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Removal of Hazardous Anions from Aqueous Solutions by La(III)- and Y(III)-Impregnated Alumina

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ABSTRACT

New adsorbents, La(III)- and Y(III)-impregnated alumina, were prepared for the removal of hazardous anions from aqueous solutions. A commercially available alumina was impregnated with La(III) or Y(III) ions by the adsorption process. The change in the surface charge due to the impregnation was measured by acid/base titration. The adsorption rate and the capacity of the alumina for La(III) and Y(III) ions were determined. The adsorption characteristics of the La(III)- and Y(III)-impregnated alumina and the original alumina for fluoride, phosphate, arsenate and selenite ions were analyzed under various conditions. The pH effect, dose effect, and kinetics were studied. The removal selectivity by the impregnated alumina was in the order fluoride > phosphate > arsenate > selenite. The impregnated alumina has been successfully applied for the removal of hazardous anions from synthetic and high-tech industrial wastewaters.

INTRODUCTION

Since detrimental effects of hazardous anions in the environment have attracted increasing attention during the last decade, development of an effective method to remove these anions from industrial wastewater has

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been required by relevant industries. A large amount of fluorine compounds has been consumed, particularly in high-tech industries such as those producing semiconductors and integrated circuits where hydrogen fluoride and ammonium fluoride have been used in the etching and cleaning processes (1, 2). Phosphate has been known to cause eutrophication, especially in enclosed water areas such as lakes and inland seas (3). Numerous measures have been taken, but the eutrophication problem has not yet been solved completely. It is well known that arsenic and selenium are highly toxic elements (4–8) which exist predominantly in the form of oxyanions in the aquatic environment (9, 10). Recently more stringent regulations for drinking water and natural water have been set forth in Japan for arsenic and selenium, <0.01 mg/L for both (11, 12).

To remove the above-mentioned hazardous anions from aqueous solutions, various methods have been proposed. Precipitation and coagulation processes which convert them into insoluble forms have been conventionally used (13–16), but these conventional methods have sometimes not been successful to meet the regulations due to their requirement of a large amount of chemicals and the limitation of solubility of the final product. Among the potential methods, the adsorption process seems most promising to reduce the concentration of hazardous anions to the minimum level without increasing salt concentration in the effluent. Recently, rare earth elements such as lanthanum and yttrium have been found to be effective to adsorb these hazardous anions (17–20). Rare earth elements have a specific affinity for fluoride, phosphate, arsenate, and selenite ions. In the present study the authors have tried to impregnate a porous alumina with lanthanum and yttrium ions with the aim of developing new adsorbents. The prepared materials were evaluated as adsorbents for the removal of fluoride, phosphate, arsenate [As(V)] and selenite [Se(IV)] ions. Arsenite [As(III)] and selenate [Se(VI)] ions, and other predominant forms of arsenic and selenium, were not examined in this study since the adsorption process is known to be ineffective to remove these species (21–23).

EXPERIMENTAL

Materials

A 100-mmol/L stock solution of fluoride, phosphate, arsenate, or selenite ion was prepared separately by dissolving analytical-grade sodium fluoride, potassium dihydrogen phosphate, dibasic sodium arsenate, or sodium selenite in deionized water, respectively. These solutions were further diluted to suitable concentrations. Sodium and potassium salts of chloride, bromide, iodide, nitrate, and sulfate were dissolved in deionized water and used for the interference study. A 100-mmol/L La(III) and Y(III) chloride solution was prepared in deionized water. A commercially available alumina (Aluminum oxide 90 active, neutral, activity I; Merck #1077.1000) was used as a support. The particle size and the surface area of the alumina were 0.063–0.200 mm and 96.1 m^2/g, respectively. Reagents of $La(OH)_3$ and $Y(OH)_3$ were used for the comparison of surface charge, for which the surface areas were found to be 45.1 and 74.7 m^2/g, respectively.

Preparation of La(III)- and Y(III)-Impregnated Alumina

Alumina (100 g) was added to 500 mL of a 100-mmol/L La(III) or Y(III) solution at an initial pH of 7.5. The mixture was stirred for 48 hours at room temperature and filtered through a 0.45-μm membrane filter. The recovered particles were washed repeatedly with deionized water until the conductivity of the rinse water became <3 μS/cm.

