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Original

Super Volume Reduction of ^{137}Cs-contaminated Solid Waste by Ion Chromatographic Elimination of Cs from ^{137}Cs-enriched Dust Generated by Pyroprocessing Decontamination

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Summary

Using an ion chromatography with copper ferrocyanide-loaded silica gel as an adsorbent, Cs was selectively removed from the simulated rinsing solution of Cs-enriched dust generated by pyroprocessing of solid waste that had been contaminated with radioactive Cs due to the Fukushima Daiichi Nuclear Power Plant accident. The spent adsorbent was stably solidified by using metakaolin as a hydraulic solidifier. The weight of the final radioactive waste was estimated to be less than 1/1000 of the original one.

Key Words: ^{137}Cs decontamination, Super volume reduction, Pyroprocessing, Ion chromatography, Copper ferrocyanide, Geopolymer

1. Introduction

The Fukushima Daiichi Nuclear Power Plant accident induced by the East Japan earthquake and tsunami of March 11, 2011 released approximately 1.8×10^{16} Bq of ^{134}Cs and 1.5×10^{16} Bq of ^{137}Cs[1-4], which contaminated plants, soils, houses and so on around the reactor site. Although ^{134}Cs has a short half-life of about two years and is mostly decayed into nonradioactive Ba, ^{137}Cs has a longer half-life of about 30 years and has been a target nucleus for environmental decontamination[5, 6]. The decontamination work after the accident generated an enormous amount of ^{137}Cs-enriched wastes. Removal and combustion of contaminated plants and other wastes generated ^{137}Cs-enriched municipal solid waste incinerator fly ash, since Cs is volatile at high temperature[7]. Removal of contaminated top soil from residential area and farmland collected a huge amount of ^{137}Cs-enriched soil. The total amounts of ^{137}Cs-enriched soil and ash is estimated to 22×10^6 m^3 [8]. A significant part of them are stored in temporary storage spaces and are waiting for final disposal.

Although landfill is the simplest way for the final disposal, it is quite difficult to find land space that is wide enough for the disposal. Volume reduction of contaminated waste is therefore a feasible way for the final disposal. The volume reduction causes the reduction of final storage space, which makes safer management of the waste. Pyroprocessing is a powerful method of ^{137}Cs decontamination and volume reduction of solid wastes. It does not choose the object of decontamination, the operation of the system is simple, and the decontamination factor (the ratio of initial specific radioactivity to final specific radioactivity resulting from a decontamination process) is high. Pyroprocessing decontamination uses calcium chloride and calcium carbonate as Cs expeller and evaporation agent, and heat treatment over 1300°C. Alkali metals in soil and ash are generally captured in silicate and/or aluminosilicate as bound ions. The calcium ions added react with these compounds at high temperature to expel alkali ions. Alkali

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*Corresponding author: E-mail: tsuneki@eng.hokudai.ac.jp ions then react with chloride ions to convert to alkali chlorides. Since the vapor pressures of alkali chlorides at high temperature increase in the order of NaCl < KCl < CsCl, CsCl is preferentially evaporated from the waste. The evaporated alkali chlorides are cooled and completely trapped in bag filter systems to give ^{137}Cs-enriched dust.

There are two methods of pyroprocessing, depending on the type of furnace used for decontamination. Melt processing uses melting furnaces and produces decontaminated slag[9, 10] which can be used as aggregate in construction. Calcination processing uses rotary kilns and produces decontaminated clinker which can be used as a main component of Portland cement[11]. The decontamination factor and the volume reduction factor (the volume ratio of the initial waste to final one to be disposed as radioactive waste, and is ^{137}Cs-enriched dust in the present case) are estimated to be 26 and 13 for melt processing, and are more than 1000 and 20 for calcination processing, respectively[12]. Although pyroprocessing gives high volume reduction factor, they are not enough for such a huge amount of contaminated waste. Moreover, since ^{137}Cs in the dust is highly water soluble, addition of Cs immobilizing agent is necessary for the safe disposal, which reduces the volume reduction factor.

