

EVALUATION OF FERROCYANIDE ANION EXCHANGE RESINS REGARDING THE UPTAKE OF Cs⁺ IONS AND THEIR REGENERATION

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Ferrocyanide-anion exchange resin was prepared and the prepared ion exchange resins were tested on the ability to uptake Cs⁺ ion. The prepared ion exchange resins were resin-KCoFC, resin-KNiFC, and resin-KCuFC. The three tested ion exchange resins showed ion exchange selectivity on the Cs⁺ ion of the surrogate soil decontamination solution, and resin-KCoFC showed the best Cs⁺ ion uptake capability among the tested ion exchange resins. The ion exchange behaviors were explained well by the modified Dubinin-Polanyi equation. A regeneration feasibility study of the spent ion exchange resins was also performed by the successive application of hydrogen peroxide and hydrazine. The desorption of the Cs⁺ ion from the ion exchange resin satisfied the electroneutrality condition in the oxidation step; the desorption of the Fe²⁺ ion in the reduction step could also be reduced by adding the K⁺ ion.

KEYWORDS : Cesium, Ion Exchange Resin, Regeneration, Soil, Decontamination

1. INTRODUCTION

Ion exchange materials are characterized by their stability with regards to radiation and temperature, thus they can be used in treating highly radioactive waste solutions [1]. Ammonium molybdophosphate (AMP) immobilized on polyacrylonitrile (PAN) is an engineered form of the cesium ion selective material developed at Czech Technical University. A version of the second order kinetic model was applied to the experimental AMP-PAN breakthrough data [2]. The transition metal ferrocyanides were immobilized on PAN and were used to remove the cesium ion [3]. The sorption capacity was determined to be 1.22 and 0.55 mmol/g for KZnHCF-PAN and KCuHCF-PAN, respectively [4].

The transition metal ferrocyanides immobilized on an anion exchange resin are simple and easy to prepare, and obtainable in any given conditions: they are non-hygroscopic and granular, thus they can be in a dry state [5]. Several applications of these resins to remove radioactive cesium ions have been reported. Folsom et al. [6] and Mann et al. [7] used ion exchange resins to concentrate radiocesium from a large volume of sea water. Watari et al. [5] reported that ferrocyanide resins can be successfully used to analyze and decontaminate

Cs-137 and I-131 in water and milk, respectively, as emergency countermeasures to avoid environmental contamination.

A mixed solution of AgNO₃ and NH₄NO₃ was used to study the regeneration of two loaded columns (KZnHCF-PAN and KCuHCF-PAN). The elution percentage of the loaded cesium from KZnHCF-PAN reached 85% of the total loaded amount, while that of CuKHCF-PAN reached 12%. Thus, it was concluded that the KZnHCF-PAN column could be used for another cycle [4]. Tanihara [8] invented a production and regeneration method for a granular ion exchanger with a high selectivity for cesium and easy solid-liquid separation. The ion exchanger that separates the cesium was regenerated by oxidizing the insoluble copper salt of hexacyanoferrous acid on a porous anion exchange resin using nitric acid. The reduction of the oxidized ion exchanger occurred by hydrazine. As a result, the ion exchanger with a high adsorption power for cesium was reproduced. Onodera et al. [9] reported a biopolymer composite ion exchanger with a high cesium selectivity and a method to manufacture this ion exchanger with ease and good reproducibility. This ion exchanger is a cesium separation/recovery agent comprised of a composite ion exchanger with a high cesium selectivity

by employing a calcium alginate gel as a matrix and loading the matrix with an inorganic ion-exchanger. They presented a method to manufacture a composite ion exchanger with a high cesium selectivity, which disperses at least one inorganic ion exchange powder selected from a group consisting of crystalline tetratitanic acid, insoluble ferrocyanides, ammonium molybdophosphate, silicon titanates, copper (II) potassium hexacyanoferrate (II), and ammonium tungstophosphate in a sodium alginate aqueous solution to prepare a slurry, and brings a calcium salt solution into contact with the slurry to disperse and load the inorganic ion exchanger in a calcium alginate gel substrate.