Characterization of La(III)- and Y(III)-Impregnated Alumina

The surface charge of La(III)- and Y(III)-impregnated alumina, alumina, La(III) hydroxide, and Y(III) hydroxide was measured by the acid/base titration method (24). 2.5 g of the material was added to 1 L of a 50-mmol/L sodium perchlorate solution from which dissolved air was removed by bubbling with nitrogen. Sodium perchlorate was used to adjust the ionic strength. The resulting mixture was shaken at 20°C for 5 days in a sealed bottle. The acid/base titration was carried out on both 50 mL of the filtrate and suspension while bubbling with nitrogen. The acid titration was made with 50 mmol/L $HClO_4$ solution and the base titration with 50 mmol/L NaOH solution. The net titration curve was obtained by taking the difference in the titration volumes (Δv) for the suspension and the filtrate at given pH values. The surface charge was calculated from the net titration curves using the following equation:

$$\sigma_0 = \Delta v MF/SAV \qquad (1)$$

where σ_0 is the surface charge (C/cm^2), Δv is the difference in titration volumes (mL), M is the molarity of the acid and base titrant (mmol/L), F is Faraday's constant (96,500 C/mol), S is the surface area of the material (m^2/g), A is the amount of the material used for titration (g/L), and V is the volume of filtrate and suspension used for titration (mL). The surface area of the materials was measured by the BET method (Micromeritics; Flow Sorb II 2300).

Removal of Fluoride, Phosphate, Arsenate, and Selenite Ions with La(III)- and Y(III)-Impregnated Alumina

A solution containing a suitable amount of anion was taken separately in a polycarbonate centrifuge tube. The pH was adjusted by the addition of 50 mmol/L acid or base solution, and the volume was made up to 25 mL with deionized water. 0.5 g of the La(III)- or Y(III)-impregnated alumina was added to each tube. The resulting mixture was shaken at 20°C for 24 hours. The mixture was centrifuged and the supernatant was analyzed for lanthanum, yttrium, phosphate, arsenate, and selenite by the ICP method, and fluoride, chloride, bromide, iodide, nitrate, and sulfate ions were determined by ion chromatography (Shimadzu Corp., LC-6A). Removal of these anions by the original alumina was also measured in the same way as above.

The adsorption capacity for these anions was measured at 20°C by varying the amount of adsorbents at an initial concentration of 2 mmol/L fluoride and 1 mmol/L phosphate, arsenate, and selenite ions. A kinetic study of the removal of these anions was carried out at different intervals of shaking time according to the above procedure.

RESULTS AND DISCUSSION

Impregnation of Alumina with La(III) and Y(III) Ions

Adsorption of La(III) and Y(III) Ions on Alumina

The interaction of the alumina with La(III) and Y(III) ions was studied. The concentrations of La(III) and Y(III) ions remaining in the liquid phase were measured in the 3 to 12 pH range. The results are shown in Fig. 1. The results in the absence of the alumina show that the hydrolysis of both La(III) and Y(III) ions took place at pH > 8. On the other hand, the concentrations of La(III) and Y(III) ions decreased abruptly in the 5 to 6 pH range in the presence of the alumina. Such a decrease of La(III) and Y(III) concentrations in the 6 to 8 pH range can be attributed to the adsorption of these ions on the surface of the alumina.

The surface characteristics of the materials were analyzed. The surface areas of the original alumina, La(III)-, and Y(III)-impregnated alumina were 96.1, 93.9, and 93.3 m^2/g, respectively. Therefore, the impregnation of La(III) and Y(III) ions slightly decreases the surface area of the support. The surface charge of the several materials were determined by the acid/base titration method and the results are shown in Fig. 2. The original alumina showed its zero point charge, pH_{zpc}, at 6.7, being positively charged in the lower pH range and negatively charged in the higher pH range. The two materials obtained from the interaction of the alumina

