

Decontamination of Soils Contaminated with Co and Cs by Using an Acid Leaching Process - 9185

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ABSTRACT

Acid leaching process has been adapted for the remediation of soils contaminated with heavy metals and radionuclides. This method has been reported to be simple, and economically promising. Moreover it can be applicable for on-site and off-site remediations as well. Investigations were conducted on an acid leaching process using surrogate contaminated soils. Size sieving, agglomeration and column leaching were carried out with soils artificially contaminated with Co and Cs, respectively. Size distribution was analyzed for a determination of the particle size required to be agglomerated. Because of the low water permeability of the soils due to their fine particles, they were sieved by using a sieve with a 0.075 mm size (No. 200 mesh) for an agglomeration. The soils with a size smaller than 0.075 mm were agglomerated by using 2 % sodium silicate (Na_2SiO_3), while the soils with a size larger than 0.075 mm were used directly for the column leaching test.

From the preliminary test (the batch scale leaching test), 0.1 M of HCl was determined as the effective leaching agent for Co and Cs. Finally, the soils mixed with the coarse soil and the agglomerated soil were decontaminated with 0.1M HCl within 11.3 days and the removal efficiencies of Co and Cs were 94.0 % and 82.8 %, respectively. In conclusion, an acid leaching process could be applied for a remediation of soils contaminated with radionuclides such as Co and Cs.

Key words : acid leaching, agglomeration, contaminated soil, Co, Cs

INTRODUCTION

Soil contaminated with radionuclides such as Co and Cs have been generated by a long term management of nuclear facilities and stored at temporary storage sites during several decades in Korea. Because of the limited storage capacity of temporary storage sites and the high cost for a final disposal at radioactive waste disposal sites, it might be necessary to remediate a soil contaminated with radionuclides to the level of a self-disposal.

Therefore, many researchers have made an effort to develop a proper remediation technology for soils using electro-kinetic [1], electro-kinetic flushing [2], a soil washing [3] and a soil flushing. In this study, a heap leaching process, one of the soil flushing methods which is simpler and more economic than the other technologies, was selected and tested for the decontamination of a soil. Heap leaching process is

usually used for the remediation of a uranium mining site and a contaminated soil with heavy metals such as Pb, Cd, Zn, etc[4,5]. Size sieving, agglomeration and acid leaching are the main processes of the heap leaching technology. A proper binder and addition ratio for an agglomeration of the fine soils should be determined because a fine soil can interrupt the resultant hydrological permeability during an acid leaching process.

The feasibility for the use of heap leaching technology was examined as a decontamination technology for soil contaminated with radionuclides. A proper agglomeration binder and addition ratio was determined for the fine soil such as silt and clay. Suitable leaching reagent and concentration was also determined for a more effective decontamination by an acid leaching process. Using artificially contaminated soil, a column test was carried out in order to check on the removal characteristics of Co and Cs from the soil.

MATERIALS AND METHODS

Fabricating artificially contaminated soil with Co and Cs

Soil was taken from a site around a nuclear facility in Seoul. Plants' roots and leaves contained in the soil were removed. At the laboratory, a soil sample was spread on a tray and dried for more than a week under room temperature in a natural state.

Particle size distribution of a soil sample used in this study was a sandy soil which had a larger size than 0.075 mm which represents 95.5 % of its whole size.

In order to fabricate artificially contaminated soil, 0.01 M of CoCl_2 and CsCl solution was added to a dried soil sample with 0.4 ml/g of a liquid to solid ratio. The artificially contaminated soil was used for the acid leaching test after a drying. The concentrations of Co and Cs in the soil were measured by an atomic adsorption spectrometer (AAS; AAnalyzer 300, Perkin Elmer co.) through conducting a content analysis of the soil and used as an initial value for a calculation of the removal efficiencies after an acid leaching process.

Size sieving and a determination of the agglomeration binder

In order to determine a proper agglomerating binder for the acid leaching process of the fine soils such as silt and clay, several binders (Starch, Vinyl acetate, Vinyl amide, Acryl amide, Sodium carboxymethylcellulose and Sodium silicate) were used for the agglomeration tests and the re-dissolution tests.

Tests for a determination of the proper agglomerating binder and the addition ratio for the fine soils were conducted. Each binder for the fine soils (~ 0.075 mm) was added at a weight volume of 0.5, 1, 2, 5, 10 and 20 %. Mixed soil with a binder was sprayed with de-ionized water and agglomerated. After drying

the agglomerated soil, a soaking test to check on the re-dissolution of the agglomerates was carried out by using de-ionized water and 0.1M HCl for 10 days.

