Mechanism of As(V) Removal in Wastewater Treatment Using Fe(III)-Supported Exchange Resins

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Abstract

As(V) removal mechanism using Fe(III)-supported ion exchange resins was elucidated by adsorption isotherms and X-ray absorption fine structure spectroscopy. Fe(III) substituted almost all counter ions in the resin at pH 2 during 1-h reaction; the Langmuir exchange capacity was 1.87 mmol Fe/g-resin. The Fe(III)-supported resin comprised ≈65% Fe(III) and ≈35% ferrihydrite. Under pH 3 and arsenic concentration of 10 mg/dm³ conditions, a sorption density of 0.035 mmol As/g-resin was obtained after 1-h reaction, and increased to 0.74 mmol As/g-resin after 120-h reaction. As(V) removal proceeded mainly via surface precipitation of poorly crystalline ferric arsenate via co-precipitation between As(V) and Fe(III); also, some As(V) adsorbed on ferrihydrite.

Key words: Arsenate, Ferrihydrite, Wastewater treatment, Ion exchange resin, X-ray absorption fine structure

1. Introduction

Arsenic has been extensively used in the metal smelting, pharmaceutical products, and petroleum industries. Additionally, arsenic and its derived compounds have been produced and used commercially in the agricultural industry as wood preservatives and agricultural chemicals, and in the high-technology field such as semiconductors and IC (integrated circuit). Although very small amounts of specific arsenic compounds are known to have beneficial effects, arsenic is widely renowned for its toxic effect on animals and humans. Long-term exposure to arsenic via drinking water can result in skin, lungs, bladder, and kidney cancer, and other skin conditions such as changes in pigmentation and thickening¹.

Elevated levels of arsenic are often present in the environment as a result of weathering and dissolution of minerals, and numerous anthropogenic sources that include mine wastes, coal fly ash, and arsenic-containing pesticides²,³. Many people in the world are exposed to potentially harmful levels of arsenic through drinking water or food. For example, several millions people in Bangladesh and India are at risk of exposure via drinking water obtained from contaminated groundwater sources³,⁴.

Acid mine drainage (AMD) contains harmful heavy metals such as arsenic and lead, and it is one of the serious global environmental problems⁵,⁶. For instance, Japan has about 80 abandoned or closed mines that have been generating AMD for the last few decades⁷–⁹. Several such mines in Japan have produced AMD that contains dilute concentrations of arsenic that exceed Japanese effluent standards (0.1 mg/dm³)⁰. For example, AMD from the abandoned Horobetsu sulfur mine, which is the largest AMD source in Japan, contains approximately 10 mg/dm³ arsenic. Because the outflow of AMD is generated from abandoned and closed mines, AMD is treated on a semi-permanent basis.

The conventional method for treating arsenic in AMD is via co-precipitation. This method involves the addition of an alkaline compound, such as calcium hydroxide or calcium carbonate, to increase the pH, resulting in the precipitation of heavy metal pollutants¹¹. Arsenate (As(V)) and arsenite (As(III)), which are the predominant inorganic arsenic species present in natural aquatic
systems\textsuperscript{12}, also co-precipitate with the hydroxides of heavy metals such as ferrihydrite and aluminum hydroxide\textsuperscript{13}. This co-precipitation method can remove As(V) from wastewater but results in a large amount of sludge that is problematic in solid–liquid separation. For AMD treatment, it is essential to minimize the amount of sludge generated because toxic substances in the sludge can potentially be re-dissolved.

This study aims to investigate As(V) removal using Fe(III)-supported ion exchange resins. Fe(III) salts was widely known as most economical reagent for As(V) removal. On the other hand, ion exchange resins-packed columns have been widely used in wastewater treatment sites because no additional solid–liquid separation is required. In some cases, ion exchange resins can be recovered and reused, thereby reducing costs and sludge generation. In the case of this study, Fe(III)-supported and As(V) removed could be recovered and reused as a virgin resin by acid treatment. This method is suitable for treatment of small volume wastewater because of the relatively high-operating cost and large installation space.

