Removal of 137Cs from contaminated soil using pilot electrokinetic decontamination equipment

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***Abstract*—*The removal efficiencies of 137Cs for 10 days were 50-70%. The removal efficiencies according to the elapsed time after 10 days were reduced. When an electric current density of 25-75 mA/cm2, sulfuric acid, nitric acid, acetic acid as electrolyte, a 0.5-2.0 cm/min hydraulic conductivity of soil were applied, respectively, the time required for the removal efficiency of 137Cs to reach 80% was 20-30 days. For improving removal efficiency of 137Cs from contaminated soil, it is necessary to increase an electric current density using sulfuric acid as an electrolyte and to decrease hydraulic conductivity of soil in the soil cell.*

Keywords*—*Decontamination, Electrokinetic equipment, 137Cs, Soil, Removal Efficiency.

I. INTRODUCTION

The contaminated soil around the underground radioactive waste solution tank of the TRIGA reactor has a slightly high hydro-conductivity and has been mainly contaminated with 137Cs due to corrosion of a connection pipe over the past 30 years. Recently, a soil washing method has been applied to remove 137Cs from the radioactive soil, but it appears that the removal efficiency of 137Cs from fine soil is lower and a lot of reagent effluent was generated [1,2,3]. An electrokinetic (EK) decontamination method provides a high removal efficiency of 137Cs and generates a little waste effluent. It was suggested that an EK decontamination method is a suitable technology for the removal of 137Cs from soil in consideration of the soil characteristics near the TRIGA reactor [4].

Most recently, EK field has been developed to enhance the remediation of contaminated soils [5,6,7]. Many extraction solutions such as diluted salt or diluted acid only dissolve a limited portion of the liable metals and metalloids in contaminated soils [8]. Recent studies reported that the EK, as an assisted method, was effective on promoting the release of heavy metals from solid-phase components and reducing the usage of washing solution [6,9]. The EK process involves introducing EK by inserting the electrodes into the contaminated soils [10].

Once the EK remediation process is over, extraction and removal of heavy metal contaminants are accomplished by electroplating at the cathode, precipitating at the cathode, pumping water near the cathode or anode, or reacting with ion exchange resins [11]. The efficiency of the EK method can be enhanced by coupling with other remediation technologies such as oxidation-reduction, bioremediation, permeable reactive barrier, and so forth [12,13]. While previous studies have shown that EK technique is effective for remediation of Cs in low-permeability soil such as clays [14, 15], a number of recent studies have also investigated the performance of EK remediation for Cs contaminated sandy soil around nuclear facilities in South Korea [16,17].

EK involves using a direct or alternating current with electrodes inserted into contaminated soils. When a low intensity electric field is applied, H+ is generated around the anode electrode through the effect of water electrolysis. As a result, more metals are demobilized under the acid condition around the anode electrode [18]. In addition, enhanced mobilization processes occur in soils resulting in the transport of metal ions from the anode to the cathode electrode [19]. Water present in soil is able to move towards the cathode through soil pores by electroosmosis while cations move to the cathode through electromigration [20]. The migration of ions makes it possible for the subsequent removal of soluble metals or immobilization with oxides, hydroxide and carbonates during the phytoremediation [21]. However, the detailed mechanisms of releasing/mobilization of metals/metalloids with EK coupled with phytoremediation are not clearly understood.

In this study, pilot EK decontamination equipment suitable for the geological characteristics of the TRIGA reactor site was developed for the removal of 137Cs from contaminated soil. The influences in which the hydraulic conductivity of soil in the soil cell, the electric current between electrodes, and the electrolyte in chambers reach the removal efficiency of 137Cs from soil were investigated experimentally using the manufactured pilot EK decontamination equipment in order to obtain a high removal efficiency of 137 Cs during a short period.

II. MATERIAL AND METHOD

2.1 Radioactive soil parameter measurement

South Korea currently has about 25 nuclear power plants and a research reactor. Because most of the nuclear facilities were constructed on hard sandstone rock, the contaminated soil around such facilities contains a lot of sand, which has a slightly high hydro-conductivity. Table 1 shows the soil properties near the TRIGA facility. The water content of soil at a nuclear facility site is about 26.7%, and the hydro-conductivity of the soil is a slightly higher.

