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REMOVAL OF ARSENIC(V) BY ZIRCONIUM(IV)-LOADED PHOSPHORIC ACID CHELATING RESIN

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ABSTRACT

Properties of Zr(IV)-loaded phosphoric acid chelating resin (RGP) as an adsorbent of As(V) were studied by means of the columnar method. Little leakage of Zr(IV) from RGP loaded with Zr(IV) (Zr-RGP) was observed within a wide pH range (pH 1.1–0.7 mol/L NaOH). Uptake of As(V) by Zr-RGP decreased with an increase in feeding-solution pH. The maximum capacity for As(V) was approximately 0.20 mmol/mL of wet resin (0.67 mmol/g of dry resin). Sodium chloride and sodium nitrate enhanced adsorption of As(V), and electrolytes in seawater also promoted the adsorption of As(V). Arsenic(V) adsorbed on Zr-RGP was quantitatively eluted with 0.4 mol/L sodium hydroxide, and repeated use of Zr-RGP was possible. In addition, Zr-RGP showed high selectivity to As(III). A large volume of 0.7 mol/L sodium hydroxide was needed to elute adsorbed As(III) quantitatively, indicating that As(III) was more strongly adsorbed by Zr-RGP than was As(V).

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INTRODUCTION

Recently phosphoric acid resin RGP (Fig. 1) with a large acid capacity (8 mEq/g) was developed by the addition of phosphoric acid to epoxy groups of macroreticular poly(glycidyl methacrylate-co-divinylbenzene) (1), and its metal ion selectivity was shown to be significantly different from that of conventional cation-exchange resins with sulfonic acid groups (1,2). RGP exhibits extremely high affinity to hard Lewis-acid metal ions, such as Ti(IV), U(VI), and Zr(IV), and shows much higher selectivity for Pb(II) than to alkaline earth metal ions (1–3). Thus, RGP is expected to be useful for the purification of water contaminated with heavy metal ions. In addition, its use in selective removal of a specific anion in water will also be possible through ligand exchange through the use of RGP loaded with metal ions that bind selectively to the target anion. Recently, we reported about the use of RGP in the removal of Pb(II) from seawater (4) and ligand exchange elimination of phosphate from river water by Zr(IV)-loaded RGP (Zr-RGP) (3). Zirconium (IV) strongly binds to phosphate resulting in sparingly soluble salt $Zr(HPO_4)_2$, which is an often used inorganic ion-exchanger (5). Because of the strong binding of Zr(IV) with phosphate, complete elution of Zr(IV) from Zr-RGP, even with nitric acid solutions at high concentration levels (1–6 mol/L), was difficult to achieve (3). Thus, Zr-RGP is expected to be one of ideal ligand exchangers for Zr(IV) strongly retained by RGP.

Since the beginning of the 1980s, many workers have shown that Zr(IV) exhibits high affinity to both arsenate and arsenite. Yoshida et al. studied the uptake of arsenite by Zr(IV)-loaded weak-acid cation-exchange and iminodiacetic acid resins. They found that Zr(IV) is more suitable for the ligand exchange adsorption of arsenite than Fe(III) (6). However, even the highest breakthrough capacity of these ligand exchangers for arsenite was less than 0.02 mmol/mL of wet resin. In 1984, Kobayashi, Sugai, and Imajyo reported the adsorption of arsenate and arsenite by a complex of zirconium oxide hydrate with active carbon that can indirectly adsorb arsenite through catalytic oxidation of arsenite into arsenate (7). However, the mechanism of the active carbon–catalyzed oxidation of arsenite with dissolved oxygen was not fully explained, and its catalytic activity changed with solution conditions (7,8). Thereafter, similar zirconium oxide–based adsorbents for arsenate and arsenite have been reported by several authors (9–12), and

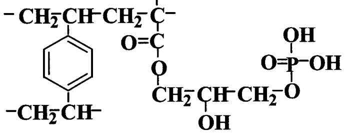

Figure 1. Chemical structure of phosphoric acid resin RGP.

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most recently, Suzuki et al. reported the behavior of a porous resin (Amberlite XAD-7) loaded with crystalline zirconium oxide in the adsorption of arsenate and arsenite (12). This adsorbent gives large equilibrium capacities of 1.5 and 1.2 mmol/g for arsenite and arsenate, respectively, whereas its breakthrough capacity for arsenate was approximately 0.20 mmol/mL of wet resin. However, its column-mode adsorption of arsenite was not described (12).

