Contents lists available at ScienceDirect





Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Selective adsorption behavior of phosphate onto aluminum hydroxide gel

Naohito Kawasaki*, Fumihiko Ogata, Hisato Tominaga

Faculty of Pharmacy, Department of Pharmacy, Kinki University, Kowake 3-4-1, Higashi-Osaka 577-8502, Japan

ARTICLE INFO

Article history: Received 26 March 2010 Received in revised form 8 May 2010 Accepted 12 May 2010 Available online 20 May 2010

Keywords: Adsorption isotherms Ion–solid interactions Aluminum hydroxide gel Phosphate

1. Introduction

Phosphorus is one of the major nutrients responsible for eutrophication. While phosphate ores are expected to be depleted within approximately 50 years [1,2]. Since phosphate ores account for approximately 80% of fertilizer raw materials [3], the depletion of these ores will greatly impact agricultural production and may potentially even lead to a food crisis. Advanced treatments are typically required to remove phosphorus from wastewater. Since the biological method yields phosphorus-containing sediments with high water content, further concentration and dehydration processes are necessary for the recovery of phosphorus [4]. In an adsorption method, phosphorus is recovered in the form of highly pure phosphate alkali during the regeneration of the adsorbent. In addition, after desorption of phosphorus, the adsorbent can be reused for phosphorus recovery.

Phosphorus adsorbents made from zeolites [5], lanthanum and yttrium compounds [6,7], aluminum compounds [8–11] and acid type F fly ash [12] have been reported thus far. However, the adsorbed phosphate has to be desorbed from adsorbents for the recovery of phosphate. For instance, only approximately 60% of adsorbed phosphate is desorbed when boehmite is used for phosphorus recovery [9]. Multi-component adsorbents comprising of mixtures of metal oxides and clay are ubiquitous in water environment. They have been shown to be significant in determining the environmental distribution of various contaminants [13]. These

ABSTRACT

The specific surface area and X-ray diffraction patterns for an aluminum hydroxide gel (AHG) calcined at 300–1150 °C, the number of surface hydroxyl groups in the AHG, and the adsorption isotherms of phosphate on AHG were measured in order to develop a phosphate recovery agent. AHG was transformed into γ - and α -alumina by the calcinations treatment. The amount of phosphate adsorbed onto AHG increases at calcining temperatures of 300–700 °C and decreases above a calcining temperature of 800 °C. It was found that AHG selectively adsorbs phosphate ions, but not other anions, and shows the highest adsorption capacity at pH 4–6. Further, the alkali resistance of AHG increased with calcination, and more than 80% of the phosphate adsorbed with an NaOH aqueous solution underwent desorption. The addition of colloidal alumina and colloidal silica resulted in the formation of granules of 500–840 μ m size. The amount of phosphate adsorbed onto AHG after granulation was similar to that before granulation. Thus, the phosphate adsorbed onto AHG did not decrease after granulation suggesting that AHG can be used as a phosphate adsorbent.

Published by Elsevier B.V.

adsorbents exhibit different adsorption behaviors compared with single-component ones [14]. Characterization studies of phosphate adsorption onto many kinds of adsorbents have been investigated [15–17]. Many studies have shown that in water environment aluminum often control the biogeochemical cycling of phosphate [18,19].

Dried aluminum hydroxide gel (AHG) is widely used as an antacid. It can be therefore safely be used as a phosphate adsorbent for the treatment of hyperphosphatemia. However, patients with renal dysfunction show impairment in aluminum excrement, and hence, the use of this gel as a treatment leads to an increase in the plasma concentration of aluminum, eventually leading to aluminum intoxication. Therefore, the dried AHG with high phosphate adsorption capacity as well as a stable structure are required. Furthermore, increasing the particle size of the adsorbent leads to a decrease in its adsorption capacity, thereby limiting practical applications.

In this study, AHG was calcined at different temperatures to obtain a stable AHG structure, and the characteristics of the calcined AHG, including surface area, XRD, phosphate adsorption–desorption, and the influence of granulation on phosphate adsorption capacity, were investigated to evaluate the stability of AHG.