TABLE 1
HYDRAULIC PARAMETERS OF TRIGA SOIL

	A	B	C
Bulk density (g/ cm^3)	1.57	1.55	1.51
Porosity (%)	41.2	39.7	40.8
Hydraulic conductivity (cm/sec)	1.8×10^{-2}	3.7×10^{-2}	7.4×10^{-2}
Water content (%)	26.7	25.7	24.5
pH	6.3	5.9	6.1

Fig. 1 shows the particle size distribution of the radioactive soil stored in the TRIGA facility, where the average particle size of the excavated radioactive soil is about 3.0 mm. The main nuclides of the radioactive soil are 137Cs, and this radioactive soil has been stored in many 200L drums in a storage facility for more than 30 years. Table 2 shows the contents of TRIGA soil components, which contain a lot of SiO2 and Al2O3.

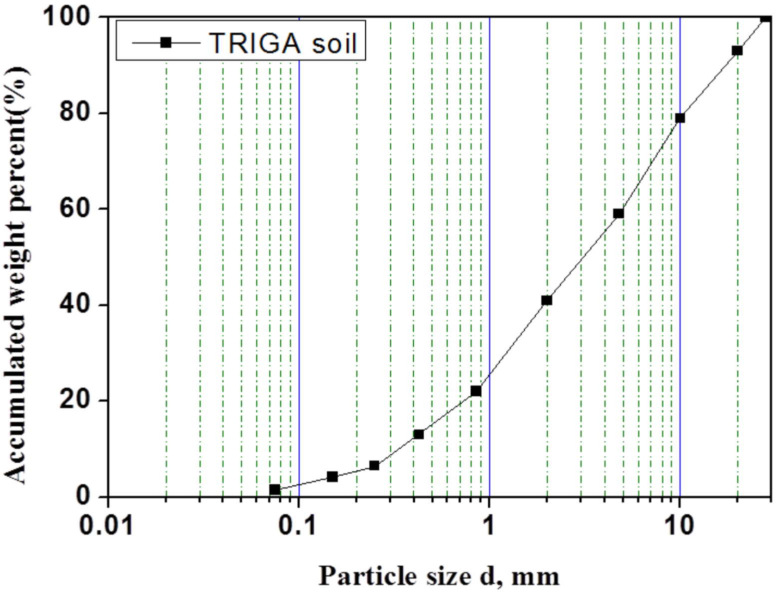

FIG. 1. THE PARTICLE SIZE DISTRIBUTION OF RADIOACTIVE SOIL STORED AT THE TRIGA FACILITY.

TABLE 2
CONTENTS OF THE TRIGA SOIL COMPONENTS.

Component	Content (%)
SiO_2	68.1
Al_2O_3	16.9
K_2O	7.87
Fe_2O_3	2.4
P_2O_5	1.54
CaO	1.49
Na_2O	0.95
MgO	0.23

2.2 Manufacturing of the pilot EK decontamination equipment

The pilot EK decontamination equipment of 24 L in size was manufactured for the decontamination of radioactive soil near the TRIGA reactor, as shown in Fig. 2. This pilot EK decontamination equipment consists of an EK soil cell of 24 L (40×40×15 cm), an anode electrode chamber (40×40×12 cm), a cathode electrode chamber (40×40×15 cm), a reagent reservoir (40×30×30 cm), an equipment support system, a pH controller, a power supply, and so on. A dimensional stable anode (DSA, 40×40 cm) was established in an anode electrode chamber to prevent the dissolution of the electrodes, and a titanium electrode was established in the cathode electrode room. The thickness of the soil cell was proportional to the decontamination period. In order to finish the soil decontamination experiment within 1 month, the thickness of the soil cell was determined to be 15 cm in consideration of pre-experiment results. The injection rate of the reagent can be controlled by changing the hydraulic head difference between the solution level in the anode electrode chamber and that in the cathode electrode room. In addition, the electro-osmosis direction of the soil cell was forward to the cathode ward, and 137Cs of the positive ions moved by electro-osmosis and electro-migration to the cathode ward.