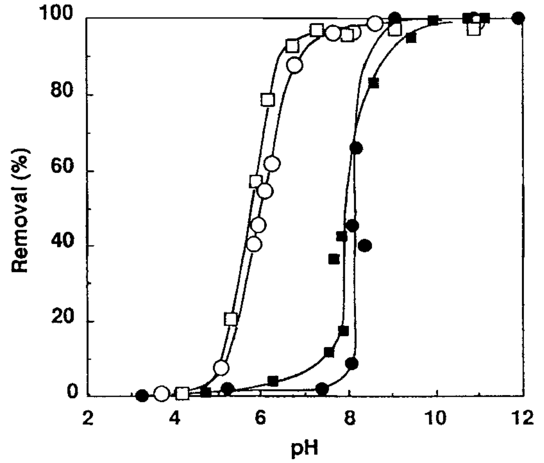

FIG. 1 Adsorption of La(III) and Y(III) ions on the alumina as a function of pH. (●) 0.1 M La(III); (○) 0.1 M La(III) + alumina; (■) 0.1 M Y(III); (□) 0.1 M Y(III) + alumina.

with La(III) and Y(III) ions showed different patterns of surface charge, giving a pH_{zpc} at 7.4 in both cases. In addition, the surface charges of La(III) and Y(III) hydroxide were measured since hydroxide was anticipated to form on the surface of the alumina by interaction with La(III) and

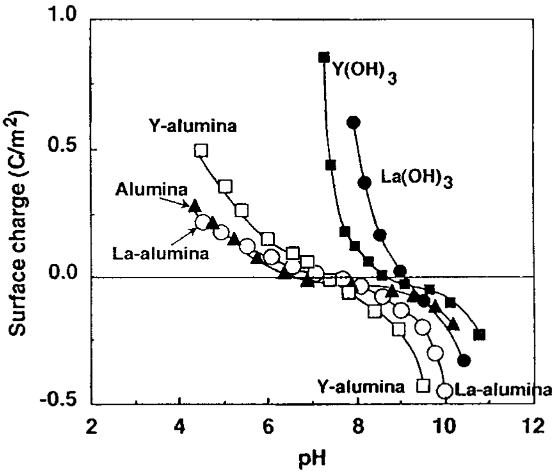

FIG. 2 Surface charge of La(III)-, Y(III)-impregnated alumina, alumina, $La(OH)_3$, and $Y(OH)_3$ as a function of pH. Ionic strength: 5×10^{-2} M $NaClO_4$.

Y(III) ions, but both $La(OH)_3$ and $Y(OH)_3$ showed substantially different patterns of surface charge, giving pH_{zpc} at 9.1 and 8.7, respectively. The above two facts indicate that La(III) and Y(III) ions are impregnated on the surface of the alumina by the adsorption process rather than by surface precipitation. This chemical adsorption reaction can be expressed as

$$>\!Al\!-\!|\!-\!OH + La(OH)_2^+ \rightarrow\ >\!Al\!-\!|\!-\!O\!-\!La(OH)_2 + H^+ \quad (2)$$

$$>\!Al\!-\!|\!-\!OH + Y(OH)_2^+ \rightarrow\ >\!Al\!-\!|\!-\!O\!-\!Y(OH)_2 + H^+ \quad (3)$$

According to the pK values of complexes of La(III) and Y(III) with the OH^- ion, $La(OH)_2^+$ and $Y(OH)_2^+$ species are predominant at around 8 and 7 pH, respectively (25). Such a concept can further be supported by our previous publication where La(III) ion was impregnated on the surface of porous silica gel (19).

Kinetics of the Adsorption of La(III) and Y(III) Ions on Alumina

The rates of adsorption of La(III) and Y(III) ions on alumina were determined at pH 7.5 where hydrolysis does not take place. The results, shown in Fig. 3, were analyzed by using the following Lagergren's equation (26):

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

where q_e and q (both in mmol/g) are the amounts of La(III) and Y(III)

