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Possibility of Fluoride Removal Using Volcano Ash Soils and Bone Charcoal as Adsorbents

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Abstract: In some regions of Southern Asia and Western Africa, it has been observed that groundwater used as a drinking water source contains more than 1.5 mg/L of fluoride, which is the standard value approved by the World Health Organization. Adsorption is one of the methods widely used for easy fluoride removal; therefore, adsorption and flow tests were conducted using volcano ash soils and bone charcoal as adsorbents. Bone charcoal has already been examined as a fluoride adsorbent; however, volcano ash soils have not been studied previously. The following results were obtained using adsorption tests: the fluoride adsorption coefficients of bone charcoal, Kanuma volcano ash soil, and Akadama volcano ash soil were 1,500, 700, and 490 mL/g, respectively. As the first step for use in a water purification plant, flow tests were conducted assuming a fixed bed adsorption tower. For a water flow rate of 2.0 m/day, 1.8 mg/L of fluoride was reduced to 0.2 and 1.0 mg/L using bone charcoal and Akadama soil, respectively. However, for the water flow rates of 3.0 or 4.0 m/day, 1.8 mg/L of fluoride was reduced to 0.2 and 1.7 mg/L using bone charcoal and Akadama soil, respectively. Therefore, it was observed that the adsorption of the Akadama soil is strongly affected by the water flow rate.

Key words: fluoride, groundwater, volcano ash soil, bone charcoal, adsorption

1. Introduction

In Japan, few groundwater sources contain over 0.8 mg F/L of fluoride, which is the tap water quality standard; groundwater containing higher fluoride content than that specified by the standard is not used for tap water. Therefore, the amount of the fluoride in the water is not particularly important. However, in Southern Asia and Western Africa, for technical and economic reasons, groundwater containing a high concentration of fluoride is often used for tap water, which may have a harmful effect on the bones of human beings.

The following methods are typically used for fluoride removal:

1) Calcium fluoride method: calcium hydroxide is added to produce calcium fluoride and flocculants are then added to precipitate calcium fluoride.
2) Hydroxide coprecipitation method: aluminum salt is added to produce aluminum hydroxide; then fluorine ions are adsorbed onto and coprecipitated with aluminum hydroxide.
3) Adsorption method: fluorine ions are adsorbed on a selective ion exchange resin made from rare-earth hydroxides.

Recently, the adsorption method using bone charcoal and the separation method using a nanofiltration (NF) membrane have been studied for fluoride removal.

A study of the adsorption method performed using bone charcoal focused on chicken bones due to the absence of religious issues involved while working with chicken bones. A previous study investigated the removal of fluoride in groundwater in Sri Lanka and examined the manufacturing process of bone charcoal based on the heating temperature and investigated the

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batch adsorption process [1, 2]. Another study focused on the separation method using nanofiltration membrane and examined the removal of fluoride in groundwater in Thailand using an NF membrane [3]. Fluoride adsorption in the soils has been studied [4, 5], and this study aims to examine fluoride adsorption of soils with different physicochemical characteristics.

2. Objectives and Methods

2.1 Objectives

Following the previous studies mentioned above, adsorption tests of fluoride removal using bone charcoal and soils were examined and flow tests were performed to examine the feasibility of the use of these materials as the adsorbents in an applied fluoride removal system.

2.2 Adsorption Tests

2.2.1 Materials

The following materials are used as adsorbents in this study.

1) Bone charcoal

Bone charcoal used in this study was obtained from Wako Chemicals Inc., Japan. Calcium phosphate is the main component of the bone charcoal.

2) Volcano ash soil

Volcano ash soils have been studied for adsorption of phosphorus and organics for wastewater and ambient water treatment. Akadama soil and Kanuma soil in Tochigi Prefecture, Japan, are porous and contain iron and aluminum. The specific surface area, iron content, and aluminum content of Akadama soil are 34 cm^2/g, 6.4-7.8%, and 8.1-12.9%, respectively. The specific surface area, iron content, and aluminum content of Kanuma soil are 112 cm^2/g, 2.1-2.3%, and 13.4-16.8%, respectively. Iron and aluminum are some of the high-performance elements for coagulation and adsorption. Akadama and Kanuma soils are used for home gardening and are available in do-it-yourself stores in Japan.

3) Sludge from water purification plant used for iron reduction bacteria (henceforth called “sludge”)

The sludge from the water purification plant using the iron reduction bacteria purification method has a high iron content. The sludge was produced by the water purification plant in Hyogo Prefecture, Japan, and was dried and levigated. The iron content of the sludge is 41%.