The removal of heavy metals or radionuclides from contaminated soil via chelation can be a valid remediation method. The important properties of the chelating agent used include the chelation bonding strength, reusability, and biostability during the remediation operation. The chelating and mobilizing agents that have been investigated include EDTA, citric acid, gluconic acid, phosphoric acid, NTA, DTPA, and ammonium acetate, in addition to a pH-adjusted aqueous solution [10, 11].

Korea depends heavily on nuclear power for its electricity generation, with 20 nuclear power units in operation sharing 40% of the total electricity production [12]. For an accurate burnup analysis of spent fuels, Cs-137 is used as a fission monitor to determine the total burnup value [13]. To prepare for nuclear incidents or accidents, the Korea Atomic Energy Research Institute (KAERI) has studied the fixation of γ -radionuclides on the soil surface using polymer solutions [14].

KAERI is preparing a soil decontamination method for soils contaminated with radionuclides such as Cs-134, Cs-137, and Co-60. The candidate chelate was citric acid. To reduce the radioactive waste volume, a feasibility study on the reuse of citric acid and a Cs⁺ ion selective ion exchange resin during the soil decontamination was performed. As Cs-137 exists as a trace ion in the soil decontamination solution, its ion exchange characteristics differ when compared with the waste solution generated by industry. Particularly, the regeneration of spent ion exchange resins by successive applications of the oxidation and reduction steps has not been well understood.

The objectives of this study are to investigate the Cs⁺ ion uptake capability for three ferrocyanide loaded ion exchange resins for a surrogate soil decontamination solution and to compare their regeneration characteristics.

2. THEORETICAL BACKGROUND

The Dubinin-Polanyi adsorption model for a single component system is derived from the difference in free energy between an adsorbed phase and a saturated liquid sorbate at the same temperature [15]. The difference in

free energy is referred to as the adsorption potential ϵ :

$$\epsilon = -RT \ln(C/C_s), \quad (1)$$

where C_s is the saturation concentration for the liquid sorbate, C is the equilibrium concentration, R is the gas constant, and T is the absolute temperature. To correlate the adsorption equilibrium data for some adsorbents, a characteristic curve can often be approximated using the following Gaussian expression:

$$W = W_o e^{-k\epsilon^2}, \quad (2)$$

where W is the volume adsorbed and the saturation limit (W_o) may be considered to represent the total specific micropore volume of the adsorbent. k is a parameter of the solid phase only. If equations (1) and (2) are combined and rearranged,

$$\frac{W}{W_o} = \frac{qV_m}{q_sV_m} = \frac{q}{q_s} = \exp\left(-kR^2T^2\left[\ln\left(\frac{C_s}{C}\right)\right]^2\right). \quad (3)$$

In these equations, q and q_s are the equilibrium and saturation concentrations in the solid phase, respectively; V_m is the molar volume of the sorbate. The later part of equation (3) is the Dubinin-Polanyi equation.

The Dubinin-Polanyi model can be extended to describe a multicomponent equilibrium [16]. The multicomponent form of the Dubinin-Polanyi equation for liquids is

$$\frac{(q_1 + q_2 + \dots)V_m}{W_o} = \exp(-k\epsilon^2), \quad (4)$$

where $W_o/V_m = q_s$. The molar volume of the sorbate is the sum of the partial molar volume multiplied by the mole fraction of each component.