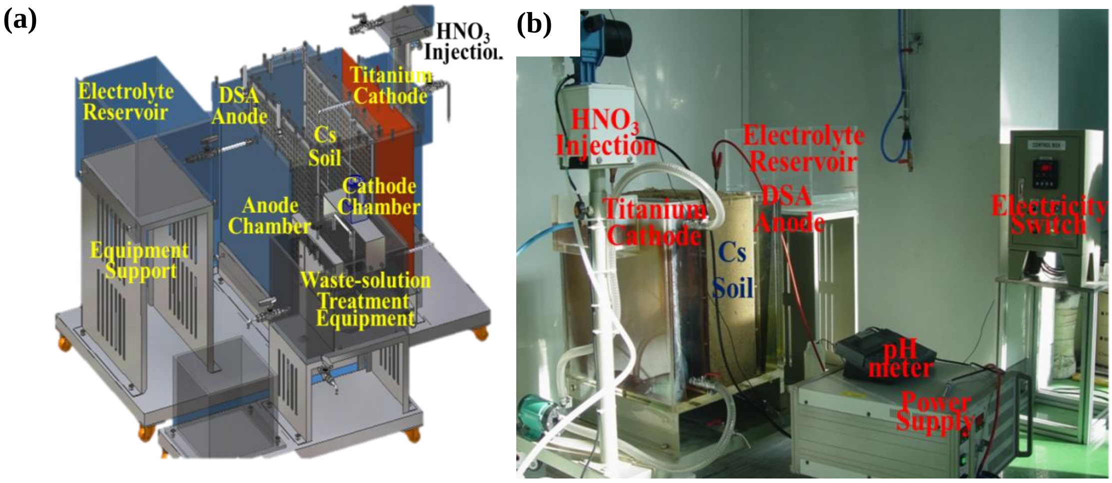

FIG. 2. (A) 3-D DESIGN DRAWING AND (B) MANUFACTURED PILOT SIZE OF ELECTROKINETIC DECONTAMINATION EQUIPMENT.

2.3 Decontamination experiment for radioactive soil

To verify the capability of the manufactured pilot EK decontamination equipment to decontaminate radioactive soil, radioactive soils that had been stored in a storage facility for more than 30 years were used for the experiments. This experiment was carried out under the following experiment conditions. The initial concentration of the soil contaminated with 137Cs is about 40 Bq/g. The hydraulic conductivity of soil in the soil cell is 0.5 cm/min, the electric current is 50 mA/cm2. The electric current between electrodes is 68-82 A, and the electric voltage between electrodes is 23-26 V. The temperature in the cathode chamber was 68-71℃. In addition, 0.3 M of H2SO4 was used as an electrolyte to accelerate the desorption of 137Cs from the soil. The period of electrokintic decontamination operation was 15-30 days. The EK decontamination experiment was stopped at the appointed times, and the rate and concentration of electrolyte effluent discharged from the cathode chamber were measured. After the decontamination experiments were completed, the concentrations of residual 137Cs of the decontaminated soil were measured. In addition, the removal efficiencies of 137Cs from the contaminated soil according to the lapsed decontamination time were calculated in consideration of the 137Cs flux rates discharged from the cathode chamber of the pilot EK equipment. The radioactivity concentrations of the electrolyte effluents were measured using an MCA.

First, the removal efficiencies according to the electric current change, i.e., 25, 50, and 75 mA/cm^2, were analyzed. Second, the removal efficiencies according to the change of electrolyte type, i.e., sulfuric acid, nitric acid, and acetic acid in chambers, were analyzed. Third, the removal efficiencies according to the change of hydraulic conductivity of soil in the soil cell, i.e., 0.5, 1.0, and 2.0 cm/min, were analyzed.

