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Vertical Electrokinetic-Flushing Remediation

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Abstract: An optimum reagent was selected to decontaminate the radioactive soil near nuclear facilities by using the developed vertical electrokinetic-flushing equipment, and the optimum remediation conditions were established to obtain a higher remediation efficiency and a minimum waste-solution volume. Namely, the acetic acid was selected as an optimum reagent due to its higher remediation efficiency. The average removal efficiency by a vertical electrokinetic-flushing remediation was 4.6% more than that by a vertical electrokinetic remediation. The optimum remediation conditions were suggested to be when the acetic concentration is 0.01 M, the injection rate of the reagent is 2.4 ml/g and the remediation period is 20 days for the vertical electrokinetic-flushing equipment of a 8.3 L volume. In these conditions, the removal efficiencies of Co^{2+} and Cs^{+} were 98.3% and 88.8%.

Keywords: Radioactive soil, removal efficiency, vertical electrokinetic-flushing

INTRODUCTION

The soil around the nuclear facilities in South Korea might be contaminated with radionuclides from a long-term operation of these facilities. Because these Korean nuclear facilities were constructed on a sandstone layer, the hydro-conductivity of the radioactive soil excavated from a nuclear site is high. It has been suggested that an electrokinetic-flushing remediation is a suitable technology in consideration of the soil characteristics near a Korean nuclear facility, which has merits of both an electrokinetic remediation and a soil flushing method (1).

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The electrokinetic process holds great promise for a remediation of contaminated soils, as it has a higher removal efficiency and it is effective for a low permeability. An electrokinetic remediation can be used to treat soils contaminated with inorganic species and radionuclides (2). The main mechanisms of a contaminant's movement in an electrical field involved in electrokinetic technology are an electromigration of ionic species and an electroosmosis. Electromigration probably contributes significantly to the removal of contaminants, especially at high concentrations of the ionic contaminants and/or a higher hydraulic permeability of a soil (3). The cathode reaction should be depolarized to avoid the generation of hydroxides and their transport in a soil. The selected liquids, also known as purging solutions, should induce favorable pH conditions in a soil, and/or or interact with the heavy metals, so that these heavy metals are removed from a soil (4). Most radioactive facility sites have been contaminated by the leakage of a radioactive waste-solution due to a corrosion of their concrete and pipes by a long-term operation of the waste-solution tanks and connection pipes, set up in the underground around nuclear power plants.

Electroosmosis moves a pore solution in response to an electric field, typically towards a cathode because of the negative surface charge of a soil. The magnitude of the transport velocity due to an electromigration and an electroosmosis is directly related to the electric voltage gradient. Also, the resulted remediation efficiency can be improved through an increase of the hydraulic potential gradient by a pumping. Recently, researchers have been investigating whether this method can be used to remove subsurface contaminants and they have compiled published research on the use of electrokinetic techniques to decontaminate fine-grained soils, and discussed some of the problems that occur during this process (5–7). Meanwhile, other researchers have also tried to develop soil flushing techniques in which soil-bound contaminants are transferred to a liquid phase by a desorption and a solubilization. Several flushing reagents have been investigated, such as water, acids, bases, chelating agents, alcohols and other additives (8). In practice, an acid washing and a chelator soil washing are the two most prevalent removal methods (9–10). Recently, the acetic acid or the sodium dodecyl sulfate were used as an electrolyte for an electrokinetic remediation to increase the removal efficiency of a metal (12–13). Ethylenediaminetetraacetic acid (EDTA) can attach to a metal ion for up to six sites, because each of the acetate groups and two nitrogen atoms have the free electron pairs necessary for a coordinate bond formation. The feasibility of this compound as a solubilizing or complexing agent has been reported in several works, especially due to its strong chelating ability for a variety of heavy metals (14). Also, because citric acid is relatively inexpensive, rather easy to handle, and has a comparatively low affinity for alkaline earth metals (Ca, K, and Mg), it is a suitable candidate for a soil washing (15). The resulting complexes are called chelates, which are usually more stable than those complexes with a single bond metal-ligand (16). HNO_3 and HCl show a significant potential to extract metal ions from a soil. However, their use is associated with a number of disturbing physical, chemical and biological properties (17). Oxalate was tested as a soil metal extractant because it is biodegradable, naturally occurring and relatively inexpensive, and forms moderately stable metal complexes (18). Moreover, oxalate is one of the strongest organic acids and therefore, it is able to attack and dissolve hydrous oxides (19). The acetic acid has been used for an electrokinetic-flushing, which consists of two carbons, four oxygens, and four hydrogens. And it can biodegrade easily in a soil (20). In order to select an optimum reagent suitable to a soil's characteristics, EDTA, oxalate, citric acid, and acetic acid have been selected as the candidate reagents.