$$V_m = X_1\bar{V}_1 + X_2\bar{V}_2 + \dots, \quad (5)$$

where \bar{V}_1 and \bar{V}_2 refer to the partial molar volumes of components 1 and 2, respectively. X_1 and X_2 refer to the mole fractions of components 1 and 2, respectively. The k parameter of the solid phase is expressed as the sum of the parameters resulting from each component:

$$\frac{1}{\sqrt{k}} = \frac{X_1}{\sqrt{k_1}} + \frac{X_2}{\sqrt{k_2}} + \dots, \quad (6)$$

where k_1 and k_2 refer to the solid phase parameters assigned

for components 1 and 2, respectively. Furthermore,

$$\varepsilon = -RT \ln \left(\frac{C_1 + C_2 + \dots}{C_s} \right). \quad (7)$$

Equation (4) can then be rewritten as:

$$\sum q_i = \exp \left[b_o + b_1 \ln \left(\sum C_i \right) + b_2 \left\{ \ln \left(\sum C_i \right) \right\}^2 \right], \quad (8)$$

where

$$b_o = \ln(q_s) - kR^2T^2 \left\{ \ln(C_s) \right\}^2, \quad (9)$$

$$b_1 = 2kR^2T^2 \ln(C_s), \quad (10)$$

$$\text{and } b_2 = -kR^2T^2. \quad (11)$$

As for the multicomponent system, the total amount of adsorbed ions on a solid surface is expressed as the sum of each ion in a liquid state.

3. EXPERIMENTAL

3.1 Preparation of the Ferrocyanide-Anion Exchange Resins

Nuclear grade OH⁻ ion type strong base anion exchange resin supplied by Supelco Co. (IRN-78) was used. 300 g of IRN-78 was contacted with 900 ml of 0.5 M potassium ferrocyanide solution for 24 hours. The ion exchange resins (resin-potassium ferrocyanide) were washed with distilled water three times. The resins were divided into three sections and each resin was dipped into 1 M cuprous nitrate, cobalt nitrate, and nickel nitrate solutions: the final ion exchange resins were resin-potassium copper ferrocyanide (resin-KCuFC), resin-potassium nickel ferrocyanide (resin-KNiFC), and resin-potassium cobalt ferrocyanide (resin-KCoFC).

Table 1. Specific Activity of Soil

Sample name	Radionuclide	Specific activity (Bq/kg)
KRR Soil	Cs-134, Cs-137	61.8 ± 1.5
	Co-60	423.4 ± 6.2
	Cr-51	< 5.2
	Fe-59	< 3.1
	K-40	1782.6 ± 8.9

3.2 Preparation of a Surrogate Soil Decontamination Solution

Radioactive waste soils that had been stored in a temporary storage site were dried in a shady area. The dried soil was sieved with a 2 mm sieve for 30 minutes. The radioactivity of the soil was measured using MCA (EG & G Co. HPGe; counting time: 100,000 seconds) and is listed in Table 1. To remove the organic compounds, soil particles of less than 2 mm were placed in contact with 1 M sodium acetate and a 30% H₂O₂ solution. Then, they were washed with distilled water. 50 g of the soil between 0.063 mm and 1 mm was placed in contact with 500 ml of a mixed solution of 0.05 M citric acid and 0.03 M nitric acid for 3 hours at room temperature. The solution pH was 4.0. The soil particles were removed from the solution by passing the solution through a 0.1 μm Whatman filter. The metal ion concentration in the remnant solution was analyzed using an atomic absorption spectrometer (Perkin Elmer Co., Analyst 300). The metal ion concentration is listed in Table 2.

3.3 Cs⁺ Ion Uptake Test and Regeneration of the Spent Ion Exchange Resin

The resin-KCoFC, resin-KNiFC, and resin-KCuFC with different amounts of 1, 2, 3, 4, and 5 g were poured into 100 ml of the surrogate solution. Then, they were stirred for 24 hrs at room temperature using a 300 rpm stirrer. The pH of the solution phase before and after the ion exchange test was monitored.

Resin-KCoFC, resin-KNiFC, and resin-KCuFC were saturated with the Cs⁺ ion. 2 g of the ion exchange resin was contacted with 500 ml of H₂O₂ for 10 hrs in the concentration range from 0.025 to 5.60 M. The resins were then washed with distilled water and reduced using hydrazine. 2 g of the ion exchange resin was contacted with 500 ml of a N₂H₄ solution for 10 hrs in a concentration range from 0.04 to 8.72 M. At each step, the concentration of the metal ions desorbed from the ion exchange resins was analyzed.

