

An Analysis of the Ageing Effect on the Removal of Cesium and Cobalt from Radioactive Soil by the Electrokinetic Method

Gye-Nam Kim, Won-Zin Oh, Hui-Zun Won, and Chong-Hun Jung

Korea Atomic Energy Research Institute
150, Deokjin-dong, Yuseong-gu, Daejeon, 305-353, Korea
kjs@kaeri.re.kr

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Abstract

The ageing effects of radionuclides in radioactive soil on remediation using the electrokinetic method were analyzed. Comparative experiments were conducted for the reactive soil around a TRIGA research reactor contaminated with ^{137}Cs and ^{60}Co for 15 years and the non-reactive soil that was intentionally contaminated with Cs^+ and Co^{2+} for 3 days. It was observed that because of an aging effect on ^{137}Cs , the efficiency of removing it decreased. H_2SO_4 used as an additive to increase the removal efficiency showed a higher removal capability than other chemicals for both ^{137}Cs and ^{60}Co . The efficiency of removing radionuclides from the radioactive soil in the column was proportional to the capability of the added chemical to extract radionuclides. It took 10 days to achieve a 54% removal of ^{137}Cs and a 97% removal of ^{60}Co from the soil. The volume of the soil wastewater discharged from the soil column by the electrokinetic method was 20% below that for soil washing.

Key Words : ageing effect, cesium, cobalt, electrokinetic

1. Introduction

South Korea has many nuclear power plants and two research reactors. As such, the soil around the nuclear facilities can become contaminated with radionuclides over the long-term operation of these facilities. The Korea Atomic Energy Research Institute (KAERI) removed soil contaminated with radionuclides from the area around the TRIGA research reactor in 1988, placed the soil in 4000 sets of 200 L waste drums, and has been storing the drums in a

radioactive waste storage in KAERI since 1988. The contamination characteristics of the TRIGA soil are shown in Table 1. The smaller the size of the soil particles is, the higher the radioactive concentration in the soil particles is. Also, the contamination concentrations of ^{137}Cs are higher than those of ^{60}Co . Meanwhile, the radioactivities of ^{137}Cs and ^{60}Co in parts of the contaminated soil have been found to be higher than the restoration target (0.2 Bq/g). Therefore, it is necessary to develop a technology for decontaminating the TRIGA soil presently contaminated with

radionuclides.

While the ^{137}Cs and ^{60}Co in the large particles ($>0.063\text{mm}$) of the TRIGA soil can be removed by using the soil washing method, the ^{137}Cs and ^{60}Co in the minute particles ($<0.063\text{mm}$) of soil are not effectively removed by this method. In particular, the efficiency of removing ^{137}Cs from minute particles ($<0.063\text{mm}$) by soil washing is extremely low. Accordingly, many researchers have applied the electrokinetic method to remove radionuclides from minute particles of soil and have investigated the optimum experimental conditions for maximizing the efficiency of radionuclide removal.

Heavy metals incorporated in soils take different chemical forms, including a dissolved ionic form, an electrostatically absorbed form, and a surface complex form [1]. With electrokinetic remediation, the pH of the cathode side in the soil column increases, due to the production of OH^- in the cathode electrode. As such, a hydroxide precipitate forms on the cathode side and the efficiency of contaminant removal is reduced [2]. Li et al. [3,4] reported that a decrease in both the conductivity and the metal precipitation can be

avoided by injecting a 0.01 M KNO_3 solution into the soil or by inserting a cation selective membrane between the cathode and the soil to stop the advancement of the hydroxyl front toward the anode. The electrolyte pH is then controlled by the addition of 0.05 M KOH at the anode and 0.05 M HCl at the cathode [5]. Lee and Yang [6] also developed a new method of controlling the electrolyte pH, using a circulation system in electrokinetic soil remediation. In addition, to avoid the precipitation of copper hydroxide in a sand column, Yu and Neretnieks [2] inserted a conductive solution between the sand and the cathode. The removal rates of heavy metals from soil have also been increased with the addition of salts of competing noncontaminant cations, soil acidification, and catholyte drainage [7].

