aluminum oxide,

which accounts for 38.05%, followed by iron (III) oxide and magnesia. Fig. 1d clearly

shows that arsenic atoms had adhered to the surface of adsorbent by comparing with

the pristine Akadama. Fig. 2 depicts the XRD patterns of pristine and adsorbed

Akadama mud with an initial As(V) concentration of 100mg/L. According to XRD

analyses, the acicular or prismatic partials described above might be defined as

boehmite (JCPDS 74-1895) or goethite (JCPDS 81-0463). It could be inferred that the

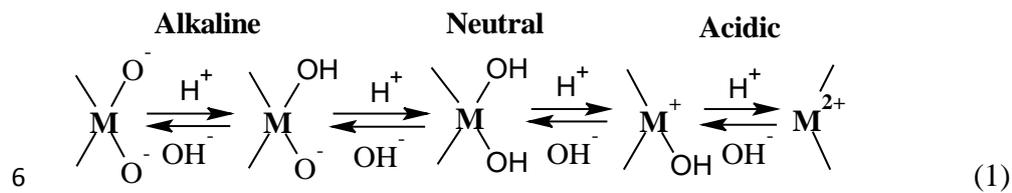reaction with the arsenate had slightly raised the peaks of boehmite and goethite.

3.2. Batch adsorption study

3.2.1. Effect of pH

The results presented in Fig. 3 indicate that the sorption capacity greatly depends

on the solution pH. The optimal pH for arsenic uptake appeared to be pH 3, with a

removal capacity of 4.98mg/g. The removal ability fluctuated slightly over the pH

range of 3 to 11, which means that Akadama mud can adapt to a wide range of pH for

As(V) adsorption and is suitable for practical applications under domestic conditions.

The equilibrium pH (pH_{final}) values were measured after 24h adsorption. It is clear

that pH_{final} increases smoothly with increasing $pH_{initial}$ (Fig. 3). However, it was

observed that Akadama mud had a significant capacity to buffer highly acidic and

alkaline solutions.

The pH of zero point charge (pH_{zpc}) of Akadama mud was 6.9 provided by the

manufacturer (Table 1), which greatly impacts multivalent ions adsorption. As

reported in previous work [33], anionic pollutant adsorption from aqueous medium

depends heavily on the protonation pH range of the predominant metallic oxidic

group, which affects the surface charge of the adsorbent particles and the ionization

degree [33]. The metallic oxidic groups were supposed to be the reactive sites that

developed at the edges of Akadama particles. The mechanism for the surface-charge

property of the metallic oxidic groups can be described as follows [34]:

$$
\underset{\text{Alkaline}}{\overset{}{}}\qquad\underset{\text{Neutral}}{\overset{}{}}\qquad\underset{\text{Acidic}}{\overset{}{}}
$$

$$
\begin{array}{c}\diagdown \raisebox{1ex}{O$^-$}\\ M\\ \diagup \raisebox{-1ex}{O$^-$}\end{array} \underset{OH^-}{\overset{H^+}{\rightleftharpoons}} \begin{array}{c}\diagdown \raisebox{1ex}{OH}\\ M\\ \diagup \raisebox{-1ex}{O$^-$}\end{array} \underset{OH^-}{\overset{H^+}{\rightleftharpoons}} \begin{array}{c}\diagdown \raisebox{1ex}{OH}\\ M\\ \diagup \raisebox{-1ex}{OH}\end{array} \underset{OH^-}{\overset{H^+}{\rightleftharpoons}} \begin{array}{c}\diagdown \\ M^+\\ \diagup \raisebox{-1ex}{OH}\end{array} \underset{OH^-}{\overset{H^+}{\rightleftharpoons}} \begin{array}{c}\diagup \\ M^{2+}\\ \diagdown \end{array} \qquad (1)
$$

where M represents the metallic oxidic adsorbent surfaces. The hydroxylated surface

of the adsorbent develops charge in aqueous solution through amphoteric dissociation,

resulting in a $pH_{initial}$ that tends to be neutral somewhat.

$MOH^+ + OH^- \leftrightarrow \quad M(OH)_2, \qquad pH < pH_{zpc} \qquad (2)$

$M(OH)O^- + H^+ \leftrightarrow \quad M(OH)_2 \qquad pH > pH_{zpc} \qquad (3)$

The decrease in the adsorption efficiency at pH <3 (Fig. 3) was probably due to

metallic-oxidic group loss caused by solubilization and/or degradation. In contrast, the

decreasing trend at pH >11 could be explained by repulsion of the negative charge of

the anionic species in the As(V) solution ($HAsO_4^{2-}$) and the negative surface charge of

the metallic oxide when $pH > pH_{zpc}$. The experiment data agree with the results

obtained by Escudero et al. [35] and Jeon et al. [36].