In a previous study, we demonstrated that As(V) uptake mechanism via co-precipitation using ferrihydrite was a combination of surface complexation and surface precipitation\textsuperscript{14}. Surface complexation was associated with two-dimensional adsorption of As(V) onto the surface of ferrihydrite, whereas surface precipitation was used to describe the three-dimensional uptake of As(V) onto ferrihydrite. Thus, As(V) removal mechanism using Fe(III)-supported ion exchange resins is expected to be similar to As(V) co-precipitation using ferrihydrite. However, comprehensive studies are required to validate this hypothesis.

The objective of this study is to assess As(V) removal mechanism using Fe(III)-supported ion exchange resins; the results were compared with conventional removal methods such as co-precipitation. Fe(III)-supported ion exchange resins were first prepared onto which batch As(V) removal studies were conducted. To discuss the details of As(V) removal mechanism, both filtrates and residues following batch removal experiments were analyzed, generating sorption isotherms. Also, the chemical states of the iron and arsenic ions in the ion exchange resins were evaluated by X-ray absorption fine structure (XAFS) spectroscopy.

2. Experimental

2.1. Standards and reagents

All chemicals and solutions used in this study were of analytical grade and purchased from Kanto Chemicals Inc., Tokyo, Japan. The As(V) and Fe(III) solutions were respectively prepared from Na\(_2\)HAsO\(_4\)·7H\(_2\)O and Fe(NO\(_3\))\(_3\)·9H\(_2\)O. In this study, gel-type and Na-form strong acid cation resin (DIAION, SK104, Mitsubishi Chemicals, Tokyo, Japan) was used.

For the Fe(III) support experiments, the pH was adjusted to 2 by addition of 1 M HNO\(_3\). For the As(V) removal studies, the pH and ionic strength were respectively adjusted to 3 and 0.05 mg/dm\(^3\) by addition of 1 M HNO\(_3\), NaOH, and NaNO\(_3\). All experiments were conducted at 25°C and at least in triplicate; the error was confirmed to be within 5%.

2.2. Fe(III) support experiments

A 1000 mg/dm\(^3\) Fe(III) solution was first prepared by dissolving Fe(NO\(_3\))\(_3\)·9H\(_2\)O in 1.0 dm\(^3\) deionized (DI) water and the pH was adjusted to 2.0. Then, 1.25–50 mg/dm\(^3\) of ion exchange resins was immersed in the Fe(III) solution, and the resulting mixture was stirred using a magnetic stirrer for 1 h. After stirring, the solution was filtered through a 0.45-μm membrane filter. The Fe(III) concentration in the filtrate was analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES) using a SPS-4000 atomic emission spectrometer (Hitachi high-technology science, Tokyo, Japan) equipped with a hydride generator accessory (HYD-10, Hitachi high-technology science, Tokyo, Japan). The residue was dried at 45°C for 24 h and subsequently used for the As(V) removal experiments and subjected to XAFS analysis.

Prolonged reactions were also conducted to evaluate the mechanism of the Fe(III)-supported resins process and compared with the As(V) uptake mechanism onto the Fe(III)-supported resins. For the prolonged reactions experiments, the initial Fe(III) concentration and pH were set at 1000 mg/dm\(^3\) and 2, respectively. The Fe(III) solution was stirred using a magnetic stirrer for 24 h following immersion of 5 g/dm\(^3\) ion exchange resins in the Fe(III) solution.

2.3. As(V) removal studies

A 10 mg/dm\(^3\) As(V) solution was prepared by dissolving Na\(_2\)HAsO\(_4\)·7H\(_2\)O in 0.1 dm\(^3\) DI water, and the pH and ion ionic strength were adjusted to 3 and 0.05 mg/dm\(^3\), respectively. Then, 1–50 mg/dm\(^3\) Fe(III)-supported ion exchange resins were immersed in the As(V) solution. The result-
ing solution was stirred using a magnetic stirrer for 1 h. After stirring, the solution was filtered through a 0.45-μm membrane filter. The As(V) concentration in the filtrate was analyzed by ICP-AES, as above. The residue was dried at 45°C for 24 h and analyzed via XAFS.

