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Performance Test of Alpha Spectrometry for Environmental Radioactivity Analysis

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1. Introduction

Environmental sampling is the most effective method in safeguards the detection of undeclared nuclear activities. [1]

Environmental samples are analyzed by various methods such as, ICP-MS (inductively coupled plasma mass spectrometry), AMS (accelerator mass spectrometry) TIMS (thermal ionization mass spectrometry), HRGS (high resolution gamma spectrometry) and alpha /beta particle analysis [2-5].

In this study, we will described the result of performance test using alpha spectrometry for analyzing environmental samples.

2. Experimental Instrument and Materials

2.1 Preparation of sample

In this study, we used two standard reference materials (SRM) and ^{232}U tracer. The ^{232}U standard solution was used for determination of recovery of radiochemical separation. This solution was diluted with 2mol dm^{-3} nitric acid by National Physical Laboratory (NPL). Activity concentration of principal radionuclide is 10.0 Bq^{-1} (±0.10 Bq^{-1}, k =2).

Soil samples are SRM (Standard Reference material) 4350B (River Sediment, NIST) and 4353A (Rocky Flats Soil Number2, NIST).

An acid in this experiment were the ACS (American Chemical Society Reagent) grade for the sample digestion. We used HNO_3 (Suprapur;65%, 69%), HCl(37%) from Merck, and H_2O_2, HF (48%), H_2SO_4, Ammonium water, Ascorbic Acid, Oxalic Acid from SIGMA ALDRICH. Water was deionized and purified with Milli-Q water system.

SRM samples are soil. Soil sample is pre-treatment in order to prepare the solution for the chemical separation of U [3-4]. The step of sample preparation is described in Table. 1.

Table. 1. Preparation step of sample

1	**Sample weighing**	0.5g	
2	**Burned in a muffle furnace**	0~100℃	1 h
		100~200℃	6 h
		200~350℃	5 h
		350~450℃	3 h
		450℃	more than 20 h
3	**Add to ^{232}U tracer**	0.5 g	
4	**Evaporate**	low temperature	
5	**Add to mixed acid**	1 ㎖ HNO_3+4 ㎖ HF+1 ㎖ $HClO_4$	
6	**Packing & Heating**	more than 20 h	
7	**Open & Evaporate acid**		
8	**Add to HNO_3**	1 ㎖ 5M HNO_3 + 1 ㎖ Mill-Q	
9	**Add to H_2O_2**	3~4 drop	
10	**Evaporate**	slowly & low temperature	
11	**Add to acid & Heating**	15ml 3M HNO_3+0.5M $Al(No_3)_3$	
12	**Add to Ascorbic Acid**	0.05~0.1g 0.02M Ascorbic Acid	
13	**Add to HNO_3**	3M HNO_3	
14	**Centrifugation**		

2.2 Separation of uranium

UTEVA resin were used for the extraction chromatographic. UTEVA resin does not absorb mono-, di-, and tri-valent ions. Thus, the resin effectively separates uranium isotope from any interfering elements prior to electrodeposition [5]. The column was preconditioning with 30ml 3M HNO_3. Sample had been dissolved 15ml 3M HNO_3 was loaded onto a UTEVA resin column (100~150 ㎛). Because ^{232}U is solution, It was prepared without pre-treatment. Just column only. The process is summarized in Fig.1. [5-7].

Separation procedure of Uranium by extraction chromatography

1		preconditioning with 30ml 3M HNO_3
2	Pu	**Sample loading** 15ml 3M HNO_3 (to remove Fe, Th)
		15ml 3M HNO_3
3	Th	5ml 9M HCl
		20ml (5M HCl+0.05M $C_2H_2O_4$) (to remove Pu, Np, Th)
4	**U**	**30ml 0.01M HCl**

Fig.1. Diagram of the separation procedure of Uranium by extraction chromatography with UTEVA resin.

2.3 measurements

The disk sources for alpha spectrometric measurements were prepared by electrodeposition .

Alpha spectrometry (Canberra) used in this study, is shown in figure 1 and its characteristics are described in table 1.

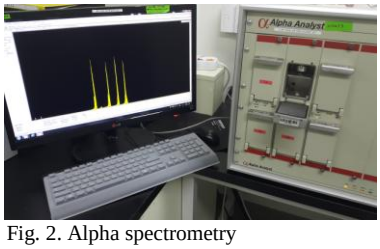

Fig. 2. Alpha spectrometry

Table. 2. Characteristics of alpha spectrometry

Sample size (detectable)	Up to 51 mm (in diameter)
Sample-to-detector spacing	1 to 45 mm (in 4 mm increments)
Energy resolution	< 18 keV (FWHM)
Energy range	3 to 15 MeV
Active area	450mm^2
Background	< 1 count/hr (above 3 MeV)
Vacuum rage	0 to > 26.7 kPa

2.4 Results

Measurement data of the uranium activity using the SRM samples are reported in Table. 3. There are also shown the Certificate of sample.

Measured recovery results are from 75.96 to 112.9%. Generally, measured recovery results are from 90 to 100% [6-8]. Concentration of U results are lack of consistency. We need to verification of experimental stage for the better results.

Table.3. Result of the activity concentration of U

Sample Name	**Time (s)**	**U**	**Certificate value** (Bq/g)	**Measured** (Bq/g)	**recovery ^{232}U (%)**
4353a -1	10 만	238	0.0396	0.0622	99.05
		235	0.0019	0.0028	
		234	0.0404	0.0694	
		232			
4353a -2	10 만	238	0.0396	0.0606	
		235	0.0019	0.0016	
		234	0.0404	0.0639	
		232			
4353a -3	10 만	238	0.0396	0.0600	
		235	0.0019	0.0020	
		234	0.0404	0.0683	
		232			
4353a -4	10 만	238	0.0396	0.0148	100.7
		235	0.0019	0.0008	
		234	0.0404	0.0165	
		232			
4350B -1	10 만	238	0.0308	0.0227	75.96
		235	0.0017	0.0011	
		234	0.0332	0.0274	
		232			
4350B -2	10 만	238	0.0308	0.0312	104.4
		235	0.0017	0.0011	
		234	0.0332	0.0345	
		232			
^{232}U_1	10 만	232			109.6
^{232}U_2	10 만	232			112.9

3. Conclusions

Measurement data of the U activity using SRM based on extraction chromatography with UTEVA resin. It should be effective way to separate of uranium isotope for the measurement of alpha spectrometry.

But, the result of this measurement data is higher than another recovery data. Also concentration of U data is lack of consistency.

We leave out of consideration many effect of factors about influence in the experiment process.

In the future work, we will try to reduce the step of experiment process and reflect the uncertainty factors.

Furthermore, we will study of the laboratory's quality assurance and quality control for the environmental sample analysis. Because the environmental sample analysis has purpose of safeguards for tracking nuclear activities of nuclear facilities.

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