As mentioned above, Zr(IV) has high affinity to both arsenate and arsenite. Therefore, the behavior of Zr-RGP in adsorption of arsenate and arsenite is of interest to researchers. In removal of trace amounts of arsenic in a huge volume of water, a breakthrough capacity is much more important than an equilibrium capacity because the latter is usually measured by the batch method at high concentration levels of arsenic. Taking into account the fact that large equilibrium capacities do not always mean large breakthrough capacities, we examined the detailed behavior of Zr-RGP in adsorption of arsenate by the columnar method. In addition, we also tested the Zr-RGP in the simultaneous adsorption of both arsenate and arsenite.

EXPERIMENTAL

Materials

The resin RGP was prepared by the functionalization of macroreticular poly(glycidyl methacrylate-co-divinylbenzene) of 32–60 mesh according to the reported method (1). The degree of cross-linking of RGP used in this work was completed with 10% (mol) of divinylbenzene. The resulting RGP was conditioned according to a conventional column method (13). In the final stage of conditioning, RGP was changed into the hydrogen form and washed with water until the washing became acid free. After air-drying, the RGP was dried at 40°C for 24 hours. Properties of RGP in the hydrogen form are given in Table 1.

A 0.01-mol/L Zr(IV) solution was prepared from zirconium (IV) nitrate of reagent grade and its pH was adjusted with nitric acid. A stock solution of arsenate was prepared by dissolving analytical grade disodium hydrogen arsenate ($Na_2HAsO_47H_2O$) in water and that of arsenite was prepared by dissolving arsenic trioxide in water and filtering the solution with a Millipore filter (0.45 μm)

Table 1. Properties of the Phosphoric Acid Resin (RGP) in the Hydrogen Form

Specific Surface Area (m^2/g)	Acid Capacity (mEq/g)	Salt Splitting Capacity (mEq/g)	Phosphorus Content (mmol/g)
29.2	6.98	2.77	3.75

The degree of cross-linking is 10% (mol) of divinylbenzene.

because of its low solubility. Exact concentrations of arsenic in these stock solutions were determined by means of an inductively coupled plasma atomic emission spectrometer (ICP-AES) with a standard solution of arsenic (Wako Chemical Co, Tokyo, Japan). Feeding solutions were prepared by the dilution of stock solutions, and their pH was adjusted with nitric acid or sodium hydroxide. Weakly alkaline feeding solutions were freshly prepared and used immediately. Hereafter, arsenate and arsenite are rendered as As(V) and As(III), respectively.

Loading of Zr(IV) onto RGP

Zirconium (IV) was loaded onto RGP by means of the columnar method. The wet settled RGP in the hydrogen form (1 mL) was applied to a glass column (i.d. 0.7 cm) and the 0.01 mol/L Zr(IV) solution ($\mathrm{pH} \approx 0.8$) was fed into the column at an hourly rate of 6 mL (6 h^{-1} in space velocity) until the concentration of Zr(IV) in the column effluent was nearly equal to that in the feeding solution. The column was then washed with water until the washing became acid free.

To calculate the amount of Zr(IV) loaded, all column effluents, including the washing, were collected on a fraction collector, and the Zr(IV) concentration in each fraction was determined by means of ICP-AES. By subtracting the sum of the Zr(IV) amount in all fractions from the amount of Zr(IV) supplied to the column, the content of Zr(IV) in RGP was calculated to be 0.22 mmol/mL of wet resin (0.72 mmol/g). The column thus prepared was used in further experiments. All results were obtained without change of column RGP.

Column-Mode Adsorption and Elution of As(V)

Feeding solutions containing As(V) were fed into an Zr-RGP column at an hourly rate of 10 mL (10 h^{-1} in space velocity) and then were washed with water (4 mL). The As(V) adsorbed was eluted with 0.4 mol/L aqueous sodium hydroxide unless otherwise noted. All column effluents, including washings, were collected on a fraction collector. Concentrations of arsenic in all fractions were measured by means of ICP-AES, and zirconium fractions were also measured in some experiments. In this work, breakthrough points are designated as feeding-solution bed volumes, where C/C_o equals up to 0.05, where C_o and C represent concentrations of arsenic in the feeding solution and in the column effluent, respectively.