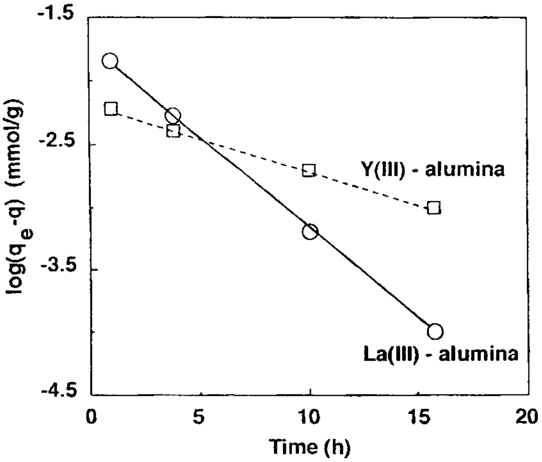

FIG. 3 Kinetic data for the adsorption of La(III) and Y(III) ions on the alumina.

ions adsorbed by the alumina at equilibrium and at time t (minutes), respectively. Regression analysis gave the following equations:

$$\log(q_e - q) = -1.73 - 0.152t \qquad (5)$$

$$\log(q_e - q) = -2.17 - 0.053t \qquad (6)$$

for La(III) and Y(III) with correlation coefficients of 0.997 and 0.999, respectively. Therefore it can be concluded that the adsorption follows a first-order reaction. K_{ad} for La(III) and Y(III) is calculated to be 0.349 and 0.123 min^{-1}, respectively.

Adsorption Capacity

The isotherms for the adsorption of La(III) and Y(III) ions by the alumina were measured at 20, 30, and 40°C by changing the initial concentrations of these metal ions. The results were analyzed by using the following rearranged Langmuir isotherm:

$$C_e/q_e = C_e/Q^\circ + 1/Q^\circ b \qquad (7)$$

where C_e (mmol/L) is the equilibrium concentration and Q° (mmol/g) and b (L/mmol) are the Langmuir constants representing the adsorption capacity and adsorption energy, respectively. The results, shown in Fig. 4, indicate

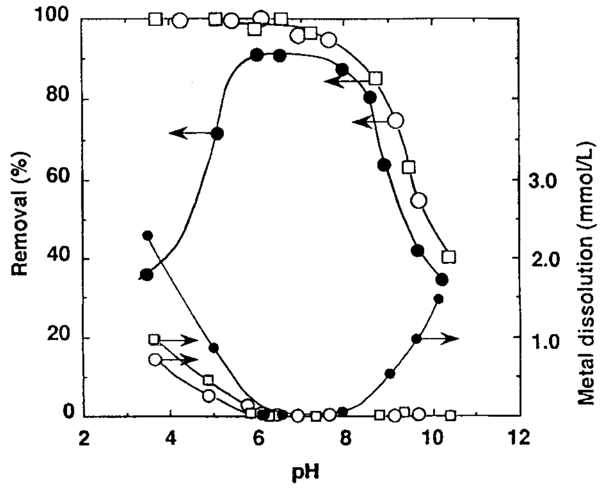

FIG. 4 Removal of fluoride ions by La(III)-, Y(III)-impregnated alumina, and alumina, and dissolution of metal as a function of pH. (○) La(III)-alumina; (□) Y(III)-alumina; (●) alumina.

that the adsorption of La(III) and Y(III) ions by the alumina follows the Langmuir adsorption model at 20, 30, and 40°C. The Langmuir constants are summarized in Table 1. The adsorption capacities for La(III) and Y(III) were 0.234–0.256 and 0.422–0.487 mmol/g, respectively. A larger amount of Y(III) ions can be impregnated in the alumina.

Removal of Fluoride, Phosphate, Arsenate, and Selenite Ions with La(III)- and Y(III)-Impregnated Alumina

Effects of pH

It has been known that alumina itself adsorbs fluoride, phosphate, arsenate, and selenite ions (27–32). Evaluation was made on the performance of prepared La(III)- and Y(III)-impregnated alumina for the removal of these anions. Figure 4 illustrates the removal of fluoride ions, as an example, by the impregnated and original alumina as a function of pH, where dissolution of La(III), Y(III), and Al(III) from these materials is also shown. The highest fluoride removal was obtained by the alumina in the 6 to 8 pH range. The dissolution of Al(III) became significant at pH <6 and >8, indicating consumption of the material. The La(III)- and Y(III)-impregnated alumina exhibited a similar pattern of fluoride removal with a higher percent removal than the original alumina, with maximum removal in the pH <8 range. Dissolution of La(III) and Y(III) from the material was negligible in the alkaline pH region. Although the dissolution of La(III) and Y(III) was less significant than that of Al(III), it became appreciable at pH < 6. Therefore, the optimum pH range is 6 to 8 for impregnated alumina where the highest fluoride removal can be attained and the dissolution of the materials can be suppressed. The pH effects on the removal of phosphate, arsenate, and selenite ions were similar to those of fluoride.