In the current study, TRIGA non-radioactive soil intentionally contaminated with Cs^+ and Co^{2+} for a short time and the TRIGA radioactive soil contaminated with ^{137}Cs and ^{60}Co for a long time were remediated with the electrokinetic method. The ageing effect was analyzed by comparing the results of the two remediation experiments. In order to increase the efficiency of

Table 1. Contamination Characteristics of TRIGA Soil

| Effective dose rate of waste drum surface | Soil Particle Size | Volume(%) | Co-60(Bq/kg) | Cs-137(Bq/kg) |
|---|--------------------|-----------|--------------|---------------|
| 0.05mR/h above (7%) | $>1.0\text{mm}$ | 28.3 | 38.9-233.7 | 1.5-886.0 |
| | 0.063-1.0mm | 61.2 | 155.2-1079.3 | 16.3-6700.2 |
| | $<0.063\text{mm}$ | 10.5 | 480.5-6645.0 | 47.1-19547.0 |
| 0.02-0.05mR/h (60%) | $>1.0\text{mm}$ | 48.5 | 2.6-24.0 | 0.3-35.1 |
| | 0.063-1.0mm | 46.5 | 6.3-217.8 | 13.9-436.7 |
| | $<0.063\text{mm}$ | 5.0 | 31.1-316.4 | 287.9-1663.7 |
| 0.02mR/h below (33%) | $>1.0\text{mm}$ | 52.4 | 5.3-15.2 | 2.5-7.0 |
| | 0.063-1.0mm | 43.1 | 14.1-69.1 | 25.4-53.0 |
| | $<0.063\text{mm}$ | 4.5 | 56.3-204.7 | 85.6-377.2 |

removing ^{137}Cs and ^{60}Co from the aged radioactive soil, many kinds of chemicals were added to the electrokinetic experiments. The most effective chemical was selected according to the results of the experiments. A device to solve the ageing effect was explored by analyzing the results of the electrokinetic experiment, which was conducted with the addition of the selected chemical.

2. Theorem and Modeling

2.1. Transport Mechanisms

The electric field drives the contaminants toward the electrodes by two mechanisms. The first is electroosmosis, an electrokinetic phenomenon in which the saturating liquid and the dissolved substances flow toward an electrode [8]. The electroosmotic flow rate is proportional to the product of the applied electric field strength and the ζ potential at the soil-liquid interface. The value of the ζ potential depends on the soil properties, as well as on the ionic strength and the pH of the saturating liquid. A typical ζ potential is about -10 to -100 V, and in a 100Vcm^{-1} electric field, water will flow toward the cathode at a velocity of around $1\text{-}10\text{ cm min}^{-1}$. The second mechanism is electromigration, by which charged ions move in the electric field strength, the charge on the ion, and the mobility of the ion. The transport of charged ions by electromigration is generally more rapid than that by electroosmosis. In cases where the ionic strength is high, due to the presence of inorganic contaminants, the ζ potential is small and transport occurs mainly by electromigration. The contribution of each transport to the species flux is derived from the corresponding phenomenological relationship [9] and is extended to an isotropic porous medium using a vadose model.

The bulk liquid velocity, u_c , in cm min^{-1} is described as the sum of the contribution due to electroosmosis, u_{eo} , in cm min^{-1} and due to any pressure gradient, u_h , in cm min^{-1} . The electroosmotic velocity is calculated from the Helmholtz-Smoluchowski relation [10]

$$u_{eo} = \frac{\varepsilon \zeta}{\tau^2 \mu} \nabla \phi \equiv k_e \nabla \phi \quad (1)$$

where u_{eo} is the pore velocity due to electroosmosis in cm min^{-1} , ε the permittivity of the pore liquid in F cm^{-1} , ζ the zeta potential of the soil in V, τ a nondimensional tortuosity factor that accounts for the capillary path and is greater than 1, μ the viscosity of the pore liquid in $\text{Pa}\cdot\text{min}$, ϕ the electrostatic potential in V, and k_e in $\text{cm}^2(\text{V}\cdot\text{min})^{-1}$ an effective electroosmotic permeability coefficient used for convenience in the numerical formulation of the model.

The contribution of the pressure gradient is calculated from Darcy's Law

$$u_h = -\frac{k_h}{n\mu} \nabla p \equiv k'_h \nabla p \quad (2)$$

where u_h is the pore velocity in cm min^{-1} resulting from the pressure gradient, k_h the hydraulic permeability of the medium in cm^2 , n the porosity, and p the pressure in Pa. Again, an effective permeability coefficient k'_h in $\text{cm}^2(\text{Pa}\cdot\text{min})^{-1}$ is defined for convenience. For an individual species, the electromigration velocity [11], u_{em} , in cm min^{-1} is given by

$$u_{em} = -v z F \frac{\nabla \phi}{\tau^2} \equiv k_m \nabla \phi \quad (3)$$

where the migration velocity is relative to the pore liquid, v is the mobility defined as the velocity resulting from a unit force per mole, z is the charge number, F is Faraday's constant, and k_m is an effective mobility coefficient in $\text{cm}^2(\text{V}\cdot\text{min})^{-1}$.