Further volume reduction and the immobilization of ^{137}Cs can be attained at once by eliminating Cs from the dust with ion chromatography. Fig. 1 shows the flow chart of the super volume reduction by pyroprocessing and ion chromatography. Pyroprocessing of ^{137}Cs-enriched waste produces ^{137}Cs-enriched dust. Washing of the dust in a vessel causes the dissolution of radioactive and nonradioactive CsCl into water. The other soluble salts such as NaCl and KCl are also dissolved. The insoluble sediment in the vessel is transferred again to a furnace to convert to construction materials. The solution containing ^{137}Cs is introduced into a column packed with a Cs adsorbent. ^{137}Cs in the solution is adsorbed in the adsorbent together with stable Cs, and the ^{137}Cs-free eluate is discharged as a nonradioactive solution. Since the concentration of Cs is very low in the dust, a significant volume reduction can be attained by the ion chromatographic process. The adsorbent is judged as Cs-saturated when ^{137}Cs is detected in the eluate by a gamma detector. The Cs-saturated adsorbent is mixed in the mixer with metakaolin and a NaOH solution to convert to solid geopolymer which has an ability of immobilizing Cs[13] and is possible to be disposed as final radioactive waste. The final volume reduction factor is then estimated as the volume ratio of the initial contaminated waste to the geopolymer. The details of the ion chromatography and the solidification process will be mentioned in the following section.

2. Materials and Methods

Transition metal ferrocyanides prepared as candidates of a Cs adsorbent were manganese (II), cobalt (II), ferric, nickel (II) and zinc ferrocyanides. Transition metal ferrocyanide MeFeCN (Me and FeCN denote transition metal and ferrocyanide ions, respectively) was prepared by mixing 0.016 mol/dm^3 of a $K_4Fe(CN)_6$ solution and an equivolume solution of 0.021 mol/dm^3 transition metal chloride or sulfate in a centrifugal tube of 15 cm^3 volume. Mixing of the two solutions gave water-insoluble MeFeCN without leaving $Fe(CN)_6^{4-}$ in the aqueous phase, which was confirmed by $KMnO_4$ redox titration of the supernatant solution. For measuring the Cs adsorption capacity of MeFeCN, three cm^3 of the solution of 2.0 mol/dm^3 KCl containing 1.0×10^{-4} mol/dm^3 of CsCl with 500 Bq/cm^3 of ^{137}CsCl was added to the suspension of MeFeCN in the tube. For measuring the Cs adsorption capacity at low KCl concentration, a pure 10 mmol/dm^3 CsCl solution was used instead of the 2 mol/dm^3 KCl solution. The suspension was stirred with a tube rotator for two days, centrifuged at a speed of 4000 rpm for 15 minutes, and then filtrated with a 0.2 μm pore syringe filter. Two cm^3 of the filtrate was transferred in a plastic petri dish with inside dimensions of 34 mm diameter and 10 mm height. For preventing uneven distribution of the filtrate due to surface tension between the filtrate and the plastic bottom, a paper filter of 33 mm in diameter was inserted in the dish before the transfer. The petri dish with the filtrate was then transferred into a γ-ray measuring apparatus with a NaI crystal scintillator of 50.8 mm diameter and 50.8 mm height. The radioactivity of ^{137}Cs was determined under the secular equilibrium condition. The Cs removal ratio R of a Cs adsorbent defined by

$$R = \frac{[\text{X-Cs}]}{[\text{Cs}^+]_0} \quad \cdots\cdots\cdots\cdots \quad (1)$$

where $[\text{Cs}^+]_0$ is the total molar amount of Cs and [X-Cs] is the molar amount of Cs adsorbed by the adsorbent, was determined by comparing the radioactivity of the solution with and without the adsorbent.

A Cs adsorbent for ion chromatography was prepared by embedding CuFeCN in silica gel matrix[14, 15]. Fifty cm^3