2. Materials and methods

2.1. Material

The AHG used in this study was in the form of a white amorphous powder (Tomita Pharmaceutical Co., Ltd.). 20 g of AHG was placed in

^{*} Corresponding author. Tel.: +81 6 6730 5880x5556; fax: +81 6 6721 2505. *E-mail address*: kawasaki@phar.kindai.ac.jp (N. Kawasaki).

a magnetic crucible and heated to the target temperature over 2 h in a muffle furnace. The temperature was maintained at 300–1150 °C for 2 h (AHG30–AHG115). The granulated AHG (G-AHG) was prepared as follows. After adding 0.5 g of ethyl cellulose and 18 ml of ethanol to 9.5 g of AHG45 calcined at 450 °C and drying at 110 °C for 24 h, particles with sizes of 500–840 μ m (EC-AHG) were obtained. After adding 8 g of virgin AHG into 10 ml of 20% colloidal alumina or 20% colloidal silica (Nissan Chemical Industries, Japan) and maintaining the temperature at 450 °C for 2 h, particles with diameters in the range of 500–840 μ m (AS-AHG and ST-AHG) were obtained. Granulated AHG (0.1 g) was added to 50 ml of KH₂PO₄ solution of 127–1130 mg-PO₄/l and shaken for 15 h at 25 °C, and the phosphate concentration in the filtrate was then measured. The amount of phosphate adsorbed was calculated from the difference between the initial concentration and equilibrium concentration.

Differential thermal analysis of AHG was conducted using a simultaneous thermogravimetry/differential thermal analyzer DTG-60AH (Shimadzu, Japan). XRD analysis was carried out using a RINT2100V (Rigaku, Japan) diffractometer. Electron microscopy was carried out using a scanning electron microscope JSM-5200 (JEOL, Japan), and the specific surface area of AHG was measured using a specific surface analyzer, NOVA e4200 (Yuasa Ionics, Japan).

2.2. Hydroxyl groups of AHG

The number of surface hydroxyl groups in AHG was calculated by fluoride ion adsorption [20]. After adding 0.125 g AHG to 50 ml of 420 mg/l NaF solution, whose pH was adjusted to 4.6 using a 0.2 mol/l acetic acid–0.2 mol/l sodium acetate buffer, and shaking for 15 h, the fluoride ion concentration in the filtrate was then measured using a UV–vis spectrophotometer DR/4000 (Hach, USA). The number of hydroxyl groups was calculated from the difference between the initial and equilibrium fluoride ion concentrations.

2.3. Gelation percentage of AHG

The gelation percentage of AHG was measured according to the Japan Pharmacopoeia standards. Approximately 2.0 g of AHG was added to 15 ml of HCl and heated for 30 min. After cooling, 500 ml total volume was obtained by the addition of distilled water. 20 ml of the obtained solution was added to 30 ml of 0.05 mol/l ethylenediaminetetraacetic acid solution and 20 ml of pH 4.8 acetic acid–ammonium acetate buffer and heated for 5 min. After cooling, 55 ml of ethanol was added to this solution and titrated by a 0.05 mol/l zinc acetate solution. The gelation percentage of AHG was calculated using the formula $p = (30.0 - a) \times 2.549 \times (b \times 20/500)$, where *p* indicates the gelation percentage (%), *a* is the amount of zinc acetate required for titration (ml) and *b* is the amount of AHG (g).

2.4. Phosphate adsorption onto AHG

After adding 0.1 g of AHG to 50 ml of KH₂PO₄ with 127 mg-PO₄/l and shaking the solution for 15 h at 25 °C, the phosphate concentration in the filtrate was measured and the amount of phosphate adsorbed was calculated from the difference between the phosphate concentration before and after adsorption. The phosphate concentration was measured using a phosphate test kit (Hach, USA). The amount of phosphate adsorbed onto AHG at equilibrium was calculated using the formula $q = (Co - Ce) \times V/Ws$, where q denotes the amount of phosphate adsorbed per gram (mg-PO₄/g), Co is the initial concentration of phosphate (mg-PO₄/l), V is the volume of the solution (l) and Ws is the amount of AHG (g).

