Chemical Leaching of Radionuclide from Contaminated Fine Concrete Waste

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1. Introduction

In Korea, the decontamination and decommissioning of the retired research reactors and uranium conversion plant has been under way. Hundreds of tons of concrete wastes are expected from the D&D of these facilities [1]. Typically, the contaminated layer is only 1~10mm thick because cement materials are porous media, the penetration of radionuclides may occur up to several centimenters from the surface of a material [2]. Surface activity can be a loose contamination arising from a deposition of nuclides from an interfacing medium, and it also can be tightly bound. Most of the radionuclides are significantly concentrated on pastes material under 1mm. Radionuclides can be easily removed during the course of a leaching test from pastes.

Radionuclides solubility is one of the factors, along with sorption and inventory that can determine the 'source term' for potential migration to the far filed of a contaminated concrete waste. Several processes can affect the concentration of radionuclide in cement paste phase, e.g.:

- a) coprecipitation with other phase
- b) lattice incorporation in the major cement hydration products (solid solution).
- c) sorption at hydrous surfaces (chemisorption, adsorption)
- d) complex and colloid formation in the aqueous phase [3]

The best case for retardation of radionuclide mobility is to have a low aqueous solubility. An understanding of interaction mechanisms in important where data are required for performance assessment, that are outside the range of experimental systems studied such as pH range, timescales.

The aim of this study was to establish the optimized leaching conditions for the treatment of concrete paste contaminated with radioniclides. The leaching tests had been performed using the pastes which were contaminated with the radionuclide (i.e. ⁶⁰Co, U) generated from TRIGA MARK II and uranium conversion plant. To minimize the total radioactive waste volume of the radioactive concrete as a paste, the pastes were leached at a various condition to HCl and H₂C₂O₃ leachent.

2. Method and Result

For the experimental test, radioactively contaminated concrete pastes are used generated from dismantling the retired TRIGA MARK II and III research reactors and uranium conversion plant. Figure 1 shows the separation process flow chart. Sorption is a generic term, which covers the process of absorption, physical adsorption and chemical adsorption. Sorption mechanisms depend on the chemical speciation of the sorbate, and the chemical and physical properties of the sorbent. Absorption is the incorporation of a sorbate within the physical or molecular structure of a sorbent, i.e. the sorbate has access to the bulk, not just the surface, of the sorbent, Physical adsorption can be divided into three types:

- (a) Surface complex, which is mixture of chemical and electrostatic interactions:
- (b) Ion exchange, which is an example of electrostatic adsorption due to coulombic forces of attraction between charged sorbate species and the sorbent:
- (c) Other mechanism, e.g. solid-solution formation, coprecipitation.

In the first step concrete parts are crushed for a size reduction. The large cement paste pieces were then broken into smaller <2mm diameter. This <2mm pulverized cement paste was then sieved below <1mm using 8-in. diameter brass wire cloth sieves and an electric vibratory shaker for 5min in batches. The radionuclide tracer spike was added to the weighed amount of cement paste powder contained in a 80mL capacity cylindrical vessel. After air-drying overnight, the cement paste powder was homogenized by mixing thoroughly. The contaminated paste was taken by a standard cylindrical vial at 20ml for a quantitative analysis. The pastes were analyzed with a MCA (multichannel-analysis) of a high-purity germanium detector for evaluate the radioisotope distribution.



Fig.1. Experimental procedure

Figures 1, 2 show the specific activity of the paste for light and heavy concrete paste on $1M H_2C_2O_4$ leaching solution. As shown in this figure, leached specific activity from contaminated paste was increased by increasing the agitation time. The removed radionuclide from paste was achieved by up to 66%. Contaminated paste should be a possible removal.

The most importation reactions of cement with waste are those which lead to binding [4], i.e. a chemical reaction. The nature of the binding forces in a cement matrix is species, so it is difficult to produce a general description. Many specific also form one or more solubility-limiting phases by reaction with cement components. "Solubility-limiting" implies the formation of a phase which would not ordinarily exist in cement, because it contains one or more radioactive species.



Fig. 1. Specific activity of light concrete paste on 1M $H_2C_2O_4$



Fig. 2. Specific activity of heavy concrete paste on 1M $H_2C_2O_4$

Figures 3, 4 show the specific activity of the paste for light and heavy concrete paste on 1M HCl leaching solution. As shown in this figure, leached specific activity from paste was significantly decrease increased by up to 99%. HCl had higher extraction efficiency higher than oxalic acid for ⁶⁰Co from concrete paste. Removed efficiency rate of ⁶⁰Co in the 1M-HCl leaching solution was decontaminated by up to 99%. Only at temperatures 25°C of leaching solution, was there any significant water-soluble or cation-exchangeable ⁶⁰Co leached from spiked pulverized cement paste.



Fig. 3. Specific activity of light concrete paste on 1M HCl



Fig. 4. Specific activity of heavy concrete paste on 1M HCl

3. Conclusions

HCl and oxalic acid had higher extraction efficiency of ⁶⁰Co. About 99% of a total of ⁶⁰Co was decontaminated from light and heavy concrete paste in 1M-HCl leaching solution.

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