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ORIGINAL PAPER

Decontamination of radioactive concrete using electrokinetic technology

Gye-Nam Kim · Wang-Kyu Choi · Kune-Woo Lee

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Abstract The experimental results of the electrokinetic decontamination with 0.01 M of nitric acid were that the cesium ions were removed by up to 52% from the concrete after 15 days, also the cobalt ions were only removed by up to 0.7%. The concrete should be washed with H_2SO_4 as a pretreatment before electrokintic decontamination to lower its pH below 4.0. The removal efficiencies of ^{60}Co and ^{137}Cs by nitric acid were increased by 3.1 and 2.5% more than those by acetic acid. The larger the particle size of the concrete, the more the removal efficiencies of ^{60}Co and ^{137}Cs were increased. Also, the removal efficiencies of ^{60}Co and ^{137}Cs by the application of an electric current of 20 mA/cm^2 were increased by 1.6 and 3.9% more than those by the application of 10 mA/cm^2. The removal efficiencies of ^{60}Co and ^{137}Cs from the radioactive concrete of 1,940 Bq/kg were 99.8 and 92.3% by electrokinetic decontamination after pretreatment by the application of an electric current of 20 mA/cm^2 for 25 days.

Keywords Electrokinetic decontamination · Concrete · Cobalt · Cesium · Nitric acid

1 Introduction

A great volume of radioactive concrete is generated during the operation and the decommissioning of nuclear facilities in the Republic of Korea. It has been taken care of in a radioactive waste storage house. The main radionuclides in the concrete are cobalt and cesium, and their concentrations are below 3,000 Bq/kg. If the radioactive concentration of the concrete is decontaminated below a self-disposal basis concentration (about 100 Bq/kg), the radioactive concrete can be disposed of in reclaimed land and this is cheaper than the disposal costs in a middle-low level radioactive repository. In this study, washing-electrokinetic technology was developed to decontaminate the concrete generated in nuclear facilities, which combined an electrokinetic and a washing method.

Even though many methods have been developed for the removal of radionuclides from contaminated concrete, their removal efficiencies of nuclides from a concrete are generally low. The electrokinetic process holds great promise for the decontamination of contaminated soils, as it has a high removal efficiency and it is time effective for a low permeability. The specific aspects of decontamination of concrete and mortar surfaces are analyzed such as the effect of chelating agents (EDTA, citric acid), zeta-potential on a concrete surface, effects of sorption and complex formation equilibrium, and specific advantages and problems of the electrokinetic decontamination process [1]. An electrokinetic technology has been used to treat concrete contaminated with radionuclides and to study its effect on chloride transport through cement-based materials [2]. It was shown that the combination of an electrokinetic treatment and the action of Trilon B solution made it possible to considerably enhance the efficiency of the removal of cesium from the surface of a hardened cement mortal [3]. Also, the electrokinetic decontamination experiments of concrete have determined that the loading of cesium and strontium on concrete may be decreased by using electrolyte solutions containing competing cations, while solubilization of uranium and cobalt require lixiviants containing complexing agents [4]. Nondestructive decontamination of mortar and concrete was performed by an electrokinetic method.

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The removal efficiency of Cs^+ was high, while that of Co^{2+} was low due to its precipitation in the high pH of concrete [5]. Therefore it was presented by results of this research that the pre-treatment of concrete by acetic solution was needed to dissolve cobalt precipitate. Radioactive concrete contaminated with long-lived nuclides, especially, uranium isotopes, can be cleaned by means of a liquid decontamination of fragmented wastes by solutions of sodium carbonate and hydrocarbonate. Increasing the temperature of the decontamination solution and imposing an electric field greatly accelerate the decontamination of wastes [6].

The main mechanisms of a contaminant's movement in an electrical field involved in electrokinetic technology are an electro-migration of ionic species and electro-osmosis of an effluent. Electro-migration probably contributes significantly to the removal of contaminants, especially at high concentrations of the ionic contaminants and/or a high hydraulic permeability of concrete [7]. The cathode reaction should be depolarized to avoid the generation of hydroxides and their transport in concrete. The selected liquids, also known as purging solutions, should induce favorable pH conditions in concrete, and/or interact with the heavy metals, so that these heavy metals are removed from concrete [8]. Electro-osmosis moves a pore solution in response to an electric field; in a soil the movement goes towards the cathode because of a negative surface charge of a soil. The magnitude of the transport velocity due to an electro-migration and electro-osmosis is directly related to the electric potential gradient. Recently, researchers have been investigating whether this method can be used to remove subsurface contaminants and they have published their research results on the use of electrokinetic techniques to decontaminate fine-grained soils, and discussed some of the problems that occurred during this process [9–11]. An equation for an electrokinetic process is as follows. The molar flux of the species is proportional to the electro-osmotic permeability (k_o) and the electro-migration coefficient (k_m). Also, the electro-osmotic permeability (k_o) is proportional to the zeta potential of the concrete (ζ).