Batch scale leaching test

In order to determine a proper leaching reagent and the concentration of the reagent for the contaminated soil, a batch scale leaching test was carried out using 5 reagents (de-ionized water, HCl, oxalic acid, acetic acid and citric acid) at different concentrations (0.001, 0.005, 0.01, 0.05, 0.1, 0.5M) with 10 ml/g of a liquid to solid ratio. After being shaken for 6 hrs and filtrated by a 0.2 μm membrane filter, the concentrations of Co and Cs were analyzed by AAS. The results indicated that 0.1 M HCl is a proper leaching reagent and concentration for the decontamination of the soil.

Acid leaching test for the mixture of coarse soil (0.075 mm ~ 4.75 mm) and the agglomerated soil (~ 0.075 mm) using a column test

Acryl column with a 20 cm ID and 70 cm height was used for a large-scale acid leaching test. In order to evaluate the acid leaching process for a decontamination of the soil, a column test was carried out by using 10 kg of mixed soil at a ratio of the initial composition with fine soil (450 g) after an agglomeration and the coarse soil (9,550 g) after a size sieving. Leaching reagent (0.1 M HCl) was continuously supplied to the column with a mean flow rate of 3.0 ml/min. Column effluents were sampled periodically. Effluents' volume was also measured. The concentrations of Co and Cs were analyzed by AAS after a filtration.

Table 1. Results of the dissolution tests for the agglomerated soils with different binders and addition ratios

Binders	Addition ratio (wt; %)					
	0.5	1	2	5	10	20
Starch	X	X	X	X	X	X
Vinyl acetate	X	X	X	X	X	X
Vinyl amide	X	X	X	X	X	X
Acryl amide	X	X	X	X	X	X
Sodium carboxymethylcellulose	X	X	X	X	X	X
Sodium silicate	X	Δ	O	O	O	O

RESULTS AND DISCUSSION

Size distribution after the agglomeration

From the results of the soaking test (Table 1), sodium silicate with a 2 % volume (wt) was determined as a proper binder and addition ratio for the agglomeration of the fine soil. It was found that the mean diameter of a soil particle after an agglomeration was increased by about 100 times than before an agglomeration (Fig. 1). This significant increase of a soil particle's size may improve the water permeability of the soil during the acid leaching process and make the management of this leaching treatment process easier.

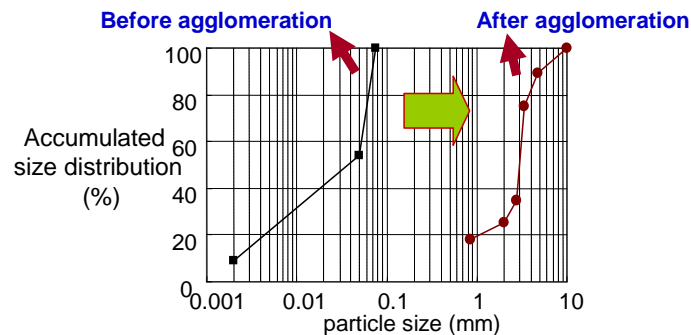


Figure 1. The variation of the particle size distribution by an agglomeration using a soil smaller than 0.075 mm

Determination of the proper leaching reagents and concentration

The results of the batch scale leaching test for determining the proper leaching reagent are shown in Figure 2. It was observed that Co and Cs were only leached out at 26.1 % and 15.4 % from the soil by using de-ionized water (d.w.), respectively. Co and Cs were also leached out at around 40 % and 30 %, respectively, by using a low molecular organic acid such as oxalic acid, acetic acid and citric acid. In the case of HCl as an acid leaching reagent, 98.7 % of Co was leached out under a higher concentration than 0.01 M showing 99.6 % of Co was leached out under 0.05 M, and 52.5 % of Cs was leached out under a higher concentration than 0.01 M showing 63.2 % of Cs was leached out under 0.1 M . It was confirmed that HCl had a 2 or 3 times higher leaching capability for Co and Cs from the soil, when comparing it with a low molecular organic acid. Leaching ratio was almost similar under a higher concentration of HCl than 0.05 M for Co and 0.1 M for Cs. Therefore, 0.1 M HCl was used for the soil decontamination experiment by using the acid leaching process.