4) Filter sand for water purification plant (henceforth called “sand”)

The sand used for the sand filtration in water purification plants was used as the control material in our tests. The sand used in this study is obtained from Kagoshima Prefecture, Japan.

2.2.2 Methods

The test water used in this study was the commercially available natural mineral water produced in France with a fluoride standard solution (Wako Chemicals Inc., Japan) mixed into the water; this was used to simulate the groundwater. 0-10 mg/L of fluoride with 50 mL of water was placed into each plastic bottle, along with 0-5.0 grams, respectively, of materials. All bottles were sealed and stored for one week at 25°C in the dark. Then, the water was filtrated using a paper filter (5C, Advantec Inc., Japan), and the fluoride concentration was analyzed via an ion chromatograph (Dionex ICS-1100 with AS23 column). The pH of the water is equal to 7.2. A schematic of the adsorption test is shown in Fig. 1.

It was assumed that fluoride was sufficiently adsorbed to the materials after one week. Hence, the fluoride concentration after one week was considered to be the equilibrium concentration.

The mass of fluoride initially present in the water in the plastic bottle is calculated as

$$M_0 = C_0 \times V \qquad (1)$$

where

M_0 = Mass of fluoride initially present in the water (mg)

C_0 = Initial fluoride concentration (mg/L)

V = Mass of water in the plastic bottle (L).

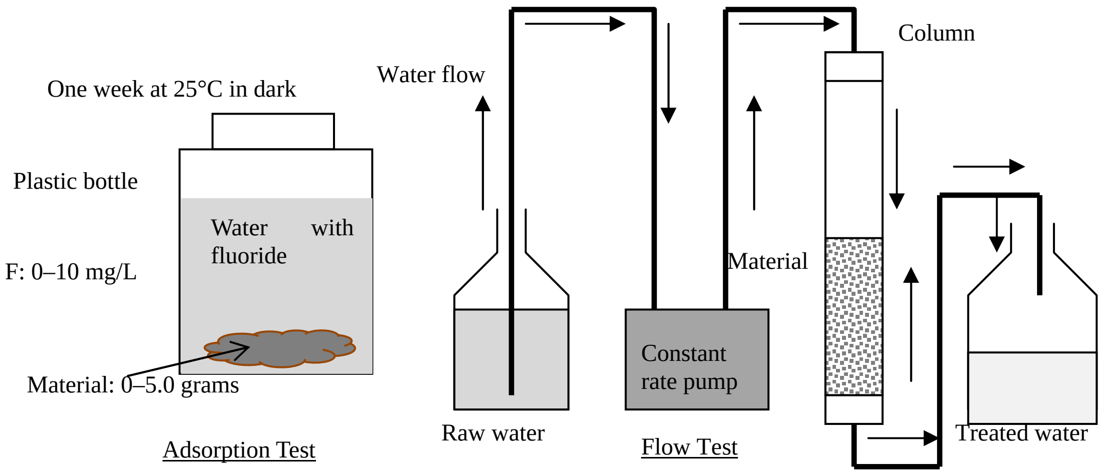

Fig. 1 Schematic of the adsorption test and the flow test.

The mass of fluoride in the water at adsorption equilibrium in the plastic bottle is calculated as

$$M = C \times V \tag{2}$$

where

M = Mass of fluoride in the water at adsorption equilibrium (mg)

C = Equilibrium concentration of fluoride (mg/L).

The mass of the fluoride adsorbed on the material is calculated by

$$M_c = M_0 - M \tag{3}$$

where

M_c = Mass of the fluoride adsorbed on the material (mg).

Adsorption quantity of the material is calculated as

$$q_c = \frac{M_c}{m} = \frac{V}{m}(C_0 - C) \tag{4}$$

where

q_c = Adsorption amount (mg/g)

m = Mass of the material (g).

The adsorption is evaluated using the Freundlich adsorption isotherm with q_c and C.

The adsorption coefficient is calculated as

$$K_d = \frac{q_c}{C} = \frac{V}{m}\left(\frac{C_0 - C}{C}\right) = \frac{V}{m}\left(\frac{C_0}{C} - 1\right) \tag{5}$$

where

K_d = Adsorption coefficient (L/g)

Fluoride removal fraction is calculated as

$$R = \frac{C_0 - C}{C_0} \times 100 \tag{6}$$

where

R = Fluoride removal fraction (%).