Finally, the contribution of dispersion to the

species flux for the dilute solutions is given by Fick's law

$$j^d = -\frac{D}{\tau^2} \nabla C \quad (4)$$

where j^d is the molar flux due to dispersion in $\text{mol}(\text{cm}^2 \cdot \text{min})^{-1}$, C is the molar concentration in the pore liquid in mol , and D is the dispersion coefficient in $\text{cm}^2 \text{min}^{-1}$. Superposing the different mass-transfer mechanisms, the total flux of an individual species is given by

$$j = (u_{eo} + u_h + u_{em})C - \frac{D}{\tau^2} \nabla C \quad (5)$$

or in terms of the gradient of pressure, the electrostatic potential, and the concentration by

$$j = [(k_e + k_m) \nabla \phi + k_h \nabla p]C - \frac{D}{\tau^2} \nabla C \quad (6)$$

where j is the molar flux of species per unit pore area in $\text{mol}(\text{cm}^2 \cdot \text{min})^{-1}$. The coefficients k_e , k_m , k_h , and D/τ^2 may vary in space and time.

2.2. Change Equation and Charge Balance

The equation for concentration change is derived from the application of the conservation of mass in a control volume of a porous medium, which contains both the liquid and solid phases. Neglecting transport of adsorbed species, applying Gauss's divergence theorem, and taking the limit of infinitesimal control volume leads to

$$\frac{\partial(C + C^a)}{\partial t} + \nabla \cdot j = R \quad (7)$$

C^a in mol^{-3} is the molar concentration in the adsorbed phase, and R is the net volumetric rates of production of chemical species due to chemical reactions.

But since the total mass of each element is conserved

$$R = 0 \quad (8)$$

Then, Eq.7 can be rewritten as follows.

$$\frac{\partial(C + C^a)}{\partial t} + \nabla \cdot ((u_{eo} + u_h + u_{em})C - \frac{D}{\tau^2} \nabla C) = 0 \quad (9)$$

The molar concentration of an individual species in the adsorbed phase is demonstrated by

$$C^a = \frac{\rho}{n} K_d C \quad (10)$$

where K_d is the equilibrium sorption coefficient.

Substituting the pore velocity due to electroosmosis and the pressure gradient, as well as the electromigration velocity in Eq. 9 [12] yields

$$\frac{\partial C}{\partial t} (1 + \frac{\rho}{n} K_d) = -(\frac{\epsilon \zeta}{\tau^2 \mu} \nabla \phi - \frac{k_h}{n \mu} \nabla p - v_z F \frac{\nabla \phi}{\tau^2}) \frac{\partial C}{\partial x} + \frac{D}{\tau^2} \frac{\partial^2 C}{\partial x^2} \quad (11)$$

Assuming that the soil is neutral prior to contact with the pore liquid, the total charge density in the system can be obtained from the concentrations of species in the solution and in the solid phase. Here, since the solid phase is taken to include the thin double layer, the net charge in the soil and in the double layer must be zero.

2.3. Initial and Boundary Conditions

Suitable initial and boundary conditions are needed to obtain the numerical solution of Eq.11. ^{137}Cs and ^{60}Co were initially uniformly distributed in the soil column. Since the ^{137}Cs and ^{60}Co concentrations in the solution in the anode reservoir were zero and the solution constantly flowed through the soil column, the initial and boundary conditions were as follows.

$$C(0, x) = C_0 \quad 0 < x < 20 \text{ (cm)}$$

$$C(t, 0) = 0$$

$$\frac{\partial C}{\partial x}(t, 20) = 0$$

Also, each electrode was given an electric potential of 100 voltages.

$$\phi(t, 0) = 100 \text{ (V)}$$

2.4. Code Development

A numerical code was developed to predict the residual contaminant concentration in the soil in the column after the soil remediation by the electrokinetic method. An equation based on the governing equation, Eq.11, was rewritten in the FORTRAN 77 language. The Galerkin finite element method with a linear basis function, an implicit difference scheme for the space derivative, and a backward difference scheme for the time derivative were used. In addition, the Thomas algorithm was used for the matrix calculation. Meanwhile, to avoid any numerical oscillation, a space step (Δx) was selected so that P_e (Pecret number) > 2 , and a time step (Δt) was selected so that Cu_e (Courant number) < 1 .

3. Soil Remediation Experiment

3.1. Measurement of Input Parameters for Simulation

The main components in the TRIGA soil were SiO_2 , Al_2O_3 , and K_2O . In addition, the physical characteristics of the soil, such as the dry bulk density, the porosity, and the water content are listed in Table 2. The experimental computations

are as follows: dry bulk density (ρ_b) is the oven-dried mass of the sample divided by its field volume. Porosity (n) is obtained by $1 - (\rho_b / \rho_s)$. Particle mass density (ρ_s) is the oven-dried mass divided by the volume of the solid particles. Water content (θ) is volume of the water (V_w) divided by the total volume (V_T).