III. RESULTS AND DISCUSSION

The first paragraph under each heading or subheading should be flush left, and subsequent paragraphs should have a five-space indentation. A colon is inserted before an equation is presented, but there is no punctuation following the equation. All equations are numbered and referred to in the text solely by a number enclosed in a round bracket (i.e., (3) reads as "equation 3"). Ensure that any miscellaneous numbering system you use in your paper cannot be confused with a reference [4] or an equation (3) designation. Cesium (137Cs+) in the contaminated soil in the EK separation equipment was removed by electro-osmosis, electro-migration, and a hydraulic pressure flow, as in the following equation:

$$j = [\,(k_o + k_m)R\nabla I + k_h \nabla p]C - \frac{D}{\tau^2}\nabla C \qquad (1)$$

where j is the molar flux of the species per unit pore area, ko is the electro-osmotic permeability, km is the electro-migration coefficient, R is the electric resistance, I is the electric current, kh is the hydraulic permeability, p is the pressure, C is the molar concentration, D is the diffusion coefficient, and is non-dimensional tortuosity.

3.1 Experiment results according to the electric current

The results of EK experiment operated at an electric current density of 25, 50, 75 mA/cm2 are shown in Fig. 3. The removal efficiencies were proportional to the elapsed time. The removal efficiencies of 137Cs for 10 days were 60-70%. In addition, the removal efficiencies according to the elapsed time after 10 days were reduced. When 25 mA/cm2 was applied, the time required for the removal efficiency of 137Cs to reach 80% was 25 days. When 50 mA/cm2 was applied, the time required for the removal efficiency of 137Cs to reach 80% was 20 days. When 75 mA/cm2 was applied, the time required for the removal efficiency of 137Cs to reach 80% was 15 days. Even if a high electric current appears a high removal efficiency, the consumption rate of electricity for operation of the EK equipment increases. In addition, a gap between anode and cathode electrodes should be narrow to generate a high electric current. However, the narrower the width of the soil cell is, the smaller the soil volume per batch to be treated by the EK equipment is. Conclusively, it is considered that when the electric current increases, the 137Cs ion transport velocity is fast due to the increase of electro-osmosis and electro-migration in equation (1).