2.3 Flow Tests

The adsorption tower process is more suitable for the continuous water purification necessary to obtain stable water resources than the batch adsorption process. Therefore, flow tests using laboratory size column were performed for bone charcoal and volcano ash soil. Akadama soil was selected from volcano ash soils for the test. A schematic of the flow test is shown in Fig. 1. The column is made from glass, has a diameter and height of 25 and 200 mm, respectively, and was manufactured by BIO-RAD Inc. The height of each material is 100 mm, and its volume is approximately 49 cm^3. The raw water used for the test was groundwater that exceeds the Japanese environmental standard for fluoride, with a fluoride concentration of approximately 1.8 mg/L. Its pH was 7.3. Raw water was flowed in the sealed column using the constant rate pump at the rates of 2.0, 3.0, or 4.0 m/day; then the water flowed out of the column as treated water. Fluoride concentrations were analyzed following the same procedure as in the adsorption test.

3. Results and Discussion

3.1 Adsorption Tests

The results of the adsorption tests are expressed in the Freundlich adsorption isotherm and are shown in Fig. 2. The adsorption isotherm is given by

$$q_c = kC^n \quad (7)$$

and can also written as

$$\log q_c = \log k + n \log C \quad (8)$$

where

k = constant; If k is larger, adsorption ability is higher.

n = constant; If n is smaller, fluoride can be similarly adsorbed in a large range of concentrations.

Table 1 shows the values of the k, n, chi-square value, and average coefficient of adsorption for each Freundlich adsorption isotherm.

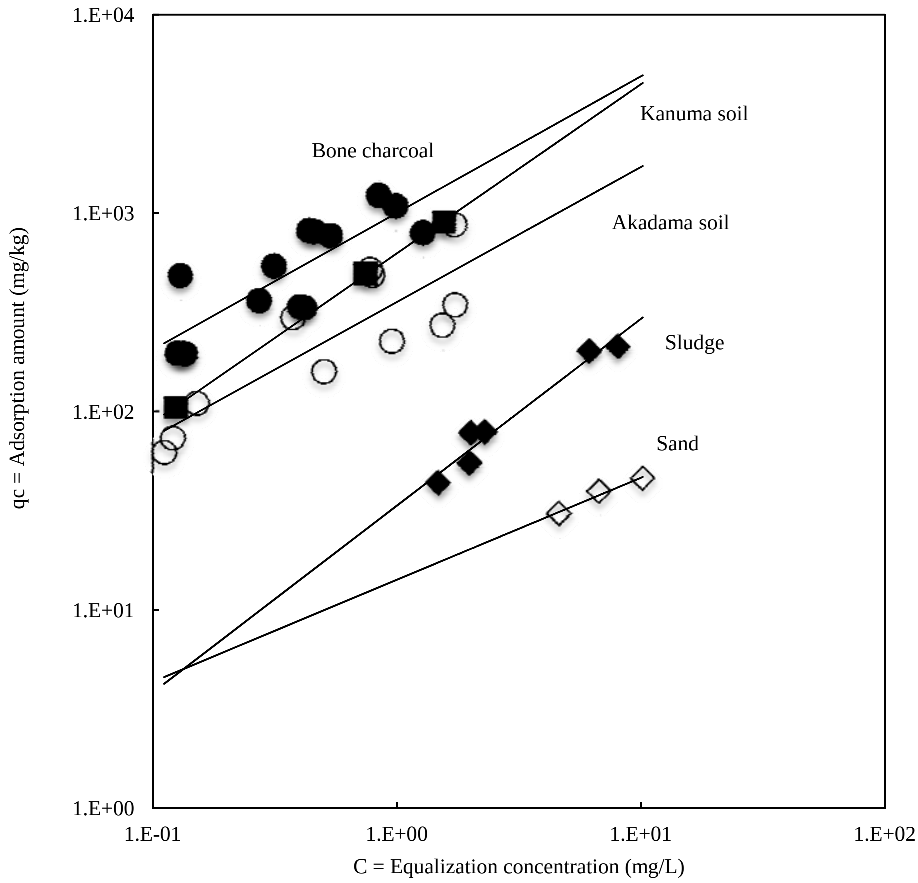

Fig. 2 Adsorption isotherm graph of adsorption tests.

Table 1 Results of adsorption tests.