In addition, the amounts of phosphate adsorbed in solutions with different pH values were measured in order to assess the effect of the pH on phosphate adsorption. The pH values, measured using a digital pH meter (Mettler Toledo, Japan), were adjusted to between 2 and 9 using HCl and NaOH solution. After adding 0.1 g AHG to 50 ml of 127 mg/l phosphate aqueous solution, adjusting the pH of the solution, and shaking it for 15 h at 25 °C, the phosphate concentration in the filtrate and its pH value were measured.

2.5. Selective adsorption of phosphate onto AHG

The amounts of chloride ions, nitrate ions, and sulfate ions adsorbed on AHG were measured to assess the selectivity of phosphate adsorption on AHG. After adding 0.1 g of AHG to 50 ml of 58 mg/l sodium chloride solution, 144 mg/l sodium sulfate solution, and 85 mg/l sodium nitrate solution or to a mixed solution consisting of these 4 species of anions, and shaking the solution for 15 h at 25 °C, the concentration of each ion in the filtrate was measured. The concentrations of chloride and sulfate ions were measured using a UV–vis spectrophotometer, the concentration of nitrate ions was measured using a nitrate test kit (Hach, USA), and the amount of phosphate adsorbed was calculated from the difference between its initial and equilibrium concentrations.

2.6. Phosphate adsorption-desorption onto AHG

After adding 0.5 g of AHG to 50 ml of KH₂PO₄ aqueous solution with 1100 mg-PO₄/l and shaking the solution for 15 h at 25 °C, the phosphate concentration in the filtrate was measured. Then, after adding 0.3 g of AHG (amount of phosphate adsorbed + amount of AHG) that had been dried at 110 °C following adsorption to 500 ml of 400 mg/l NaOH and shaking the solution for 15 h at 25 °C, the phosphate concentration in the filtrate was measured, and the amount of phosphate desorbed on AHG was calculated for the desorption analysis. In addition, the mass of phosphate was measured after drying by adding 0.5 g of AHG to 400 mg/l NaOH solution and shaking the solution for 15 h at 25 °C in order to determine the amount of aluminum dissolved after the treatment of AHG with the NaOH solution. The aluminum ion concentration in the filtrate was measured using UV-vis spectrophotometry. The aluminum concentration in the alkaline solutions was also measured. The AHG meets the Japan and US pharmacopoeia standards, with the total concentration of aluminum being less than 10 mg/l. Further, AHG has an unstable structure, and undergoes dissolution in basic solutions, resulting in an increase in the aluminum ion concentration in the solutions.

2.7. Adsorption isotherms of phosphate onto P-AHG and G-AHG

After adding 0.1 g of P-AHG or G-AHG to 50 ml of KH_2PO_4 with 127–1130 mg-PO₄/l and shaking the solution for 15 h at 25 °C, the phosphate concentration in the filtrate was measured, and the amount of phosphate adsorbed was calculated from the difference between phosphate concentrations before and after adsorption. The phosphate concentration was measured using the phosphate test kit.

3. Results and discussions

3.1. Physical properties of AHG

The results of differential thermal analysis of AHG are shown in Fig. 1. An endothermic reaction occurred at a calcining temperature of approximately 100 °C, which was accompanied by a significant decrease in quality. From these results, AHG is assumed to have undergone dissociation to release the water of crystallization at a temperature near 100 °C. The results of XRD analysis of AHG are shown in Fig. 2. These results confirmed that AHG30 and virgin



Fig. 1. Differential thermal analysis of virgin AHG.



Fig. 2. XRD of calcined AHG. (\blacktriangle) γ -Alumina and (\bigcirc) α -alumina

AHG have similar structures and AHG40–AHG70 have an amorphous structure. Compared to virgin AHG, the crystalline structure of AHG80–AHG115 changed significantly to form a crystalline aluminum compound. The results of the XRD analysis indicate that AHG100–AHG115 have γ - and α -type structures, and calcining AHG above 600 °C transforms its structure into the γ -type structure. A highly porous platinum-alumina catalyst is produced at 1200 °C, with alumina having a γ -type structure [21]. Alumina produced from unstable aluminum compounds has been reported to have a γ -type structure.