Regeneration of the Zr-RGP Column

After the elution of As(V), a dilute nitric acid solution (pH 3) was fed into the column to neutralize hydroxide ions bound to the loaded Zr(IV), and the column was washed with water until the column effluent became acid free. A new

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0.01 mol/L Zr(IV) solution (pH ≈ 0.8, 4 mL) was fed into the column to compensate for Zr(IV) lost during adsorption, elution, and washing operations (the insignificant loss of Zr(IV) will be subsequently addressed). After the column was washed with water until the washing was found to be acid free, the next adsorption experiment was started. Uptake of As(V) gradually increased with each repetition of up to 8–10 cycles of adsorption-elution-regeneration as observed by Chanda, O'Driscoll, and Rempel in the adsorption of As(III) and As(V) by Fe(III)-loaded iminodiacetic acid resin (14).

Removal of As(V) from River- and Seawater

River water was taken from the Shirakawa River (Kumamoto-shi, Japan) and seawater was provided by the Aitsu Marine Biological Station of Kumamoto University (Amakusa, Kumamoto, Japan). Both were filtered with a Millipore filter (0.45 μm) to remove suspending particles. Main metal ions and anions in river and the seawater samples were determined by means of ICP-AES and ion chromatography, respectively. To the filtered river water and seawater samples, As(V) was added, and resulting solutions were used as feeding solutions after adjustment of pH with nitric acid or sodium hydroxide.

RESULTS AND DISCUSSION

Stability of Zr(IV) in Zr-RGP

In effective ligand exchange, metal ions loaded on resins should not be eluted with solutions containing target anions. As already reported (3), little Zr(IV) dissolved into the column effluent, even when phosphate solutions at various pH values were fed into Zr-RGP column. In this work, concentrations of Zr(IV) in column effluents were also monitored in adsorption, elution, and washing operations. Figure 2 shows an example of concentration profiles of Zr(IV) of the column effluent when 2.5 mmol As(V) solution at pH 2.0 was fed into Zr-RGP column. The concentration of Zr(IV) in the column effluent was less than 2.2 μmol and the loss of Zr(IV) from the Zr-RGP column was negligible. When a dilute nitric acid of pH 2 or a sodium hydroxide solution (0.4 mol/L) without As(V) was fed into the column, no Zr was detected in column effluents. This result indicates that Zr(IV) is very strongly retained by RGP.

Properties of P(V) and As(V) are similar to each other. Why does no Zr(IV) Zr-RGP elute with As(V)? The density of phosphate groups in RGP is as high as 3.75 mmol/g (Table 1) but those of As(V) in feeding solutions were less than 10 mmol/L in this work. Probably, As(V) and phosphate (3), present at less than 10 mmol/L in solutions, cannot effectively take up Zr(IV) from Zr-RGP, which is maintained by relatively high amounts of phosphate groups in the RGP phase.

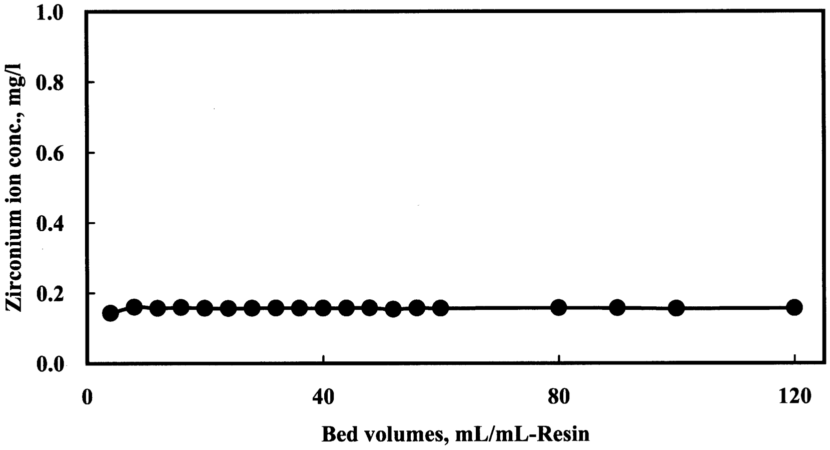

Figure 2. Typical profiles of Zr(IV) dissolution from the Zr(IV)-loaded RGP column during the adsorption of As(V). Flow rate 10 h^{-1} in space velocity, As(V) 2.5 mmol/L, pH 2.0. Volume of each feeding solution supplied to the column was 120 bed volumes.