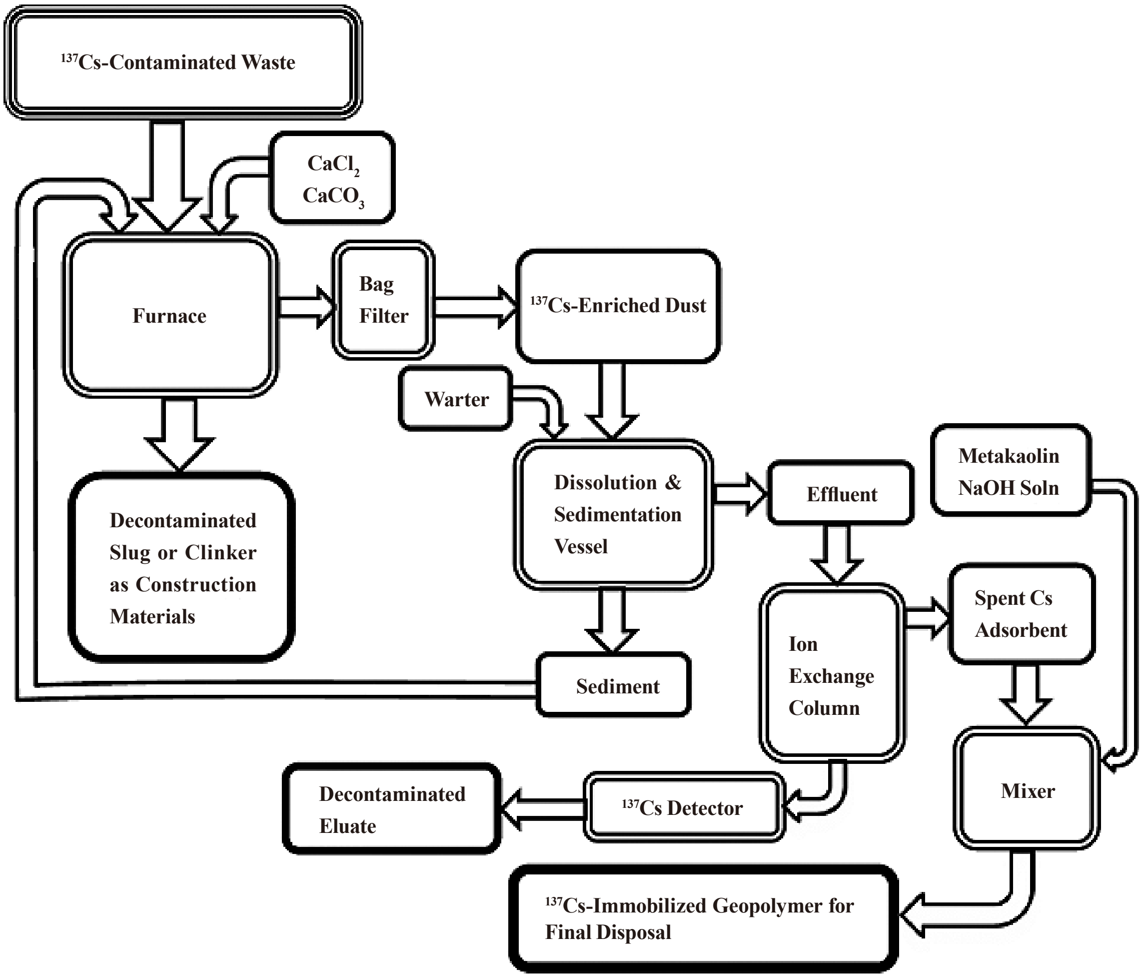

Fig. 1 Flow chart for super volume reduction of ^{137}Cs-contaminated waste by pyroprocessing and ion chromatography

of a 2 mol/dm^3 HCl solution was added to 100 g of water glass (composition: SiO_2 = 6.11 mol/kg, Na_2O = 2.84 mol/kg, H_2O = 45.7% by mass) while stirring vigorously. Forty five mmol of $K_4Fe(CN)_6$ was then dissolved in the mixed solution. The solution was added in dropwise into a 500 cm^3 solution of 20 wt% H_2SO_4 containing 125 mmol of $CuSO_4$. CuFeCN-loaded silica gel was gradually formed in the solution. After standing for one day, the silica gel was recovered from the solution and was washed with water and dried at 60ºC for overnight. After crushing, the silica gel was sieved to collect particles suitable for ion chromatography. The particles with the diameter of 0.15 mm to 0.30 mm were used as a Cs adsorbent. The Cs removal ratio of the CuFeCN-loaded silica gel was measured in the same manner as that for MeFeCN.

An ion exchange column was prepared by pouring the aqueous slurry of 1 g of CuFeCN-loaded silica gel into an ion exchange column with an inner diameter of 9 mm. The height of the resultant bed was 28 mm so that the bulk density of the adsorbent was 0.56. The breakthrough curves for Cs adsorption were acquired by passing a solution containing 2 mol/dm^3 of KCl and 2 mmol/dm^3 of CsCl with 500 Bq/cm^3 of C^{137} through the column at constant linear velocities of 9.4 cm/h to 34.6 cm/h. The radioactivity of the effluent was measured in the same manner as that for MeFeCN.

The spent Cs adsorbent was solidified with metakaolin (Satintone S P33 from Hayashi Kasei Co. Ltd. Composition: Al_2O_3 = 4.37 mol/kg, SiO_2 = 8.65 mol/kg, Na_2O = 0.32 mol/kg) NaOH and water in a plastic petri dish of inside dimensions of 34 mm diameter and 10 mm height. After curing for one month at ambient temperature, the solidified adsorbent with 10 mm height was immersed in 200 cm^3 of distilled water or sea water at 40ºC, and the radioactivity of the solid was periodically measured for examining the Cs leachability of the solid.

