

Sequential determination of $^{239,240}\text{Pu}$, ^{238}Pu , ^{90}Sr and ^{241}Am Isotopes in Environmental Samples

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1. Introduction

Recently, several studies have been reported on the combined procedure for the determination of radionuclides in soils or sediment samples with extraction chromatographic materials such as the TRU, Sr and TEVA Spec resins [1,2]. These methods are somewhat difficult to apply for very low levels of fallout radionuclides without using large amount of extraction chromatographic resin. It was, also, reported that Pu, ^{90}Sr , ^{237}Np and ^{241}Am isotopes were purified in the soil and sediment with the anion exchange resin, Sr Spec resin and TRU resin [3,4]. However, these methods have a disadvantage in that extraction chromatographic materials are somewhat expensive, therefore, it is not suitable to determine Pu, Sr and Am isotopes for routine environmental samples. In order to reduce the analysis cost and turnaround time in conventional methods, a simultaneous method to determine Pu, Sr and Am nuclides in environmental samples has become increasingly important. A combination of chromatographic elution techniques with the use of anion exchange resin provides a powerful tool for separating and purifying actinides from other hindrance nuclides. In this study, the rapid and reliable determination of Pu, Sr and Am nuclides was developed in low levels of Pu, ^{90}Sr and ^{241}Am isotopes in environmental samples. The developed analytical method for Pu, ^{90}Sr and ^{241}Am isotopes was validated by application to IAEA Reference materials and environmental soil samples.

2. Experiments and Results

A flow chart of the radiochemical procedure for Pu, ^{90}Sr and ^{241}Am isotopes in the nitric acid medium is shown in Fig. 1. The determination of the actinides at low levels in environmental materials has always been time consuming and involved the use of large amounts of hazardous materials. However, plutonium presents fewer problems than other actinides like Am, because it is relatively easily separated by the anion exchange resin. Pu(IV) and Th(IV) are strongly adsorbed on the resin as anionic nitrate complexes while U(VI) is only weakly adsorbed. Also, Am(III) and Sr(II) as well as Fe(II), Ca(II), Mg(II), Al(III), Ti(IV), REEs and other soil matrix components are not retained on the anion exchange column and therefore pass completely through it. The column was then washed with 50 mL of 8M HNO_3 for removing hindrance nuclides such as trace levels of U isotopes and Fe ions weakly adsorbed on the column. Thorium was desorbed from the column by passing through 50 mL of 9M HCl. Plutonium(IV) is retained by the column as anionic chloride complexes. Finally, plutonium was stripped from the column by passing through with 20 mL of 0.1M $\text{NH}_4\text{I}/12\text{M HCl}$.

The separation of the actinides from environmental samples, especially in the trivalent state like ^{241}Am has been a complicated and time-consuming task. Recently, the chromatographic resins for extraction such as TRU, UTEVA and Diphonex resins have given an opportunity to develop fast and simple separation methods for many radionuclides [5,6]. The TRU Spec resin has been used for separation of Am isotopes from the Pu, U and Th isotopes. However, for high concentrations of inorganic elements such as Fe, Al, P and Ca as well as natural nuclides such as U and Th in the soil sample, it is necessary to remove these elements from the sample solution before passing the TRU Spec column. It was reported that actinides and lanthanides are coprecipitated with calcium oxalate effectively from acidic solutions at pH 1.5, while other major ions such as iron, aluminum, titanium, and phosphate are left in the solution. So, in this study, iron, aluminum and phosphate were removed from Am fraction with the oxalate precipitation method.

The determination of Pu, Am and Sr isotopes updated in this study was validated by application to the IAEA-Reference samples. For the IAEA-375 soil sample, the activity concentrations of $^{239,240}\text{Pu}$, ^{90}Sr and ^{241}Am with the updated method in this study were 0.26 ± 0.5 Bq/kg, 113.7 ± 7.0 Bq/kg and 0.11 ± 0.004 Bq/kg, respectively. These values were consistent with the reference values reported by the IAEA. Also, for

the IAEA-300 sediment sample, the activity concentrations of $^{239,240}\text{Pu}$, ^{90}Sr and ^{241}Am were consistent with the reference values reported by the IAEA.

3. Summary

In this work, a quantitative method of the sequential separation of Pu, ^{90}Sr and ^{241}Am nuclides was developed in environmental soil samples using by an anion exchange resin connected with a Sr Spec resin. The sequential methods of Pu, ^{90}Sr and ^{241}Am nuclides can reduce analysis cost and time. With the sequential method updated in this study, the activity concentrations of $^{239,240}\text{Pu}$, ^{90}Sr and ^{241}Am in the IAEA reference materials were close to the reference values reported by the IAEA.

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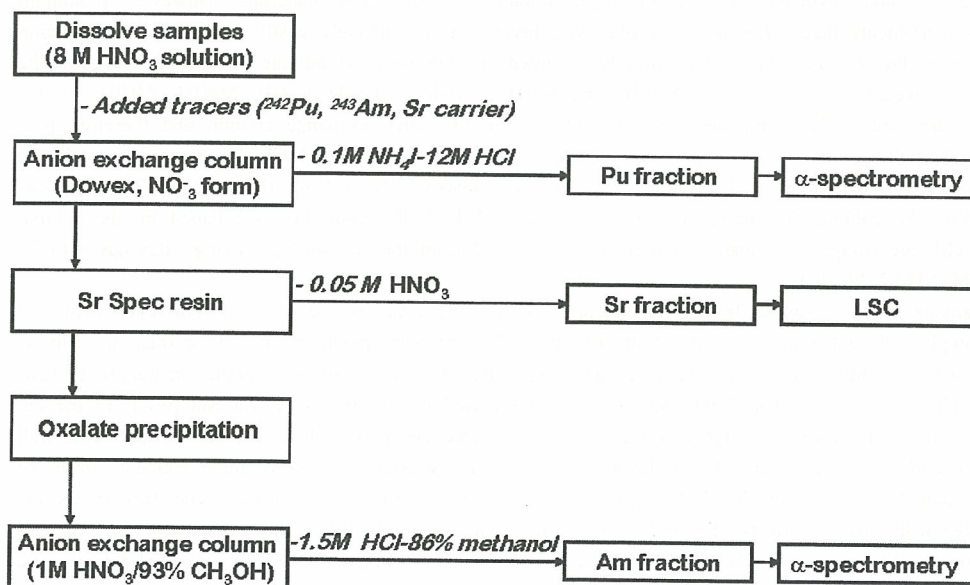


Fig. 1. Separation scheme of Pu, ^{90}Sr and ^{241}Am isotopes in environmental samples