Effect of pH on Adsorption of As(V)

Figure 3 shows the effect of the pH of the feeding solution on adsorption of As(V). Table 2 summarizes uptake of As(V) in up to 100 bed volumes, breakthrough points, and breakthrough capacities. When the pH of the feeding solution

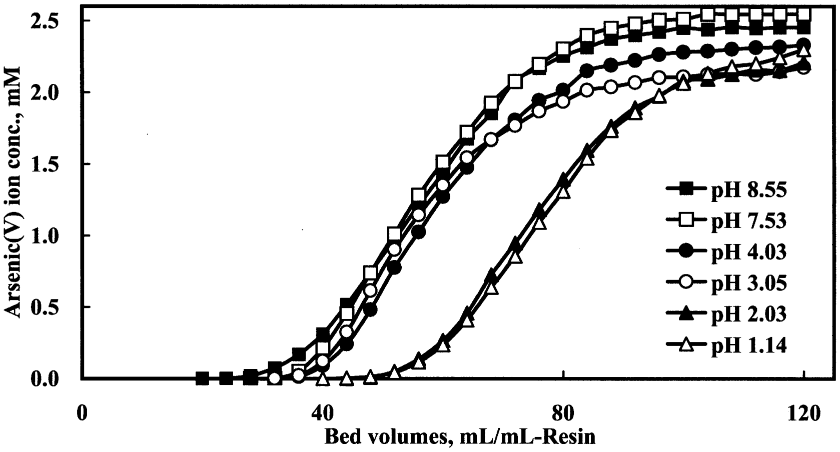

Figure 3. Effect of pH on As(V) adsorption. Flow rate 10 h^{-1} in space velocity, As(V) 2.5 mmol/L. Volume of each feeding solution supplied to the column was 120 bed volumes.

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Table 2. Dependence of As(V) Uptake on pH of Feeding Solutions

pH	As(V) Uptake (mmol/mL)	Breakthrough Point (Bed Volumes)	Breakthrough Capacity (mmol/mL)
1.14	0.184(0.613)*	56	0.132(0.440)*
2.03	0.176(0.587)	54	0.120(0.400)
3.05	0.142(0.473)	40	0.093(0.310)
4.03	0.144(0.480)	42	0.094(0.313)
7.53	0.142(0.473)	38	0.090(0.300)
8.55	0.136(0.453)	34	0.077(0.257)

Uptake up to 100 bed volumes of feeding solutions.
Breakthrough points were designated as bed volumes where $C/C_o = 0.05$.
* Uptake or breakthrough capacity in mmol/g.

is less than 2, the uptake of As(V) up to 120 bed volumes is approximately 0.19 mmol/mL of wet resin, and the breakthrough points are approximately 55 bed volumes. Between pH 2 and 3, breakthrough points decreased markedly, whereas the decrease in the breakthrough points between pH 3 and 7.5 was not so remarkable. The breakthrough capacity at pH 1.14 was 0.132 mmol/mL of wet resin, and that at pH 8.55 decreased to 0.077 mmol/mL of wet resin. Because the dry weight of 1 mL of wet settled RGP was 0.3 g, breakthrough capacities per unit weight of dry resin are approximately 0.25–0.44 mmol/g.

We would like to compare the capacities of Zr-RGP for uptake of As(V) with those of other ligand exchangers. Lanthanide-impregnated silica gel had been shown to have the maximum adsorption capacity of 0.1–0.13 mmol/g (15,16). Fe(III)-loaded iminodiacetic acid resin exhibited poor kinetic adsorption of As(V) even at a flow rate of 2 h^{-1} in space velocity, and its breakthrough capacity was approximately 0.085 mmol/g (14). However, Kobayashi, Sugai, and Imajyo reported that the breakthrough capacity of zirconium oxide hydrate–activated carbon complex was 0.17 mmol/g (7) although the Zr(IV) content was as high as 1.6 mmol/g, which is 20% (wt), as ZrO_2. Similar situations are also observed in Amberlite XAD-7 loaded with crystalline zirconium oxide (12). Its breakthrough capacity for As(V) was 0.24 mmol/mL of wet resin at an hourly flow rate of 6 bed volumes, whereas its Zr(IV) content was as large as 2.1–2.6 mmol/g (12). Therefore, it seems that Zr(IV) in zirconium oxide crystalline cannot contribute to rapid uptake of As(III) and As(V) because diffusion of species in the crystalline solid is very slow. However, these zirconium oxide–based adsorbents possess breakthrough capacities for As(V) that are comparable to those of Zr-RGP.