The number of surface hydroxyl groups calculated from the specific surface area of AHG and the amount of fluoride ions adsorbed is presented in Table 1. The specific surface area of AHG30–AHG100 did not change significantly, however that of AHG110 and AHG115 changed slightly. It has been reported that the specific surface area of gibbsite, an aluminum compound, becomes $254 \text{ m}^2/\text{g}$ at a calcining temperature of $300 \,^{\circ}\text{C}$ and $18 \,\text{m}^2/\text{g}$ at $1150 \,^{\circ}\text{C}$ [11]. However, no apparent decrease in the specific surface area of AHG was significantly observed after calcining. Also, the adsorption of phosphate by various aluminum compounds is related to their specific surface

| Table 2 | | |
|----------------------------|--------------------------------|----------------|
| Amount of anions adsorbed, | , and removal percentage of an | ions onto AHGs |

Table 1

Physical and chemical properties of AHGs calcined at different temperatures.

| Samples | Specific surface area (m²/g) | Amount of hydroxyl group (mmol/g) | Number of hydroxyl groups (site/nm ²) | Gelation (%) |
|---------|---------------------------------|--|--|--------------|
| Virgin | 94.9 | 1.00 | 6.3 | 51.9 |
| AHG30 | 96.5 | 1.12 | 7.0 | 67.9 |
| AHG40 | 92.3 | 1.12 | 7.3 | 81.7 |
| AHG50 | 89.0 | 1.00 | 6.8 | 78.3 |
| AHG60 | 107.2 | 1.12 | 6.3 | 85.9 |
| AHG70 | 96.9 | 1.06 | 6.6 | 86.7 |
| AHG80 | 91.1 | 0.76 | 5.0 | 44.5 |
| AHG90 | 101.5 | 0.59 | 3.5 | 26.1 |
| AHG100 | 88.0 | 0.53 | 3.6 | 15.0 |
| AHG110 | 80.3 | 0.41 | 3.1 | 9.9 |
| AHG115 | 80.2 | 0.35 | 2.6 | 11.2 |

area [11]. The number of surface hydroxyl groups in AHG30–AHG70 and AHG80–AHG115 is 1.00–1.12 and 0.35–0.76 mmol/g, respectively, decreasing with increasing calcining temperature. AHG40 has the highest number of surface hydroxyl groups. The number of surface hydroxyl groups in AHG decreases with increasing calcining temperature. The calculated number of surface hydroxyl groups in AHG is similar to that reported in literature (<25 sites/nm², obtained using various methods) [22]. The gelation percentage increased with increasing calcining temperature up to 700 °C, but deceased above 800 °C. This indicates that the structure of AHG becomes unstable by calcining AHG below 700 °C.

SEM micrographs of virgin AHG, AHG40, AHG70, AHG80, and AHG115 are shown in Fig. 3. The images indicate that the particles did not undergo disintegration or a change during the calcining process, and only crystal-type was changed from γ - to α -alumina by the calcining treatment.

3.2. Phosphate adsorption onto AHG

The amounts of phosphate and other anions adsorbed onto AHG treated at different calcining temperatures are listed in Table 2. The amount of phosphate adsorbed on AHG30–AHG70 increased minimally compared to that on virgin AHG, and the amount of phosphate adsorbed on AHG80–AHG115 decreased. This indicates that phosphate adsorption depends on the number of surface hydroxyl groups in AHG and the specific surface area of AHG. It has been reported that phosphate adsorption onto an aluminum compound depends on ion exchange with the surface hydroxyl groups of the compound [9,10], consistent with the results of our study. Therefore, AHG30–AHG70 that show higher adsorption capacity compared to virgin AHG were used in the following experiment.