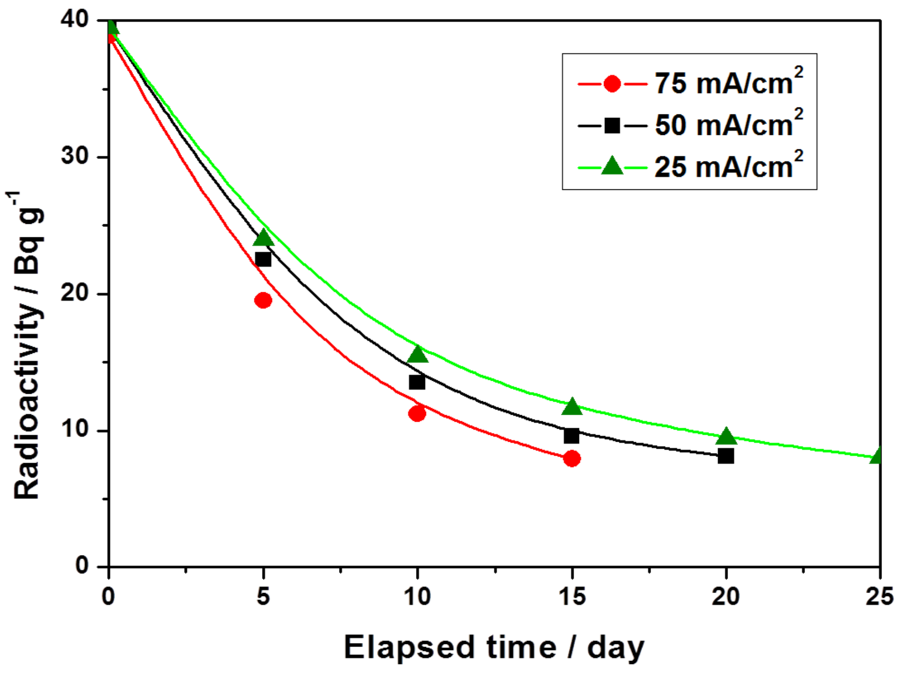

FIG. 3. EK EXPERIMENT RESULTS ACCORDING TO THE ELECTRIC CURRENT

3.2 Experiment results according to the electrolyte type in chambers

The results of the EK experiment operated by different electrolyte type in the anode and cathode chambers are shown in Fig. 4. The removal efficiencies were proportional to the elapsed time. The removal efficiencies of 137Cs for 10 days were 50-60%. In addition, the removal efficiencies according to the elapsed time after 10 days were reduced. When sulfuric acid was applied, the time required for the removal efficiency of 137Cs to reach about 80% was 20 days. When nitric acid was applied, the time required for the removal efficiency of 137Cs to reach about 80% was 25 days. When acetic acid was applied, the time required for the removal efficiency of 137Cs to reach about 80% was 30 days. Even if sulfuric acid shows the high removal efficiency, the sulfate gas was discharged due to the high temperature of electrolyte in the anode and cathode chambers. In addition, when the acetic acid as an electrolyte in the anode/cathode chambers is used, the electric current between the anode and cathode electrodes and the removal efficiency of 137Cs from soil decrease. Also, sulfuric acid desorbs more 137Cs from soil than nitric acid and acetic acid. Conclusively, it is considered that when the sulfuric acid as an electrolyte in the anode/cathode chambers is used, the 137Cs ion transport velocity is fast due to the increase of the electric current (I) and 137Cs concentration (C) in Eq. (1) by a strong acid.

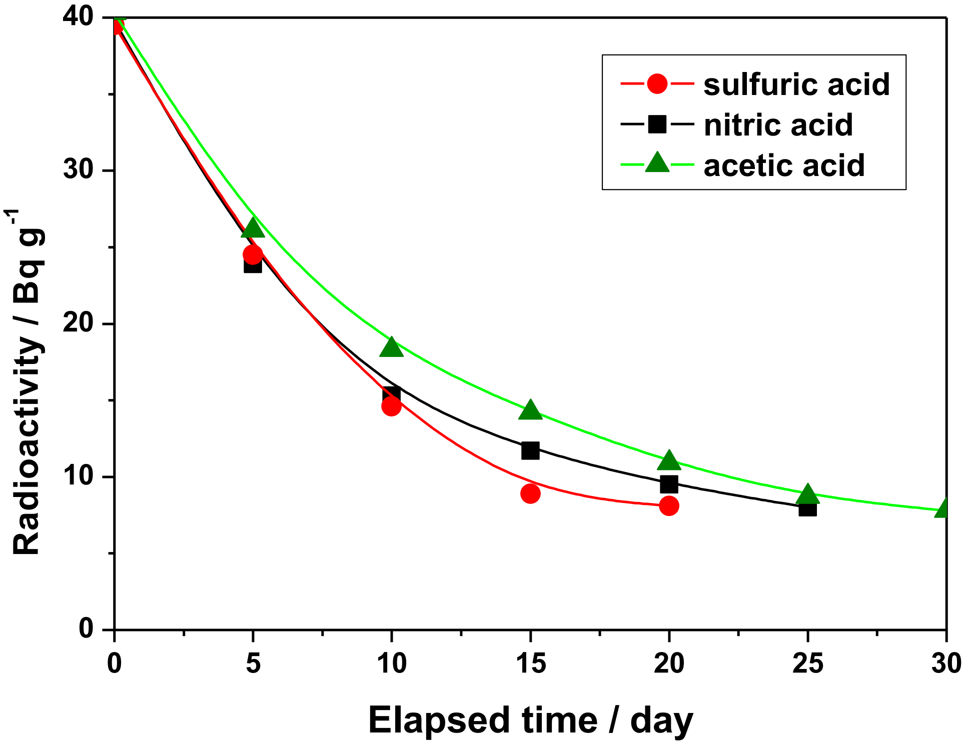

FIG. 4. EK EXPERIMENT RESULTS ACCORDING TO THE DIFFERENT ELECTROLYTE IN CHAMBERS.