Because the As(V) uptake increased gradually, up to 8–10 cycles of the column operation as already mentioned, we can say that the content of Zr(IV) in Zr-RGP is probably increased by the repeated adsorption-elution-regeneration operations. Because of extreme difficulty in the quantitative elution of Zr(IV) from

Zr-RGP, even with solutions of a strong acid solution at high concentrations of 1–6 mol/L, we are now developing a suitable analytical method for determination of Zr(IV) through the use of Zr-RGP.

In the adsorption of As(V) with Zr-RGP, breakthrough capacities decreased with an increase in pH. This result is due the high affinity of Zr(IV) for the hydroxide ion. Similar pH dependency was reported in batch-mode uptake of As(V) by zirconium oxide–based adsorbents (6,7,12). Other adsorbents, such as Fe(III)-loaded iminodiacetic acid resin and lanthanide-loaded silica gel, exhibited the optimum pH range for maximum uptake of As(V): pH 6.4–7.5 for the lanthanide-loaded silica gel (16) and pH 2 for Fe(III)-loaded iminodiacetic acid (14). The decrease in uptake of As(V) by these ligand exchangers at the low pH regions is ascribable to the elution of loaded metal ions with hydrogen ions.

Effect of As(V) Concentration

Figure 4 shows the adsorption of As(V) from feeding solutions containing As(V) at different concentrations. In the case of the 5 mmol/L As(V) feeding solution, the arsenic concentration in the column effluent was nearly equal to that in the feeding solution at 120 bed volumes. Thus, the equilibrium capacity at pH 3 was calculated to be 0.18 mmol/mL of wet resin. The breakthrough point of As(V) was located at approximately 20 bed volumes and the breakthrough capacity (0.10 mmol/mL of wet resin) was approximately one-half the equilibrium capacity.

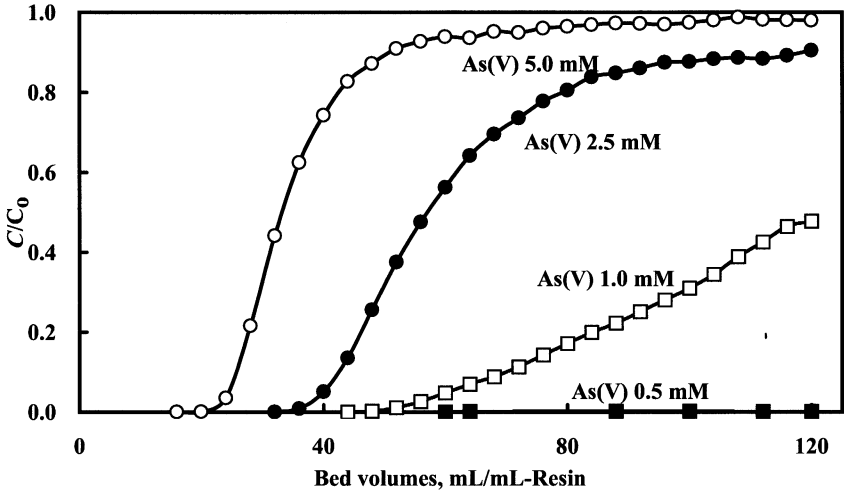

Figure 4. Effect of As(V) concentration. Flow rate 10 h^{-1} in space velocity, pH 3.0. Volume of each feeding solution supplied to the column was 120 bed volumes.