In order to obtain an equilibrium sorption coefficient, 6 sets of Cs^+ and Co^{2+} solutions, with different concentrations, were prepared and put into individual plastic bottles containing 10 g of soil. The temperature was 25 °C, 50mL of a Cs^+ solution for 10 g of soil was used, and the shaking time was 24 hours. Next, the concentration of Cs^+ remaining in each solution was measured by

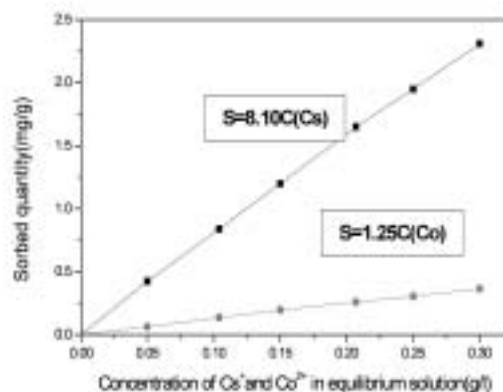


Fig. 1. Equilibrium Sorption Coefficients of Cs^+ and Co^{2+} for TRIGA Soil

Table 2. Parameters for Simulation

| Parameter | Values |
|---|--|
| Cell dimensions, L | $\phi 2.8\text{cm} \times 20\text{cm}$ |
| Bulk density, ρ_b | 0.76gcm^{-3} |
| Faraday 's constant, F | 96487C mol^{-1} |
| Potential at anode well, ϕ_1 | 100.0 V |
| Potential at cathode well, ϕ_2 | 0.0 V |
| Porosity, n | 0.61 |
| Tortuosity, τ | 1.2 |
| Equilibrium sorption coefficient, K_d | $8.10(Cs^+), 1.25(Co^{2+}) (\text{lk g}^{-1})$ |
| Dispersion coefficient, D | $0.08\text{ cm}^2/\text{min}$ |
| pH of soil | 4.3 |

means of atomic absorption spectroscopy (Analyst 300, from the Perkin Elmer Company). The quantity of Cs^+ sorbed in the soil was calculated. Under the above experimental conditions, the equilibrium sorption coefficient for Cs^+ was 8.10 kg^{-1} , while that for Co^{2+} was 1.25 kg^{-1} , as shown in Figure 1.

3.2. Remediation with Acetic Acid of TRIGA Non-radioactive Soil Intentionally Contaminated with Cs^+ and Co^{2+} for 3 Days

Non-radioactive soil around the TRIGA research reactor was sampled, and then soil particles with diameters of less than 0.063 mm were picked out by means of sieving. The minute soil was intentionally contaminated with 0.01 M of Cs^+ and Co^{2+} solution, and then it was dried for 3 days. The contaminated soil was remediated using the electrokinetic method. Figure 2 shows the electrokinetic experimental equipment on a

laboratory scale. The soil column was cylindrical in shape, with a 2.8 cm diameter and a 20.0 cm length. A titanium electrode, filter, stainless steel net, and reservoir were all established at both ends of the column and the volume of the column was 123 cm^3 .

The option chosen for restraining the pH increase in the soil column was to inject an acetate buffer into the soil column, while simultaneously adding an acidic solution to the cathode reservoir and the anode reservoir with an alkaline solution. CH_3COONa and CH_3COOH solutions were chosen as the alkaline and the acidic solutions, respectively.

Accordingly, a compound solution of 0.5 M of CH_3COONa and CH_3COOH was mixed with the contaminated soil to form an acetate buffer solution in the soil column. The TRIGA soil contaminated with Cs^+ and Co^{2+} was placed in a large bowl and mixed with the acetate buffer solution. Then, the saturated soil was injected into the soil column. The soil column was tapped