Seawater contains many types of anions. Therefore, in order to recover phosphate from seawater by selective adsorption of phosphate on AHG, it is very important to assess the influence of

| Samples | Amount of phosphate ad | Amount of phosphate adsorbed (mg-PO ₄ /g) | | Removal percentage of anion (%) | | | | |
|---------|------------------------|--|-----------|---------------------------------|---------|---------|--|--|
| | In single solution | In mixed solution | Phosphate | Chloride | Sulfate | Nitrate | | |
| Virgin | 29.3 | 23.6 | 55.1 | 0.9 | 1.0 | 1.1 | | |
| AHG30 | 33.2 | 27.9 | 62.6 | 0.9 | 1.2 | 0.9 | | |
| AHG40 | 31.5 | 26.5 | 58.3 | 0.5 | 2.1 | 0.6 | | |
| AHG50 | 33.4 | 25.1 | 62.6 | 0.9 | 2.8 | 1.3 | | |
| AHG60 | 34.5 | 26.9 | 63.9 | 0.9 | 2.6 | 0.9 | | |
| AHG70 | 31.2 | 23.5 | 58.3 | 0.9 | 2.1 | 2.0 | | |
| AHG80 | 28.5 | - | _ | - | - | - | | |
| AHG90 | 26.5 | - | - | - | - | - | | |
| AHG100 | 21.1 | - | - | - | - | - | | |
| AHG110 | 20.5 | - | - | - | - | - | | |
| AHG115 | 20.5 | - | - | - | - | - | | |
| | | | | | | | | |



Fig. 3. SEM micrographs of AHG.

other anions on the phosphate adsorption capacity of AHG. The removal percentage of chloride, sulfate, nitrate and phosphate ions from each anion is in the following order: chloride ions < sulfate ions < nitrate ions < phosphate ions on Table 2. This indicates that AHG selectively adsorbs phosphate. The removal percentage of phosphate from a mixed anion solution is calculated in order to assess the selectivity of phosphate adsorption on AHG. The above result indicates that AHG selectively adsorbs phosphate from the mixed anion solution. Although the adsorption of phosphate from the mixed anion solution on AHG decreased slightly as compared to that from the phosphate solution, the other anions had little influence on the adsorption. This result is also consistent with those observed for boehmite [8] and gibbsite [9] with different crystal types. All aluminum compounds may probably exhibit a similar phenomenon, that is, phosphate would be adsorb onto both the hydroxyl groups and the surface of AHG.

Since the adsorption of ions is usually influenced by the pH value of the solution, it is important to assess the adsorption behavior of phosphate in solutions with different pH values. Phosphate adsorption onto AHG in solutions with different pH values is shown in Fig. 4. The results indicate that adsorption of phosphate onto AHG is not dependent on the calcining temperature, and a large amount of phosphate is adsorbed in the weak acid range of pH 4–6, decreasing with increasing pH value. This is because phosphate

cannot exchange ions with AHG in alkaline aqueous solutions. In addition, it has been suggested that the phosphate adsorbed on AHG might undergo desorption with high efficiency after treatment with an alkaline solution. Furthermore, the mechanism of



Fig. 4. Amount of phosphate adsorbed onto AHG in different pH solutions.



Fig. 5. Saturated amount of phosphate adsorbed-desorbed onto calcined AHG in NaOH.

phosphate adsorption onto AHG is assumed to be similar to that for boehmite [8] and gibbsite [11]. However, the amount of phosphate adsorbed onto AHG is higher than boehmite and gibbsite. Therefore, AHG is considered to have a crystal structure suitable for the adsorption–desorption of phosphate. Phosphate would be adsorbed onto the hydroxyl groups and the surface of AHG, because the amount of adsorbed of phosphate depended upon the pH in solution.

The saturated amount of phosphate adsorbed-desorbed onto AHG is shown in Fig. 5. The desorption rate of phosphate onto AHG in an NaOH aqueous solution is 80% or higher, which indicates that phosphate can be adsorbed-desorbed with high efficiency. AHG is known to undergo dissolution in alkaline aqueous solutions, and therefore, it is necessary to assess the alkali resistance of AHG for studying the adsorption-desorption of phosphate onto AHG. Moreover, it has been reported in the Japanese Pharmacopoeia that AHG dissolves in sodium hydroxide [23]. The recovery rate of AHG after treatment with NaOH solution is shown in Fig. 6. These results indicate that the recovery rate increases with an increase in calcining temperature, and the recovery rate of AHG40 is 1.5 times higher than that of virgin AHG. In addition, the recovery rate of AHG100–AHG115 is approximately 90%. This can be explained by the structure becoming stable after calcining, on the basis of the XRD results, eventually resulting in the formation of a crystalline aluminum compound (α -Al₂O₃). This indicates that calcined AHG is suitable of use as an adsorbent.