TABLE 1
Langmuir Constants for the Adsorption of La(III) and Y(III) Ions on the Alumina

	Langmuir constants			
	La(III)-alumina		Y(III)-alumina	
Temperature (°C)	Q° (mmol/g)	b (L/mmol)	Q° (mmol/g)	b (L/mmol)
20	0.256	19.7	0.487	19.9
30	0.234	10.9	0.505	20.8
40	0.237	11.2	0.422	16.7

Effects of Dose

To emphasize the effects of the impregnation of alumina to remove anions, the doses of the adsorbents were changed at a fixed anion concentration. Figure 5 shows the percent removal of anions as a function of dose of the adsorbents. It is clearly shown that both La(III)- and Y(III)-impregnated alumina gave a higher percent removal for fluoride and phosphate ions, especially at low doses of the adsorbent. There was no significant difference in the removal of arsenate and selenite ions between the impregnated alumina and the original alumina.

Adsorption Capacity

The adsorption capacities of impregnated alumina and original alumina for fluoride, phosphate, arsenate, and selenite ions were determined by changing the dose. The results were analyzed by using the Langmuir

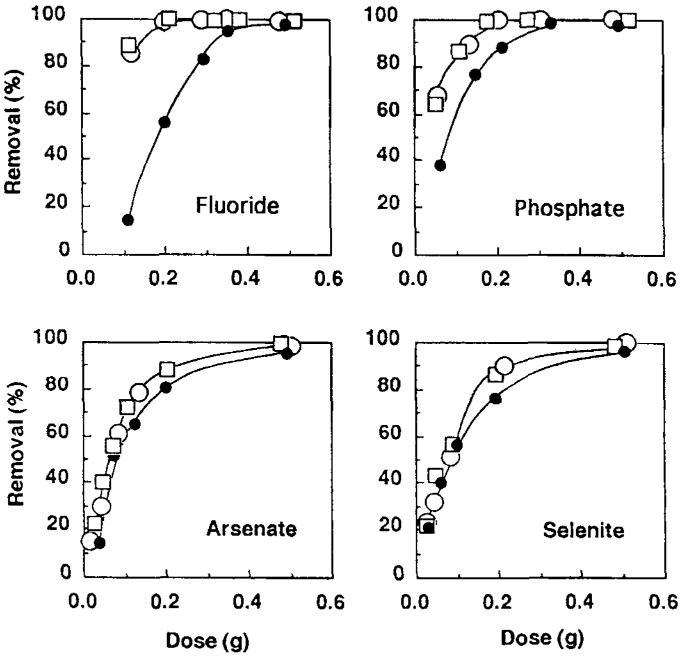

FIG. 5 Removal of fluoride, phosphate, arsenate, and selenite ions by La(III)- and Y(III)-impregnated alumina as a function of dose. (○) La(III)-alumina; (□) Y(III)-alumina; (●) alumina.

model, Eq. (7). The values of the adsorption capacity, Q°, are incorporated in Table 2 and compared with those of the original alumina. The original alumina showed similar adsorption capacities for each anion in the 0.174 to 0.186 mmol/g range. The impregnated alumina showed a significant increase of the adsorption capacity for fluoride and phosphate ions. On the other hand, impregnation of alumina with La(III) and Y(III) did not significantly increase the adsorption capacity for arsenate and selenite ions.