3.2.2. Adsorption Kinetic

Fig. 4 shows that As(V) adsorbed increased with an increase in contact time and

the pseudo-equilibrium was achieved after approximately 48 h. Such a long

adsorption time was probably due to the accompanying reaction caused by the

complex composition of the nature materials. The experimental data were analyzed

using a pseudo-first order Lagergren equation:

$\log (q_e - q_t) = \log (q_e) - t*K_{ad} / 2.303$ (4)

where q_e and q_t are the amount of As adsorbed at pseudo-equilibrium condition and at

time t, respectively, and K_{ad} is the adsorption rate constant. A linear relationship with

a correlation coefficient of 0.9418 was found between $\log (qe - qt)$ and time, that

indicates the first-order nature of As(V) adsorption.

3.2.3. Isotherm study

The As(V) adsorption isotherms were studied in initial arsenic concentrations of

5, 10, 20, 50, and 100 mg/L with an Akadama dosage of 10g/L (Fig. 5). The obtained

data were analyzed according to linearized forms of Langmuir, Freundlich, and

Redlich–Peterson isotherms. The results indicate that the experiment data best fitted

the Langmuir isotherm, which assumes monolayer adsorption onto a homogeneous

surface and no interaction between sorbed molecules at any of the concentrations or

temperatures [37]. The linearized form of the Langmuir isotherm equation is:

$1/q_e = (1/Q*b*Ce) + 1/Q,$ (5)

where q_e is the amount of solute adsorbed (mg/g) at equilibrium and C_e is the

equilibrium concentration (mg/L), The values of the empirical constants Q and b,

denoting the monolayer capacity and energy of adsorption, were calculated from the

slope and intercept of plot between C_e/q_e and C_e.

Langmuir plots for As(V) adsorption onto Akadama mud are presented in Fig. 5.

The values of the Langmuir correlation coefficients were calculated using linear

1   regression and are given in Table 2. The estimated value of Q for As(V) adsorption

2   increases with a rise in temperature until 50°C then decreases at 65°C. A similar trend

3   can also be seen in the calculated Langmuir parameter b (Table 2). This could be

4   interpreted that As(V) adsorption is endothermic at low temperatures but exothermic

5   in high megathermal aqueous medium.

6   The constant b, which is related to the energy of adsorption, indicates that As(V)

7   adsorption onto Akadama mud was favorable at all concentrations and temperatures in

8   this study. The data are provided in Table 2. (Only the results for an initial

9   concentration of 50mg/L are given.)

10   *3.2.4. Effects of water mineralization*

11   In this study, the effects of water mineralization, mainly focused on the

12   coexisting ions and ionic strength, on the arsenic removal efficiency was investigated.

13   The effect of coexisting ions is presented in Fig. 6. For coexisting anions, the

14   presence of sulfate (in terms of S) and nitrate (in terms of N) did not perceptibly

15   interfere with As (V) removal even at the concentration of 50 mg/L. However, both

16   fluoride (in terms of F) and phosphate (in terms of P) exhibited obvious adverse

17   effects on As (V) removal. The adsorption efficiency decreased quickly from 87.64%

18   to 67.31% with an increase of fluoride concentration from 0 to 50 mg/L. The

19   competitive ability of phosphate was much higher than that of fluoride; As (V)

20   removal dramatically decreased from 87.64% to 36.57% with the same concentration

21   increase of phosphate. This phenomenon suggested that some adsorption sites on the

22   surface of Akadama mud can be occupied by the arsenate, phosphate and fluoride, and

that the adsorption sites had a stronger affinity for phosphate than for arsenate or

fluoride. Sun et al. [23] reported that phosphate evidently inhibited arsenate removal

due to the competitive effect between arsenate and phosphate species.

All of the investigated coexisting cations elevated the As (V) removal ability of

Akadama mud (Fig. 6b). When ferric ions coexisted in the aqueous solution, the As(V)

adsorption efficiency increased significantly from 87.64% to 93.58% with an addition

of 5mg/L of Fe^{3+}, then increased slightly with further addition. The univalent cation

K^+ and bivalent cation Ca^{2+} exhibited a similar trend but with an inferior effect

compared with trivalent cation Fe^{3+}. Multivalent metallic cations carrying a large

quantity of charge can thus enhance the electrostatic attraction with anions ($HAsO_4^{2-}$).

