Production cross sections of ^{225}Ac in the ^{232}Th(^{14}N, $xnyp$) reactions at 116 and 132 MeV/nucleon

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^{225}Ac ($T_{1/2}$ = 10.0 d) is one of the most promising alpha-particle-emitting radionuclides for targeted radionuclide therapy.[1)] However, the current global availability of ^{225}Ac is too small to support large clinical trials, and a stable supply system for ^{225}Ac has not yet been established in Japan even at the basic research scale of 100 MBq. A spallation reaction of ^{232}Th with high-energy protons is expected to be a potential production route for ^{225}Ac.[2)] At RIKEN, radionuclides of a large number of elements, called multitracer, have been produced by the spallation of metallic targets such as natTi, natAg, and ^{197}Au irradiated with a 135 MeV/nucleon ^{14}N beam from the RIKEN Ring Cyclotron (RRC).[3)] In this work, we investigated the feasibility of ^{225}Ac production via the ^{232}Th(^{14}N, $xnyp$)^{225}Ac reaction for the future domestic supply of ^{225}Ac. We also investigated the production of ^{225}Ra ($T_{1/2}$ = 14.9 d) because it is useful as an ^{225}Ac/^{225}Ra generator to produce high-radionuclidic-purity ^{225}Ac.[2)]

A ^{14}N^{7+} beam was extracted from the RRC. Three metallic ^{232}Th foils (69 mg/cm^2), two ^{27}Al plates (415 mg/cm^2), and another three ^{232}Th foils were placed in this order from the upstream side of the beam in the multitracer production chamber.[3)] The targets were irradiated for 1 h with a 20-pnA-intensity beam.

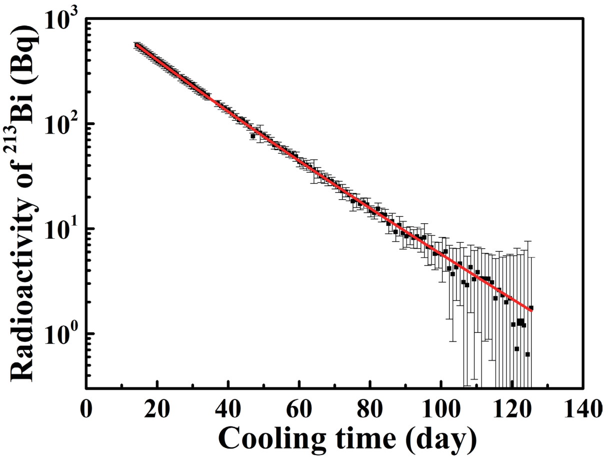

Fig. 1. Radioactive decay curve of the 440.5-keV γ-line of ^{213}Bi on the decay chain of ^{225}Ra → ^{225}Ac → ^{221}Fr → ^{227}At → ^{213}Bi → ⋯. The solid curve indicates the fitting result using the two-body successive decay equation (^{225}Ra → ^{225}Ac → ⋯).

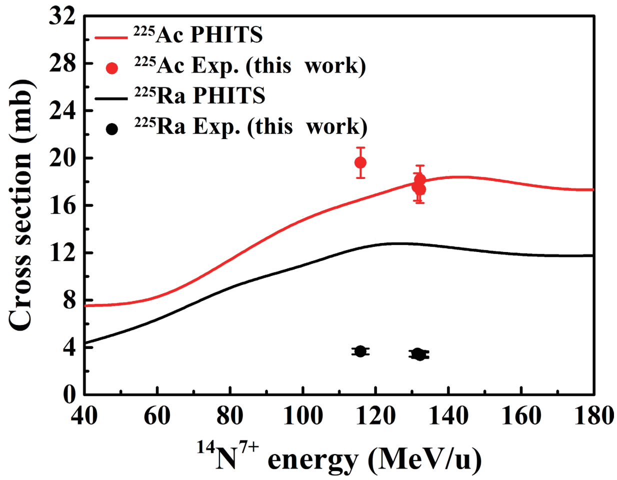

Fig. 2. Cross sections of the ^{232}Th(^{14}N, $xnyp$)^{225}Ac, ^{225}Ra reactions in comparison with the PHITS calculations.

After the irradiation, the second foil of each set of three ^{232}Th foils was subjected to γ-ray spectrometry with Ge detectors to determine the production cross sections of ^{225}Ac and ^{225}Ra. The ^{27}Al plates were used as beam-energy degraders. The beam energies on the measured ^{232}Th targets were calculated to be 132 and 116 MeV/nucleon using the stopping power model[4)] in the LISE^{++} program.[5)]

The radioactivities of ^{225}Ac and ^{225}Ra at the end of the irradiation were determined by following the activity of ^{213}Bi ($T_{1/2}$ = 45.59 min), which was in radioactive equilibrium as the great granddaughter of ^{225}Ac. Figure 1 shows a typical decay curve of the 440.5-keV γ-line of ^{213}Bi. The two-body successive decay equation (^{225}Ra → ^{225}Ac → ⋯) was applied to fit the decay curve after subtracting the small contribution of the 440.4-keV γ-ray of ^{228}Ac, which originally existed in the ^{232}Th target as the granddaughter of ^{232}Th. Some short-lived parents of ^{225}Ac and ^{225}Ra were produced in the reactions; therefore the measured cross sections of ^{225}Ac and ^{225}Ra are cumulative for electron-capture decay and β^- decay, respectively. The cross sections of the ^{232}Th(^{14}N, $xnyp$)^{225}Ac,^{225}Ra reaction are shown in Fig. 2. The cross sections of ^{225}Ac are larger than those of ^{225}Ra by a factor of 5. The experimental results were compared with those calculated by the Particle and Heavy Ion Transport code System (PHITS).[6)] The PHITS code reproduces the cross sections of ^{225}Ac, while it overestimates those of ^{225}Ra by a factor of 4. The production yield of ^{225}Ac was tentatively evaluated to be 3.3 MBq/pμA·h

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at 132–80 MeV/nucleon by normalizing the PHITS calculations to the experimental cross sections. Based on our typical experimental conditions (incident beam energy: 132 MeV; beam intensity: 1 pμA; target thickness: 4.5 g/cm^2; irradiation time: 2 d), approximately 150 MBq of ^{225}Ac can be produced at the end of the irradiation. In the near future, we will measure the cross sections of ^{225}Ac and ^{225}Ra at lower energies of 80 and 100 MeV/nucleon to evaluate their yields more reliably.

References

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