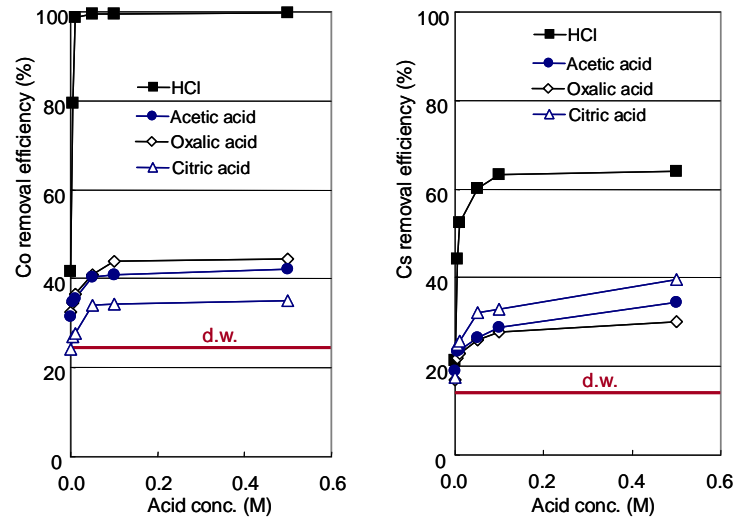


Figure 2. Removal efficiency of Co and Cs in Hydrochloric acid, oxalic acid, acetic acid and citric acid with various concentration of 0.001, 0.005, 0.01, 0.05, 0.1 and 0.5M

Results of the column leaching tests for the mixed soil (coarse and agglomerated)

Leaching concentration with time and the removal efficiency with an accumulated liquid to solid ratio of 0.1 M HCl in the column test are shown in Figures 3 and 4. After 2 days, Co was leached out at more than 80 % from the column while Cs was leached out at only more than 40 %. In conclusion, the mixed soil was decontaminated by 0.1M HCl within 11.3 days and the removal efficiencies of Co and Cs were 94.0 % and 82.8 %, respectively. An acid leaching process could possibly be applied for a remediation of soils contaminated with radionuclides such as Co and Cs.

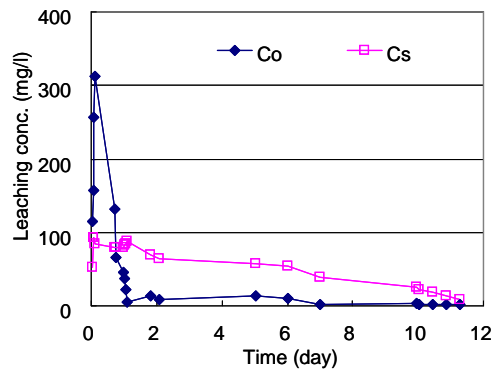


Figure 3. Leaching concentrations of Co and Cs with time from the column test using the mixed soil

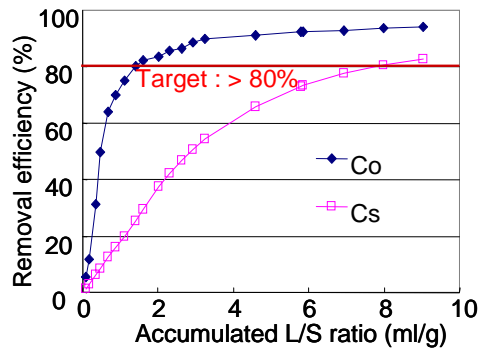


Figure 4. Removal efficiencies of Co and Cs with the liquid to solid ratio from the column test using the mixed soil

CONCLUSIONS

Size sieving, an agglomeration and a column leaching were carried out with soils artificially contaminated with 236 mg of Co per kg of soils and 532 mg of Cs per kg of soils, respectively. Size distribution was analyzed for a determination of the particle size required to be agglomerated. Because of the low water permeability of the soils due to their fine particles, they were sieved by using a sieve with a 0.075 mm size (No. 200 mesh) for an agglomeration. The soils with a size smaller than 0.075 mm were agglomerated by using 2 % sodium silicate (Na_2SiO_3), while the soils with a size larger than 0.075 mm were used directly for the column leaching test. From the preliminary test (the batch scale leaching test), 0.1 M of HCl was determined as an effective leaching reagent for Co and Cs.

Finally, the soils mixed with the coarse soil and the agglomerated soil were decontaminated by 0.1M HCl within 11.3 days and the removal efficiencies of Co and Cs were 94.0 % and 82.8 %, respectively. In conclusion, an acid leaching process could possibly be applied to a remediation of soils contaminated with radionuclides such as Co and Cs.

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