The reaction time was set at 1 h, because typical reaction times for AMD treatment are less than 1 h. Prolonged reaction studies were also conducted to confirm the rate-determining process for As(V) removal by Fe(III)-supported ion exchange resin. The initial As(V) concentration and pH were set at 10 mg/dm$^3$ and 3, respectively, and the As(V) solution was stirred using a magnetic stirrer for 120 h following immersion of 0.1 g/dm$^3$ Fe(III)-supported ion exchange resins in the As(V) solution.

2.4. XAFS analysis

X-ray absorption near-edge structure (XANES) analysis was performed at the beam line BL-12C in the Photon Factory (PF) of the National Laboratory for High-Energy Physics, Tsukuba, Japan. K-edge of As was obtained in the range of 11360–12 965 eV. For analysis, Ar gas (100%) and N$_2$ gas (100%) were used for the incident and transmission beams, respectively. Also, K-edge of Fe was recorded in the range of 6781–7856 eV. For analysis, the ionization chamber was filled with N$_2$ gas (50%) and Ar gas (50%) to monitor the incident beam, whereas N$_2$ gas (100%) was used for the transmission beam. The beam was untuned by 30% to avoid higher order harmonics generation in the K-edge analysis of Fe.

The residue obtained following the 1-h reaction experiments was dried at 45°C for 24 h and subjected to XAFS analysis. For As K-edge analysis, the fluorescence method was used because As concentration in the residue was less than 0.5% and the residue was provided in powder form. In contrast, for Fe K-edge analysis, the transmission method was used because Fe concentration in the residue was sufficient and the residue was provided in pellet form that was prepared by mixing with BN powder and compacting under 20 kN.

Ferrihydrite, poorly crystalline ferric arsenate and Fe(III) ions were analyzed as reference materials in Fe K-edge. Ferrihydrite was synthesized at pH 5 from Fe(NO$_3$)$_3$·9H$_2$O solution. As(V)-adsorbed ferrihydrite and poorly crystalline ferric arsenate were also analyzed as reference materials in As K-edge. As(V)-adsorbed ferrihydrite was synthesized from the As(V) adsorption experiments at pH 5 using the above-mentioned ferrihydrite. Poorly crystalline ferric arsenate was synthesized using the procedure described by Jia et al.$^{16}$, with slight modification. These materials were also used as reference in XAFS analysis in our previous studies that included the preparation procedures.$^{14,15}$

3. Results and Discussion

3.1. Fe(III) support experiments

Figure 1 shows the relationship between residual Fe(III) concentration and exchange capacity obtained from the Fe(III) support experiments. The Fe(III) concentration was set at 1000 mg/dm$^3$ while the dosage of the ion exchange resin was varied from 1.25 to 50 mg/dm$^3$. The pH of the solution that was initially set at 2 changed minimally during the Fe(III) supporting experiments.

The experimental data were fitted accordingly, generating a Langmuir-type isotherm (Figure 1); the maximum exchange capacity of the resin was calculated at 1.87 mmol Fe/g-resin. The neutral salts splitting capacity of the resin used in this study was estimated at <5 meq/g-resin. It is believed that Fe(III) were exchanged with as much counter ions in the ion exchange resin as possible.

As observed in Figure 1, the Fe(III) supporting amount was saturated when the dosage of the resin was under 10 mg/dm$^3$ and the residual Fe(III) concentration was above 14.9 mmol/dm$^3$. Therefore, Fe(III)-supported ion exchange resins at a resin dosage of 10 mg/dm$^3$ was used for the subsequent As(V) removal studies.