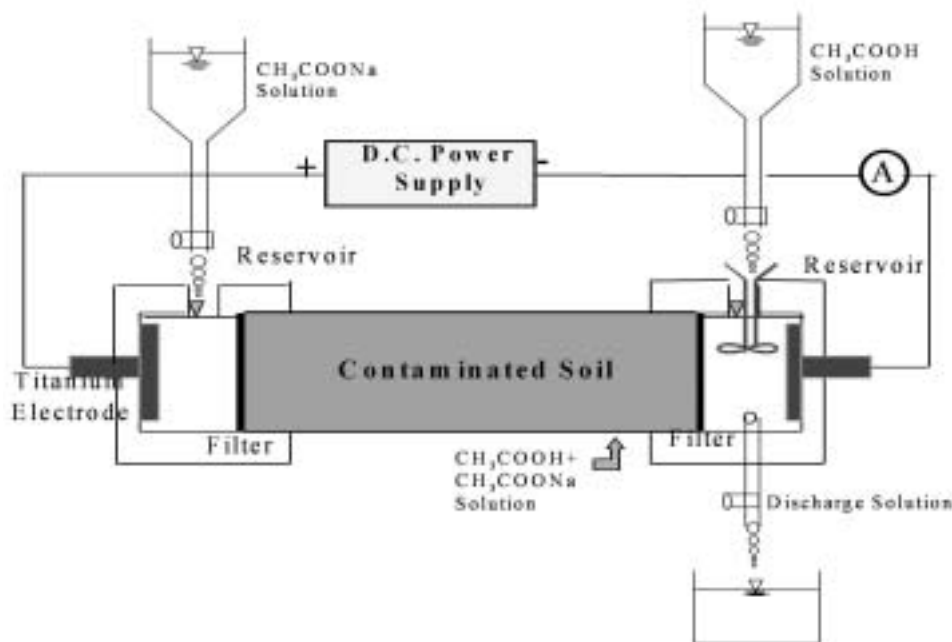


Fig. 2. Schematic Diagram of Cylindrical Column for Electrokinetic Remediation

continually until almost all the air bubbles had been removed and the weight of the TRIGA soil was 110 g. The volume of the buffer solution injected into the column was 77 ml. One side of the column was connected to an anode, while the other side was connected to a cathode. The voltage difference between the two electrodes was 100 V and the electric current was about 10 mA. Due to this pH control, a regular electrical potential gradient was expected. The experimental process was conducted as follows. After the mixed soil, including the acetate buffer solution, was injected into the column, the experiment was initiated under a constant electric voltage gradient. The volumes of the anode and cathode reservoirs were both 20 ml. 19 ml of a 0.5 M CH_3COONa solution was initially injected into the anode reservoir, while 13 ml of a 0.5 M CH_3COOH solution was injected into the cathode reservoir. The pore solution in the column flowed toward the cathode reservoir by electroosmosis. When the cathode reservoir was filled with about 19 ml of the pore solution released from the soil column, the experiment was suspended, and the volume of the pore solution in the cathode reservoir was accurately measured. Because the CH_3COONa solution in the anode reservoir continuously moved to the cathode by electroosmosis, this solution was continuously injected through the upper hole of the anode reservoir. This process was repeated many times until the experiment was completed. Thereafter, the TRIGA soil was divided into several sections, which were then analyzed for their Cs^+ and Co^{2+} concentrations using Atomic Absorption Spectroscopy.

3.3. Remediation with Acetic Acid of TRIGA Radioactive Soil Contaminated with ^{137}Cs and ^{60}Co for 15 Years

Radioactive soil from the TRIGA site

contaminated with ^{137}Cs and ^{60}Co , which has been stored in a radioactive waste storage at KAERI since 1988, was used for this experiment. The radioactive concentrations of ^{137}Cs and ^{60}Co in the soil were 14.7 Bq/g and 3.2 Bq/g, respectively. The radioactive soil was sampled from a drum, and then the soil particles with diameters of less than 0.063 mm were picked out by sieving. The option chosen for restraining the pH increase in the soil column is the same as that in the previous section. TRIGA soil contaminated with ^{137}Cs and ^{60}Co was placed in a large bowl and mixed with a compound solution of 0.5 M of CH_3COONa and CH_3COOH . Then, the saturated soil was injected into the soil column by tapping it in. A 0.5 M CH_3COOH solution was continuously injected into the anode reservoir, because the solution in the anode reservoir moved to the cathode by electroosmosis. A 0.5 M solution was injected into the cathode reservoir to restrain the pH increase. The experiment was initiated under a constant electric voltage gradient. After completion of the experiment, the TRIGA reactive soil in the column was divided into several sections, the ^{137}Cs and ^{60}Co radioactive concentrations of which were then analyzed using a multi channel analyzer (MCA)

3.4. Remediation with Sulfuric Acid of TRIGA Radioactive Soil Contaminated with ^{137}Cs and ^{60}Co for 15 Years

To increase the efficiency of remediation of the radioactive soil contaminated with ^{137}Cs and ^{60}Co , experiments were conducted to select an optimum additive by mixing the soil with many kinds of chemicals as follows.

Radioactive soil was sampled from a drum and then the soil particles with diameters of 0.063-1.0 mm were picked out by sieving. 20 g of soil was put into several plastic bowls and 20 ml of 0.5 M

of H_2O , Citric acid, NH_4NO_3 , FeCl_3 , $(\text{COOK})_2\cdot\text{H}_2\text{O}$, $(\text{NH}_4)_2\cdot\text{SO}_4$, $\text{H}_2\text{C}_2\text{O}_4\cdot\text{H}_2\text{O}$, Na_3PO_4 , Na_2SO_4 , Citric acid+ HNO_3 , HCl , and H_2SO_4 was added to each bowl. The bowls were attached to the scrubber equipment and were been scrubbing for 4hours. The soil remaining after screening solution from the scrubbing mixture was dried, and then radioactive concentrations of those were measured using a Multi Channel Analyzer (MCA). The efficiency of ^{137}Cs and ^{60}Co removal of each additive was analyzed by comparison of the radioactive concentrations of the initial soil with that of the scrubbing soil, and analysis results are shown in Figure 3. H_2SO_4 shows the highest efficiency for removal of ^{137}Cs and ^{60}Co from the soil, as shown in Figure 3. Therefore, H_2SO_4 was selected as the additive for this experiment.