Meanwhile, most of the electrokinetic equipments have been manufactured as a horizontal type. The horizontal type has two problems. First, the middle part of a soil cell forms an unsaturated zone, so that it reduces the removal efficiency of a radionuclide from a soil in the soil cell. Second, because an electrolyte in the anode compartment of a horizontal electrokinetic equipment is contaminated with radionuclide by a backward flow of an electroosmosis due to a change of the pH and the reagent type in a soil cell, the generation volume of the soil waste-solution is increased. In order to resolve such problems, Jing-Yuan Wang (2007) started to develop vertical electrokinetic equipment to remove the contaminants accumulated in a cathode, easily. This equipment has a cathode in the upper side so that the reagent in a soil cell might flow upward and the contaminants in a soil cell might be accumulated at the upper side. Upward vertical electrokinetic equipment has already been used to remove a heavy metal from kaolin (21) and to remove an organic material from a soil (22).

In this study, the ex-situ vertical electrokinetic-flushing equipment suitable to the geological characteristics of Korean nuclear facility sites was developed for a remediation of a contaminated soil. In order to obtain a higher removal efficiency during a short period, the optimum remediation conditions for the developed vertical electrokinetic-flushing equipment were obtained though ex-situ experiments with a change of various parameters.

MATERIALS AND METHODS

Soil Parameter Measurement

Korea currently has about 20 nuclear power plants and a research reactor. Because most nuclear facilities in Korea have been constructed on a hard sandstone rock, the contaminated soil around a nuclear facility contains a lot of sand, which has a higher hydro-conductivity. Soil was extracted from around a research reactor site for the experiments, which was not contaminated. And then it was artificially contaminated with Co^{2+} and Cs^{+} for the experiments. Hydraulic conductivity of the soil was measured by a constant head test method and the zeta potential of the soil according to the concentration of the acetic acid was measured with ELS-8000 ($-100 \sim 100\,\text{mV}$). Each parameter was obtained by the following equations.

$$\mathrm{n} = 1 - \frac{\rho_b}{\rho_s}$$

$$\theta = V_w / V_T$$

$$K = \frac{VL}{Ath}$$

Here, n is the porosity, ρ_b is the bulk density, ρ_s is the particle mass density, θ is the water content, V_w is the volume of water, and V_T is the total unit volume. Meanwhile, ρ_b is the oven-dried mass of a sample divided by it's the field volume. K is the hydraulic conductivity, and h is the hydraulic head.

The measurement results of the properties of the soil near the nuclear facility used in the experiments are shown in Table 1. Also, Table 2 shows the particle size percent of the soil near the nuclear facility. The saturation degree of the surface at the nuclear facility site was about 24 ~ 32 % and the hydro-conductivity of the soil was a little higher.

Table 1. Properties of the soil near the nuclear facility

Soil properties	Value
Saturation degree (pH)	
Surface	32.2% (6.9)
10 cm depth	27.3% (6.9)
30 cm depth	25.4% (6.1)
40 cm depth	23.7% (5.9)
50 cm depth	24.4% (5.9)
Porosity	0.40
Bulk density	1.54 (g/cm^3)
Hydraulic conductivity	1.8×10^{-3} cm/sec

Table 2. Particle size percent of soil near a nuclear facility

Soil class	Sieve no.	Diameter	%	Cumulative (%)
Clay + Silt	200	0.075	5.55	5.55
	100	0.15	4.87	10.42
	60	0.25	2.33	12.75
Sand	40	0.425	9.41	22.16
	20	0.85	8.88	31.04
	10	2	21.22	52.26
Gravel	4	4.75	38.93	91.19
	2	10	8.81	100.00

Manufacturing the Vertical Electrokinetic-Flushing Equipment

Electrokinetic-flushing equipment was manufactured, because an electrokinetic-flushing method is suitable for a soil near the domestic nuclear facilities, which has a little higher hydro-conductivity. This electrokinetic-flushing equipment is a combination of electrokinetic equipment and soil flushing equipment. Namely, the difference between the electrokinetic-flushing equipment and the electrokinetic equipment is that the electrokinetic-flushing equipment obtains a flushing remediation effect by an acid reagent as well as an electrokinetic remediation effect. The injection rate of a purging reagent can be controlled by changing the hydraulic head difference by an adjustment of the location of a waste-solution reservoir. The vertical electrokinetic-flushing equipment consists of a reagent reservoir, an electrokinetic soil cell, a soil waste-solution reservoir, a waste-solution treatment equipment, an equipment support, a quantitative pump, a power supply and so on shown as Fig. 1. Because the soil in a soil cell of the developed vertical electrokinetic-flushing equipment was located below the anode compartment, the anode electrolyte in this equipment flows downward by its gravitation, such an anode electrolyte should not be contaminated by radionuclides and the soil in a soil cell should be completely saturated with electrolyte.

Thus, the electro-osmosis direction in a soil cell of the vertical equipment is downward. In order to remove the hydrogen gas generated at the cathode electrode plate, the cathode electrode plate was slanted to 15°, so that the hydrogen gas may pass below the cathode electrode plate and then through a vertical discharge pipe into the atmosphere. This equipment can be operated for 24 hours and the resultant soil waste-solution is transferred to a waste-solution reservoir.