Table 2. Concentration of Metal Ion

Metal ion	Concentration (M)
Mg	4.54 X 10 ⁻⁴
Ca	1.51 X 10 ⁻³
Al	1.81 X 10 ⁻³
Si	1.63 X 10 ⁻³
Cs	2.29 X 10 ⁻⁵
Co	8.46 X 10 ⁻⁵
Fe	1.38 X 10 ⁻³

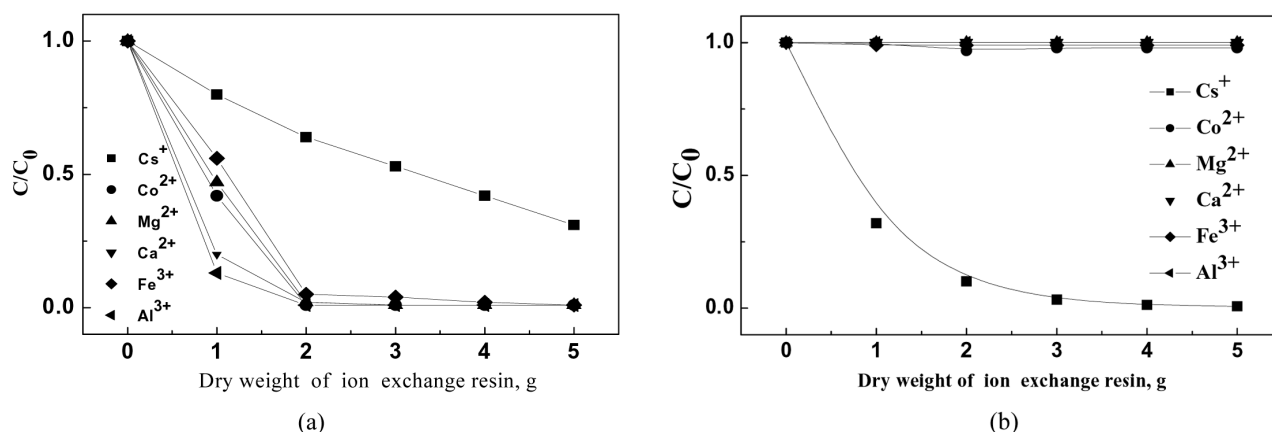


Fig. 1. Change of the Metal Ion Concentration in a Surrogate Solution: (a) IRN-77 and (b) Resin-KCuFC

4. RESULTS AND DISCUSSION

4.1 Cs⁺ Ion Uptake Behavior

Comparative tests were performed with a surrogate solution using a commercial cation exchange resin (IRN-77) and resin-KCuFC for 24 hrs. The results are shown in Fig. 1. The selectivity of the metal ions on IRN-77 is in the order of $Al^{3+} > Ca^{2+} > Co^{2+} > Mg^{2+} > Fe^{2+} > Cs^{+}$, while it is in the order of $Cs^{+} \gg Co^{2+}, Fe^{2+} \geq Mg^{2+}, Ca^{2+}, Al^{3+}$ on resin-KCuFC in the experimental range. For the commercial cation exchange resins, the Cs⁺ ion is difficult to remove in very low concentrations.

The uptake capability of the three different ion exchange resins against time was also investigated. The results are shown in Fig. 2. The ion exchange adsorption of the Cs⁺ ion onto the Cs⁺ ion selective ion exchange

resin was assumed to be a process with at least two different stages. The first stage was a very fast intake corresponding to approximately the first 20 min. Then, the uptake of the Cs⁺ ion slowed with time and attained a constant value within 2 hrs. For all tested conditions, the order of the Cs⁺ ion uptake capability was resin-KCoFC > resin-KNiFC > resin-KCuFC.