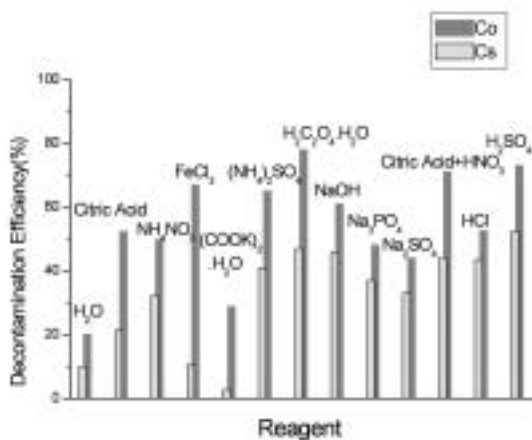


Fig. 3. Soil Decontamination Efficiency Versus Reagent

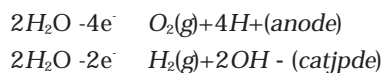
Next, radioactive soil was sampled from a drum and then the soil particles with diameters of less than 0.063 mm was picked out by sieving. TRIGA soil contaminated with ^{137}Cs and ^{60}Co was placed in a large bowl and mixed with a solution of 0.5 M of H_2SO_4 . Then, the saturated soil was injected into the soil column by tapping it in. A 0.5 M H_2SO_4 solution was injected into the cathode

reservoir to restrain the pH increase. The experiment was initiated under a constant electric voltage gradient. After completion of the experiment, the TRIGA reactive soil in the column was divided into several sections, the ^{137}Cs and ^{60}Co radioactive concentrations of which were then analyzed using MCA.

4. Results and Discussion

4.1. Remediation with Acetic Acid of TRIGA Non-radioactive Soil Intentionally Contaminated with Cs^+ and Co^{2+} for 3 Days

When the TRIGA soil was mixed with a NaCl solution instead of an acetate buffer solution to raise the electric field strength, the pH of the electrolyte in the anode reservoir decreased to a low value immediately after electrokinetic remediation, while that in the cathode reservoir increased to a high value. After remediation for 0.8days, the pH at a point 14 cm away from the anode filter in the soil column increased from the initial 4.0 to 11.7, and a precipitate with metal ions started to form at that point. Hydrogen ions evolved at the anode, while hydroxide ions evolved directly at the cathode during the experiment period. Metal ions migrated from the front of the column by electrokinetic remediation, formed a precipitate, and continuously accumulated at a 14 cm point in the column. The remediation efficiency was significantly decreased after 6.7 days, due to the small volume and lower concentration of the effluent.



When the TRIGA soil was mixed with an acetate buffer solution and a 0.5 M CH_3COOH solution was periodically injected into the cathode reservoir,

the pH of the effluent in the anode reservoir remained below 5.0. Consequently, no $\text{Co}(\text{OH})_2$ formed in the soil column, due to the lower pH.

About 3.54 pore volumes of effluent were discharged for 5.9 days, and the average effluent volume rate was 0.6 pore volume/day.

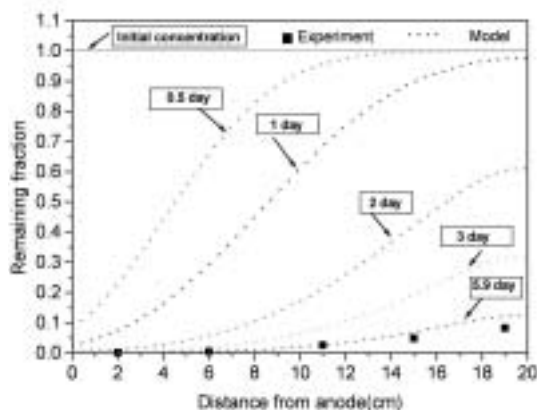


Fig. 4. Remaining Fraction of Cs^+ Concentration in Soil Column Versus Time

The total Cs^+ residual concentration in the column versus each remediation time is shown in Figure 4. Since no precipitate was formed in the column, due to the restraint of any pH increase, Fig. 4 showed a high remediation efficiency. Namely, 25.1% of the