The possible mechanism of the removal of these anions, especially fluoride and phosphate ions, by impregnated alumina can be explained by an ion-exchange process between these anions and the hydroxide group on the surface of the material as shown in Eqs. (8) and (9):

$$>\mathrm{Al}{-}|{-}\mathrm{O}{-}\mathrm{La(OH)_2} + 2\mathrm{A}^- \rightarrow\ >\mathrm{Al}{-}|{-}\mathrm{O}{-}\mathrm{LaA_2} + 2\mathrm{OH}^- \quad (8)$$

$$>\mathrm{Al}{-}|{-}\mathrm{O}{-}\mathrm{Y(OH)_2} + 2\mathrm{A}^- \rightarrow\ >\mathrm{Al}{-}|{-}\mathrm{O}{-}\mathrm{YA_2} + 2\mathrm{OH}^- \quad (9)$$

where A^- is a monovalent anion. At $\mathrm{pH} < \mathrm{pH_{zpc}}$, the surface of the impregnated alumina is positively charged. Consequently, coulombic attraction can readily take place in conjunction with specific chemical adsorption due to an exchange reaction between anions and hydroxide ion. When $\mathrm{pH} > \mathrm{pH_{zpc}}$, the surface is negative, and the adsorption is suppressed due to coulombic repulsion.

Kinetic Study

Our kinetic study shows that removal of these anions follows a first-order reaction. The rate constants were calculated by using Eq. (4) and are shown in Table 2. The La(III)- and Y(III)-impregnated alumina gave lower K_{ad} values for the adsorption of fluoride, arsenate, and selenite ions, indicating that these anions were removed faster than by the original

TABLE 2
The Rate Constants and Adsorption Capacities of the Impregnated and Original Alumina for the Removal of Fluoride, Phosphate, Arsenate, and Selenite Ions

	K_{ad} (min^{-1})				Adsorption capacity (mmol/g)			
Material	F	P	As(V)	Se(IV)	F	P	As(V)	Se(IV)
La(III)-alumina	0.046	0.084	0.024	0.062	0.328	0.332	0.172	0.194
Y(III)-alumina	0.036	0.071	0.018	0.092	0.368	0.319	0.193	0.182
Alumina	0.080	0.056	0.135	0.150	0.174	0.184	0.182	0.186

alumina. On the other hand, the impregnated alumina gave higher K_{ad} values for the adsorption of phosphate ion, indicating slower adsorption.

Selectivity and Interference

One of the important characteristics of an adsorbent is selectivity to remove only the pollutant of interest among various chemical species. In many cases, industrial wastewaters contain various kinds of ions which it is not necessary to remove. The performance of La(III)- and Y(III)-impregnated alumina was evaluated with two types of synthetic wastewaters and a real wastewater which contained various anions. The results are summarized in Table 3. In the two synthetic wastewaters, chloride, bromide, iodide, nitrate, and sulfate ions were not adsorbed by both La(III)- and Y(III)-impregnated alumina, and significant amounts of fluoride, phosphate, arsenate, and selenite ions were removed. Therefore, the impregnated alumina is highly selective for the adsorption of fluoride, phosphate, arsenate, and selenite ions and free from interference by common anions. In the second synthetic wastewater with a higher anion concentration, fluoride ion was still removed at a high percent removal, but the removal of the other anions decreased significantly due to the limitation of adsorption capacity and probably due to interference from the high

TABLE 3

Removal of Fluoride, Phosphate, Arsenate, and Selenite Ions with the Lanthanum- and Yttrium-Impregnated Alumina

	Equilibrium concentration		% Removal	
Composition of wastewater	La-alumina	Y-alumina	La-alumina	Y-alumina
Synthetic wastewater:				
F, P, As(V), Se(IV): 2.0 mM Cl, Br, I, NO_3, SO_4: 5.0 mM	F: 0.001 P: 0.010 As: 0.020 Se: 0.100	F: 0.001 P: 0.012 As: 0.060 Se: 0.130	F: 99.95 P: 99.5 As: 99.0 Se: 95.0	F: 99.95 P: 99.4 As: 97.0 Se: 93.5
F, P, As(V), Se(IV): 5.0 mM Cl, Br, I, NO_3, SO_4: 8.0 mM	F: 0.001 P: 0.100 As: 0.980 Se: 1.502	F: 0.001 P: 0.130 As: 1.000 Se: 1.120	F: 99.98 P: 98.0 As: 80.4 Se: 69.6	F: 99.98 P: 97.4 As: 80.0 Se: 77.6
Real wastewater:[a]				
F: 2.19; P: 0.09; Cl: 1.05 mM NO_3: 7.49; SO_4: 0.87 mM	F: 0.007	F: 0.002	F: 99.7	F: 99.91

[a] From a semiconductor manufacturing industry.

concentration of common anions. Based on the equilibrium concentrations, the order of the adsorption selectivity of these two impregnated alumina is fluoride > phosphate > arsenate > selenite. The impregnated alumina were also effective for removing fluoride ion from the real wastewater which contained various types of pollutants.