3.2. As(V) removal studies

Figure 2 shows the relationship between residual As(V) concentration and As(V) sorption density. The initial As(V) concentration was fixed at

![Fig. 1](image-url) Relationship between residual Fe(III) concentration and exchange capacity. Initial Fe(III) concentration = 1000 mg/dm$^3$; initial pH = 2; and reaction time = 1 h.
10 mg/dm³ (0.15 mmol/dm³) and the dosage of the Fe(III)-supported resin was varied from 1 to 50 mg/dm³. It should be noted that the studied As(V) concentration (i.e., 10 mg/dm³) is comparable to that in AMD from the abandoned Horobetsu sulfur mine in Japan. The pH that was initially set at 3 slightly dropped to 2.5 following addition of the Fe(III)-supported resins. The experimental data were fitted accordingly, generating a Langmuir-type sorption isotherm from which the maximum sorption density was obtained at 0.035 mmol As/g-resin. The sorption density corresponded to 0.019 mmol As/mmol Fe in the resin. The As(V) concentration in solution could be reduced to below the Japanese effluent standard (0.1 mg/dm³) upon addition of 50 g/dm³ Fe(III)-supported ion exchange resins, which corresponded to the addition of 0.49 mmol/dm³ Fe(III).

As(V) removal mechanism by conventional ferrihydrite-based co-precipitation was previously demonstrated to proceed via a combination of surface complexation and surface precipitation. A maximum sorption density of 1.0 mmol As/mmol Fe was achieved when the initial As/Fe molar ratio was large and the primary As(V) removal mechanism proceeded via surface precipitation. In contrast, a sorption density of ≈0.4 mmol As/mmol Fe was obtained when the initial As/Fe molar ratio was small and the primary As(V) removal mechanism occurred via surface complexation. Conversely, the maximum As(V) sorption density on ferrihydrite in simple adsorption processes has been widely reported at ≈0.205 mmol As/mmol Fe.

Relative to the results obtained via conventional As(V) removal methods using ferrihydrite, the sorption density obtained in this experiment was lower. The low sorption density is believed to be due to the short reaction time employed (i.e., 1 h) that was likely insufficient for complete As(V) removal by the Fe(III)-supported ion exchange resin. Therefore, prolonged reaction studies were investigated and discussed as follows.

3.3. Long-time reaction studies

Figure 3 shows the residual Fe(III) concentration and Fe(III) exchange density following 24-h reaction of the Fe(III) supporting experiments and Figure 4 shows the residual As(V) concentration and As(V) sorption density after 120-h reaction in the As(V) removal experiments using Fe(III)-supported ion exchange resins.

In the Fe(III) supporting experiments, the reaction rapidly reached equilibrium, as shown in Figure 3. This indicated that diffusion of Fe(III) ions into the resin and subsequent exchange with the counter ions present in the resin were relatively fast.

In contrast, in the As(V) removal experiments, reaction was slow, requiring 96 h to reach equilibrium. When compared with the sorption density obtained for the 1-h study (Figure 2), the sorption density obtained after 120 h significantly improved to 0.74 mmol As/g-resin. This value corresponded to 0.40 mmol As/mmol Fe, which is comparable with the results obtained in conventional As(V) removal experiments using a ferrihydrite-based co-precipitation process. The slow reaction suggested that As(V) removal mechanism by Fe(III)-supported ion exchange
resins is complex and differs from that occurring in the Fe(III) supporting resins processes, thereby suggesting the occurrence of restricted access/diffusion inside resins.

