Measurement of the isotopic ratio of Np-236 to Np-237 in Th-232 + Li-7 reaction products by accelerator mass spectrometry

A. Nakajima,,[*1,*2] K. Teranishi,[*1,*3] A. Yokoyama,[*1,*4] A. Sakaguch,[*1,*5] K. Hain,[*6] R. Morita,[*1,*3] A. Seto,[*1,*4] A. Nagai,[*1,*4] D. Mori,[*1] Y. Komori,[*1] T. Yokokita,[*1] Y. Wang,[*1] and H. Haba[*1]

Neptunium-236 can be a useful tracer in the determination of Np-237, an isotope of the minor actinide Np, which exists in tiny quantities in the environment owing to its release from nuclear facilities and nuclear tests. Such determination is of practical use in various earth science fields such as surface material circulation and environmental pollution assessment.[1] The measurement of Np-237 is expected to be quantified by accelerator mass spectrometry (AMS), although an internal standard method for Np needs an appropriate spike. Chemical yield tracers for several elements are available now, but the spike for neptunium has not been developed yet. We aim to devise an efficient method for the production of Np-236 in the ground state of half-life 1.54×10^5 y as a candidate for the spike nuclide, with minimal contamination by Np-237.

In this study, Np-236 tracer production was implemented in the reaction of Th-232 + Li-7 to measure Np-237. The resulting yields of Np-236m and Np-236g were presented in the previous report.[2] The yield in the ground state (6^-) was found to be larger than that in the excited state (1^+) in the studied energy range. This demonstrates that the reaction system adopted is promising. In addition, we have to measure the isotopic ratios of Np-236g to Np-237 in the same nuclear system.

We irradiated Th foil targets with 42 MeV Li-7 ions from the RIKEN AVF cyclotron while integrating the beam current with a Faraday cup in the irradiation course. Chemical procedures were performed to isolate Np atoms from the target. Details of the procedures were described previously.[2]

For the Np-236g and Np-237 measurement, samples for AMS and inductively coupled plasma mass spectrometry (ICP-MS) were prepared through purification with UTEVA resin after waiting for Np-236m to decay out, and they were brought to the VERA facility in the University of Vienna, with which this project is in collaboration.

Figure 1 shows the preliminary isotopic ratios of Np-236g to Np-237 isotopes measured using AMS. The yield of Np-236 was found to be approximately 10 times larger than that of Np-237 in the studied energy range. The figure also shows the ratios calculated under a simple assumption with the nuclear reaction model code EMPIRE2[3] comprising various nuclear models. A large discrepancy is observed between the observation and prediction. It may be due to difficulty in the yield reproduction of used models at the energy region beyond the maximum yield energy for the products. The observation indicates that the reaction system adopted is promising thus far, although the data may need to be modified because of contamination by uranium nuclides and other nuclides with a mass number of 236.

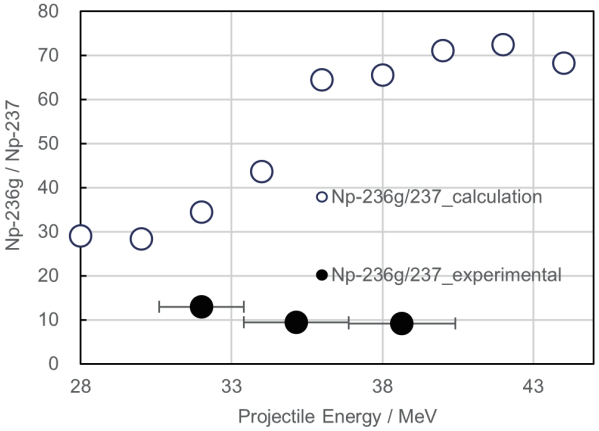

Fig. 1. Preliminary isotopic ratios of Np-236g to Np-237 in the Th-232 + Li-7 reaction system compared with the theoretical prediction. Closed circles represent the observation, while open circles indicate data from calculation.

The analysis of the result is in progress, and additional experiments including mass spectrometry are in planning. We aim to confirm the precision and reproducibility of the data in consideration of the contamination by isobars, which affects the mass measurement and is a major concern.

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

References

1) W. Rund, G. S. Goff, *Radionuclide in the Environment*, edited by D. A. Atwood (Wiley, 2013).
2) Y. Hayakawa *et al.*, RIKEN Accel. Prog. Rep. **53**, 179 (2020).
3) M. Herman *et al.*, Nucl. Data Sheets **108**, 2655 (2007).