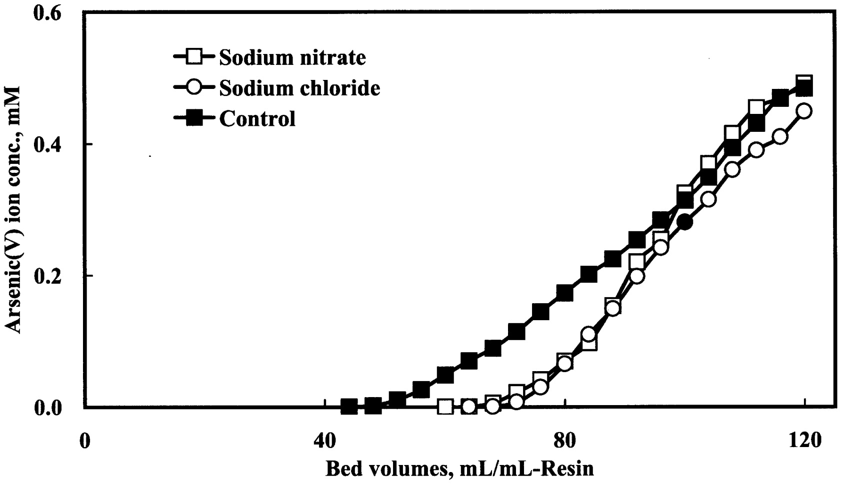

Figure 5. Effect of electrolytes. Flow rate 10 h^{-1} in space velocity, pH 3.0. As(V) 2.5 mmol/L. Concentration of sodium chloride or sodium nitrate was 0.01 mol/L. Volume of each feeding solution supplied to the column was 120 bed volumes.

Although breakthrough points increased with a decrease in the concentration of As(V) in the feeding solutions, they were not constant. This inconsistency is due to factors, such as concentration of As(V) and total ionic strength of feeding solutions, that affect breakthrough capacities in ligand exchange adsorption of anionic species even when pH is held constant. The breakthrough point is beyond 120 bed volumes when 0.5 mmol/L As(V) solution was supplied to the column and indicates that the rate of As(V) uptake by Zr-RGP is satisfactory even in sub-mmol/L levels of As(V).

Effect of Electrolytes

Figure 5 shows the effects of sodium chloride and sodium nitrate on the adsorption of As(V). Clearly, the anions from these electrolytes in large excess did not interfere with the adsorption of As(V). On the contrary, these electrolytes enhanced the breakthrough point of As(V). A similar enhancing effect of electrolytes was already reported in the ligand exchange adsorption of As(V) (14), phosphate (3), and fluoride (17). The results in Fig. 5 show that the binding of Zr(IV) with chloride and nitrate is very weak. This enhancing effect of electrolytes is difficult to explain by the usual anion exchange mechanism, which leads to the decrease in uptake of the target anion in the presence of competitive anions in large excess. In a similar work on the uptake of fluoride by Zr-RGP, column effluents during adsorption of fluoride were analyzed by means of ion chromatography (18). Although Zr-RGP was regenerated with a dilute nitric acid solution, no nitrate was detected in the column effluents. This means that no exchangeable anion exists in the Zr-RGP column because Zr-RGP is thoroughly washed with water after its regeneration. This situation is quite different from the cases with zirconium oxide–based adsorbents, in which chloride from hydrochloric acid used in the regeneration of the adsorbent binds to zirconium oxide such that $(ZrO_2Zr(OH)_2(H_2O)_n)Cl_2$ and the chloride bound to Zr(IV) is replaced by As(V) (7). In the case with Zr-RGP, in the pH region under which this work was conducted, most As(V) existed as $H_2AsO_4^-$ and/or $HAsO_4^{2-}$. A possible adsorption mechanism for the adsorption of As(V) is estimated as follows:

$$(RO{-\!\!-}PO_3)_2Zr(IV) + Na^+ + H_2AsO_4^- \Leftrightarrow (RO{-\!\!-}PO_3)_2Zr(IV)(H_2AsO_4)Na$$
$$(RO{-\!\!-}PO_3)_2Zr(IV) + 2Na^+ + HAsO_4^{2-} \Leftrightarrow (RO{-\!\!-}PO_3)_2Zr(IV)(HAsO_4)Na_2$$

The water molecules coordinated with Zr(IV) were omitted because the detailed structure for the coordination sphere of Zr(IV) in Zr-RGP is not yet known. However, the proposed mechanism qualitatively explains the enhancing effect of electrolytes that do not have a high affinity to Zr(IV) on the adsorption of As(V). An increased sodium ion concentration would be represented as a shift in the equilibrium to the right-hand side of the equations.

Adsorption of Both As(III) and As(V)

Suzuki et al. found that Amberlite XAD-7 loaded with crystalline zirconium oxide exhibits equilibrium capacity for As(III) that is higher than that for As(V) and also reported that the optimum pH range for the adsorption of As(III) is from 8 to 10 (12). Thus, the pH of the feeding solutions containing both As(III) and As(V) was adjusted to 8. Figure 6 shows the results. Breakthrough capacity for arsenic was approximately 0.10 mmol/mL of wet resin (0.33 mmol/g). From this breakthrough capacity, we estimated that the breakthrough point for a 0.5 mmol/L arsenic solution would be approximately 200 bed volumes. Indeed, no breakthrough point was observed within the tested range (120 bed volumes) and arsenic was not detected in the column effluent. Thus, Zr-RGP exhibits satisfactory adsorption rates for both As(III) and As(V) even at an hourly flow rate of 10 h^{-1}, which is nearly twice that adopted in the study of Suzuki et al. (12).