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

$$k_o = \frac{\varepsilon\zeta}{\tau^2\mu} \tag{2}$$

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 pressure, C is the molar concentration, D is the diffusion coefficient, τ is the non-dimensional tortuosity, ε is the permittivity of the pore liquid, ζ is the zeta potential of the concrete, φ is the electric potential of the concrete and μ is the viscosity of the pore liquid.

Meanwhile, researchers have tried to develop washing techniques in which soil-bound contaminants are transferred to the liquid phase by desorption and solubilization. Several washing solutions have been investigated, such as water, acids, bases, chelating agents, alcohols and other additives [12, 13]. In practice, acid washing and a chelator soil washing are the two most prevalent removal methods used [14, 15]. Recently, acetic acid or sodium dodecyl sulfate has been used as an electrolyte for electrokinetic decontamination to increase the removal efficiency of a metal [16–18].

In this study, the electrokinetic decontamination of radioactive concrete was carried out, and then the removal efficiencies of ^{60}Co and ^{137}Cs were analyzed. In order to increase the removal efficiencies of ^{60}Co and ^{137}Cs from concrete, washing with a high concentration of H_2SO_4 has been executed as a pretreatment to lower its pH below 4. Firstly, when the concrete powder was washed with a high concentration of H_2SO_4, a lot of cobalt and cesium was removed and simultaneously, the pH of the concrete dropped to below 4. Secondly, the electrokinetic decontamination was applied to increase the removal efficiencies of the cobalt and cesium more, which resulted in high removal efficiencies of about 90%. Also, optimum decontamination conditions for electrokinetic decontamination were obtained through several experiments and the decontamination period needed for reducing the concrete radioactive concentration to a clearance concentration for self disposal was estimated.

2 Materials and equipment

2.1 Manufacturing of artificially contaminated concrete

The radioactive waste storage house of nuclear facilities has taken care of concrete debris having a particle size range of 20–50 cm in 200 L drums. Non-radioactive concretes in 200 L drums were selected and crushed by a crusher to below 0.5 cm (Fig. 1). The measurement results of hydraulic parameters of the crushed concrete were: bulk density (g/cm^3) was 1.66, porosity (%) was 39.5, and hydraulic conductivity (cm/s) was 2.4×10^{-3}. In order to fabricate an artificially contaminated concrete, the crushed concrete was combined and shaken with 0.01 M of Co^{2+} and Cs^+ and then it was dried for more than a week to create a uniform concentration. A simulated crushed concrete was used in this study.

2.2 Sampling and particle size distribution of the radioactive concrete

Figure 2 shows the particle size distribution of the radioactive concrete stored in a nuclear facility and the average

Fig. 1 **a** Concrete debris in a 200 L radioactive drum, **b** concrete particles ground up by a crusher

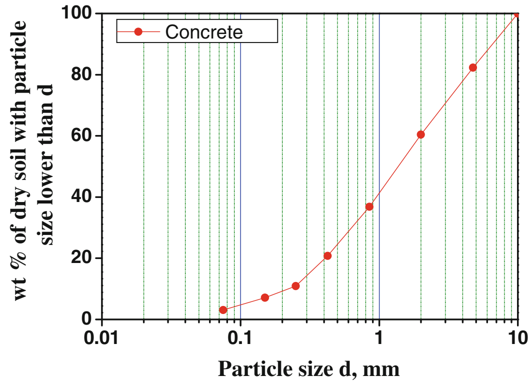

Fig. 2 Particle size distribution curve of the crushed radioactive concrete

particle size of the crushed radioactive concrete is 1.2 mm. The main nuclides of the radioactive concrete are ^{60}Co and ^{137}Cs and this radioactive concrete has been stored in many 200 L drums in a storage facility for more than 20 years. The smaller the concrete particle was, the higher its radioactivity concentration.