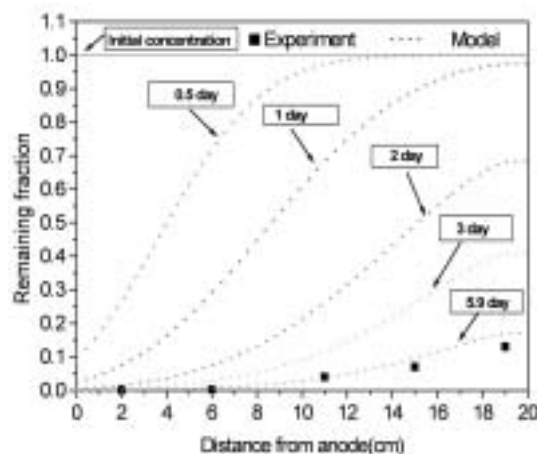


Fig. 5. Remaining Fraction of Cs^{2+} Concentration in Soil Column Versus Time

total amount of Cs^+ in the column was removed after remediation for a 0.5 day, 44.4% after 1.0 day, 76.1% after 2.0 days, 89.0% after 3.0 days, and 96% after 5.9 days. At the end of the run, the TRIGA soil was drawn out from the soil column and divided into several sections. The concentration of the pore solution was measured using atomic absorption spectroscopy. Meanwhile, Figure 5 shows the results of the Co^{2+} remediation, which exhibited a similar pattern to that of the Cs^+ remediation. Namely, 22.7% of the total amount of Co^{2+} in the column was removed after remediation for a 0.5 day, 44.4% after 1.0 day, 71.8% after 2.0 days, 85.2% after 3.0 days, and 94.4% after 5.9 days. Meanwhile, the values predicted by the proposed model for the residual concentration after remediation were similar to those obtained by experiment.

4.2. Remediation with Acetic Acid of TRIGA Radioactive Soil Contaminated With ^{137}Cs and ^{60}Co for 15 Years

When the TRIGA soil was mixed with an acetate buffer solution and a 0.5 M of CH_3COOH

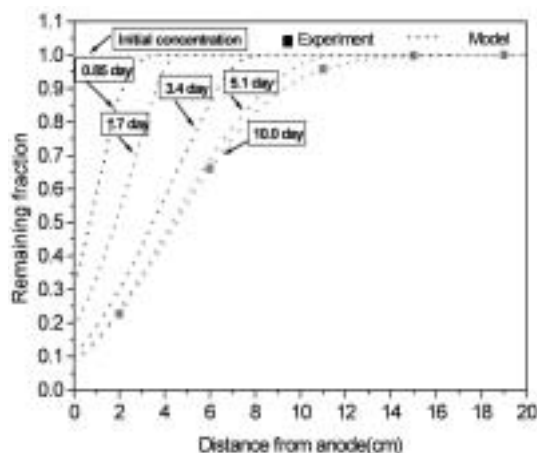


Fig. 6. Remaining Fraction of ^{60}Cs Concentration in Soil Column Versus Time (on using acetic acid)

solution was periodically injected into the cathode reservoir, the pH of the effluent in the anode reservoir remained below 5.0. Consequently, no $\text{Co}(\text{OH})_2$ formed in the soil column, due to the lower pH.

About 3.65 pore volumes of effluent were discharged for 10 days. The average effluent volume rates were 0.42 pore volume/day for 2 days, a 0.38 pore volume/day from 2 to 6 days, and a 0.32 pore volume/day from 6 to 10 days.

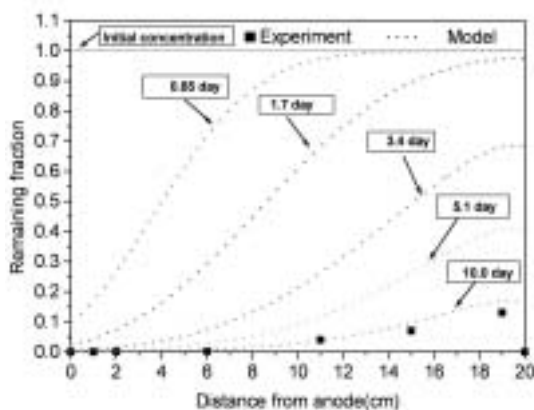


Fig. 7. Remaining Fraction of ^{60}Co Concentration in Soil Column Versus Time (on using acetic acid)

The remaining fraction of ^{137}Cs radioactive concentration in the column versus each remediation time is shown in Figure 6. Even if no precipitate formed in the column, due to the restraint of any pH increase, Figure 6 showed lower remediation efficiency. Namely, 6% of the total radioactive amount of ^{137}Cs in the column was removed after remediation for a 0.85 day, 11% after 1.7 days, 18% after 3.4 days, 23% after 5.1 days, and 25% after 10.0 days. At the end of the run, the TRIGA soil was drawn out from the soil column and divided into several sections. The radioactive concentration of the remediated soil was measured using MCA. The results show that the efficiency of removing ^{137}Cs

in the radioactive soil decreased somewhat, due to the ageing effect.