Table 1 Main components of Cs-enriched dust

Water-insoluble components (% by mass)			Water-soluble components (mol/kg)					
$CaCO_3$	Carbonaceous residue	Inorganic residue	Cs^+	Na^+	K^+	Ca^{2+}	Cl^-	SO_4^{2-}
46.5	3.3	2.5	3.2×10^{-5}	4.76	2.18	0.28	6.94	0.27

3. Results and Discussion

(1) Ion chromatography

The purpose of this study is to reduce the volume or weight of ^{137}Cs contaminated waste as much as possible. To find an ion exchanger with high Cs adsorption capacity is therefore very important for achieving this purpose. An ion exchange reaction between Cs^+ ions in an aqueous phase and the other ions, for example K^+ ions, in ion exchange sites is expressed as

$$\text{X-K} + Cs^+ \rightarrow \text{X-Cs} + K^+ \quad \cdots\cdots\cdots\cdots\cdots\cdots \quad (2)$$

Under the equilibrium condition, we obtain

$$K_{Cs/K} = \frac{[K^+][\text{X-Cs}]}{[Cs^+][\text{X-K}]} \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \quad (3)$$

Here $K_{Cs/K}$ is the Cs selectivity coefficient of the ion exchanger with respect to K^+ ions, [X-Cs] and [X-K] are the molar amounts of Cs^+ and K^+ ions in the ion exchanger, and $[Cs^+]$ and $[K^+]$ are molar amounts of Cs^+ and K^+ ions in the aqueous phase, respectively. Due to inhomogeneity of ion exchange sites, the selectivity coefficient generally depends on [X-Cs]/[X-K] and monotonously decreases with increasing [X-Cs]/[X-K], because ion exchange sites with high $K_{Cs/K}$ are preferentially occupied by Cs^+ ions. The law of conservation of mass gives the relations of

$$[K^+] + [\text{X-K}] = [K^+]_0 \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \quad (4)$$

$$[Cs^+] + [\text{X-Cs}] = [Cs^+]_0 \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots \quad (5)$$

$$[\text{X-K}] + [\text{X-Cs}] = [\text{CEC}] \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots \quad (6)$$

where $[K^+]_0$ and $[Cs^+]_0$ are the total molar amounts of ion-exchangeable K^+ and Cs^+ ions, respectively, and [CEC] is the cation exchange capacity of the ion exchanger.

The Cs adsorption capacity is defined by [X-Cs]/[CEC]. Using the relations of Eq. (2) to Eq. (6), the Cs adsorption capacity is expressed as

$$\frac{[\text{X-Cs}]}{[\text{CEC}]} = \frac{1}{1 + ([K^+]/[Cs^+])/K_{Cs/K}} \quad \cdots\cdots\cdots\cdots \quad (7)$$

It is evident from Eq. (7) that the Cs adsorption capacity decreases with increasing $[K^+]/[Cs^+]$ and increases with increasing $K_{Cs/K}$. Table 1 shows an example of the main components of dust generated by pyroprocessing. The concentration of K and Na in the dust is more than four orders of magnitude higher than that of Cs. An ion exchanger with very high Cs selectivity coefficient of $K_{Cs/K} > 10^4$ is therefore necessary for the volume reduction. Since the selectivity coefficients of Cs adsorbents other than MeFeCN, such as zeolites and clay minerals, are less than one thousand[16], MeFeCN were used for the preparation of a Cs adsorbent for ion chromatography. In alkali and alkaline earth metal ions, only K^+ ions significantly suppresses the Cs adsorption[16, 17], so that the Cs adsorption capacities of MeFeCN were measured as a function of $[K^+]/[Cs^+]$.