2.3 Manufacturing and experiment of a 1 L electrokinetic equipment size

Electrokinetic equipment of a 1 L size was manufactured for the experiment. It consists of an acryl concrete cell, two electrode compartments, an electrolyte solution reservoir, an effluent storage reservoir, and a power supply (Fig. 3). The contaminated concrete was placed into an acryl concrete cell of 10.0 × 11.5 × 15.0 cm. A 0.2 μm size paper filter was inserted between the electrode compartments to prevent an influx of the concrete particles into the electrode compartments. The electrolyte solution reservoir supplies the reagent solution to the anode compartment. The effluent storage reservoir receives the concrete waste-solution. An outlet was constructed at the bottom of the left electrode compartment to discharge the contaminated solution. The cathode electrode used titanium, and the anode electrode used a dimensional stable anode (DSA, titanium coated with platinum) to prevent the electrode from melting. The concrete in an acryl concrete cell was saturated with a reagent by solid–liquid ratio, 4:1. The pH in the cathode compartment and the generated waste solution (effluent) volumes in both electrode compartments were periodically measured.

3 Decontamination experiment

The experiment conditions and results with the manufactured electrokinetic equipment are shown in Tables 1 and 2. Effluent volume mL/g in Tables means the waste solution volume, mL, generated through an electrokinetic experiment per unit gram of concrete.

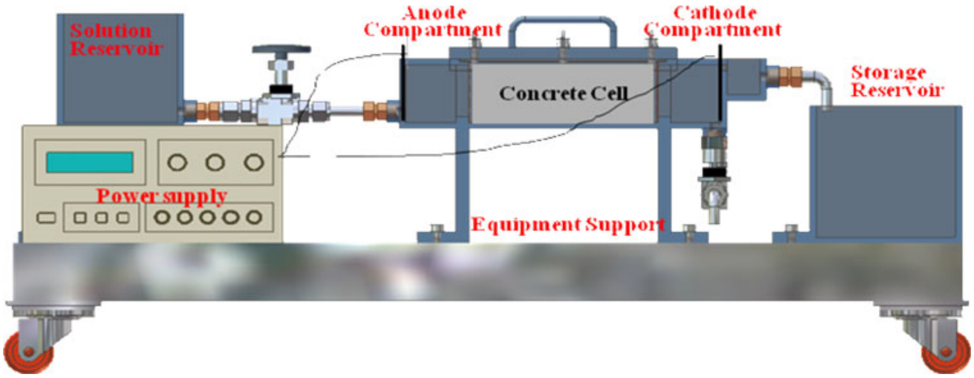

Fig. 3 The electrokinetic equipment of 1 L for concrete

Table 1 The Co^{2+} and Cs^{+} removal efficiencies and the effluent volumes for the electrokinetic decontamination without or with washing

Reagent (M)	Decon. time (days)	Initial con. (mg/kg)		Removal efficiency (%)		Electric current (mA/cm^2)	Particle size (mm)	Effluent volume (ml/g)	Remark
Nitric acid (0.01 M)	15	Co	247	Co	0.7	20	0.6	2.2	
		Cs	382	Cs	52.0				
Nitric acid (0.01 M)	15	Co	245	Co	99.6	20	0.6	2.3	After washing with H_2SO_4
		Cs	387	Cs	99.3				

Table 2 The ^{60}Co and ^{137}Cs removal efficiencies and the effluent volumes for the electrokinetic decontamination with different experimental conditions

Reagent (M)	Decon time (days)	Initial con. (Bq/kg)		Removal efficiency (%)		Electric current (mA/cm^2)	Particle size (mm)	Effluent volume (ml/g)	Removal efficiency after H_2SO_4 washing (%)
Nitric acid (0.01 M)	20	Co	420	Co	99.6	20	0.6	2.8	Co
		Cs	560	Cs	88.2				Cs
Acetic acid (0.01 M)	20	Co	420	Co	96.5	20	0.6	2.7	Co
		Cs	560	Cs	85.7				Cs
Nitric acid (0.01 M)	15 20	Co	420	Co	99.7	20	1.2	2.9	Co
		Cs	560	Cs	90.5				Cs
Nitric acid (0.01 M)	20	Co	420	Co	98.0	10	0.6	2.7	Co
		Cs	560	Cs	84.3				Cs