Meanwhile, Figure 7 shows the results of ^{60}Co remediation. Namely, 23% of the total amount of ^{60}Co in the column was removed after remediation for 0.85 day, 44% after 1.7 days, 72% after 3.4 days, 85% after 5.1 days, and 94% after 10.0 days. The efficiency of removing ^{60}Cs in the radioactive soil decreased slightly, due to the ageing effect.

4.3. Remediation with Sulfuric Acid of TRIGA Radioactive Soil Contaminated with ^{137}Cs and ^{60}Co for 15 Years

To increase the efficiency of the remediation of the radioactive soil contaminated with ^{137}Cs and ^{60}Co , a H_2SO_4 solution was used as an additive. When the TRIGA soil was mixed with a 0.5M of the H_2SO_4 solution and the H_2SO_4 solution was periodically injected into the cathode reservoir, the pH of the effluent in the anode reservoir remained below 5.0. Consequently, no formed in the soil column, due to a lower pH.

About 3.7 pore volumes of effluent were discharged for 10 days. The average effluent

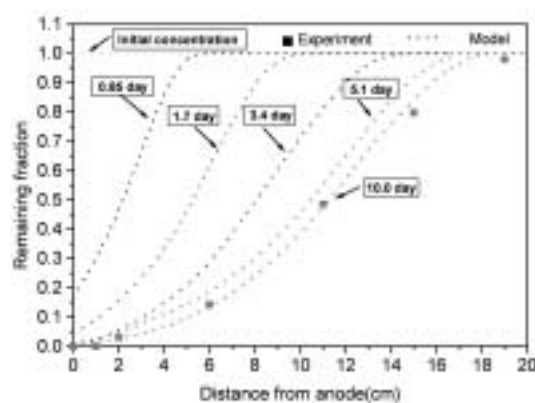


Fig. 8. Remaining Fraction of ^{137}Cs Concentration in Soil Column Versus Time (on using acetic acid)

volume rate was 0.42 pore volume/day for 2 days, 0.37 pore volume/day from 2 to 6 days, and 0.33 pore volume/day from 6 to 10 days.

The remaining fraction of ^{137}Cs radioactive concentration in the column versus each remediation time is shown in Figure 8. No precipitate formed in the column, due to the restraint of any pH increase. Figure 8 showed a higher remediation efficiency than Figure 6. Namely, 12.1% of the total radioactive amount of ^{137}Cs in the column was removed after remediation for 0.85 day, 25.8% after 1.7 days, 39.1% after 3.4 days, 50.0% after 5.1 days, and 53.7% after 10.0 days.

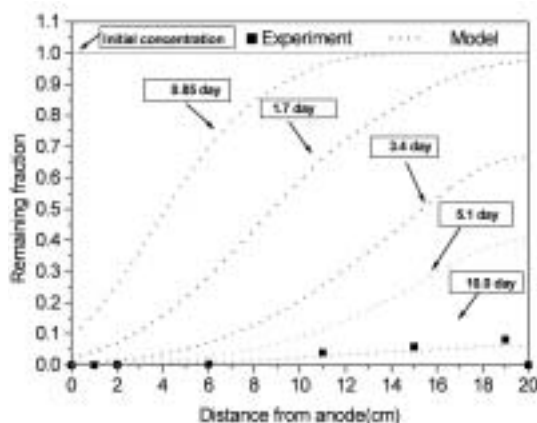


Fig. 9. Remaining Fraction of ^{60}Co Concentration in Soil Column Versus Time(on using acetic acid)

Figure 9 shows the results of ^{60}Co remediation. Namely, 23.7% of the total amount of ^{60}Co in the column was removed after remediation for 0.85 day, 45.4% after 1.7 days, 72.8% after 3.4 days, 87.7% after 5.1 days, and 97.0% after 10.0 days. Figure 9 showed a little higher remediation efficiency than Figure 7. Therefore, it was found that the efficiency of removing radionuclides from the radioactive soil in the column is proportional to the capability of the added chemical to extract the radionuclides.

5. Conclusions

Comparative experiments on radioactive soil contaminated with ^{137}Cs and ^{60}Co and non-radioactive soil intentionally contaminated with Cs^+ and Co^{2+} for a short time showed that the efficiency of removing ^{137}Cs in the radioactive soil decreased somewhat due to the ageing effect. H_2SO_4 used as an additive showed a higher removal capability for ^{137}Cs and ^{60}Co than other chemicals. The efficiency of removing radionuclides from the radioactive soil in the column was proportional to the capability of the added chemical to extract radionuclides. The volume of the soil wastewater discharged from the soil column by the electrokinetic method was 20% lower than that for soil washing.

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