Fig. 2 shows the Cs adsorption capacities of MeFeCN as a function of $[K^+]/[Cs^+]$. Since the cation exchange capacity [CEC] is proportional to [MeFeCN], where [MeFeCN] is the molar amount of $Fe(CN)_6^{4-}$ ions in MeFeCN, the Cs adsorption capacity in Fig. 2 is defined

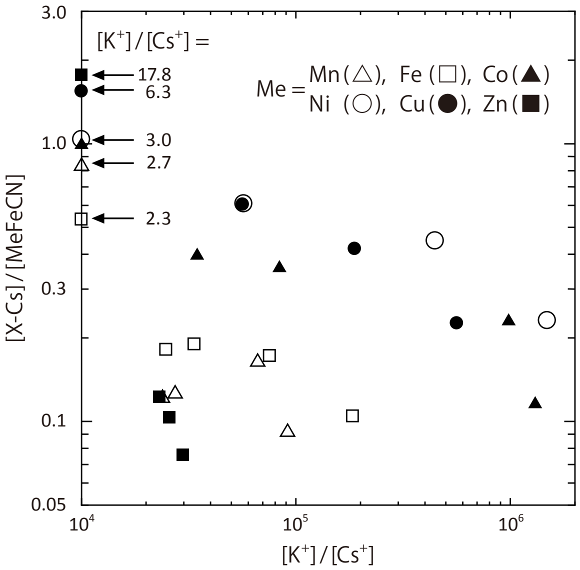

Fig. 2 Cs adsorption capacity of transition metal ferrocyanide MeFeCN as a function of K^+ ion to Cs^+ ion concentration ratio $[K^+]/[Cs^+]$ in solution

as [X-Cs]/[MeFeCN] instead of [X-Cs]/[CEC]. The capacities at low $[\mathrm{K}^+]/[\mathrm{Cs}^+]$ are also shown on the left abscissa. Although FeFeCN or Prussian blue is a well-known Cs adsorbent that has been widely used for the elimination of radioactive Cs, compared to the other MeFeCN, the Cs adsorption capacity is not necessarily high. ZnFeCN shows the highest Cs adsorption capacity at low $[\mathrm{K}^+]/[\mathrm{Cs}^+]$ molar ratio, though the capacity at high $[\mathrm{K}^+]/[\mathrm{Cs}^+]$ is the lowest. NiFeCN and CuFeCN show the highest capacity near $[\mathrm{K}^+]/[\mathrm{Cs}^+] = 10^5$, a typical condition for the chromatographic elimination of Cs. CuFeCN was finally selected as an ion exchanger for Cs removal because of its low toxicity and low cost.

Although CuFeCN has high ability of Cs adsorption, it has the form of a fine powder and is not suitable for column application. Sorbent particles for column chromatography should be large and rigid enough for maintaining the smooth flow of an aqueous solution, and have high specific surface area for fast and efficient ion exchange reaction. For overcoming the drawback of CuFeCN powder, CuFeCN-loaded silica gel was used as a Cs adsorbent for ion chromatography. The cation exchange capacity of the silica gel was 0.29 eq/kg, which indicates that the percentage of CuFeCN loading was about 10% by mass. The Cs selectivity coefficient of the silica gel was derived from Cs removal ratio R that was defined by Eq. (1). Substitution of Eq. (1) into Eq. (3) leads to

$$\frac{R}{(1-R)(1-R[\mathrm{Cs}^+]_0/[\mathrm{CEC}])} = K_{Cs/K}\frac{[\mathrm{CEC}]}{[\mathrm{K}^+]} \cdots\cdots \quad (8)$$

The Cs selectivity coefficient is then obtained by plotting the value of $R/\{(1\text{-}R)(1\text{-}R[\mathrm{Cs}^+]_0/[\mathrm{CEC}])\}$ as a function of $[\mathrm{CEC}]/[\mathrm{K}^+]$. Fig. 3 shows the plot of Eq. (8) within the range of $[\mathrm{K}^+]/[\mathrm{Cs}^+] = 4.8\times10^4$ to 31.4×10^4. The slope of the plot gives a constant value of $K_{\mathrm{Cs/K}} = 2.75\times10^4$, which implies that the adsorbent maintains a high Cs adsorption capacity of [X-Cs]/[CEC] = 0.22 even $[\mathrm{CEC}]/[\mathrm{K}^+] = 1\times10^5$.

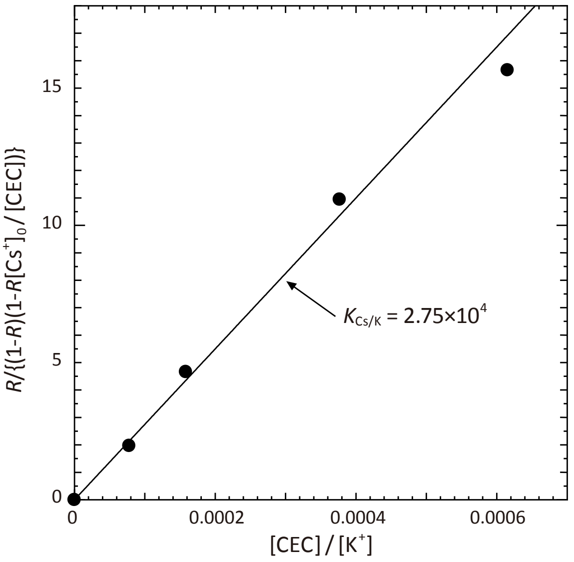

Fig. 3 Cs selectivity coefficient $K_{\mathrm{Cs/K}}$ of copper ferrocyanide-loaded silica gel determined from the relation of Eq. (8)

