Age-dating of UO_2 Based on $^{230}Th/^{234}U$ Determination by New Chemical Separation Method Using UTEVA Resin

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1. Introduction

Nuclear forensic deals with the history of nuclear and radioactive materials. The history of nuclear and radioactive materials is including the separation method, purification method, production type and age, etc. Age-determination of uranium samples is an important technique for nuclear forensics by analyzing a daughter-mother radionuclide pair such as $^{230}Th/^{234}U$, $^{231}Pa/^{235}U$. $^{230}Th/^{234}U$ is one of the most commonly used isotope pair due to relatively rapid ingrowth of ^{230}Th in comparison with others and availability of high precision. For accurate and precise determination of $^{230}Th/^{234}U$ ratio, Th must be purified by removing interfering species from uranium samples prior to analysis as much as possible. Additionally, ^{232}Th impurities during chemical procedures should be minimized and well evaluated because standard ^{232}Th is used as a spike for isotope dilution mass spectrometry (IDMS). For this purpose, we developed relatively simple and effective chemical separation method based on UTEVA resin and optimized separation conditions.

2. Experimental

2.1 Analytical Procedure

We prepared simulated samples to obtain elution curve and recovery yield. Uranium standard solution 500 μg/mL and thorium standard solution 10 ng/mL (SPEX Certiprep®) were mixed in 5 M HNO_3. To verify this method, we prepared UO_2 samples dissolved in HNO_3. This sample was diluted in 5 M HNO_3 for using chemical separation. After separation isotopic ratio measurements of uranium and thorium for IDMS were performed using MC-ICP-MS.
For chemical separation, we used UTEVA resin (100-150 mesh, Eichrom) 1 mL and 5 M HCl as thorium eluent solution. One step was added for purification of thorium in separated samples. Total analytical procedure is shown in Fig. 1.

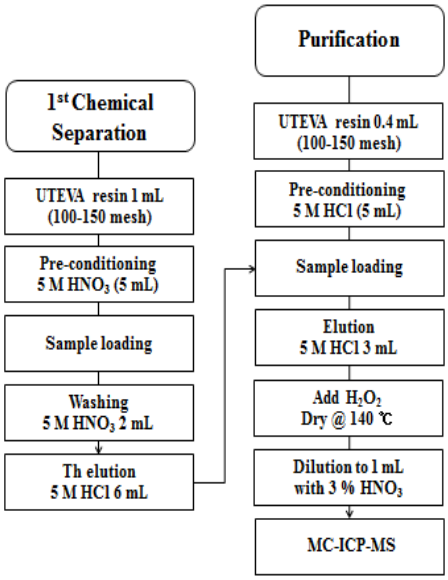

Fig. 1. Total chemical separation steps to separate uranium and thorium.

2.2 Instrumentation

Uranium and thorium isotopic ratio measurements were performed using MC-ICP-MS (Neptune Plus, Thermo Scientific Inc., Germany) equipped with desolvation system (Aridus II, CETAC, USA). For IDMS, Th isotopes were determined based on the simultaneous measurements using SEM and Faraday cup.
Since desolvation system reduced the formation of hydride and oxide form of thorium, it enhanced signal sensitivities. The typical operating conditions of MC-ICP-MS and a desolvation system are summarized in Table 1.

3. Results and Discussion

First, we optimized chemical separation condition to separate uranium and thorium. Elution curve and recovery yield were obtained from newly developed separation schemes. As shown in Fig.2 the optimal volumes of thorium eluent (5 M HCl) was 6 mL and the recovery yield was 99.5 %.

Table 1: Optimized operating conditions of MC-ICP-MS and a desolvation system

	Parameter	Setting
MC-ICP-MS	RF power	1200 W
	Cooling gas flow rate	15 L/min
	Auxiliary gas flow rate	0.8 L/min
	Mass resolution (ΔM/M)	400
	Number of spectra acquired	10 x 3
Desolvation system	Solution take rate	180 μL/min
	Spray chamber temp.	110 ℃
	Membrane temp.	160 ℃
	Ar flow rate	~ 4.3 L/min
	N_2 flow rate	~ 7 mL/min

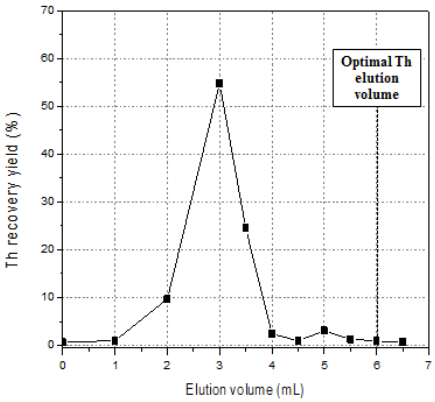

Fig.2 The elution curve of thorium.

This optimized condition is applied to separate uranium and thorium in UO_2 reference materials. As a result we confirmed that uranium and thorium were separated successfully and estimated amount of ^{230}Th and ^{234}U respectively. The ratio of ^{230}Th and ^{234}U is shown in Table 2.

Table 2: The ratio of ^{230}Th/^{234}U after chemical separation using UTEVA resin

UO_2 material	#1	#2	#3	Average	RSD (%)
^{230}Th/^{234}U	3.46 x 10^{-5}	3.55 x 10^{-5}	3.55 x 10^{-5}	3.42 x 10^{-5}	1.4

This new chemical separation method is expected to be more versatile than previous method using TEVA resin or ion exchange chromatography because it is possible to evaluate Pu age by analyzing ^{241}Am/^{241}Pu as well as ^{230}Th/^{234}U at the same time.

REFERENCES

[1] Lim, Sang Ho, et al. "Improvement in the chemical separation and determination of uncertainties for bulk analysis of Pu isotopes at ultra-trace levels by using MC-ICP-MS." Journal of Radioanalytical and Nuclear Chemistry 307.3 (2016): 1853-1859.