3.1 Electrokinetic decontamination of the artificially contaminated concrete without washing

Acetic acid has been used for electrokinetic decontamination which consists of two carbons, four oxygens, and four hydrogens [19]. The difference of decontamination efficiencies by nitric acid and acetic acid is not significant as shown in Fig. 4. But the electricity consumption on usage of nitric acid can be reduced due to its higher electrolytic conductivity. Namely, at the same concentration (0.01 M) and voltage (17 V), an electric current in a concrete cell on the usage of nitric acid (20 mA/cm^2) was generated at about 20 times higher than that on the usage of acetic acid (1 mA/cm^2) as shown in Fig. 4. The electricity consumption can be reduced by almost 20 times on the usage of nitric acid. Therefore, nitric acid was selected as an optimum reagent for this electrokinetic decontamination.

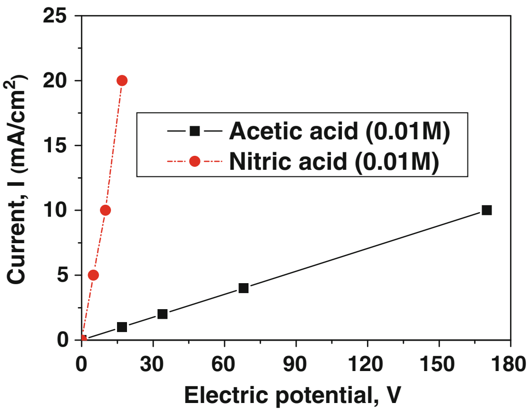

Fig. 4 A relation between electric potential and electric current with acetic acid and nitric acid in the electrokinetic equipment

Electrokinetic decontamination was executed with nitric acid as a reagent for the artificially contaminated concrete for 15 days. The artificially contaminated concrete was saturated with 0.01 M of nitric acid and the pH in a concrete cell was 12.0. The crushed concrete was put into a concrete cell of the 1 L electrokinetic equipment like that shown in Fig. 3. When the concentration of nitric acid was more than 0.01 M, the zeta potential of the concrete was positive and the nitric acid solution in the concrete cell flowed to the anode compartment due to an electro-osmosis. Therefore, the electrode locations in this experiment are the reverse of those of the concrete electrokinetic decontamination. The electric current across a concrete cell, the flow rates of the waste solution (effluent) and the concentrations of the waste solution in both electrode compartments, and the pH in the cathode compartment were measured periodically throughout the duration of the experiment. The pH in the cathode compartment increases with time elapse due to the generation of hydroxide ion. On the other hand, the pH in the anode compartment decreases with time elapse due to the generation of hydrogen ion as the following equations show:

$$2H_2O \rightarrow 4e + O_2(g) + 4H^+ : \text{Anode} \tag{3}$$

$$2H_2O \rightarrow 4e + H_2(g) + 2OH^- : \text{Cathode} \tag{4}$$

If the pH in the concrete cell increases to more than six, it is difficult to remove Co^{2+} from the concrete due to the formation of $Co(OH)_2$. Therefore, in order to maintain the pH of the concrete near the cathode to lower than four, nitric acid was periodically injected into the cathode compartment. Also, after the completion of an experiment for 15 days, the concrete in a concrete cell was divided into six sections and dried for more than 3 days to analyze the removal efficiency of the cobalt and cesium ions from the concrete. Each 5 g of dry concrete and about 10 mL of undiluted nitric acid solution were combined, and heated at 150 °C on a hot plate for 3 days, and filtered by a 0.2 μm size filter, and diluted to 50 mL, and then the concentrations of the cobalt and cesium ions in the diluted solution were measured by atomic absorption spectrophotometer (AAS).