The maximum Cs adsorption capacity is attained only when ion exchange equilibrium is maintained between an adsorbent bed and an inflowing solution. It is therefore important to know the maximum flow velocity under the equilibrium condition. For determining the maximum flow velocity, 1 g of CuFeCN-loaded silica gel was packed in a cylindrical column with an inner diameter of 9 mm, and the breakthrough curves for Cs adsorption were acquired by passing a solution containing 2 mol/dm^3 of KCl and 2 mmol/dm^3 of CsCl through the column at constant linear velocities (volumetric flow rate divided by the cross section of the flow) of 9.4 cm/h to 34.6 cm/h. Fig. 4 compares the breakthrough curves under different linear velocities. The Cs adsorption capacity at the break through point was 0.85, which was very close to the maximum capacity of 0.96 for the present solution with $[\mathrm{K}^+]/[\mathrm{Cs}^+] = 10^3$. As shown in Fig. 5, using a solution composed of 1 mol/dm^3 of KCl, 1 mol/dm^3 of NaCl 1 mol/dm^3 of CaCl_2 and 1 mmol/dm^3 of

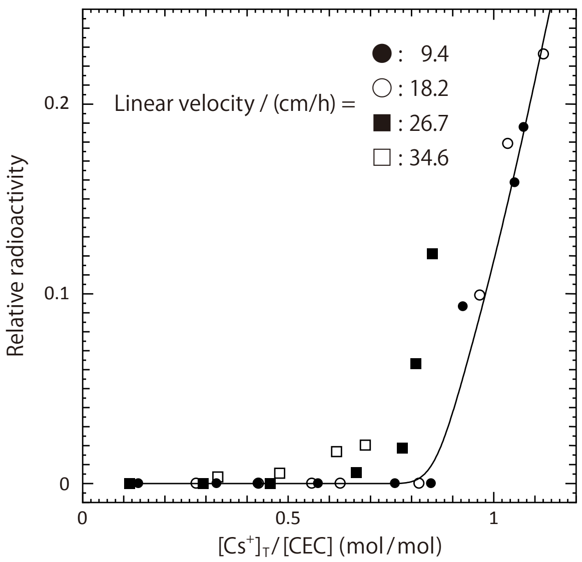

Fig. 4 Breakthrough curve in Cs^+ ion adsorption of copper ferrocyanide-loaded silica gel from solution containing 2 mol/dm^3 of KCl and 2 mmol/dm^3 of CsCl as a function of linear fluid velocity. $[\mathrm{Cs}^+]$T denotes the total molar amount of Cs^+ ions passing through the silica gel column with cation exchange capacity [CEC]

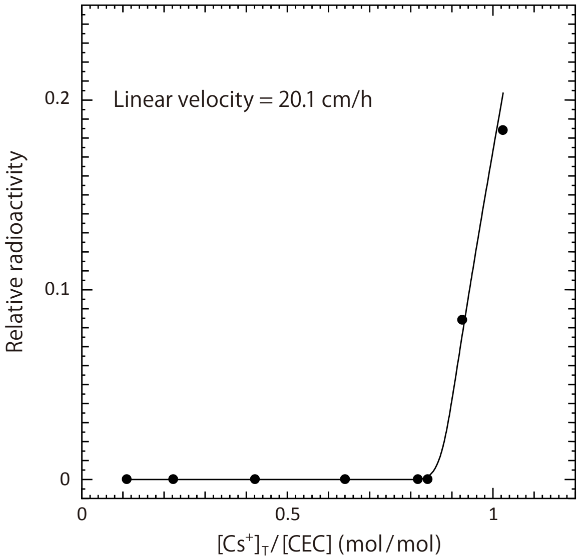

Fig. 5 **Breakthrough curve in Cs^+ ion adsorption of copper ferrocyanide-loaded silica gel from solution containing 1 mol/dm^3 of KCl, 1 mol/dm^3 of NaCl, 1 mol/dm^3 of $CaCl_2$ and 1 mmol/dm^3 of CsCl. $[Cs^+]_T$ denotes the total molar amount of Cs^+ ions passing through the silica gel column with cation exchange capacity [CEC]**

CsCl also gave the same breakthrough point, which certified that alkali and alkaline metal ions other than K^+ scarcely suppressed the Cs adsorption.