Fig. 6. Recovery rate of AHG after NaOH treatment for desorption of phosphate.



Fig. 7. Adsorption isotherms of phosphate onto P-AHG and G-AHG.

The adsorption isotherms of phosphate onto P-AHG and G-AHG are shown in Fig. 7. Granulated adsorbents with non-uniform quality are obtained for calcining temperatures less than 400 °C. Granulated adsorbents with uniform quality can be obtained when calcined at higher temperatures. More energy is required to produce G-AHG than P-AHG. Thus, the calcining temperature is selected to be 450 °C. The results indicate that phosphate adsorption on 3 types of G-AHG is comparable to that on P-AHG at all concentrations. This shows that there is almost no decrease in the amount of phosphate adsorbed after granulation due to the addition of organic binders such as ethyl cellulose or inorganic binders including colloidal alumina and colloidal silica. It is generally expected that the amount of phosphate adsorbed decreases due to the reduction in the apparent contact area with solvents following granulation. However, granulated AHG, especially AHG treated with colloidal alumina and colloidal silica reported to exhibit phosphate adsorption [24,25], shows no decrease in the phosphate adsorption capacity.

4. Summary

hydroxide gel Aluminum can be used for the adsorption-desorption of phosphate if the crystal structure AHG is stabilized by calcining at 300-700 °C and the alkali resistance of AHG increases with increasing calcining temperature. AHG60 was found to be the best adsorbent, as its specific surface area, the number of hydroxyl groups, stability, and amount of phosphate adsorbed are the highest. We have thus successfully prepared a novel type of granular adsorbent that does not exhibit a decrease in its adsorption capacity after granulation due to the addition of ethyl cellulose, colloidal alumina, and colloidal silica.

Acknowledgments

This work was financially supported by the "Antiaging Center Project" for Private Universities from the Ministry of Education, Culture, Sports, Science and Technology, Japan, 2008–2012.

References

- C.R. Burdick, D.R. Refling, D.H. Stensel, Advanced biological treatment to achieve nutrient removal, J. Water Pollut. Control Feder. 54 (1982) 1078–1086.
- [2] T. Hano, H. Takahashi, M. Hirata, K. Urano, S. Eto, Removal of phosphorus from wastewater by activated alumina adsorbent, Water Sci. Technol. 35 (1997) 39–46.
- [3] D. Zhano, A.K. Sengputa, Ultimate removal of phosphate from wastewater using a new class of polymeric ion exchangers, Water Res. 32 (1997) 1613–1625.