CONCLUSION

A commercially available alumina was impregnated with La(III) or Y(III) ion by the adsorption process. The rates of adsorption of La(III) and Y(III) ions followed a first-order reaction. According to the Langmuir model, the adsorption capacities of the alumina for La(III) and Y(III) were 0.234–0.256 and 0.422–0.487 mmol/g at 20–40°C, respectively. The surface charges of La(III)- and Y(III)-impregnated alumina, alumina, La(III) hydroxide, and Y(III) hydroxide were measured by the acid/base titration method and their pH_{zpc}'s were 6.7, 7.4, 7.4, 9.1, and 8.7, respectively.

The adsorption characteristics of La(III)- and Y(III)-impregnated alumina and the original alumina for fluoride, phosphate, arsenate, and selenite ions were analyzed under various conditions. These anions were removed efficiently in the 6 to 8 pH range. The dissolution of La(III) and Y(III) from the impregnated alumina became appreciable at $pH < 6$. The original alumina showed 0.174 to 0.186 mmol/g of adsorption capacities for each anion at neutral pH. The impregnated alumina showed a significant increase in the adsorption capacity for fluoride and phosphate ions. The impregnation did not enhance the adsorption capacity for arsenate and selenite ions. The adsorption rate followed a first-order reaction. The removal selectivity by the impregnated alumina was found to be in the order fluoride > phosphate > arsenate > selenite. Impregnated alumina has been successfully applied for the removal of hazardous anions from synthetic and high-tech industrial wastewaters.

REFERENCES

1. K. Yabe, "Wastewater Treatment System for Semiconductor Factories," *Kuki Seijo*, *31*(4), 218–224 (1993).
2. D. G. Gilles and R. C. Loehr, "Waste Generation and Minimization in Semiconductor Industry," *J. Environ. Eng.*, *120*(1), 72–86 (1994).
3. Editorial Board of Japan Society on Water Environment, "Feature: Technology for Control of Eutrophication in Marine Area," *J. Jpn. Soc. Water Environ.*, *18*(7), 538–558 (1995).
4. F. W. Pontius, K. G. Brown, and C. J. Chen, "Health Implications of Arsenic in Drinking Water," *J. Am. Water Works Assoc.*, *86*(9), 52–63 (1994).