3.4. XAFS analysis

XAFS analysis was conducted to further evaluate As(V) removal mechanism by the Fe(III)-supported resins. Figure 5 shows the Fe K-edge XANES spectra of the residues before and after As(V) removal. Fe(III) ions in solution, ferrihydrite, and poorly crystalline ferric arsenate were also analyzed by XAFS and the spectra were used as reference materials for comparison with the XANES spectra of the ion exchange resins. The XANES spectrum of As(V)-adsorbed ferrihydrite was similar to that of ferrihydrite. Comparison of spectra of the reference materials showed that the spectra of ferrihydrite and poorly crystalline ferric arsenate featured a pre-edge peak at 7112 Å that was not clearly observed in the spectrum of Fe(III) ion. Moreover, the shape of the main peak following adsorption was different: a relatively sharp peak was obtained in the spectrum of Fe(III) ion, whereas a broad peak was obtained in the spectra of ferrihydrite and poorly crystalline ferric arsenate. The position of the small shoulder after the main peak was also different: the shoulder peak was observed at 7137 Å in Fe(III) ion, 7135 Å in poorly crystalline ferric arsenate, and 7146 Å in ferrihydrite. XANES analyses were successfully applied to clearly identify these three ferric chemical and mineralogical forms.

XANES analysis of the Fe(III)-supported resin before the As(V) removal experiments revealed the presence of a weak pre-edge peak at 7112 Å and a weak shoulder peak (after the main peak) at 7137 Å. A considerably weak shoulder peak at 7146 Å was also found. Furthermore, the main peak corresponding to the resin, after the adsorption edge, was not as sharp as that corresponding to the Fe(III) ion. These results suggested that the main chemical form of ferric in the Fe(III)-supported resin was Fe(III) ion; a small amount of ferrihydrite was also present.

The XANES spectra of the resin after As(V) removal bear similarities to those of the resin before As(V) removal. However, the main peak became slightly broader and a shoulder peak (after the main peak) was observed at 7135 Å. These results suggested that the main chemical form of ferric in the resin after As(V) removal remained a combination of Fe(III) ion and ferrihydrite; also poorly crystalline ferric arsenate was present.

To obtain the weight ratio of the chemical/mineralogical form of ferric in the ion exchange resin, XANES spectra of the residue before and after the As(V) removal experiments were fitted using the spectra corresponding to ferrihydrite, poorly crystalline ferric arsenate, and Fe(III) ion between 7100 and 7300 Å, using XAFS analysis software, REX2000.

As shown in Table 1, the major chemical form of ferric in the Fe(III)-supported resins was Fe(III) ion, and ≈35% of ferric was present as the precipitated ferrihydrite form. Thus, when Fe(III) ion was supported onto the resins, 65% of Fe(III) ion...
was driven into the resin and exchanged with the counter ions, whereas others precipitated as ferrihydrite around the surface of the resin.

Following As(V) removal, the content of Fe(III) ion decreased whereas the content of ferrihydrite increased; the presence of a small amount of poorly crystalline ferric arsenate was also noted. The increased formation of ferrihydrite and poorly crystalline ferric arsenate was attributed to the higher pH used (i.e., 3) for the As(V) removal experiments relative to that in the Fe(III) supporting studies that was sufficiently high to instigate precipitation of ferrihydrite and poorly ferric arsenate. The formation of poorly crystalline ferric arsenate only was previously observed in co-precipitation processes, whereby sorption of As(V) proceeded on precipitates concurrently with the precipitation of ferrihydrite. Therefore, these results suggest that mechanism of As(V) removal by Fe(III)-supported resins is a combination of co-precipitation and simple adsorption processes. Some As(V) ions are adsorbed onto ferrihydrite that formed around the surface of the resin during the Fe(III) supporting process or by the exchange counter ions during the As(V) removal experiments, whereas other As(V) ions are adsorbed onto poorly crystalline ferric arsenate during co-precipitation with Fe(III) ions. Precipitation of poorly crystalline ferric arsenate or ferrihydrite during the As(V) removal experiments requires diffusion of the Fe(III) ion from the interior of the resin through precipitates located around the surface of the resin. This step is likely to be rate-determining, thereby requiring longer times (i.e., 96 hours, Figure 4) to achieve an equilibrium state.