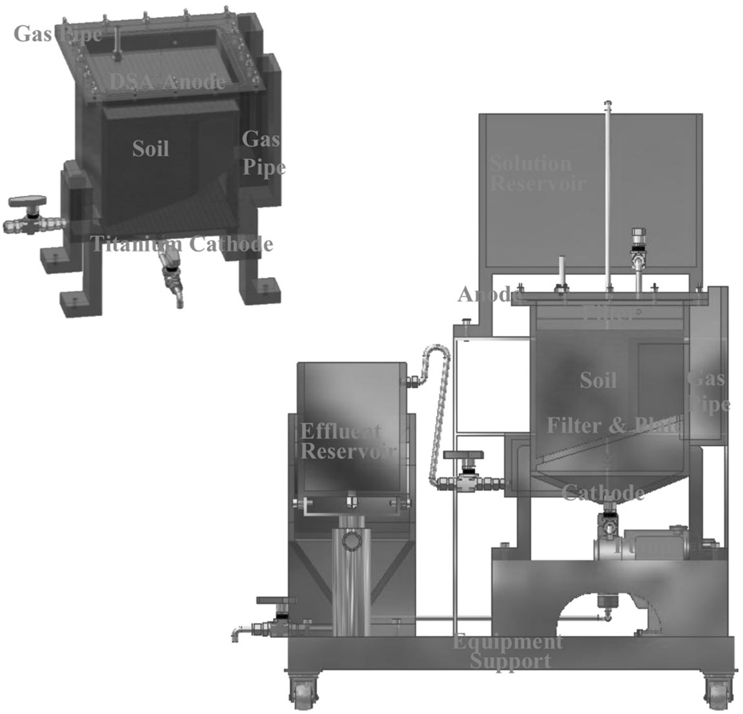

Figure 1. Schematic diagram of vertical electrokinetic-flushing equipment.

Experiments to Select an Optimum Reagent for a Vertical Electrokinetic Remediation

The soil near a nuclear facility which was not contaminated was also excavated, and it was artificially contaminated with 0.01 M of Co^{2+} and Cs^{+} for the experiments. This soil was mixed at a ratio of 1 g: 0.4 ml with a 0.01 M solution of Co^{2+} and Cs^{+} and the mixed soil was dried for more than one week. Each 5 g of dry soil was mixed with 10 ml of undiluted nitric acid solution, The mixed soil was heated at 150°C on a hot plate for 3 days, filtered by a 0.2 μm size filter, diluted to 50 ml, and then the concentrations of the Co^{2+} and Cs^{+} in the diluted solution were measured by AAS. The concentrations of Co^{2+} and Cs^{+} in the artificial soil were 238 mg/kg and 514 mg/kg, respectively. In order to maintain an equality of the soil size for several experiments, the soil near the nuclear facility was sieved by a No. 200 sieve (75 μm) and a No. 10 sieve (2000 μm), and then the soil with particles of $75 \sim 2000$ μm in size was used for experiments.

Electrokinetic-flushing remediation equation for a removal of metal ions from a soil cell is regarded as the following equation.

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

Where j is the molar flux of the species per unit pore area, k_o is the electro-osmotic permeability, k_m is the electro-migration coefficient, k_h is the hydraulic permeability, p is the hydraulic pressure, ϕ is the voltage potential, C is the molar concentration, D is the diffusion coefficient, and τ is the non-dimensional tortuosity.

Vertical electrokinetic-flushing equipment can perform both an electrokinetic remediation and an electrokinetic-flushing remediation by controlling the location of the soil waste-solution reservoir. Namely, in the case of the electrokinetic experiment, the effluent level in the soil waste-solution reservoir nearly accords with the reagent level in the anode compartment. But in the case of the electrokinetic-flushing experiment, the effluent level in the soil waste-solution reservoir is lower than the reagent level in the anode compartment, so that the injection rate of the reagent from the anode compartment may be increased. Namely, the reagent flushing process is generated in a soil cell by the hydraulic head difference. And the injection rate of a reagent from the anode compartment can be controlled by the hydraulic head difference between the reagent level in the anode compartment and the effluent level in the soil waste-solution reservoir.

Four candidate reagents, EDTA, oxalate, citric acid, and acetic acid, were used for a vertical electrokinetic remediation as shown in Table 3. An optimum reagent was determined from a comparison of the removal

Table 3. Removal efficiencies of Co^{2+} and Cs^{+} and effluent volume for vertical electrokinetic remediation with different reagent

Purging reagent	Experiment time (days)	Removal efficiency		Injection rate of reagent	Voltage gradient (V/cm)
EDTA	20	Co (%)	84.6	17294 ml	2
(8.3 L, 0.01 M)		Cs (%)	59.3	(1.4 ml/g)	
Oxalic acid	20	Co (%)	91.6	19444 ml	2
(8.3 L, 0.01 M)		Cs (%)	83.2	(1.6 ml/g)	
Citric acid	20	Co (%)	88.3	23450 ml	2
(8.3 L, 0.01 M)		Cs (%)	68.7	(1.9 ml/g)	
Acetic acid	20	Co (%)	94.2	19730 ml	2
(8.3 L, 0.01 M)		Cs (%)	83.7	(1.6 ml/g)	

efficiencies of Co^{2+} and Cs^{+} by vertical electrokinetic experiments with different reagent. The average pH of the soil in a soil cell for an electrokinetic remediation was changed to $2 \sim 3$.

