

Synthesis of Np isotopes via the $^{232}\text{Th} + ^7\text{Li}$ nuclear reaction

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Human nuclear activities have released long-lived actinides, including neptunium (Np)-237 ($T_{1/2} = 2.1$ My), into the environment. Monitoring of this nuclide in the environment is essential not only from the perspective of radiation protection but also for its usage as an environmental dynamic tracer. To use Np-236g as a spike for the mass spectrometric analysis of Np-237, the synthesis of Np-236g via the Th-232 + Li-7 nuclear reaction has been attempted.¹⁾ In this study, we performed gamma spectrometry and determined the nuclear-reaction cross sections of Np-234 and Np-236m produced by side reactions to validate the experimental system used for Np-236g synthesis, and to deepen our understanding of the Th-232 + Li-7 nuclear-reaction mechanism.

The irradiation experiment was based on the method described by Teranishi *et al.*²⁾ as follows: Thorium metal foils (purity 99.5%, area density 9–16 mg/cm²) or thorium nitrate electrodeposits on an Al foil (purity >99%, area density 0.5–1 mg-Th/cm²) were used to compose a stacked target for irradiation. At the RIKEN RIBF, the stacked target was irradiated with a ⁷Li beam (approximately 1 particle μA , approximately 42 MeV) for approximately 10 hours. The energy incident on each target was adjusted using an aluminum foil sandwiched between the targets as a degrader. The irradiated samples were dissolved in acid, and the target material of Th and fission products were separated via TEVA resin. Additionally, Pa isotopes remaining in the Np fraction obtained by chemical separation with TEVA resin were removed using TK400 resin. The gamma rays emitted from Np-234 and Np-236m present in the Np fraction were measured using a Ge semiconductor detector to quantify the radioactivity. In these experiments, a known amount of the Np-239 tracer was added to the initial sample solution before chemical separation to determine the separation yield.

Chemical separation significantly removed interfering nuclides, resulting in gamma-ray spectra showing clear peaks of the target nuclides Np-234 and Np-236m. Within the irradiation energy range of 34.2–42.2 MeV, the minimum nuclear-reaction cross section of Np-234 was 0.0344 ± 0.0013 mb at an irradiation energy of 34.2 MeV, and the maximum value was obtained to be 5.26 ± 0.28 mb at an irradiation energy

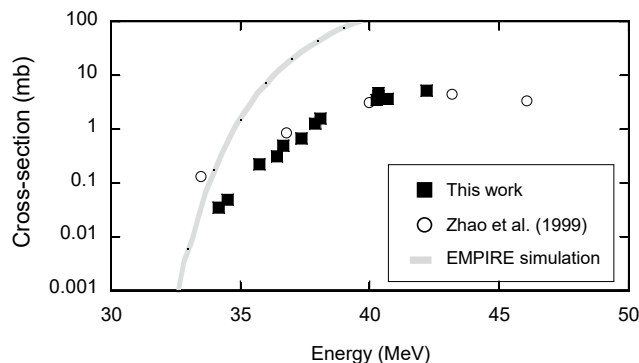


Fig. 1. Cross sections of ^{234}Np obtained from Th-232 + Li-7 reactions.

of 42.2 MeV (Fig. 1). The excitation function obtained here for the $^{232}\text{Th}(^7\text{Li}, 5n)^{234}\text{Np}$ reaction reproduces very well the cross sections obtained by Zhao *et al.*³⁾ very well, thus providing an excitation function based on detailed data with higher accuracy. This result demonstrates the validity of the method of using Np-234 as a standard for the determination of cross sections for other Np-producing reactions. However, these nuclear-reaction cross sections were one to two orders of magnitude smaller than those simulated by EMPIRE II.⁴⁾ This is probably due to the significant influence of side reactions as shown by Teranishi *et al.*²⁾ and the low accuracy of the simulation. Additionally, the proposed method succeeded in obtaining the nuclear-reaction cross section of Np-236m at irradiation energies of 28.6–42.2 MeV, ranging from 0.12 mb (28.6 MeV) to 0.41 mb (35.7 MeV). This result suggests that Np-236g is also synthesized in this reaction system based on the theoretical calculations by EMPIRE II.^{2,4)} The production amounts of Np-236g and Np-237, which interferes with the analysis of environmental samples, will be quantified by mass spectrometry.

In conclusion, we found that the chemical separation method for measuring the gamma rays of Np isotopes synthesized by the Th-232 + Li-7 nuclear reaction works well. Np-234 and Np-236m were synthesized, and their nuclear-reaction cross sections and excitation functions were obtained. This reveals the possibility of spike contamination by Np isotopes other than Np-236g, which we aim to synthesize. The effect of these isotopes on Np-237 quantification will be discussed in the future.

References

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