The ionic radii of the Co²⁺, Ni²⁺, and Cu²⁺ ions are 0.078, 0.072, and 0.069 nm, respectively. If the Cs⁺ ion uptake behavior of the three Cs⁺ ion selective ion exchange resins in Fig. 2 is compared with the ionic radii of the substituted metal ions, the order of the uptake capability of the two cases (1 and 3 g) coincides precisely with the size of the transition metal ions. Two explanations are possible: after the Co²⁺ ion is substituted with the K⁺ ions, the remaining K⁺ ion in resin-KCoFC is

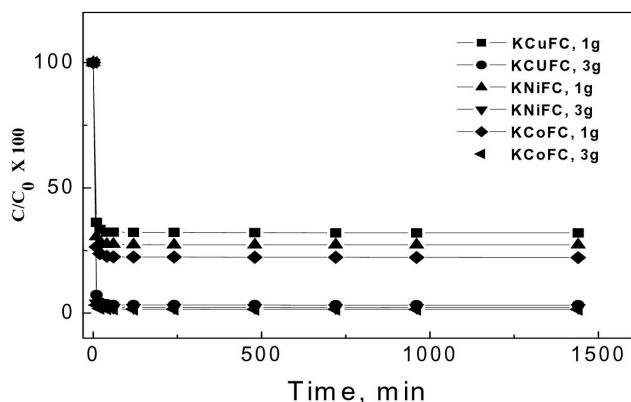


Fig. 2. Uptake of the Cs⁺ Ion from the Surrogate Solution over Time

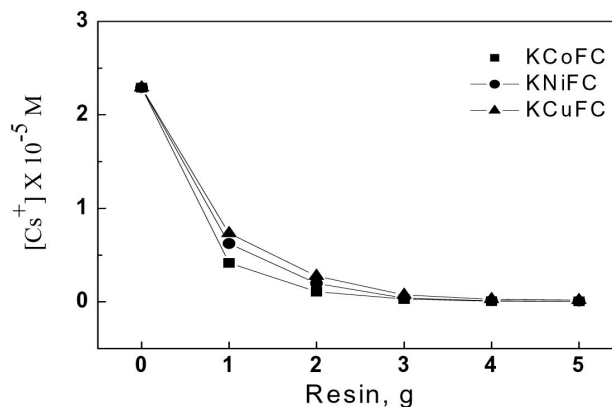


Fig. 3. Uptake Characteristics of the Three Cs⁺ Ion Selective Ion Exchange Resins

more easily substituted with the Cs⁺ ion as a result of the size effect, or the negative charge of the electrons in the Co²⁺ ion is more widely distributed than in the Ni²⁺ and Cu²⁺ ions.

The equilibrium of the Cs⁺ ion in the surrogate soil decontamination solution after 24 hrs of reaction with the three Cs⁺ ion selective ion exchange resins was investigated and the concentration of the Cs⁺ ion remaining is plotted against the amount of ion exchange resins in Fig. 3. The calculated distribution coefficient of the Cs⁺ ion for resin-KCoFC changes from 348 to 9.98 X10³, for resin-KNiFC changes from 268 to 6.65 X10³, and for resin-KCuFC changes from 212 to 2.84 X10³ ml/g.

Moon et al. [17] obtained the ion exchange isotherms for the 4A/Cs-Sr and PAN-4A/Cs-Sr systems and evaluated the ion exchange capacities of a PAN-zeolite composite ion exchanger using the Dubinin-Polanyi equation. As the Dubinin-Polanyi model was adapted for a single component system, deviations from the experimental data occurred. To obtain the ion exchange isotherms of Cs⁺ ion selective ion exchange resin systems, it was assumed that a specific component ion of the multicomponent system is predominantly adsorbed on a solid surface and the uptake of the other ions is negligible. In this case, the predominantly adsorbed ion in the multicomponent system of equation (8) can be expressed as the following modified Dubinin-Polanyi equation:

$$q_i = \exp[b_o + b_1 \ln(c_i) + b_2 \{\ln(C_i)\}^2]. \quad (12)$$

The experimental data and modeling results are shown in Fig. 4. Figure 4 indicates that the ion exchange systems are well described by the modified Dubinin-Polanyi equation. The small deviation from the experimental data is ascribed to the assumption that the uptake of other ions in the solution is negligible. The parameter values are listed in Table 3. The values, however, do not coincide with each other for the three ion exchange resin systems. This can be explained by the difference of the saturation concentration between the three ion exchange resins, the parameter k being characteristic of the ion exchange resins, and the effect of coexisting cation and anions.