Table 3 shows the experimental conditions for a vertical electrokinetic remediation with different reagent. For the experiment, a constant voltage of 31.4 was applied, so that the potential gradient may be 2 V/cm. The injected concentration of EDTA, oxalic acid, citric acid, and acetic acid into the electrode compartments was 1×10^{-2} M respectively, so the pH for both electrode compartments was around 3. Due to the electrolysis reaction at the electrodes, H^{+} at the anode and OH^{-} at the cathode compartment were generated, and the pH of the solution in the cathode compartment was increased up to 12. The top level of the acetic acid solution in the anode electrode compartments was maintained at the same level with that of the effluent in the soil waste-solution reservoir to avoid the formation of a hydraulic gradient across a soil, and the pore liquid was transported across a soil cell by only an electro-osmosis (electrokinetic remediation). About 8,300 $(23 \times 23 \times 15.7)\,cm^3$ of the soil saturated with 0.01 M acetic acid was placed in a soil cell and the total weight of the soil in a soil cell was 12,330 g. The electric current across a soil cell, as well as the flow rate and concentration of the pore solution, and the pH in the anode and the cathode compartments were measured periodically throughout the duration of an experiment. If the pH in a soil increases to more than 8, it is difficult to remove Co^{2+} from that soil due to the formation of $Co(OH)_2$. So HNO_3 was injected periodically into the cathode compartment in order to maintain the pH of the soil near the cathode at lower than 6. Also, after the completion of an experiment of 20 days, the soil in a soil cell was divided into 6 sections and dried for more than 7 days to analyze the removal efficiencies of Co^{2+} and Cs^{+} from the soil. Also, to measure the pH distribution in a soil cell, 10 g of a sample from each segment of a soil was mixed with 25 ml of distilled water, and the resultant slurry was stirred thoroughly and allowed to stand for a few minutes. The pH of the supernatant was then measured using a calibrated pH meter.

Fixation of the Remediation Conditions for a Vertical Electrokinetic-Flushing Remediation

For a vertical electrokinetic-flushing remediation, the position of the soil waste-solution reservoir was lowered to produce a difference between the reagent level in the anode compartment and the effluent level in the soil waste-solution reservoir and to increase the injection rate of a reagent (electrokinetic-flushing remediation). The injection rate of a reagent can

Table 4. Removal efficiencies of Co^{2+} and Cs^{+} and effluent volume for vertical electrokinetic-flushing under different remediation conditions

Purging reagent	Treatment time (days)	Removal efficiency		Injection rate of reagent	Voltage gradient (V/cm)	Remediation type
Acetic acid	20	Co (%)	94.2	19730 ml	2	Electrokinetic
(8.3 L, 0.01 M)		Cs (%)	83.7	(1.6 ml/g)		
Acetic acid	20	Co (%)	98.3	29596 ml	2	Electrokinetic-flushing
(8.3 L, 0.01 M)		Cs (%)	88.8	(2.4 ml/g)		
Acetic acid	20	Co (%)	99.2	59347 ml	2	Electrokinetic-flushing
(8.3 L, 0.01 M)		Cs (%)	90.1	(4.8 ml/g)		
Acetic acid	20	Co (%)	97.6	29468 ml	2	Electrokinetic-flushing
(8.3 L, 0.002 M)		Cs (%)	88.2	(2.4 ml/g)		
Acetic acid	20	Co (%)	98.0	29690 ml	2	Electrokinetic-flushing
(8.3 L, 0.05 M)		Cs (%)	86.7	(2.4 ml/g)		

be controlled by changing the hydraulic head difference by an adjustment of the position of the waste-solution reservoir. Three experiments were carried out with a change of the reagent injection rate, so that the optimum reagent injection rate may be determined for a higher removal efficiency and a minimum generation rate of a waste-solution as shown in Table 4. Vertical electrokinetic-flushing experiments were executed with 0.01 M acetic acid which was selected as a result of the following experiments under a potential gradient of 2 V/cm for 20 days. Next, the electrokinetic-flushing experiments were executed with the acetic acid of 0.002 M, 0.01 M and 0.05 M concentration to select the optimum concentration for a higher removal efficiency.

RESULTS AND DISCUSSION

Experiments to Select an Optimum Reagent for a Vertical Electrokinetic Remediation

Table 3 shows the experimental results with different reagents such as the removal efficiencies of Co^{2+} and Cs^{+} and the effluent volume. The initial pHs in the electrode compartments were near 3, because the concentration of the washing solution was 0.01 M. Also, because H^{+} was generated at the anode due to the electrolysis reactions, the pH in the anode compartment decreased to about 2.3. While the OH^{-} generated in the cathode compartment resulted in a pH increase from about the 1st day. If the pH in the cathode compartment increases to more than 8, hydroxides are formed in the soil and the pore electrolyte flow rate decreases. Generally, under low pH values, Co^{2+} exists in an ionic form in a solution and migrates towards a cathode due to a positive charge. Accordingly, in order to maintain the pH of the soil lower than 6, HNO_3 was injected periodically into the cathode compartment. After an experiment for 20 days, the pH distribution values in a soil cell were between 2.5 and 3.5 and the pHs at a 0.15 normalized distance showed slightly higher values. When the acetic acid was used as an electrolyte reagent, the pH in the anode compartment appeared to be the lowest value as shown in Fig. 2.