Elution of Adsorbed As(V) and As(III)

Sodium hydroxide is frequently used to elute both As(III) and As(V) adsorbed on zirconium oxide–based adsorbents (7,12) and Fe(III)-loaded chelating resins (14). Then, elution of both As(V) and As(V) was tested with sodium

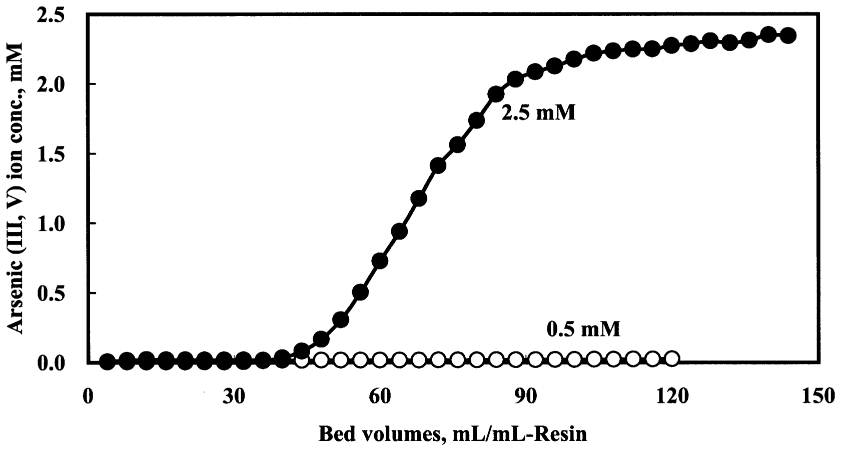

Figure 6. Simultaneous removal of arsenic (III) and arsenic (V) at different concentrations. Flow rate 10 h^{-1} in space velocity, pH 8.0. Volumes of feeding solutions containing 2.5 mmol/L and 0.5 mmol/L total arsenic were 144 and 120 bed volumes, respectively. The molar ratio of As(V)/As(III) was unity.

hydroxide solutions. Figure 7 shows 2 examples of elution profiles of arsenic. Arsenic (V) adsorbed on Zr-RGP was quantitatively eluted with a 0.4 mol/L hydroxide solution within 24 bed volumes at a flow rate of 6 h^{-1} in space velocity. Figure 7 also shows the simultaneous elution of both As(III) and As(V), which

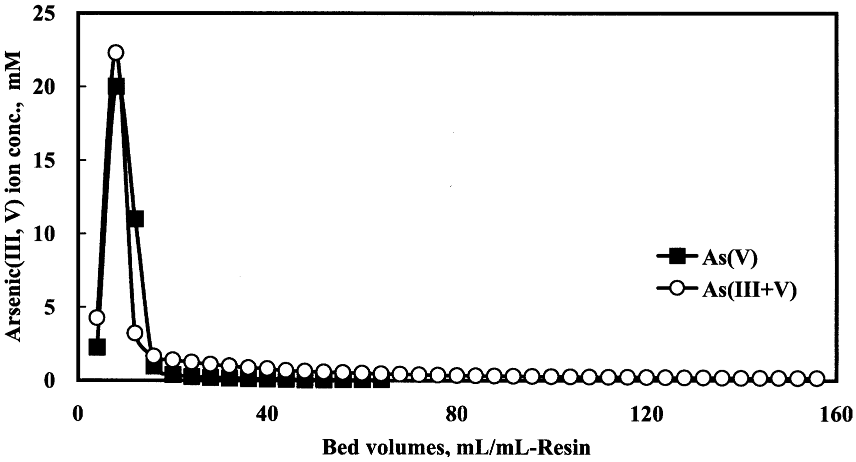

Figure 7. Typical elution curves of As(III) and As(V) with sodium hydroxide solutions. Flow rate 6 h^{-1} in space velocity. Concentrations of NaOH were 0.4 mol/L (64 bed volumes) for elution of arsenic (V) and 0.7 mol/L (156 bed volumes) for elution of arsenic (III) and arsenic (V).