For calculating the degree of volume reduction by decontamination processes, it is convenient to use weight reduction factor instead of volume reduction factor. Weight reduction factor is defined as the weight ratio of the initial waste to final one to be disposed as radioactive waste. The weight reduction factor of ^{137}Cs-contaminatd waste by pyroprocessing and ion chromatography using CuFeCN-loaded silica gel can be calculated in the following manner. Suppose ^{137}Cs-contaminated soil contains 13600 ppm of K (0.35 mol/kg) and 5 ppm of Cs (0.38×10^{-4} mol/kg)[18], and the molar ratio of K to Cs in ^{137}Cs-enriched dust is the same as that in the soil, the amount of adsorbent necessary for fixing Cs from 106 kg of soil, w, is estimated from [CEC], $K_{Cs/K}$ and the molar ratio of K to Cs in the soil, as

$$w = \frac{38}{[\text{X-Cs}]} = \frac{38[1+([\text{K}^+]/[\text{Cs}^+])/K_{Cs/K}]}{[\text{CEC}]} = 175\,(\text{kg}) \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \quad (9)$$

The weight reduction factor of 5714 is thus attained. The time for Cs separation can be estimated from the maximum linear velocity, the volume of a Cs solution generated from Cs-enriched dust, and the cross section of a column for Cs adsorption. Assuming that the solution contains 2 mol/dm^3 of KCl, the volume of the solution generated from 106 kg of ^{137}Cs-contaminated soil is estimated to be 175×10^3 dm^3. Using a cylindrical column with an inner diameter of 60 cm, the time necessary for flowing the solution with the maximum linear velocity of 20 cm/h is estimated to be 3094 h or 129 days. The height of spent adsorbent in the column is estimated from bulk density 0.56 of CuFeCN-loaded silica gel to be 110 cm.

(2) Solidification of spent adsorbent

The spent adsorbent is fine particles with high specific radioactivity, so that the solidification of the adsorbent is desirable for the final disposal. Cement solidification is a simple and low cost solidification processes and has been widely used for radiologically contaminated waste. The treatment involves mixing of waste with a hydraulic binder and water. The solidification reaction generally proceeds at ambient temperature under an alkaline condition. Under an alkaline condition, however, MeFeCN decomposes into metal hydroxide and ferrocyanide to release Cs^+ ions, so that Cs adsorption ability is required to the resultant solid. Although Portland cement is commonly used as a binder, it has no ability of adsorbing Cs^+ ions. In the present study, metakaolin was used as a hydraulic binder because the resultant solid, so-called geopolymer, has an ability of adsorbing cations in its matrix[13].

The solidification was carried out at ambient temperature by mixing the spent adsorbent, metakaolin NaOH and water. The mixing ratio by weight was 1 : 2 : 0.7 : 1.8, respectively. The amount of NaOH was adjusted in such a way that the molar amount was the same as that of Al atoms in metakaolin. The resultant mixture gave a sound solid within one day at ambient temperature after the mixing. Decrease of the content of metakaolin from 2 to 1 caused the formation of a brittle solid. Chemical reactions taking place in the mixture were polymerization reaction to form aluminosilicate as geopolymer,

$$\text{mAl}_2\text{O}_3 + 2\text{mNaOH} + \text{nSiO}_2 \rightarrow (\text{NaAlO}_2)_{2m}(\text{SiO}_2)_n + \text{mH}_2\text{O} \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots \quad (10)$$

and CuFeCN decomposition reaction to release Cs^+ and $Fe(CN)_6^{4-}$ ions. The released Cs^+ ions were trapped in aluminosilicate by ion exchanging with Na^+ ions. The Cs immobilization ability of the geopolymer was measured by immersing a geopolymer disc in a petri dish in 200 cm^3