Figure 3 shows the total Co^{2+} removal efficiency versus the remediation time. The removal efficiency rate until 10 days were fast, while those after 10 days were slow. The removal efficiency of Co^{2+} from a soil cell with EDTA was 84.6%, that of cobalt with oxalic acid was 91.6%, that of Co^{2+} with citric acid was 88.3%, and that of Co^{2+} with acetic acid was 94.2%. The removal efficiency of Co^{2+} from the contaminated soil with the acetic acid for an electrokinetic remediation was the highest.

Figure 4 shows the total Cs^{+} removal efficiency versus the remediation time. The removal efficiency rates were almost constant for 20 days. Therefore it is predicted that the Cs^{+} desorption time was longer than the Co^{2+} desorption time and the total Cs^{+} removal efficiency increases lineally with an increase of the remediation days. Namely, in order to obtain a remediation efficiency of more than 90%, more remediation days are required. The removal efficiency of cesium from a soil cell with EDTA was 59.3%, that of Cs^{+} with oxalic acid was 83.2%, that of Cs^{+} with citric

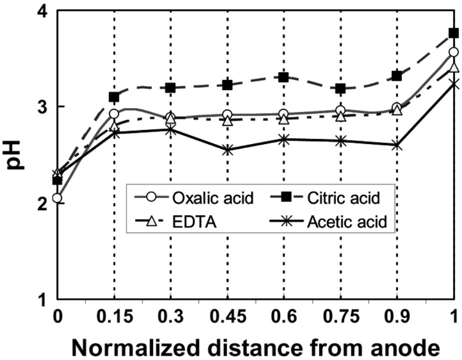

Figure 2. pH variation in a soil cell after a completion of vertical electrokinetic remediation versus reagent.

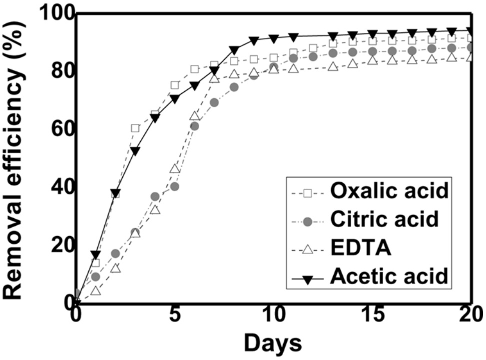

Figure 3. Co^{2+} remediation efficiency during vertical electrokinetic remediation versus reagent.

acid was 68.7%, and that of Cs^+ with acetic acid was 83.7%. Therefore, it was found that the removal efficiency of Cs^+ from a soil cell with the acetic acid was the highest. Results of the above experiments show that the removal efficiencies of Co^{2+} and Cs^+ with the acetic acid were the highest. It may be the reason that the pHs of the soil in the soil cell after a remediation using the acetic acid as an electrolyte reagent appeared to be the lowest value. Therefore, the acetic acid was selected as the optimum reagent and it was used for the following experiments. Also, the injection rate from the anode compartment for the electrokinetic remediation with the acetic acid was 1.6 ml/g, which means a ratio of the reagent of 1.6 ml per soil 1 g.

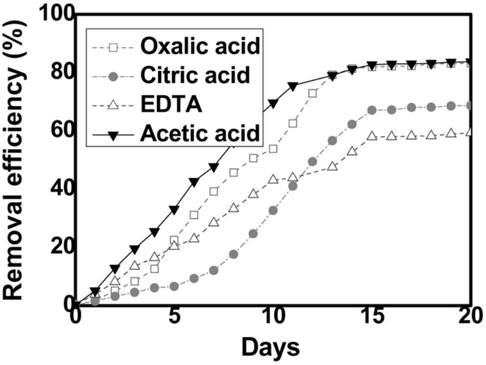

Figure 4. Cs^+ remediation efficiency during vertical electrokinetic remediation versus reagent.

Fixation of the Remediation Conditions for a Vertical Electrokinetic-Flushing Remediation

Table 4 shows the removal efficiencies of Co^{2+} and Cs^{+} and the effluent volume by a vertical electrokinetic-flushing under different remediation conditions. A comparison of the removal efficiencies between the vertical electrokinetic remediation and the vertical electrokinetic-flushing remediation for 20 days is shown in Fig. 5. The average removal efficiency of Co^{2+} and Cs^{+} by the vertical electrokinetic-flushing remediation was 4.6% more than that by the vertical electrokinetic remediation. Even if the reagent injection rate for the vertical electrokinetic-flushing remediation is increased to 4.8 ml/g from 2.4 ml/g, the average removal efficiency of Co^{2+} and Cs^{+} might be increased below 1% but a lot of effluent volume be generated as shown in Table 4 and Fig. 5. Therefore, it is suggested that the optimum reagent injection rate for a vertical electrokinetic-flushing remediation is 2.4 ml/g.