Figure 6 shows the As K-edge XANES spectra of the residue following As(V) removal studies. As(V)-adsorbed ferrihydrite and poorly crystalline ferric arsenate were analyzed as reference materials. These two chemical arsenate forms were distinguished by the shape of the first main peak: poorly crystalline ferric arsenate featured a sharper peak than that of As(V)-adsorbed ferrihydrite.

As observed in Figure 6, the main first peak of the resin after As(V) removal as relatively sharp and similar to that of poorly crystalline ferric arsenate. Thus, the main mechanism of As(V) removal by the Fe(III)-supported resins is expected to proceed via a co-precipitation process between As(V) and Fe(III). As previously demonstrated, poorly crystalline ferric arsenate precipitates via a surface precipitation mechanism when the initial As/Fe molar ratio is above 0.4, even if the concentrations of As(V) and Fe(III) in bulk solution are unsaturated thermodynamically. In the case of As(V) removal by the Fe(III)-supported resin, Fe(III) ion located in the inner regions of the resin diffused slowly thus, a high As/Fe molar ratio would be achieved around the surface of resin, followed by precipitation of poorly crystalline ferric arsenate. Table 2 shows the weight ratio of the chemical forms of arsenate in the ion exchange resin following As(V) removal, obtained from XANES spectra fitting using As(V)-adsorbed

| Fe(III) support | Ferrihydrite | Poorly crystalline ferric arsenate | Fe(III) ion | R value |
|----------------|-------------|----------------------------------|------------|--------|
| (Before As(V) removal) | 35.0 | — | 65.0 | 0.020 |
| After As(V) removal | 49.5 | 6.2 | 44.3 | 0.035 |

Fig. 6 As K-edge XANES spectra of ion exchange resins following As(V) removal, As(V)-adsorbed ferrihydrite, and poorly crystalline ferric arsenate.
ferrihydrite and poorly crystalline ferric arsenate as reference materials. These results suggest that the main mechanism of As(V) removal by the Fe(III)-supported resins proceeds via surface precipitation of poorly crystalline ferric arsenate.

We also conducted EPMA observation for cross-section of the resin before/after As(V) removal. EPMA results showed that Fe(III) inside the resin was gradually concentrated toward the surface and almost As(V) was existed at the surface of the resin after As(V) removal. These results suggested that As(V) removal was influenced by the diffusion of Fe(III) inside the resin.

4. Conclusions

In this study, As(V) removal using Fe(III)-supported ion exchange resins was investigated at an initial arsenic concentration of 10 mg/dm$^3$ and pH 3. Fe(III) exchanged almost all counter ions in the resin at pH 2 after 1-h reaction; the exchange capacity was calculated as 1.87 mmol Fe/g-resin based on Langmuir plots. Based on XANES analysis of the resin, ≈65% of the ferric supported on the resins existed as Fe(III) ion while the remaining existed as the ferrihydrite precipitated form. Under pH 3 and initial As(V) concentration of 10 mg/dm$^3$ conditions, the sorption density obtained following 1-h reaction was 0.035 mmol As/g-resin that increased to 0.74 mmol-As/g-resin after 120-h reaction. Based on XANES analysis, the main mechanism of As(V) removal by the Fe(III)-supported resins proceeded by surface precipitation of poorly crystalline ferric arsenate via co-precipitation between As(V) and Fe(III); also, some As(V) adsorbed on ferrihydrite via a simple adsorption process. These results suggested that Fe(III) diffusion from the inner regions of the resin through the precipitate located around the surface of resin was the rate-determining step, thereby requiring prolonged reactions for As(V) removal by the Fe(III)-supported resins.

Above results suggested that Fe(III)-supported resin could remove As(V) in densely-concentrated form by the combination of surface precipitation and surface complexation to ferrihydrite, and achieve both As(V) uptake and solid-liquid separation at once. Toward more efficient As(V) removal, most appropriate solution conditions of co-existing ions and pH for Fe(III)-supporting should be investigated, to support more Fe(III) to resin as Fe(III) ions form, not as ferrihydrite form.

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