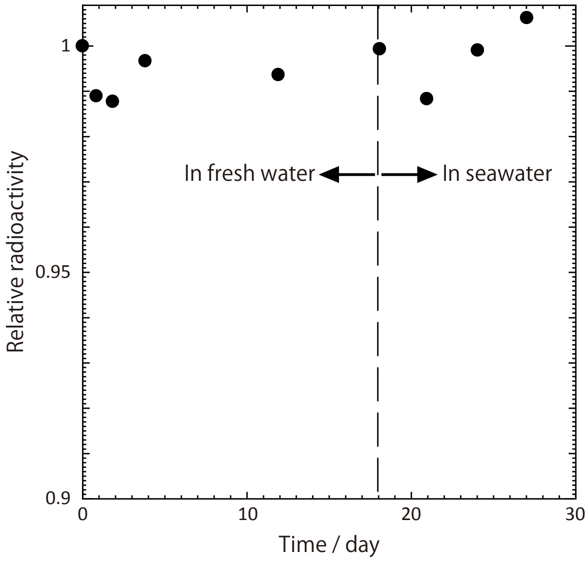

Fig. 6 **Time evolution of the radioactivity of ^{137}C in metakaolin-solidified spent Cs adsorbent of copper forrocyanide-loaded silica gel in fresh and sea waters at 40ºC**

of distilled water or sea water. Fig. 6 shows the Cs immobilization ability of the geopolymer disc of 34 mm diameter and 10 mm height in 200 cm^3 of distilled water or sea water at the temperature of 40ºC. The leaching of Cs was not observed for about one month both in fresh and sea water, which certifies that the metakaolin-solidified adsorbent can be safely disposed as a final radioactive waste.

Solidification of the spent adsorbent caused the decrease of the weight reduction factor. The final weight reduction factor F_f is given by

$$F_f = \frac{132.9\times10^3}{C_{\mathrm{Cs}}}\frac{[\mathrm{CEC}]/W_{\mathrm{S}}}{1+([\mathrm{K}^+]/[\mathrm{Cs}^+])/K_{\mathrm{Cs/K}}} \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots \ (11)$$

where C_{Cs} is the content of Cs in original waste in ppm by mass, [CEC] is the cation exchange capacity of a Cs adsorbent in eq/kg, $K_{\mathrm{Cs/K}}$ is the Cs selectivity coefficient of the adsorbent with respect to K^+ ions, $[\mathrm{K}^+]/[\mathrm{Cs}^+]$ is the molar ratio of K^+ ions in Cs-enriched dust with respect to Cs^+ ions, and W_{S} is weight increment ratio of the spent adsorbent by solidification. Solidification of the spent Cs adsorbent caused 5.5 times increase of the weight, so that it decreased the final weight reduction factor for the ^{137}Cs-contaminated soil from 5.7×10^3 to 1.0×10^3.

The weight reduction of ^{137}Cs-contaminated wastes accompanies the increase of the specific radioactivity of solidified final waste. The specific radioactivity of the final waste is equal to the specific radioactivity of the initial waste multiplied by F_f. Suppose the specific radioactivity of initial waste is 100 kBq/kg and the final weight reduction factor is 1000, the specific radioactivity of the final waste is 100 MBq/kg, which belongs to low level radioactive waste (L2 waste) and is possible to be disposed into a near-surface concrete pit disposal facilitiy. The weight reduction factor of 1.0×10^3 for 2.5×10^9 kg of ^{137}Cs-contaminated soil implies that the final radioactive waste can be disposed into a near-surface concrete pit with the size comparable to a long course swimming pool of 50 m×25 m×2 m.

4. Conclusion

It was revealed that the combination of pyroprocessing and ion chromatographic elimination of Cs is very effective for weight and volume reduction of ^{137}Cs-contaminated waste. Although the present study deals with a small amount of materials, scaling up of the proposed decontamination process may not be difficult. Equation (11) indicates that further increase of the weight reduction factor can be attained by 1) decreasing $[\mathrm{K}^+]/[\mathrm{Cs}^+]$ by improving the pyroprocessing, 2) increasing [CEC] and $K_{\mathrm{Cs/K}}$ by improving the Cs adsorption ability of adsorbent for ion chromatography, and 3) improving the solidification method to reduce the weight increment ratio. Several attempts to decrease the $[\mathrm{K}^+]/[\mathrm{Cs}^+]$ ratio and to increase the [CEC] of Cs adsorbents while keeping high $K_{\mathrm{Cs/K}}$ and high ion-exchange rate are under progress. If the final weight reduction factor of 1.0×10^4, ten times higher than the present value, is attained, all the ^{137}Cs contaminated wastes waiting for the final disposal can be converted as regular construction materials and 2.5×10^6 kg of ^{137}Cs-enriched solidified waste that can be disposed into a single concrete pit of 50 m×25 m×2 m as L2 waste.

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