Figure 6 shows a comparison of the removal efficiencies versus the acetic acid concentration during a vertical electrokinetic-flushing remediation with different acetic acid concentrations. Figure 7 is the zeta potential of the soil versus the concentration of the acetic acid. When the reagent concentration was increased from 0.01 M to 0.05 M, the removal efficiency of Co^{2+} was slightly decreased by 0.3% and that of Cs^{+} was decreased by 2.1%. That reason is thought to be that the

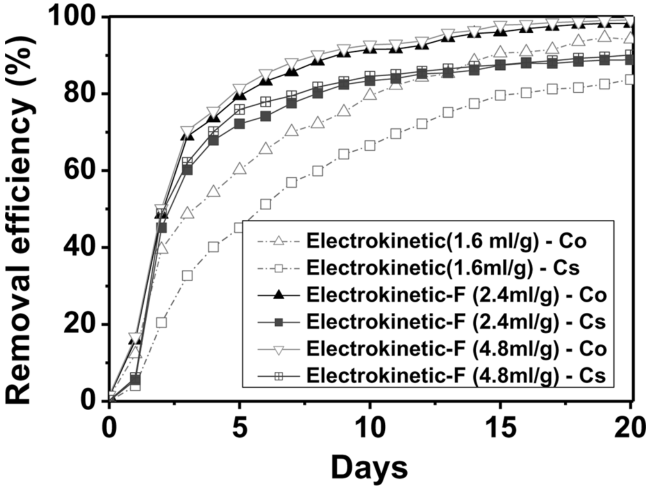

Figure 5. A comparison of removal efficiencies between a vertical electrokinetic remediation and a vertical electrokinetic-flushing remediation for 20 days.

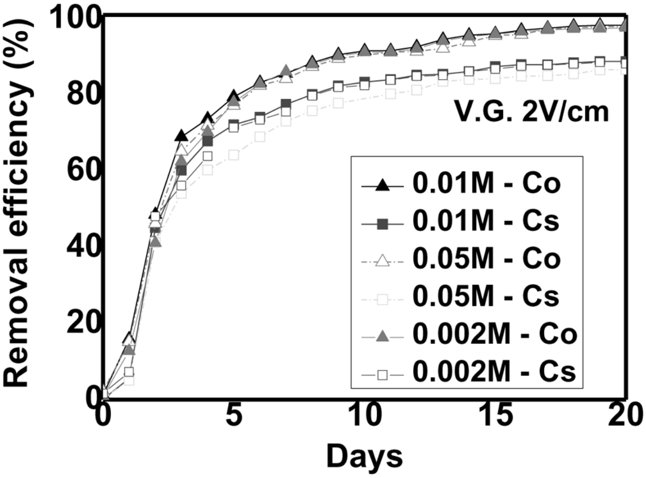

Figure 6. A comparison of removal efficiencies with time during vertical electrokinetic-flushing remediation with different acetic acid concentration.

negative zeta potential of the soil in the 0.01 M acetic acid was more than that in the 0.05 M acetic acid (23). Meanwhile when the reagent concentration was decreased from 0.01 M to 0.002 M, the average removal efficiencies of Co^{2+} and Cs^{+} were decreased by only 0.7%. Therefore, it is

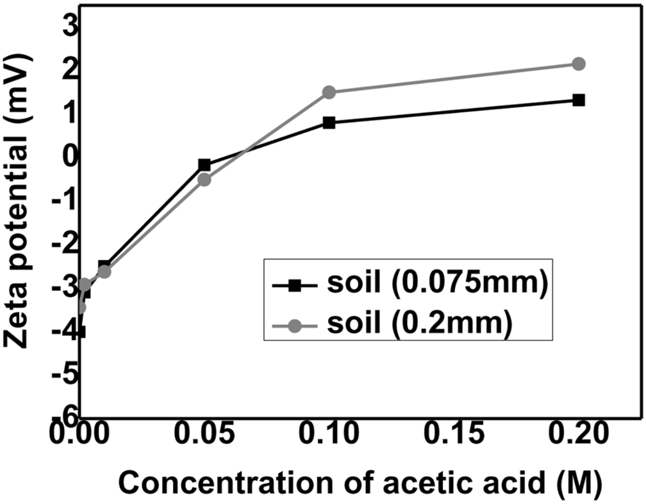

Figure 7. Zeta potential of the soil versus the concentration of acetic acid.