Furthermore, the ferric ion was prone to form hydroxylated complexes like ferric

hydroxide that can provide high affinity to As(V), thereby enhancing the adsorption

ability of Akadama mud. Zhang et al. [38] reported that metallic cations can link the

adsorbent particle with arsenate, forming a metal–arsenate complex or a

metal–H_2O–arsenate complex.

Fig.7 demonstrates that As(V) adsorption with < 150 mesh Akadama mud at

35°C results in the highest removal efficiency (87.64%), followed by < 150 mesh

Akadama mud at 20°C (82.65%) and > 14 mesh Akadama mud at 35°C (77.54%).

However, once ferric cations were added to the reactants, even in concentrations as

low as 5mg/L, experiments performed under the above conditions exhibited almost

the same high arsenic removal efficiency, demonstrating that coexisting metallic

cations (Fe^{3+}) can enhance the sorptive properties of Akadama mud, and might be the

1   key factor in determining the As(V) adsorption performance, as compared with the

2   surface area and adsorption temperature.

3   The ionic strength had a slight effect on As(V) sorption with Akadama mud

4   (Table 3). When the solution ionic strength was increased from 0.001 to 0.1M, the

5   As(V) uptake increased only from 4.74 to 5.25mg/g. However, the obtained data were

6   all higher than that of a blank experiment with DI water. These trends indicate an

7   inner-sphere sorption mechanism for As(V) [39], which suggests that an exchange of

8   aqueous ligand for surface hydroxyl complexes might be generated on the surface of

9   the inner sphere. These trends are consistent with the results reported by Goldberg et

10   al. [39] and Fuller et al. [40], in which arsenate sorption onto mineral surfaces was

11   due to the formation of inner-sphere complexes.

12   All of the above results indicate that the mineralization of aqueous medium has a

13   marked effect on the arsenic adsorption. It is important to investigate arsenic removal

14   based on actual waters instead of deionized water.

15   *3.3. Desorption of the adsorbents*

16   The desorption of the adsorbed Akadama mud was studied using the adsorbed

17   Akadama mud in an isotherm study with an As(V) load of 5.12mg/g. As indicated in

18   Fig. 3, As(V) adsorption decreased dramatically in the polar pH range, so the

19   desorption of adsorbed arsenic under polar acid and alkali conditions was considered.

20   The result (Fig. 8) indicates that at lower pH (pH 1), 3.46mg of arsenic was desorbed

21   from the used Akadama mud, accounting for 67.48% of the adsorbed As(V). In

22   contrast, at an alkaline pH of 13, the desorbed As(V) was 2.42mg, which was 47.29%

of the adsorbed As(V). It is obvious that the desorption is more efficient in the strong

acid range.

4. Conclusions

Akadama mud is a cost-effective adsorbent for arsenic removal from water

solutions, especially from low-level phosphate and fluoride solutions. The As(V)

adsorption capacity of Akadama mud, estimated by the Langmuir equation, was

5.30mg/g at 20 ± 0.5°C. The used Akadama mud could be regenerated under polar

acid or alkali conditions. Under polar acid conditions (pH 1), 67.48% of the adsorbed

arsenic was desorbed.

The relationship between the degree of water mineralization and As(V) removal

was attributed to the effect of coexisting ions on the Akadama mud and the ionic

strength in the surrounding aqueous medium. It is suggested that the deionized batch

experiment should not be extrapolated directly to all cases. It is important to design

batch experiments based on particular adsorbents and on the characteristics of actual

water.

References

[1] Agency for Toxic Substances and Disease Registry (ATSDR), 2005. Top 20

Hazardous Substances: ATSDR/EPA Priority List for 2005.

1 <http://www.atsdr.cdc.gov/cxcx3.html>.

2 [2] H. Brammer., P. Ravenscroft, Environ. Int. 35 (2009) 647–654.

3 [3] R.T Nickson, J.M. McArthur, P. Ravenscroft, W.G. Burgess, K.M. Ahmed, Appl

4 Geochem. 15 (2000) 403–413.

5 [4] H.M. Guo, Y.X. Wang, G.M. Shpeizer, S. Yan, J. Environ. Sci. Health A 38 (2003)

6 2565–2580.

7 [5] WHO. Arsenic. Environmental Health Criteria 18, IPCS International Programme

8 of Chemical Safety. Vammala (Finland): Vammalan Kõirjapaino Oy., 1981.

9 [6] C.K. Jain, I. Ali, Water Res. 34 (2000) 4304–4312.

10 [7] B.K. Mandal, K.T. Suzuki, Talanta 58 (2002) 201–235.

11 [8] A.A. Duker, E.J.M. Carranza, M. Hale, Environ. Int. 31 (5) (2005) 631–641.

