

Rapid Determination of U Isotopic Ratios in the Vicinity of Nuclear Facilities

Jong-Myoung Lim^{a*}, Ji-Young Park^{a,b}, Yoonhee Jung^a, Hyuncheol Kim^a, Won-Young Kim^a, Gun Ho Chung^a

^aKorea Atomic Energy Research Institute, 989-111 Deadeok-daero, Yuseong-gu, Dejeon, 34057, Korea

^bKorea University of Science and Technology, UST 217, Gajeong-ro, Yuseong-gu, Daejeon, 34057, Korea

*Corresponding author: jmlim@kaeri.re.kr

1. Introduction

It is important to assure the radiological safety for the general population. In general, the sources of radioactivity in environmental media can be divided into both natural (e.g., solar radiation and natural decay) and anthropogenic sources (e.g., human activities such as accidents, the nuclear power industry, and weapon tests). As an attempt to reduce the social costs and apprehension arising from the radioactivity in environmental media, an accurate and rapid assessment of radioactivity is highly desirable. Hence, it is important to develop a series of evaluation of rapid procedures for efficient radioactivity management.

Alpha-emitters (e.g., ²³⁵U, ²³⁸U, ²³⁹⁺²⁴⁰Pu, ²⁴¹Am, etc.) in the environment are known to have a relatively high toxicity. Thus these nuclides have been a focus of interest not only in the waste management field from nuclear power plants, but also in environmental studies for the origin of the nuclear materials.

In general, α -spectrometry has a prominent measurement capability at a very low activity level of ²³⁸U, ²³⁵U, and ²³⁴U, it has a major disadvantage of a long counting time (especially ²³⁵U) for the determination of α -nuclide activity. Contrary to the α -spectrometry method, a measurement technique using ICP-MS with an advanced sample introduction and mass counting system allows radioactivity in many samples to be measured within a short time period with a high degree of accuracy and precision. It should be noted that the ICP-MS technique has proved advanced capabilities to determine very low levels of activity concentration for the long half-lived radionuclides such as ²³⁸U, ²³⁵U, ²³⁷Np, ²³²Th, ⁹⁹Tc, and ²³⁹⁺²⁴⁰Pu.

However, both methods encounter the most significant difficulties during pretreatment (e.g., purification, speciation, and dilution/enrichment). Because the pretreatment process consequently plays an important role in the measurement uncertainty, a method of development and validation should be performed.

In this study, a rapid digestion and separation method for U isotopes was developed in various environmental sample media such as soil, sediment, ground water, air particulate, and food stuff. The pretreatment techniques for specific sample media were developed for rapid analysis. Furthermore, the selective

extraction chromatography process was developed for concentrating and purification of U isotopes for ICP-MS measurement. Finally, the evaluated method in this study were applied for quantitative determination of U isotopes of the environmental samples in the vicinity of research reactor facility.

2. Experimental

2.1 Apparatus and reagents

A reagent of analytical grade and deionized water (>18 M Ω) were used for each process. For the alkali-fusion of solid powder samples, lithium metaborate and lithium bromide (XRF scientific Limited) with an automatic fusion machine (K2 Prime, Katanax, Canada) were used. For an analysis of the radioactivity, ICP-MS (SPECTROMS, SPECTRO, Germany) with an APEX nebulizer, and α -spectrometry (Alpha AnalystTM, CANBERRA, USA) were used. For the concentration and purification of U isotopes UTEVA resin (EICHROM, USA) was used.

2.2 Procedure

An analysis of solid matrix samples is especially complicated owing to a destruction of the matrix and digestion into a liquid phase. These procedures are usually very time consuming and do not fully recover target isotopes. Thus, a rapid and accurate analytical method that can be used to evaluate the radioactivity of U isotopes should be developed and validated.

The analytical process for determination of U isotopes in environmental samples are shown in Fig. 1. The solid powder samples were digested using an alkali-fusion method and concentrated using iron hydroxide co-precipitation. The co-precipitation was conducted by adding an Fe-carrier and adjusting the pH to 7 using a concentrated ammonia solution. After the precipitate was allowed to settle down by centrifuging, the supernatant was discarded and the precipitate was dissolved with 20 mL of concentrated HNO₃. To determine the ²³⁸U concentration, the solution was diluted by about 200 times and initially measured using ICP-MS. For the concentration of U isotopes, the solution was passed through UTEVA resin. For the samples loading and rinsing, 20 mL of 3 M HNO₃ was used. 9 M HCl and 5 M HCl-0.05 M oxalic acid were sequentially pass through the column for Th, Np, and

Pu. Finally, the purified U solution was eluted by 0.01 M HNO₃.

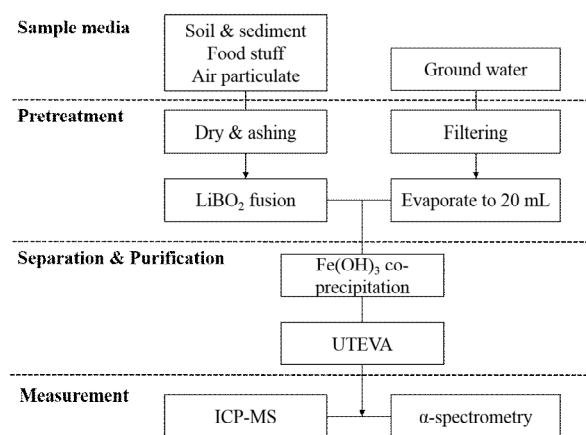


Fig. 1 Analytical procedure for U isotopes in environmental samples

3. Results and Discussions

In order to satisfy the method detectable activity (MDA) for the regulation of a radioactivity monitoring program, the analysis of U isotopes always requires an extremely large sample amount. These procedures usually remove all of the organics in the samples during the dry ashing. The ashed residues have been especially complicated into a liquid phase because of their carbonization. This process is very time consuming and does not fully recover the target isotopes. Thus, a digestion method for ashed-residue was developed for a rapid analysis of U isotopes using LiBO₂ fusion and Fe co-precipitation. The liquid samples were directly vaporized and co-precipitated by Fe(OH)₃ for concentration of target nuclides.

The 80 mL of a prepared solution was loaded into the 3 mL bed volume of a preconditioned UTEVA. The matrix elements (e.g., Fe, lanthanides, alkali earth metals, etc.) were almost removed at the stage of loading and rinsing. With the exception of U isotopes, actinides were eliminated by loading 9 M HCl and oxalic acid, in a step-wise manner. The solution of the extraction process using UTEVA effectively provide a matrix-free and high concentration of U. Finally, the mass concentrations and isotopic ratios of U were determined using a sector-field ICP-MS system with an APEX nebulizer.

For an evaluation of the accuracy and precision of the evaluated method, various reference materials (RMs) from the IAEA and NIST were analyzed. Based on the analytical results, one may conclude that the evaluated method for actinides using alkali-fusion, Fe

co-precipitation, the extraction chromatography process, and ICP-MS measurements is fast and fairly reliable.

U isotopic ratios in environmental samples were analyzed with established procedures. The radioactivity of ²³⁸U in soil, sediment, ground water, and air particulate in the vicinity of research reactor facility ranged from 27.3 to 48.2 Bq·kg⁻¹, from 21.7 to 51.0 Bq·kg⁻¹, from 5.99 to 196 Bq·L⁻¹, and 0.538~3.89 μBq·m⁻³, respectively.

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