3.2 Electrokinetic decontamination after washing as a pretreatment

The results of the previous electrokinetic decontamination showed that it is impossible to completely remove all cobalt and cesium from the concrete by only using the electrokinetic technology. So, washing-electrokinetic decontamination which combines washing technology and electrokinetic technology was used. Namely, the concrete was washed with a high concentration of sulfuric acid, and then it was decontaminated by electrokinetic technology. Firstly, the concrete was washed with 3.0 M of sulfuric acid with a 2.0 mL/g ratio in a scrubbing tank for 4 h. The $CaCO_3$ in the cement of the concrete was decomposed to CO_2 by a reaction with sulfuric acid. The pH of the concrete was reduced to below four. The sulfuric acid solution from the mixture was removed, and the remaining concrete was put into the 1 L electrokinetic equipment. The electrokinetic decontamination was executed with 0.01 M of nitric acid as an electrolyte reagent under 20 mA/cm^2 for 14.83 days (washing: 0.17 days). The electrode location in this experiment was in the same direction as that of the concrete electrokinetic decontamination. Namely, the left is the anode compartment, and the right is the cathode compartment (Fig. 3). Because the pH of the concrete was lowered to below four due to the sulfuric acid washing, the effluent reagent flowed to the cathode compartment. 1,600 g of concrete was put into the concrete cell, whose pH was lowered to below four. The electric current across the concrete cell, the flow rate and the concentration of the waste solution in the cathode compartment, and the pH in the cathode compartment were measured periodically throughout the duration of the experiment. In order to maintain the pH in the concrete near the cathode at lower than four, nitric acid was injected periodically into the anode compartment. Meanwhile, after the completion of the experiment for 15 days, the concrete in a concrete cell was divided into six sections and dried for more than 3 days to analyze the removal efficiency of the cobalt and cesium ions from the concrete.

3.3 Decontamination of radioactive concrete

In order to verify the decontamination capability for radioactive concrete by the developed electrokinetic equipment, the radioactive concrete that had been stored in a storage facility for more than 20 years was used for the experiments. The radioactivity concentration was measured by multi-channel analyzer (MCA) with a standard tube of 50 cc, QCY48 (Amersham), manufactured by Korea research institute standards and sciences (KRISS). The time required to measure the radioactivity concentration of a concrete sample of 20 g by MCA was estimated to be more than 8 h. The original radioactivity concentration of the concrete before decontamination was calculated by averaging the values measured arbitrarily at three positions on the radioactive concrete, and that of the concrete after electrokinetic decontamination was calculated by averaging the values at four sections divided by an equal-interval in a concrete cell. The electrokinetic decontamination was stopped at decided times, and four concrete samples were extracted from a concrete cell, and then the radioactivity concentration of concrete samples were measured by MCA. The concrete samples were returned to their original positions in a concrete cell, and then the experiment was started again. The removal efficiency of nuclides is calculated as a ratio of the original concrete concentration and the concrete concentrations measured at decided times. Finally, the decontamination period for reducing the concrete radioactivity concentration to a clearance concentration (about 100 Bq/kg) was estimated through the experimental results. Table 2 shows the electrokinetic decontamination conditions and the results. The verification experiments were executed for 20 days and an electric current of 20 mA/cm^2 was applied across the concrete cell. The volume of the concrete waste solution generated every day was measured as shown in Table 2, and the pH in the cathode compartment was maintained at below four by using nitric acid.

4 Results and discussion

The experimental results of the electrokinetic decontamination with 0.01 M of nitric acid are shown in Fig. 5.

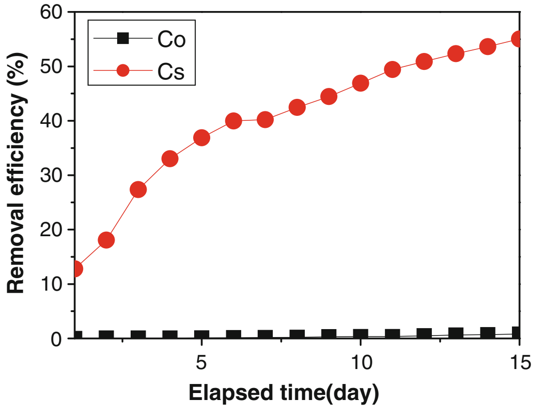

Fig. 5 Co^{2+} and Cs^{+} removal efficiencies along elapsed time with the 0.01 M nitric acid for electrokinetic decontamination without acid washing