Material	Constant *k*	Constant *n*	Chi-square value	Average coefficient of adsorption (L/g)
Bone charcoal	1,000	0.69	0.69	1.50
Akadama soil	360	0.68	0.72	0.49
Kanuma soil	630	0.85	0.99	0.70
Sludge	33	0.94	0.96	0.03
Sand	14	0.51	0.97	0.06

The adsorption coefficient of bone charcoal was the highest among the five materials examined in the adsorption test. The Kanuma and Akadama volcano ash soils also showed high fluoride adsorption coefficients. Thus, it was found that these materials may be used as the fluoride adsorbents.

3.2 Flow Tests

Fig. 3 shows the flow test results. For the water flow rate of 2.0 m/day, bone charcoal could remove approximately 90% of fluoride and Akadama soil could remove approximately 50% of fluoride. For water flow rates of 3.0 and 4.0 m/day, the amount of removed fluoride was the same for bone charcoal as that removed at the flow rate of 2.0 m/day. On the contrary, for these flow rates, the Akadama soil was almost entirely incapable of removing fluoride. Thus, it was found that the performance of the Akadama soil is affected by the water flow rate.

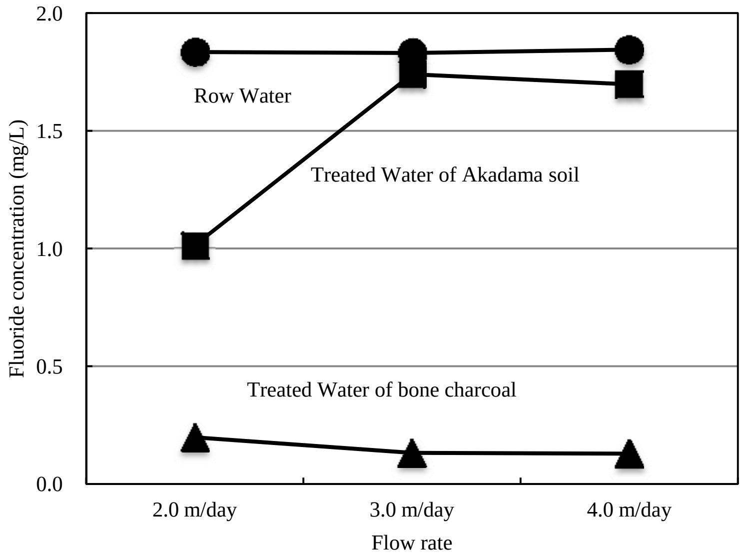

Fig. 3 Flow tests results.

4. Conclusion

According to the results of both tests, bone charcoal was the best fluoride adsorbent in groundwater. Bone charcoal can be produced from food waste and is, therefore, easily available everywhere. However, religious issues must be taken into account. On the contrary, the obtained adsorption test results for Akadama soil were not as poor as expected. However, in the case of high water flow rate, Akadama soil was almost totally incapable of removing the fluoride. Therefore, the water flow rate must be considered if Akadama soil is used as the fluoride adsorbent.

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References

[1] A. Motoyama, T. Kawamaki, S. K. Weragoda, K. Okugawa, Y. Serikawa, T. Masamoto and S. Takamatsu, Fluoride contamination in drinking water in Sri Lanka and fluoride removal by using chicken bone char, *Journal of Japan Society of Civil Engineers* 68 (2012) (7) III_517-III_523.

[2] H. Miyazaki, S. Nagasawa, A. Motoyama and T. Kawakami, Chemical reaction of fluoride removal by chicken bone char, *Journal of Japan Society of Civil Engineers* 70 (2014) (7) III_527-III_534.

[3] Y. Matsui, T. Takeda, S. Takizawa, A. Wongrueng and S. Wattanachira, Evaluation of nanofiltration process for fluoride removal from groundwater in the Ching Mai Basin, *Journal of Japan Society of Civil Engineers* 62 (2006) (4) 403-414.

[4] H. Sugita, T. Komai, S. Okita, S. Tokunaga and I. Matsunaga, Analysis on adsorption behavior of fluorine on

clay minerals using Freundlich's adsroption equation, *Journal of the Mining and Materials Processing Institute of Japan* 121 (2005) (9) 416-422.
[5] H. Sugita, T. Komai and Y. Imoto, Fundamental investigation on soil adsorption of fluorine and boron, *Journal of the Mining and Materials Processing Institute of Japan* 127 (2011) (4-5) 202-212.