- [4] M.J. Haron, S.A. Wasay, S. Tokunaga, Preparation of basic yttrium carbonate for phosphate removal, Water Environ. Res. 69 (1997) 1047–1051.
- [5] J.S. Geelhoed, T. Hiemstra, W.H. Van Riemsdijk, Phosphate and sulfate adsorption on goethite: Single anion and competitive adsorption, Geochim. Cosmochim. Acta 61 (1997) 2389–2396.
- [6] S.A. Wasay, S. Tokunaga, S.W. Park, Removal of hazardous anions from aqueous solutions by La(III)- and Y(III)-impregnated alumina, Sep. Sci. Technol. 31 (1996) 1501–1514.
- [7] S.A. Wasay, Md.J. Haron, S. Tokunaga, Adsorption of fluoride, phosphate, and arsenate ions on lanthanum-impregnated silica gel, Water Environ. Res. 68 (1996) 295–300.
- [8] S. Tanada, M. Kabayama, N. Kawasaki, T. Sakiyama, T. Nakamura, M. Araki, T. Tamura, Removal of phosphate by aluminum oxide hydroxide, J. Colloid Interface Sci. 257 (2003) 135–140.
- [9] M. Kabayama, T. Sakiyama, N. Kawasaki, T. Nakamura, M. Araki, S. Tanada, Characteristics of Phosphate ion adsorption–desorption onto aluminum oxide hydroxide, J. Chem. Eng. Japan 36 (2003) 499–504.
- [10] M. Kabayama, N. Kawasaki, N. Nakamura, S. Tanada, Adsorption/desorption characteristics of phosphate ion onto calcined boehmite surface, J. Surf. Nanotechnol. 3 (2005) 63–69.
- [11] N. Kawasaki, F. Ogata, K. Takahashi, M. Kabayama, K. Kakehi, S. Tanada, Relationship between anion adsorption and physicochemical properties of aluminum oxide, J. Health Sci. 54 (2008) 324–329.
- [12] D.G. Grubb, M.S. Guimaraes, R. Valencia, Phosphate immobilization using acidic type F fly ash, J. Hazard. Mater. 76 (2000) 217–236.
- [13] L. Weng, E.J.M. Temminghoff, S. Lofts, E. Tipping, W.H. Van Riemsdijk, Complexation with dissolved organic matter and solubility control of heavy metals in a sandy soil, Environ. Sci. Technol. 36 (2002) 4804–4810.
- [14] Y. Xu, L. Axe, Synthesis and characterization of iron oxide-coated silica and its effect on metal adsorption, J. Colloid Interface Sci. 282 (2005) 11–19.

- [15] O.R. Harvey, R.D. Rhue, Kinetics and energetics of phosphate sorption in a multicomponent Al(III)-Fe(III) hydr(oxide) sorbent system, J. Colloid Interface Sci. 322 (2008) 384–393.
- [16] D. Rodic, M. Mitric, R. Tellgren, H. Rundlof, The cation distribution and magnetic structure of Y₃Fe_(5-x)Al_xO₁₂, J. Magn. Magn. Mater. 232 (2001)1–8.
- [17] Y. Masue, R.H. Loeppert, T.A. Kramer, Arsenate and arsenite adsorption and desorption behavior on coprecipitated aluminum:iron hydroxides, Environ. Sci. Technol. 41 (2007) 837–842.
- [18] R. McDowell, L. Condron, Influence of soil constituents on soil phosphorus sorption and desorption, Commun. Soil Sci. Plant Anal. 32 (2001) 2531–2547.
 [19] J.O. Agbenin, Extractable iron and aluminum effects on phosphate sorption in
- a savanna Alfisol, Soil Sci. Soc. Am. J. 67 (2003) 589–595. [20] T. Yamashita, Y. Ozawa, N. Nakajima, T. Murata, Collection of uranium from
- [20] I. Yahdashidi, Y. Ozawa, N. Nakajina, T. Mutata, Collection of uranium from sea water with hydrous oxide adsorbents 1. Ion exchange properties and uranium adsorption of hydrous titanium(IV) oxide, Nippon Kagaku Kaishi 8 (1978) 1057–1061.
- [21] J.A. Davis, J.D. Hem, The surface chemistry of aluminum oxides and hydroxides, in: G. Sposito (Ed.), The Environmental Chemistry of Aluminum, CRC Press, Inc., Florida, 1989 (Chapter 7).
- [22] X. Yang, D. Wang, Z. Sun, H. Tang, Adsorption of phosphate at the aluminum (hydr)oxides-water interface: role of the surface acid-base properties, Colloid Surf. A: Physicochem. Eng. Aspects 297 (2007) 84–90.
- [23] The Japanese Pharmacopoeia, fourteenth ed., Part I, 2001, p. 233.
- [24] V.V. Murashov, J. Leszczynski, Adsorption of the phosphate groups on silica hydroxyls, J. Phys. Chem. A 103 (1999) 1228–1238.
- [25] C.C. Chusuei, M.J. Van Stipdonk, D.R. Justes, K.H. Loh, E.A. Schweikert, D.W. Goodman, A.A. Campbell, Book of abstracts 217th ACS National Meeting Anaheim, CA, March 21–25, 1999.