4.2 Regeneration of the Spent Ion Exchange Resins

The oxidation results for three Cs⁺ ion selective ion exchange resins saturated with the Cs⁺ ion are shown in Fig. 5. As the oxidation proceeds, the metal ions are desorbed from the ion exchange resin, and the concentration of the metal ions increases with the amount of H₂O₂ added. Particularly, the Cs⁺ ion is predominantly desorbed. The desorbed Cs⁺ ion from the ion exchange resin occurs in the order of resin-KCuFC > resin-KNiFC

> resin-KCoFC. The other metal ions, such as Fe³⁺, Co³⁺, Ni³⁺, and Cu³⁺ are also desorbed, but the amount was negligible.

Collyer et al. [18] studied the behavior of the Fe(CN)₆³⁻/Fe(CN)₆⁴⁻ redox couple in a calyx(4) resorcinarenetetra-thiol-modified gold electrode. They reported that a reversible reaction occurs between the Fe(CN)₆³⁻ and Fe(CN)₆⁴⁻ redox couple and that the reaction is affected by the steric arrangements. The oxidation of the Fe(CN)₆⁴⁻ ion on the surface of an anion

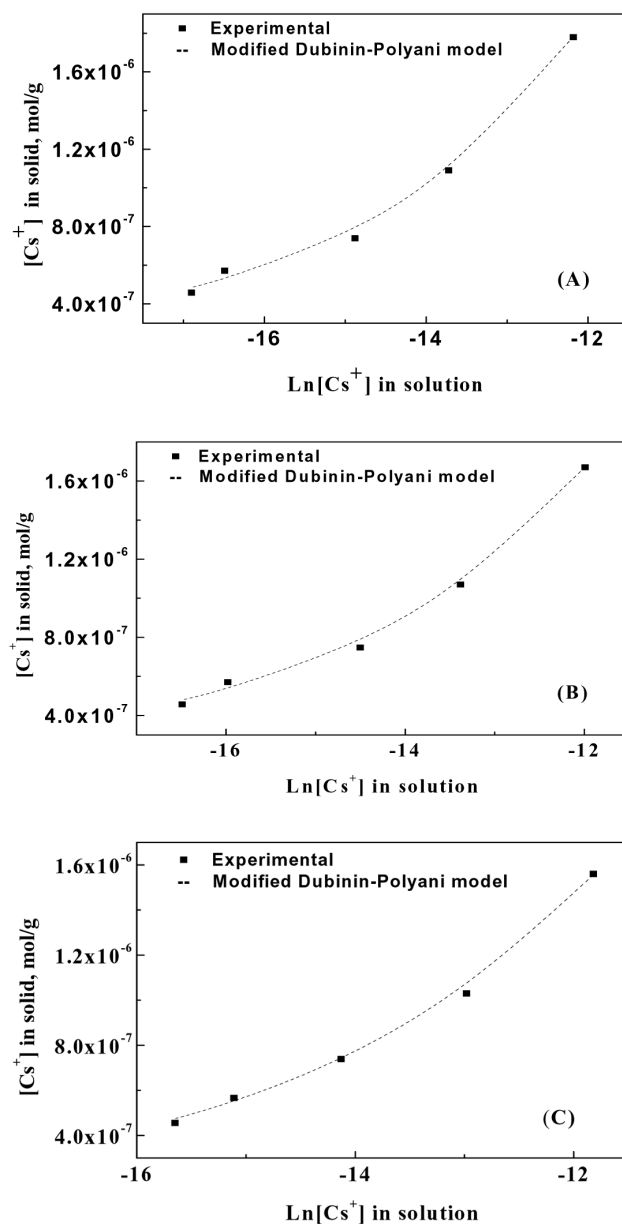
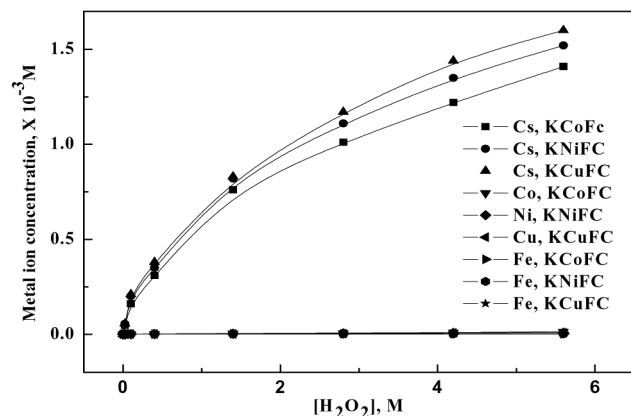
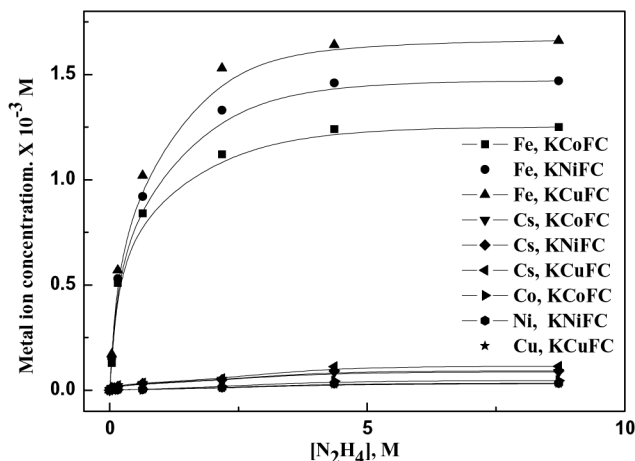