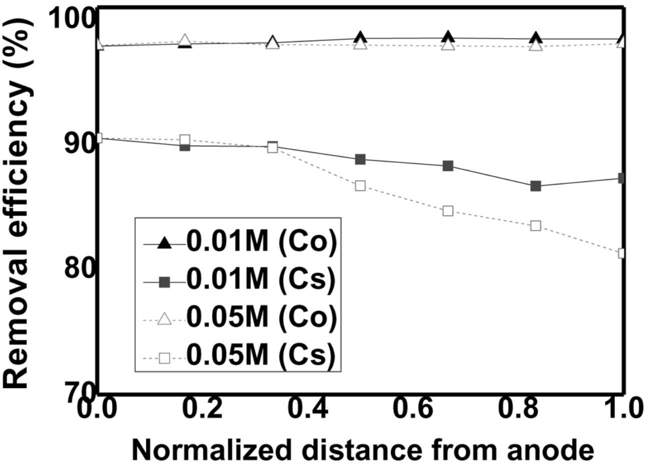

Figure 8. The cumulative removal efficiencies of Co^{2+} and Cs^{+} versus acetic acid concentration during vertical electrokinetic-flushing remediation.

suggested that the optimum concentration of the acetic acid for a vertical electrokinetic-flushing remediation is 0.01 M through Figs. 6 and 7. But in the future a comparison between the 0.01 M and 0.002 M concentrations will be reviewed from their economical aspects after the verification experiments with ^{60}Co and ^{137}Cs. Also, Fig. 8 shows the cumulative removal efficiencies of Co^{2+} and Cs^{+} versus acetic acid concentration after a vertical electrokinetic-flushing remediation for 20 days. The removal efficiency of Cs^{+} in the soil near the cathode electrode plate was decreased slightly as shown in Fig. 8. In conclusion, the optimum remediation conditions were suggested to be when the concentration of the acetic acid is 0.01 M, its injection rate is 2.4 ml/g, and the remediation period is 20 days for the vertical electrokinetic-flushing equipment of a 8.3 L volume.

Figure 9 shows the variation of the electrolyte flow rate versus the remediation time at the cathode compartment during a vertical electrokinetic-flushing. The electrolyte flow rate in a soil cell was reduced with the remediation time. Movement of the pore solution was mainly due to an electro-osmosis and the resultant hydro-head pressure. And when the 0.01 M and 0.05 M acetic acids were used as an electrolyte reagent, the average electrolyte flow rate was about 1,480 ml/day. Also, it was found that the effluent solution volume, 2.4 ml/g, generated from an electrokinetic-flushing remediation was much lower than that from a soil washing, 30 ml/g (2 times of scrubbing and a dilution for hydrocyclone work) (24).

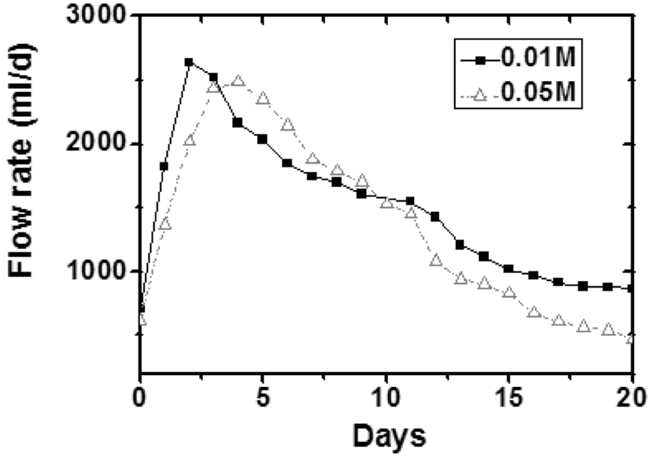

Figure 9. The effluent flow rate at the cathode compartment with time during vertical electrokinetic-flushing remediation with acetic acid.

CONCLUSION

Vertical electrokinetic-flushing equipment suitable to the geological characteristics of Korean nuclear facility sites was developed for the remediation of a contaminated soil. The optimum remediation conditions were obtained with experiments by using the developed vertical electrokinetic-flushing equipment, which can obtain a higher removal efficiency during a short period. The removal efficiencies of Co^{2+} and Cs^{+} from a soil cell with the acetic acid were the highest. It may be because the average pH in a soil cell appeared to be the lowest value, when the acetic acid was used as a purging reagent. When the results of the vertical electrokinetic remediation and the vertical electrokinetic-flushing remediation were compared, the removal efficiency by the vertical electrokinetic-flushing remediation was 4.6% more than that by the vertical electrokinetic remediation. Also, it was found that the optimum reagent injection rate for a vertical electrokinetic-flushing remediation was 2.4 ml/g. Meanwhile, when the reagent concentration was increased from 0.01 M to 0.05 M, the removal efficiencies of Co^{2+} and Cs^{+} were decreased. Therefore, the optimum remediation conditions were suggested to be when the concentration of the acetic acid is 0.01 M, its injection rate is 2.4 ml/g and the remediation period is 20 days for the vertical electrokinetic-flushing equipment of a 8.3 L volume. In these conditions, the removal efficiencies of Co^{2+} and Cs^{+} were 98.3% and 88.8%.