12 [9] J.C. Ng, J. Wang, A. Shraim, Chemosphere. 52 (9) (2003) 1353–1359.

13 [10] EPA, Implementation guidance for the arsenic rule, EPA Office of Groundwater

14 and Drinking Water, Cincinnati, 2002.

15 [11] G.L. Ghurye, D.L. Clifford, A.R. Tripp, J. Am. Water Works Assoc. 91 (10) (1999)

16 85–96.

17 [12] H. Garelick, A. Dybowska, E. Valsami-Jones, N.D. Priest, J. Soils Sediments. 5

18 (2005) 182–190.

19 [13] P. Mondal, C.B. Majumder, B. Mohanty, J. Hazard. Mater. B137 (2006) 464–479.

20 [14] J.A. Wilkie, J.G. Hering, Colloids Surf. A 107 (1996) 97–110.

21 [15] T.S. Singh, K.K. Pant, Sep. Purif. Technol. 36 (2004) 139–147.

22 [16] P.B. Bhakat, A.K. Gupta, S. Ayoob, S. Kundu, Colloid Surf. A: Physicochem.

Eng. Aspects 281 (1–3) (2006) 237–245.

[17] T. Budinova, D. Savova, B.Tsyntsarski, C.O. Ania, B. Cabal, J.B. Parra, N.

Petrov, Appl Surf. Sci. 255 (2009) 619–624.

[18] B. Daus, R. Wennrich, H. Weiss, Water Res. 38 (12) (2004) 2948–2954.

[19] Y.H. Xu, T. Nakajima, A. Ohki, J. Hazard. Mater. B 92 (2002) 275–287.

[20] S.W. Al Rmalli, C.F. Harrington, M. Ayub, P.I. Haris, J. Environ. Monit. 7 (2005)

279–282.

[21] S. Bang, G.P. Korfiatis, X.G. Meng, J. Hazard. Mater. 121 (2005) 61–67.

[22] H.M. Guo, D. Stuben, Z. Berner, Appl Geochem. 22 (2007) 1039–1051.

[23] H.W. Sun, L. Wang, R.H. Zhang, J.C. Sui, G.N. Xu, J. Hazard. Mater. B129

(2006) 297–303.

[24] W. Zhang, P. Singh, E. Paling, S. Delides, Mine Eng. 17 (2004) 517–524.

[25] F. Partey, D. I. Norman, S. Ndur, R. Nartey, Colloids and Surfaces A. 337 (2009)

164–172.

[26] S. Goldberg, C.T. Johnston, J. Colloid Interface Sci. 234 (1) (2001) 204–216.

[27] X.H. Sun, H.E. Doner, Soil Sci. 161 (12) (1996) 865–872.

[28] M. E. Pena, X. G. Meng, G. P. Korfiatis, C. Y. Jing, Environ. Sci. Technol. 40

(2006)1257–1262.

[29] M. Vithanage, R. Chandrajith, A. Bandara, R. Weerasooriya, J. Colloid Interface

Sci. 294-2 (2006) 265–272.

[30] Y. Xu, T. Nakajima, A. Ohki, J. Hazard. Mater. B92 (2002) 275–287.

[31] L.C. Roberts, S.J. Hug, T. Ruettimann, Environ. Sci. Technol. 38 (2004)

307–315.

[32] P. Chutia, S. Kato, T. Kojima, S. Satokawa, J. Hazard. Mater. 162 (2009)

440–447.

[33] S. Shevade, R.G. Ford, Water Res. 38 (2004) 3197–3204.

[34] M.S. Syed, Can J Chem. 44 (1966)1663–1670.

[35] C. Escudero, N. Fiol, I. Villaescusa, J. C. Bollinger, J. Hazard. Mater. 164 (2009)

533–541.

[36] C.S. Jeon, K. Baek, J.K. Park, Y.K.Oh, S.D. Lee, J. Hazard. Mater. 163 (2009)

804–808.

[37] M.P. Elizalde-Gonzalez, J. Mattusch, R. Wennrich, P. Morgenstern, Micropor,

Mesopor. Mater. 46 (2001) 277–286.

[38] S.W, Zhang, C.J. Liu, Z.K. Luan, X.J. Peng, H.J. Ren, J. Wang, J. Hazard.

Mater. 152 (2008) 486–492.

[39] S. Goldberg, C.T. Johnston, J. Colloid Interface Sci. 234 (1) (2001) 204–216.

[40] C.C. Fuller, J.A. Davis, G.A. Waychunas, Geochim. Cosmochim. Acta 57 (10)

(1993) 2271–2282.