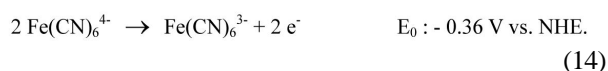
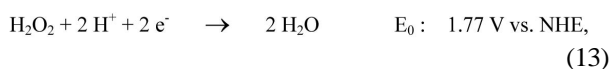
Fig. 4. Equilibrium Isotherms Modeled by the Modified Dubinin-Polanyi Equation for (a) Resin-KCoFC, (b) Resin-KNiFC, and (c) Resin-KCuFC

Table 3. Parameters of the Modified Dubinin-Polanyi Equation.

Parameter	Resin- KCoFC	Resin-KNiFC	Resin-KCuFC
b ₀	-6.37	-6.81	-7.41
b ₁	0.77	0.73	0.65
b ₂	0.02	0.02	0.12

**Fig. 5.** Release of the Metal Ion from Resin-KFC through the H₂O₂ Solution**Fig. 6.** Release of Metal Ions from Resin-KFC Using the Hydrazine Solution

exchange resin by H₂O₂ is described as the following two half reactions [19]:

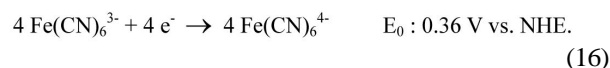
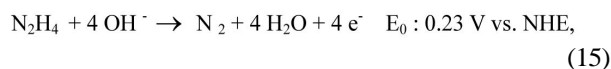


The above two reactions proceed spontaneously. As the oxidation state proceeds from Fe(CN)₆⁴⁻ to Fe(CN)₆³⁻, the positive charge of resin-KFC is increased in number. To satisfy the electroneutrality condition, the Cs⁺ ion is desorbed from the ion exchange resin. Recycling of the spent ion exchange resin via oxidation has two merits: firstly, as the final products are water and the Cs⁺ ion, the secondary waste can be managed easily and safely, and secondly, the Cs⁺ ion can be concentrated.