5. C. O. Abernathy and E. V. Ohanian, "Health Effects of Inorganic Arsenic in Drinking Water," *Proc. Water Qual. Technol. Conf. 1993*, (Pt. 2), 1969–1975 (1994).
6. T. Tsuda et al., "Inorganic Arsenic: A Dangerous Enigma for Mankind," *Appl. Organomet. Chem.*, *6*, 309–322 (1992).
7. E. Marshall, "Selenium Poisons Refuge, California Politics," *Science*, *229*, 144–146 (1985).
8. A. D. Lemly, "Guidelines for Evaluating Selenium Data from Aquatic Monitoring and Assessment Studies," *Environ. Monitor. Assess.*, *28*(1), 83–100 (1993).
9. D. A. Clifford and Z. Zhang, "Arsenic Chemistry and Speciation," *Proc. Water Qual. Technol. Conf.*, *1993*, (Pt. 2), 1955–1968 (1994).
10. R. H. Neal, G. Sposito, K. M. Holtzclaw, and S. J. Traina, "Selenite Adsorption on Alluvial Soils: I. Soil Composition and pH Effects," *Soil Sci. Soc. Am., J.*, *51*, 1161–1165 (1987).
11. *Ministerial Ordinance on Drinking Water Standards*, Ordinance No. 69 of the Ministry of Health and Welfare, Japan, December 21, 1992.
12. *Environmental Quality Standards for Water Pollution*, Notification No. 16 of the Environment Agency, Japan, March 8, 1993.
13. R. L. Culp and H. A. Stoltenberg, "Fluoride Reduction at La Crosse, Kan.," *J. Am. Water Works Assoc.*, *50*(3), 423–431 (1958).
14. P. H. Hsu, "Comparison of Iron(III) and Aluminum in Precipitation of Phosphate from Solution," *Water Res.*, *10*, 903–907 (1976).
15. T. J. Sorg, "Treatment Technology to Meet the Interim Primary Drinking Water Regulations for Inorganics," *J. Am. Water Works Assoc.*, *70*, 105–112 (1978).
16. T. J. Sorg and G. S. Logsdon, "Treatment Technology to Meet the Interim Primary Drinking Water Regulations for Inorganics: Part 2," *Ibid.*, *70*, 379–393 (1978).
17. J. Nomura, H. Imai, and T. Miyake, "Removal of Fluoride Ion from Wastewater by a Hydrous Cerium Oxide Adsorbent," *ACS Symp. Ser.*, *422*, 157–172 (1990).
18. S. Tokunaga, M. J. Haron, S. A. Wasay, K. F. Wong, K. Laosangthum, and A. Uchiumi, "Removal of Fluoride Ion from Aqueous Solution by Multivalent Metal Compounds," *Int. J. Environ. Stud.*, *48*, 17–28 (1995).
19. S. A. Wasay, M. J. Haron, and S. Tokunaga, "Adsorption of Fluoride, Phosphate and Arsenate Ions on Lanthanum-Impregnated Silica Gel," *Water Environ. Res.*, In Press.
20. M. J. Haron, W. M. Z. Wan Yunus, S. A. Wasay, A. Uchiumi, and S. Tokunaga, "Sorption of Fluoride Ions from Aqueous Solution by Yttrium-Loaded Poly(Hydroxamic Acid) Resin," *Int. J. Environ. Stud.*, *48*, 245–255 (1995).
21. M. L. Pierce and C. B. Moore, "Adsorption of Arsenite and Arsenate on Amorphous Iron Hydroxide," *Water Res.*, *16*, 1247–1253 (1982).
22. K. Kamegawa, H. Yoshida, and S. Arita, "Adsorption of Arsenic(III) and Arsenic(V) on Activated Carbon," *Nippon Kagaku Kaishi*, (10), 1365–1370 (1979).
23. P. Zhang and D. L. Sparks, "Kinetics of Selenate and Selenite Adsorption/Desorption at the Goethite/Water Interface," *Environ. Sci. Technol.*, *24* (12), 1848–1856 (1990).
24. S. W. Park and C. P. Huang, "The Surface Acidity of Hydrous CdS(s)," *J. Colloid Interface Sci.*, *117*(2), 431–441 (1986).
25. R. M. Smith and A. E. Martell, *Critical Stability Constants—Inorganic Complexes*, Vol. 4, p. 2, Plenum Press, New York, 1976.
26. S. Lagergren, *Bil. K. Svenska Venteskapsakad. Handl.*, *24* (1898), as cited by H. C. Trivedi, V. M. Patel, and R. D. Patel, "Adsorption of Cellulose Triacetate on Calcium Silicate," *Eur. Polym. J.*, *9*, 525–531 (1973).
27. Y. C. Wu, "Activated Alumina Removes Fluoride Ions from Water," *Water Sewage Works*, *125*, 76–82 (1978).

28. N. Gangoli and G. Thodos, "Phosphate Adsorption Studies," *J. Water Pollut. Control Fed.*, *45*, 842–849 (1973).
29. C. P. Huang, "Removal of Phosphate by Powdered Aluminum Oxide Adsorption," *Ibid.*, *49*, 1811–1817 (1977).
30. J. H. Gulledge and J. T. O'Connor, "Removal of Arsenic(V) from Water by Adsorption on Aluminum and Ferric Hydroxides," *J. Am. Water Works Assoc.*, *65*, 548–552 (1973).
31. S. K. Gupta and K. Y. Chen, "Arsenic Removal by Adsorption," *Ibid.*, *50*, 493–506 (1978).
32. R. R. Trussell, A. R. Trussell, and P. Kreft, *Selenium Removal from Groundwater Using Activated Alumina*, EPA Publication 600/2-80-153, Cincinnati, August 1980.

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