were adsorbed from a feeding solution containing both As(III) and As(V). In this case, the elution of As(III) is clearly more difficult than that of As(V). Although a higher concentration of sodium hydroxide (0.7 mol/L) was used with As(III), a long tail was observed. This means that As(III) more strongly binds to Zr-RGP than does As(V), as in the case of Amberlite XAD-7 impregnated with zirconium oxide (12). This result shows that oxidation of As(III) to As(V) does not significantly occur in uptake of As(III) by Zr-RGP, which is a different situation from the case of the zirconium oxide–active carbon complex (7). Suzuki et al. did not refer to the oxidation of As(III) to As(V) during the adsorption of As(III) by Amberlite XAD-7 impregnated with zirconium oxide (12).

During processes of adsorption and elution of both As(III) and As(V), the concentration of Zr in all column effluents was less than 4.4 μmol, and no turbid effluent was observed throughout. These results exhibit high durability of Zr-RGP.

Arsenic (V) Removal from River Water and Seawater

To test the possible effectiveness of Zr-RGP in the elimination of As(V) from naturally occurring water, As(V)-spiked river water and seawater samples were used as feeding solutions. Table 3 gives the concentrations of main cations and anions in the river water and seawater before being spiked with As(V). Figure 8 shows the adsorption of the spiked As(V). In the adsorption of the spiked As(V) from both river water and seawater, uptake of As(V) at pH 3.0 is greater than that at pH 8.4 or pH 6.6, as anticipated from the results on the effect of pH as previously noted. As can be seen from the data in Table 3, the total concentration

Table 3. Main Cations and Anions in River Water and Seawater Samples

			Cations			
Sample	pH (mmol/L)	Ca^{2+} (mmol/L)	Mg^{2+} (mmol/L)	K^{+} (mmol/L)	Na^{+} (mol/L)	Sum
River water	7.86	0.65	0.51	0.18	0.68	2.01
Seawater	7.87	9.97	48.8	22.7	463	544

		Anions		
	Cl^{-} (mmol/L)	NO_3^{-} (mmol/L)	SO_4^{2-} (mmol/L)	Sum (mmol/L)
River water	0.44	0.02	0.97	1.43
Seawater	521	—	25	546

— Nitrate could not be detected.

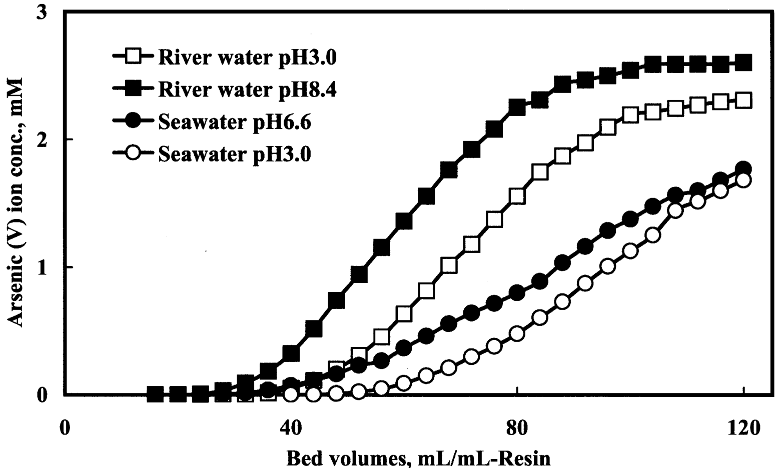

Figure 8. Application of the Zr(IV)-loaded RGP for As(V) removal from river water and seawater. Flow rate 10 h^{-1} in space velocity, As(V) 2.5 mmol/L. Volume of each feeding solution supplied to the column was 120 bed volumes.

of electrolytes in the seawater sample is much higher than that of the river water. If naturally occurring anions strongly interfere with the adsorption of As(V), uptake of As(V) from seawater should be less than that from river water. However, Fig. 8 shows that uptake of As(V) from seawater is greater than that from river water. The enhancing effect of electrolytes on the As(V) adsorption is also observed.

Zr-RGP is expected to be effective in the removal of As(V) from natural water, including seawater. In addition, we obtained evidence that Zr-RGP has a higher affinity to As(III) than to As(V). Works on detailed behavior of Zr-RGP in the adsorption of As(III) are now in progress.

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