The reduction results of the three Cs⁺ ion selective ion exchange resins used in the oxidation tests are shown in Fig. 6. Contrary to the oxidation test, the primary metal component desorbed from the resins is the Fe²⁺ ion. With an increase of the hydrazine concentration, the concentrations of the Cs⁺, Co²⁺, Ni²⁺, and Cu²⁺ ions increase continuously. However, the concentration of the Fe²⁺ ion slightly changes above 4.36 M of N₂H₄.

The reduction of the Fe(CN)₆³⁻ ion on the surface of

an anion exchange resin using N₂H₄ is described as the following two half reactions [20]:



As the reduction of Fe(CN)₆³⁻ to Fe(CN)₆⁴⁻ proceeds, the negative charge of resin-KFC is increased in number. Koichi explained that the Fe(CN)₆⁴⁻ ion desorbs from an anion exchange resin to satisfy the electroneutrality condition as follows:

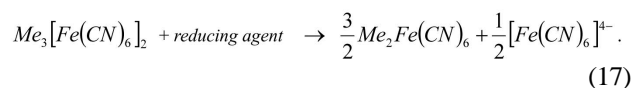


Figure 7 shows the effect of the K⁺ ion on the desorption of the Fe(CN)₆⁴⁻ ion during the reduction reaction. As the concentration of the K⁺ ion increased from 0 to 0.25 M, the concentration of the Fe(CN)₆⁴⁻ ion decreased from 1.25 to 0.72 mM for resin-KCoFC, 1.47 to 0.84 mM for resin-KNiFC, and 1.66 to 1.13 mM for resin-KCuFC. To satisfy the electroneutrality condition,

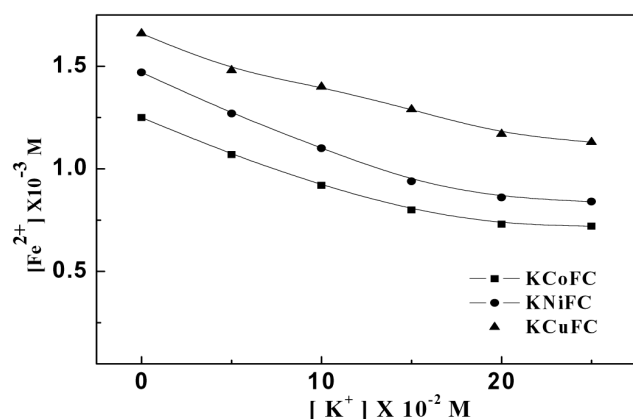


Fig. 7. Effect of the K⁺ Ion Concentration On the Desorption of the Fe²⁺ Ion

the uptake of the K⁺ ion decreased the desorption of the Fe(CN)₆⁴⁻ ion. The surface of the Cs⁺ ion selective resin was examined using a SEM after the reduction reaction and it has already been reported that the resins maintained their integrity [21]. The regenerated Cs⁺ ion selective resins were tested for use in successive cycles: more than 78% of the ion exchange resin could be recovered.

5. CONCLUSION

Decommissioning nuclear facilities requires site restoration. If the area is contaminated with radionuclides, the area must be cleaned. When the volume of soil collected from a contaminated area is very large, decontamination of the soil is required to reduce the volume. Radioactive cesium, which is the primary radionuclide for middle and long term personal radiation exposures, should be effectively removed. Resin-KCoFC, resin-KNiFC, and resin-KCuFC can be applied to separate trace amounts of the Cs⁺ ion from the surrogate soil decontamination solution. The order of the uptake capability of the Cs⁺ ion was resin-KCoFC > resin-KNiFC > resin-KCuFC. The order is related to the ionic size of the transition metal ions. The ion exchange resins were effectively regenerated by successive applications of an oxidation reaction followed by a reduction reaction. From an environmental point of view, the process is particularly useful when the regeneration of ion exchange resins needs to be achieved without using strong acids and bases.

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