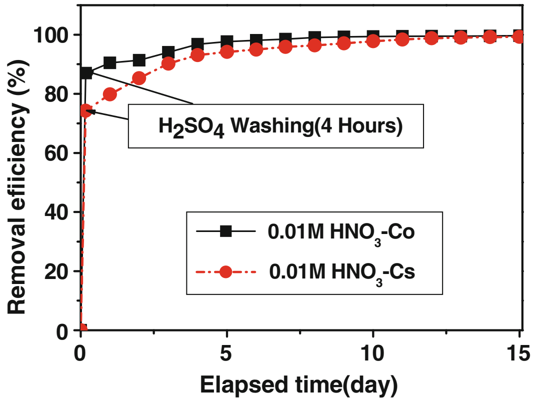

Fig. 6 Co^{2+} and Cs^{+} removal efficiencies along elapsed time with the 0.01 M nitric acid for electrokinetic decontamination after washing with 3 M of H_2SO_4 for artificially contaminated concrete particles

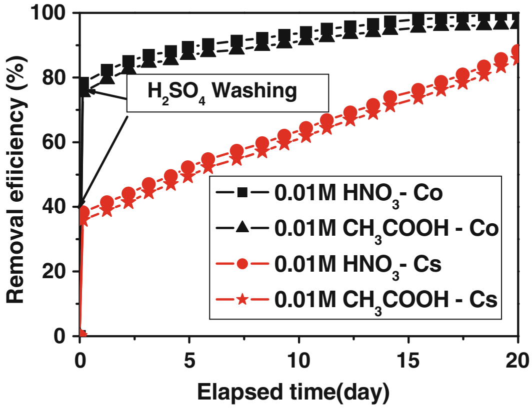

Fig. 7 ^{60}Co and ^{137}Cs removal efficiencies using different reagents in a concrete cell after washing with H_2SO_4

The surface of crushed concrete in high pH had a minus charge in the reverse of that of the soil. The nitric acid solution in the concrete cell flowed to the anode compartment due to electro-osmosis. Therefore, it was thought that the electro-osmosis direction of the cobalt and cesium ions is to the anode compartment side while the migration direction of the cobalt and cesium ions is to the cathode compartment side. Cesium ions were removed by up to 52% from the concrete, but the cobalt ions were removed by only 0.7%. The reason was that the cobalt ions were not moved by electro-osmosis and electro-migration due to their formation of a hydroxide above pH 6. But the cesium ions did not formed a hydroxide above pH 6, so were moved by electro-osmosis and electro-migration in an ion state above pH 6.

The results of the concrete washing with 3.0 M of sulfuric acid for 4 h are shown in Fig. 6. The $CaCO_3$ in the concrete reacted with sulfuric acid, and it was decomposed to CO_2, and the pH of the concrete was reduced to 3.7 after the concrete washing. Cesium ions were removed by 74.3% from the concrete, and the cobalt ions were removed by 87.0%. The sulfuric acid solution from the mixture was removed, and the remaining concrete was put into the 1 L electrokinetic equipment. The results of an execution of electrokinetic decontamination with 0.01 M of acetic acid in 20 mA/cm^2 for 14.83 days are shown in Fig. 6. Cesium ions and cobalt ions were removed slowly by electrokinetic decontamination. Cesium ions were removed by 99.3% from the concrete after 15 days and the cobalt ions were removed by 99.6%. The reason why the cobalt ions and cesium ions were mostly removed by electro-osmosis and electro-migration was thought to be due to them remaining in an ion state below pH 4. Also, the removal efficiencies of Co^{2+} and Cs^{+} were similar due to the absence of an ageing effect.

Figure 7 shows the removal efficiencies of ^{60}Co and ^{137}Cs versus the reagent type. The experimental results show that the removal efficiencies of ^{60}Co and ^{137}Cs by nitric acid were increased by 3.1 and 2.5% more than those by acetic acid. Also, when nitric acid instead of acetic acid was used, it had the advantage of reducing the electric voltage due to its higher electrolytic conductivity as shown in Fig. 4. Therefore, the electricity consumption can be reduced almost 20 times by using nitric acid. The channeling in a concrete cell did not occur during the experiment period. So, the removal efficiencies at all positions in a same cross section in a concrete cell were almost equal. Meanwhile, in order to increase the removal efficiency, electrokinetic decontamination was executed after a pretreatment. Namely, the radioactive concrete was mixed with 3.0 M of sulfuric acid with a ratio of 2.0 mL/g and it was shaken for 4 h, and then after removing the sulfuric acid solution from the concrete, electrokinetic decontamination for the remaining concrete was executed. In this case, the removal efficiencies of ^{60}Co and ^{137}Cs from

