Determination of ^{236}U in a Th target irradiated with Li ions by ICP mass spectrometry

A. Nagai,[*1,*2] K. Teranishi,[*1,*2] R. Morita,[*1,*2] H. Hosokawa,[*1,*3] A. Yokoyama,[*1,*3] A. Nakajima,[*1,*4] A. Sakaguchi,[*1,*5] Y. Shigekawa,[*1] A. Nambu,[*1] T. Yokokita,[*1] and H. Haba[*1]

Neptunium-237 ($T_{1/2} = 2.1\times10^6$ y) released from nuclear facilities is an important radioactive target nuclide that contaminates the environment.[1] The nuclide can be analyzed by accelerator mass spectrometry; however, a spike material of neptunium for the measurement has not been obtained, thus far. We previously proposed the production of neptunium-236 ($T_{1/2} = 1.5 \times 10^5$ y) in a Th + Li reaction as a candidate for the material.[2,3] However, the material prepared from the irradiated target may be contaminated with long-lived isobars of ^{236}Pu and ^{236}U, which are the decay products from ^{236m}Np, or a byproduct in the reaction. In addition, ^{235}U contained in Th as an impurity, may also absorb neutrons and produce ^{236}U. In this study, we aimed to determine U isotopes contained originally in the Th target as well as those produced in the nuclear reaction by High Resolution ICP-MS (HR-ICP-MS). The amount of ^{236}U produced in the reaction can be estimated by measuring a neutron flux during irradiation through the use of a monitoring foil of Au.

We irradiated a ^{232}Th target with 43 MeV ^{7}Li ions at the RIKEN AVF cyclotron. Au metal foils (ca. 200 mg each) and electrodeposited U samples (ca. 1 mg each) were placed at the end of the beam course to function as a neutron monitor and a reference, respectively, as illustrated in Fig. 1.

After irradiation, the neutron flux was obtained from activation of the Au foil measured by γ-ray spectroscopy using a Ge semiconductor detector. The U-electrodeposited sample was dissolved in a concentrated

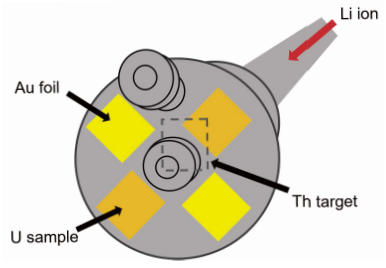

Fig. 1. Experimental setup of neutron monitor foils (Au) and U targets attached to the end of a target holder of an irradiation beam course at RIKEN.

*1 RIKEN Nishina Center
*2 Graduate School of Natural Science and Technology, Kanazawa University
*3 Institute and College of Science and Engineering, Kanazawa University
*4 Master's Program in Chemistry, University of Tsukuba
*5 Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba

Table 1. U isotopic ratios in U-electrodeposited samples by ICP-MS.

	Irradiated		Unirradiated
	U-1	U2	U-3
	Ratio%		Ratio%
235/238	0.62~0.82	0.61~0.84	0.63~0.85
236/238	0.5×0.82^{-4}	$(1–7)\times0.82^{-4}$	N.D

nitrate solution. Subsequently, the solution was diluted so that the U concentration was 0.1 or 1 ng/g, as measured by HR-ICP-MS with a reference solution of XSTC-13 supplied by SPEX Industries, Inc. for calibration.

From the measurements of photopeak intensities due to ^{198}Au and ^{196}Au in the Au foils, the thermal neutron flux and fast neutron flux were 6.5×10^5 s^{-1} and 1.6×10^7 s^{-1}, respectively. These values were calculated from induced radioactivities and the cross section data[4] of (n,γ) and $(n,2n)$ reactions. The isotope measurement results of the U-electrodeposited sample by HR-ICP-MS are shown in Table 1, where the ratios denote the range of observation due to the large dispersion of the data depending on the sample. The abundance ratios of ^{235}U to ^{238}U for all the samples are approximately 0.7%, corresponding to the natural abundance of U. Some events due to ^{236}U were observed for the samples attached to the target holder. However, the events due to the unirradiated sample was less than the detection limit. It was confirmed that ^{236}U was produced in the U-electrodeposited samples. The amount of ^{236}U produced agrees with the amount estimated from the thermal neutron flux by the monitor within some uncertainty of the data. Based on the measured concentration of U and the measured thermal flux, the number of ^{236}U atoms produced in the target sample was $(0.9 - 2.0) \times 10^2$, much less than the number of ^{236g}Np atoms estimated from the measured cross section,[2] that is, approximately 1 mb. Thus, the amount of U was concluded to be negligible to contaminate a Np tracer prepared from the irradiated target.

References

1) W. Rund, G. S. Goff, in *Radionuclide in the Environment*, edited by D. A. Atwood (Wiley, 2013).
2) Y. Hayakawa *et al.*, RIKEN Accel. Prog. Rep. **53**, 179 (2020).
3) A. Nakajima *et al.*, RIKEN Accel. Prog. Rep. **54**, 142 (2021).
4) Japanese Evaluated Nuclear Data Library, JENDL-4.0, (2010), `https://wwwndc.jaea.go.jp/jendl/Jendl_J.html` .