



## Advanced remediation of uranium-contaminated soil



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### ABSTRACT

The existing decontamination method using electrokinetic equipment after acidic washing for uranium-contaminated soil requires a long decontamination time and a significant amount of electric power. However, after soil washing, with a sulfuric acid solution and an oxidant at 65 °C, the removal of the muddy solution using a 100 mesh sieve can decrease the radioactivity of the remaining coarse soil to the clearance level. Therefore, only a small amount of fine soil collected from the muddy solution requires the electrokinetic process for its decontamination. Furthermore, it is found that the selective removal of uranium from the sulfuric washing solution is not obtained using an anion exchanger but rather using a cation exchanger, unexpectedly. More than 90% of the uranium in the soil washing solutions is adsorbed on the S-950 resin, and 87% of the uranium adsorbed on S-950 is desorbed by washing with a 0.5 M Na<sub>2</sub>CO<sub>3</sub> solution at 60 °C.

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### 1. Introduction

Many nuclear facilities such as commercial nuclear power plants will be decommissioned or dismantled in the near future. In Korea, great amounts of radioactive soil and concrete waste have already been generated from the decommissioning of two research reactors and a uranium conversion plant. The volume reduction of the radioactive soil and concrete using an appropriate treatment will decrease the amount of waste to dispose of and result in reduced disposal costs and enhanced efficiency at the disposal site (Kim et al., 2014).

The selection of an appropriate method among the soil decontamination techniques including biological, physical-chemical and thermal methods is predominantly dependent on the primary contaminants (Pavel and Gavrilescu, 2008). For a remediation of radionuclide-polluted soil, washing with an appropriate reagent is a simple and effective method. Bicarbonate, strong inorganic acids, and weak organic acids such as ascorbic acid and citric acid have been used to remove uranium from soil under ambient oxidizing conditions (Kulpa and Hughes, 2001; Ebbs et al., 1998). When only nitric acid was used for the remediation of uranium-contaminated (U-contaminated) soil in our laboratory, it was difficult to decontaminate the soil to a clearance radioactivity level (IAEA, 1996). For

more than 95% remediation of the U-contaminated soil, electrokinetic technology has been developed after a batch-type washing once or twice, as depicted in Fig. 1 (Kim et al., 2012; Kim et al., 2011). However, the electrokinetic equipment method requires a long decontamination time of almost one month and a significant amount of electric power. Thus, an improved washing method has been investigated.

A sulfuric acid solution with an added oxidant has been used to leach uranium from the uranium ore (Hague and Ritcey, 1982; KAERI, 1982), and fine soil particles were separated from coarse particles using a 100 mesh sieve in the treatment of heavy metals-contaminated soil (SEC, 2009) because a high concentration of copper, lead, and arsenic generally exists in the fine particles. In this study, the removal of the fine soil particles using a 100 mesh sieve was examined after soil washing with a sulfuric acidic solution with an added oxidant in order to decontaminate the remaining coarse soil through simple washing without an electrokinetic treatment.

In addition, when the waste solution was adjusted to near pH 9 by adding KOH to precipitate the uranyl ions, as depicted in Fig. 1, a significant amount of the secondary radioactive waste was generated by the precipitates of the dominant metals, such as iron and aluminum, in the waste solution. If uranium is selectively removed from the acidic waste solution, the volume of the secondary radioactive waste will be remarkably reduced. Thus, selective removal methods of uranium using a solvent extraction and using ion exchangers have also been investigated in this study. Rosenberg (Rosenberg et al., 2016) recently reviewed the uranium sorption by

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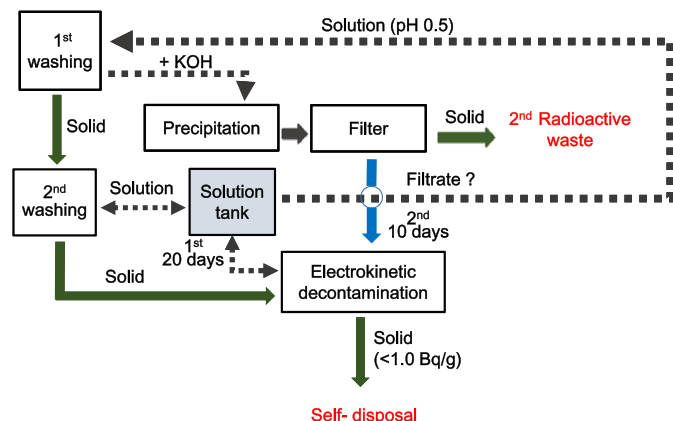


Fig. 1. A pre-existing remediation procedure for U-contaminated soil.

various types of solid phase sorbents including ion exchangers.

Alamine 336 (tri-octyl/decyl amine), a commercially available tertiary amine extractant, is known to be a useful reagent to extract uranium from acidic sulfate media in uranium ore processing (Collet et al., 2009; Kim, 2013; Kim et al., 2012; Kumar et al., 2016; Ramadevi et al., 2012). In general, tertiary amine-based extractants are the preferred reagents over other chemicals such as alkyl amides (Lapka et al., 2009), di-(2-ethylhexyl) phosphoric acid (El-Reefy et al., 1997), tri-n-butyl phosphate (TBP), 2-ethylhexyl phosphoric acid mono-2-ethyl hexyl ester (PC 88A) and tri-octylphosphine oxide (TOPO) (Singh et al., 2001) in the solvent extraction of uranium liquors with a significant presence of iron, a common impurity in leach liquors generated during the sulfuric acid leaching of ores.