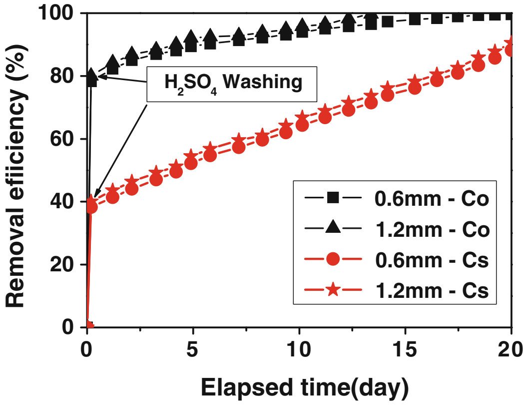

Fig. 8 ^{60}Co and ^{137}Cs removal efficiencies along the decontamination elapsed time for different particle sizes of radioactive concrete after washing with H_2SO_4

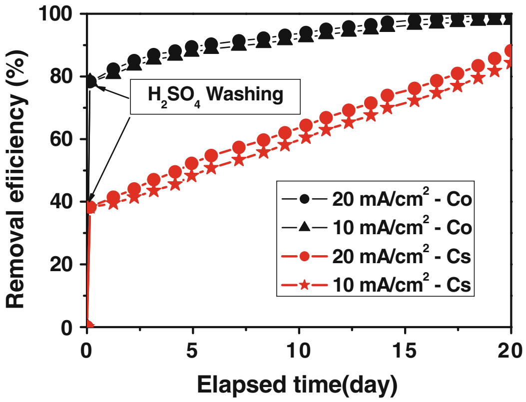

Fig. 9 ^{60}Co and ^{137}Cs removal efficiencies along the decontamination elapsed time in function of current density in a concrete cell after washing with H_2SO_4

concrete were 99.6 and 88.2% for 20 days. Also, if Figs. 6 and 7 were compared each other, the removal efficiency of ^{137}Cs with 3.0 M of sulfuric acid in Fig. 7 was reduced. The surmised reason was that the aging effect of ^{137}Cs is bigger than that of ^{60}Co due to its lager absorption coefficient.

Figure 8 shows the removal efficiency versus the particle size of concrete. The larger the particle size of the concrete, the more the removal efficiency of ^{137}Cs was increased. Namely, the removal efficiencies of ^{60}Co and ^{137}Cs from the concrete of an average 1.2 mm particle size were increased by about 3.2% (for 15 days) and 2.3% (for 20 days) more than that from the concrete of an average 0.6 mm particle size. The reason was considered to be that ^{137}Cs and ^{60}Co are stuck strongly on fine particles of concrete and the absorption coefficient of ^{137}Cs is bigger than that of ^{60}Co. It was conclusively found that the particle size did not seriously impede removal efficiency.

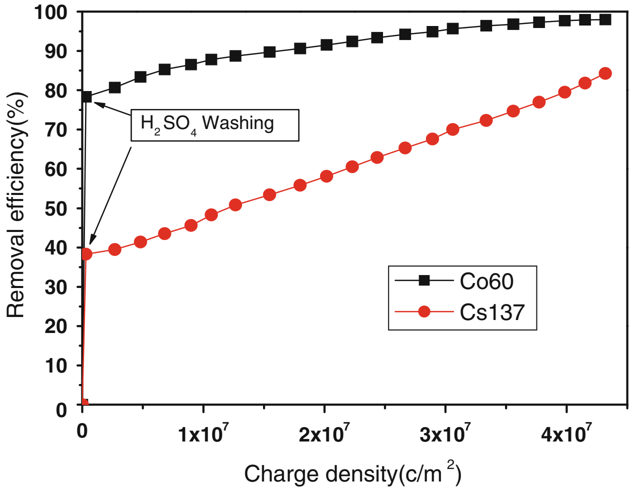

Fig. 10 ^{60}Co and ^{137}Cs removal efficiencies in function of charge density in a concrete cell after washing with H_2SO_4

Figure 9 shows the removal efficiency from the concrete versus the electric current in a concrete cell. The higher the electric current in a concrete cell, the more the removal efficiencies of ^{60}Co and ^{137}Cs was increased in accordance with Eq. 1. Namely, the removal efficiencies of ^{60}Co and ^{137}Cs by the application of an electric current of 20 mA/cm^2 were increased by 1.6 and 3.9% more than those by the application of 10 mA/cm^2. Also, Fig. 9 shows removal efficiencies of ^{60}Co and ^{137}Cs as a function of current density in a concrete cell. When current density in a concrete call was increased, removal efficiencies of ^{60}Co and ^{137}Cs were increased. Therefore the decontamination period can be shortened for an attainment to a clearance radioactivity concentration level. Figure 10 shows ^{60}Co and ^{137}Cs removal efficiencies in function of charge density in a concrete cell after washing with H_2SO_4. It was found that ^{60}Co and ^{137}Cs removal efficiencies was in proportion to chase density quantity and ^{60}Co removal efficiencies was higher than ^{137}Cs removal efficiencies due to its smaller absorption coefficient.