3.3 Experiment results according to the hydraulic conductivity of soil

The results of the EK experiment operated at a 0.5, 1.0, 2.0 cm/min hydraulic conductivity of soil in the soil cell are shown in Fig. 5. The removal efficiencies were proportional to the elapsed time. The removal efficiencies of 137Cs for 10 days were 50-60%. In addition, the removal efficiencies according to the elapsed time after 10 days were reduced. When 0.5 cm/min of soil A was applied, the time required for the removal efficiency of 137Cs to reach about 80% was 20 days. When 1.0 cm/min of soil B was applied, the time required for the removal efficiency of 137Cs to reach about 80% was 25 days. When 2.0 cm/min of soil C was applied, the time required for the removal efficiency of 137Cs to reach about 80% was 30 days. For a reduction of hydraulic conductivity of soil in the soil cell, the soil in the soil cell should be hardened. Thus, electrolyte contaminated with 137Cs in the cathode chamber should be prevented from infiltrating into the clean electrolytes in the anode chamber. Conclusively, it is considered that when the hydraulic conductivity of soil reduces, the 137Cs ion transport velocity is fast due to the increase of electro-osmosis in Eq. (1) by fine particle of soil.

Therefore, for improving removal efficiency of 137Cs from contaminated soil, it is necessary to increase an electric current density using sulfuric acid as an electrolyte, and to decrease hydraulic conductivity of soil in the soil cell.

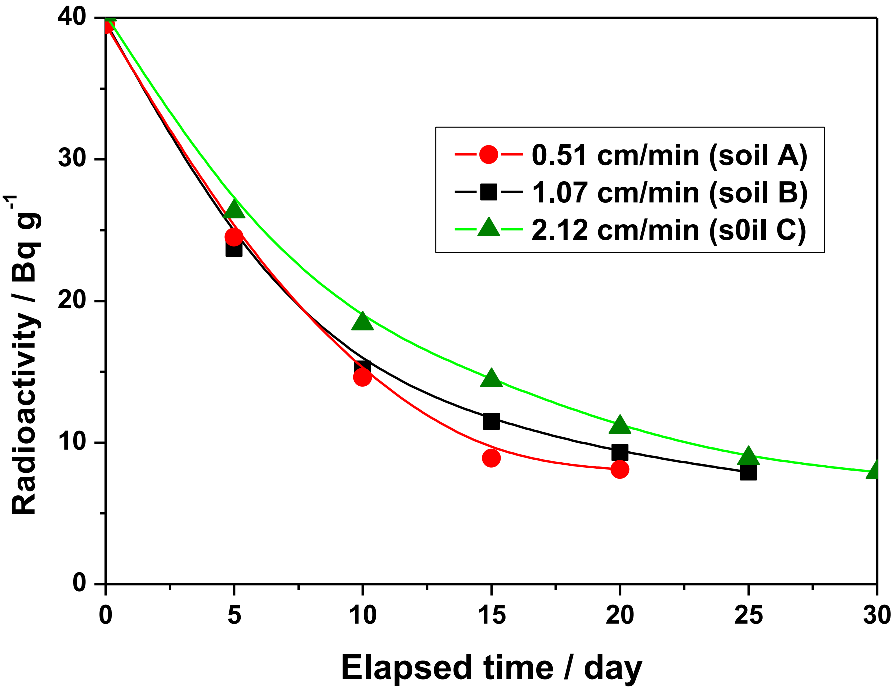

FIG. 5. EK EXPERIMENT RESULTS ACCORDING TO THE HYDRAULIC CONDUCTIVITY OF THE SOIL.

IV. CONCLUSION

To ensure a high-quality product, diagrams and lettering MUST be either computer-drafted or drawn using India ink. The removal efficiencies of 137Cs from contaminated soil in manufactured pilot EK decontamination equipment were proportional to the elapsed time. The removal efficiencies of 137Cs for 10 days were 50-70%. In addition, the removal efficiencies according to the elapsed time after 10 days were reduced. When an electric current density of 25, 50, and 75 mA/cm2 was applied, the time required for the removal efficiency of 137Cs to reach to 80% was 25, 20, and 15 days. In addition, when sulfuric acid, nitric acid, acetic acid as an electrolyte in the chambers was applied, the time required for the removal efficiency of uranium to reach 80% was 30, 25, and 20 days. Moreover, when a 0.5, 1.0, and 2.0 cm/min hydraulic conductivity of soil in the soil cell was applied, the time required for the removal efficiency of 137Cs to reach 80% was 20, 25, and 30 days.

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