A strong anion exchanger IRA 910 (Rahmati et al., 2012) and an ampholyte resin S-950 with aminophosphonic groups (Nekrasova et al., 2008; Rychkov and Norkina, 2007) were used to selectively remove uranium from a sulfuric washing solution. In addition, the Amberlite GG-400 anion exchange resin was used for U(IV) sorption in the presence of phosphate (Semnani et al., 2012).

## 2. Experiment

### 2.1. Elemental analysis of solid and solution

The concentration of uranium in solids and solution was indirectly analyzed using HPGe  $\gamma$ -spectrometry (Canberra, Genie, 2000; USA) by measuring the radioactivity of  $^{234m}\text{Pa}$  (energy: 1001 KeV). Uranium-238 undergoes alpha-particle decay to daughter  $^{234}\text{Th}$  (half-life = 24.1 days) to reach secular equilibrium in less than one year, and  $^{234}\text{Th}$  decays to  $^{234m}\text{Pa}$  (half-life = 1.17 min) by beta-particle emission (Adsley et al., 1996). Since the soil was contaminated by natural uranium decades ago, the secular equilibrium among  $^{238}\text{U}$ ,  $^{234}\text{Th}$  and  $^{234m}\text{Pa}$  in this study has already been reached. Some other elements in the solution were analyzed using inductively coupled plasma-atomic emission spectroscopy (ICP-AES, JY Ultima-2C, Jobin Yvon, France).

### 2.2. Soil washing

In order to efficiently extract uranium from uranium ore with sulfuric acid, an oxidant was added at 65 °C in KAERI (KAERI, 1982). Thus, the soil was washed by a sulfuric acid solution with  $\text{NaClO}_3$  at 65 °C. After washing, the solution was passed through a 100 mesh sieve to remove the fine particles. To find the decontamination of the remaining coarse soil through simple washing, U-contaminated

soil was washed as follows.

- ① 100 g of U-contaminated soil with 26 Bq/g was added to 100 mL of 0.3 M sulfuric acid solution in a 1.0 L Erlenmeyer flask.
- ② The solution was shaken at 150 rpm and heated to 65 °C in a water bath.
- ③ After 15 min of shaking, 1 g of  $\text{NaClO}_3$  was added to the solution.
- ④ After shaking for 3 h, the solution was passed through a 100 mesh sieve to remove the fine particles.
- ⑤ The muddy solution containing the fine soil particles was filtered with a Whatman 4 filter paper.
- ⑥ The soil remaining on the sieve was briefly washed twice with the filtrate obtained from ⑤.
- ⑦ The remaining soil was washed once more with 100 mL of a 0.3 M sulfuric acid solution at 65 °C for 3 h.
- ⑧ After washing, the solution was passed through a 100 mesh sieve.
- ⑨ The remaining coarse soil was dried, and its weight and radioactivity were measured.
- ⑩ The pH and uranium concentration in the washing solutions were measured.

The U-contaminated soil was examined in order to determine the effects of temperature and oxidant on the soil washing under various conditions, such as at room temperature and 65 °C, and with and without an oxidant ( $\text{NaClO}_3$ ), as described in Table 1. The washing and separation of the fine particles were performed using the same method described above.

From the previous study, it is known that large gravel cannot be decontaminated by washing alone. In order to limit the soil particle size for the washing, the soil was sifted with 2, 5 and 10 mm pore-sized sieves, respectively. Then, the sifted soils were washed according to the above washing method.

### 2.3. Removal of uranium from the washing solution

#### 2.3.1. Extraction of uranium by alamine 336

Alamine 336 was examined for the collection of uranium from the soil washing solution in a sulfuric acid medium (KAERI, 1982). The detailed procedure for the extraction of uranium by alamine 336 is as follows.

- ① The U-contaminated soil was washed using 0.3 M sulfuric acid.
- ② By adding small amounts of NaOH solution, the pH of the solutions was adjusted to 0.4 and 0.9, respectively.
- ③ The soil washing solution was filtered with a Whatman 4 filter paper.
- ④ Various amounts of alamine 336 and 1-dodecanol were dissolved in Exxsol™ D40 (Table 2).
- ⑤ 5 mL of solution from ④ was added to 50 mL of the soil washing solution.
- ⑥ The mixed solution was vigorously shaken for 10 min.
- ⑦ After shaking, the solution was transferred to a separation funnel.
- ⑧ The aqueous layer was separated from the organic layer.
- ⑨ The concentration of uranium in the aqueous solution before and after extraction was analyzed.

#### 2.3.2. Sorption of uranium by ion exchangers

A strong anion exchanger, IRA 910, is known to adsorb uranyl sulfate anion complexes in 0.02–9.0 M of a sulfuric acid solution

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