Figure 11 shows the decontamination period needed for reducing the concrete radioactive concentration to a clearance concentration for self disposal. The Korea Institute of Nuclear Safety suggested a clearance criterion of radioactive concrete for self disposal as being about 100 Bq/kg. Namely, about 10 days was required for the radioactive concrete of 500 Bq/kg to be reduced to 100 Bq/kg. About 15 days was required for the radioactive concrete of 1,000 Bq/kg to be reduced to 100 Bq/kg. About 25 days was required for the radioactive concrete of 2,000 Bq/kg to be reduced to 100 Bq/kg. Also, about 35 days was required for the radioactive concrete of 3,000 Bq/kg to be reduced to 100 Bq/kg. Meanwhile, for the radioactive concrete of more than 3,000 Bq/kg, after reducing its concentration

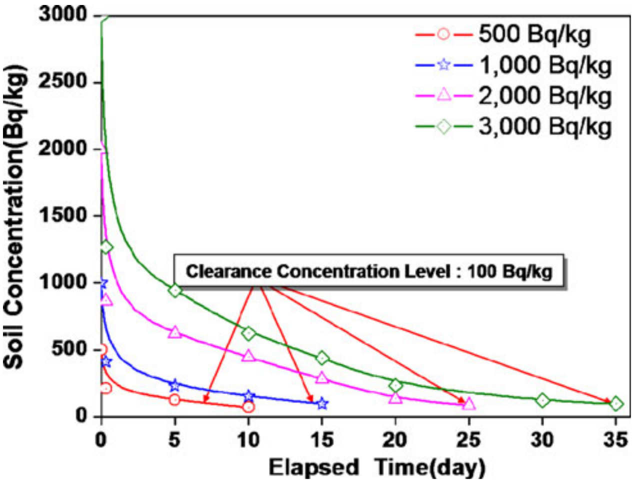

Fig. 11 Decontamination period required for decontaminating radioactive concrete to a clearance radioactivity concentration level (100 Bq/kg)

through a sieve separation until 3,000 Bq/kg, and then an electrokinetic decontamination should be executed to decontaminate it to below 100 Bq/kg. Therefore, the removal efficiencies of ^{60}Co and ^{137}Cs from the radioactive concrete of 1,940 Bq/kg were 99.8 and 92.3% by an electrokinetic decontamination by the application of an electric current of 20 mA/cm^2 for 25 days. The radionuclide residual concentration at the above part of a concrete cell was a little lower than that at the below part of a concrete cell at the end of experiment.

5 Conclusions

The concrete should be washed with 3.0 M of H_2SO_4 as a pretreatment before electrokintic decontamination to lower its pH below 4.0 for higher removal efficiencies. The removal efficiencies of ^{60}Co and ^{137}Cs by nitric acid were increased by 3.1 and 2.5% more than those by acetic acid. And, moreover, when nitric acid instead of acetic acid was used, it had the advantage of a reduction of the electricity consumption due to its higher electrolytic conductivity. The larger the particle size of the concrete, the more the removal efficiencies of ^{60}Co and ^{137}Cs were increased. Namely, the removal efficiency of ^{137}Cs from the concrete of an average 1.2 mm particle size was increased by about 2.3% more than that from the concrete of an average 0.6 mm particle size. Also, the removal efficiencies of ^{60}Co and ^{137}Cs by the application of an electric current of 20 mA/cm^2 were increased by 1.6 and 3.9% more than those by the application of 10 mA/cm^2. The removal efficiencies of ^{60}Co and ^{137}Cs from the radioactive concrete of 1,940 Bq/kg were 99.8 and 92.3% by electrokinetic decontamination after pretreatment by the application of an electric current of 20 mA/cm^2 for 25 days.

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