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REFERENCES

1. Kim, G.N.; Jung, Y.H.; Lee, J.J.; Moon, J.K.; Jung, C.H.; Chung, U.S. (2008) Development of electrokinetic-flushing equipment for a remediation of soil contaminated with radionuclides. *Journal of the Korean Radioactive Waste Society*, *6* (1): 1.
2. Reddy, K.; Xu, C.Y.; Chinthamreddy, S. (2001) Assessment of electrokinetic removal of heavy metals from soils by sequential extraction analysis. *J. Hazard. Mater.*, *B84*: 279.
3. Pamukcu, S.; Wittle, J.K. (1992) Electrokinetic removal of selected heavy metals from soil. *Environ. Prog.*, *11* (3): 241.
4. Reddy, K.; Chinthamreddy, S. (2003) Sequentially enhanced electrokinetic remediation of heavy metals in low buffering clayey soils. *J. Geotech. Geoenviron. Eng.*, *129* (3): 263.
5. Braud, F.; Tellier, S.; Astruc, M. (1998) Modeling of decontamination rate in an electrokinetic soil processing. *International Journal of Environmental Analytical Chemistry*, *68*: 105.
6. Kim, S.O.; Moon, S.H.; Kim, K.W. (2001) Removal of heavy metals from soils using enhanced electrokinetic soil processing. *Water, Air, and Soil Pollution*, *125*: 259.
7. Page, M.M.; Page, C.L. (2002) Electroremediation of contaminated soils. *Journal of Environmental Engineering. ASCE*, *128*: 208.
8. Chaiyaraksa, C.; Sriwiriyanuphap, N. (2004) Bach washing of cadmium, from soil and sludge by mixture of $Na_2S_2O_5$, and Na_2 EDTA. *Chemosphere*, *546*: 129.
9. Rampley, C.G.; Ogden, K.L. (1988) Preliminary studies for removal of lead from surrogate and real soils using a water soluble chelator, adsorption and batch extraction. *Environ. Sci. Technol.*, *32* (7): 987.
10. Sun, B.; Zhao, F.J.; Lombi, E.; McGrath, S. (2001) P. Leaching of heavy metals from contaminated soil using EDTA. *Environ. Pollut.*, *113*: 111.
11. Abumaizar, R.; Khan, L.L. (1996) Laboratory investigation of heavy metal removal by soil washing. *J. Air Waste Manage. Assoc.*, *46*: 765.
12. Kornilovich, B.; Mishchuk, N.; Abbruzzese, K.; Pshinko, G.; Klishchenko, R. (2005) Enhanced eletrokinetic remediation of metals-contaminated clay. *Colloids and Surfaces A:Physicochem. Eng. Aspects.*, *265*: 114.
13. Giannis, A.; Gidarakos, E.; Skouta, A. (2007) Application of sodium dodecyl sulfate and humic acid as surfactants on electrokinetic remediation of cadmium-contaminated soil. *Desalination*, *211*: 249.
14. Barona, A.; Aranguiz, I.; Elias, A. (2001) Metal associations in soils before and after EDTA extractive decontamination: Implications for the effectiveness of further clean-up procedures. *Environ. Pollut.*, *113*: 75.

15. Bassi, R.; Prasher, S.O.; Simpson, B.K. (2000) Extraction of metals from a contaminated sandy soil using citric acid. *Environ. Prog.*, *19* (4): 275.
16. Chao, J.C.; Hong, A.; Peters, R.W. (1998) Selection of Chelating Agents for Remediation of Radionuclide-Contaminated Soil. *Proceedings of the Conference on Hazardous Wastes Research, Utah, U.S.A.*
17. Tampouris, S.N.; Papassiopi. Paspaliaris, I. (2001) Removal of contaminant metals from fine grained soils, using agglomeration, chloride solution and pile leaching techniques. *J. Hazard. Mater.*, *B84*: 297.
18. Elliot, H.A.; Shastri, N.L. (1999) Extractive decontamination of metal-polluted soils using oxalate. *Water Air Soil Pollut.*, *110*: 335.
19. Stumm, W. (1992) *Chemistry of the Solid-Water Interface*; Wiley-Interscience: New York.
20. Kim, G.N.; Oh, W.Z.; Won, H.J.; Choi, W.K. (2003) Removal of cesium and cobalt from soil around TRIGA reactor using electrokinetic method. *J. Ind. Eng. Chem.*, *9* (3): 306.
21. Wang, J.Y.; Huang, X.J.; Kao, J.C.M.; Stabnikova, O. (2006) Removal of heavy metals from kaolin using an upward electrokinetic soil remedial (USER) technology. *Journal of Hazardous Materials*, *B136*: 532.
22. Wang, J.Y.; Huang, X.J.; Kao, J.C.M.; Stabnikova, O. (2007) Simultaneous removal of organic contaminants and heavy metals from kaolin using an upward electrokinetic soil remediation process. *Journal of Hazardous Materials*, *B144*: 292.
23. Vane, L.M.; Zang, G.M. (1997) Effect of aqueous phase properties on clay particle zeta potential and electroosmotic permeability: Implication for electrokinetic soil remediation process. *J. of Hazardous Materials*, *55*: 1.
24. Kim, G.N.; Choi, W.K.; Bung, C.H.; Moon, J.K. (2007) Development of a washing system for soil contaminated with radionuclides around TRIGA reactors. *J. Ind. Eng. Chem.*, *13*: 406.