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Technical Note

Reduction of Radioactive Waste from Remediation of Uranium-Contaminated Soil

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ABSTRACT

Great amounts of solid radioactive waste (second waste) and waste solution are generated from the remediation of uranium-contaminated soil. To reduce these, we investigated washing with a less acidic solution and recycling the waste solution after removal of the dominant elements and uranium. Increasing the pH of the washing solution from 0.5 to 1.5 would be beneficial in terms of economics. A high content of calcium in the waste solution was precipitated by adding sulfuric acid. The second waste can be significantly reduced by using sorption and desorption techniques on ampholyte resin S-950 prior to the precipitation of uranium at pH 3.0.

1. Introduction

Many nuclear facilities will have to be decommissioned or dismantled in the near future. In Korea, great amounts of radioactive soil and concrete waste had been generated from the decommissioning of two research reactors and a uranium-conversion plant. Volume reduction by an appropriate treatment will decrease the amount of waste to be disposed of, resulting in a reduction in the disposal cost and enhanced efficiency of the disposal site [1].

For the remediation of radionuclide-polluted soil, washing with an appropriate reagent is a simple and effective method. Bicarbonate, strong inorganic acids, and weak organic acids (ascorbic, citric) have been used to remove U(VI) from soil under ambient oxidizing conditions [2,3]. In our laboratory, nitric acid is used for the remediation of uranium-contaminated (U-contaminated) soil. However, it is difficult to decontaminate soil to a clearance radioactivity level by washing with nitric acid alone [4]. For greater than 95% remediation of U-contaminated soil, electrokinetic

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technology has been developed to be applied after batch-type washing one or two times. In addition, electrokinetic equipment at a practical scale (512 L of soil/batch) was recently manufactured [5,6]. However, a great amount of acidic waste solution has been generated from the washing processes and the operation of the electrokinetic decontamination equipment [7,8]; it has to be reduced in volume or be recycled. To reduce its volume, the evaporation by heating of water in the waste solution requires too much energy. Thus, a proper method for removing uranium from the waste solution in order to recycle it has been studied. Fig. 1 shows the remediation procedure of U-contaminated soil developed in our laboratory.

To precipitate uranium in the first washing solution shown in Fig. 1, the pH of the waste solution was adjusted to neutral or weak alkaline, and alum and magnetite were also added [9,10]. CaO and NaOH have been considered to increase the pH of the waste solution. While NaOH rapidly dissolves and can easily control the pH of a solution, accumulated sodium ions are hard to remove when the solution is recycled. If CaO is substituted for NaOH, calcium ions can be simply removed from the solution since calcium ions form complexes more easily than sodium ions do, although it takes a longer dissolution time to increase the pH of the solution up to 8.0 because of the lower dissolution rate of CaO.

However, the uranium precipitation method by adding CaO or NaOH generates a great amount of solid radioactive waste (second waste), approximately 10% of the initial soil volume, because the dominant metal ions such as iron and aluminum are also precipitated in the neutral or weak alkaline solution. If uranium is selectively removed from the waste solution, the volume of the second waste will be greatly reduced.

To reduce the volume of the second waste from the remediation of U-contaminated soil, this work examined the use of less acidic washing solution, the removal of dominant ions from the second waste, and the extraction of uranium from the waste solution.

2. Materials and methods

2.1. Elemental analysis of solid and solution

The concentration of uranium in a solid was indirectly analyzed using HPGe γ-spectrometry (Canberra, Genie 2000, Meriden, USA) by measuring the radioactivity of ^{234m}Pa (energy 1001 KeV). Uranium-238 undergoes alpha-particle decay to daughter ^{234}Th (half-life = 24.1 days) to reach secular equilibrium in less than 1 year, and ^{234}Th decays to ^{234m}Pa (half-life = 1.17 minutes) by beta-particle emission [11]. Since the soil was contaminated by natural uranium decades ago, the secular equilibrium among ^{238}U, ^{234}Th, and ^{234m}Pa in this study has already been reached. The radioactivity of ^{238}U corresponds to 48% of the total radioactivity for natural uranium, and 1 Bq/g radioactivity of uranium indicates that 40 mg of natural uranium is contained in 1 kg of sample [12]. The elements dissolved in the solution were analyzed using inductively coupled plasma-atomic emission spectroscopy (ICP-AES, JY Ultima-2C, Jobin Yvon, Palaiseau, France).

2.2. Using less acidic washing solution

If a less acidic solution is used in the first and second washing processes shown in Fig. 1, the second waste will be reduced. To identify the difference in decontamination efficiency at a pH range of 0.5–1.5, U-contaminated soil was washed with various pH solutions in the following sequence.

(1) Four solutions were prepared by adding 150 g of U-contaminated soil with 29 Bq/g and 300 g of the washing solution into a 1.0 L Erlenmeyer flask.
(2) The pH of the solutions was adjusted to 0.47, 0.86, 1.16, and 1.49 by adding less than 1 mL of concentrated nitric acid or sodium hydroxide solution.
(3) The solutions were shaken for 10 minutes at 100 rpm.
(4) The pH of the solutions was readjusted to 0.54, 0.85, 1.06, and 1.58 in order, respectively.

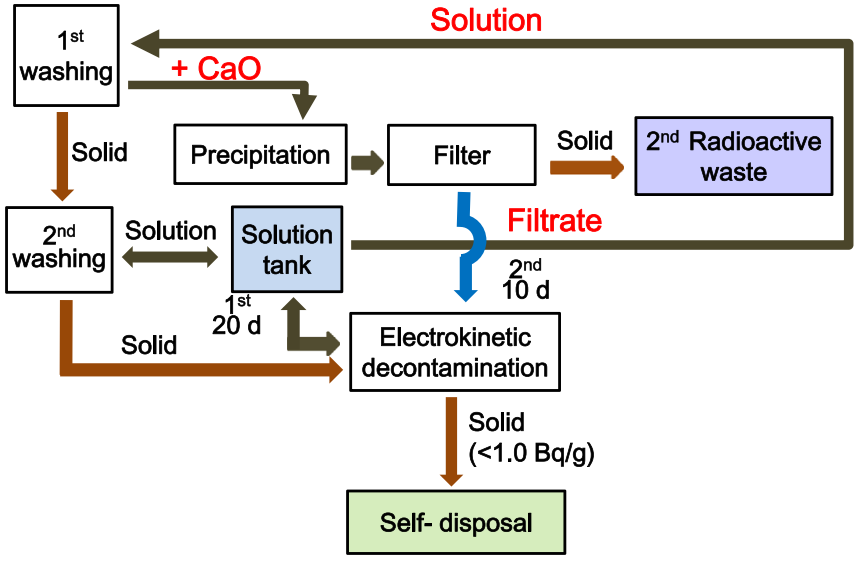

Fig. 1 – A remediation procedure for U-contaminated soil.