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 $^{222}$Ra ($T_{1/2} = 14.9$ d) because it is useful as an $^{225}$Ac/$^{225}$Ra generator to produce high-radiouclidic-purity $^{225}$Ac.2)

A $^{14}$N$^+$ beam was extracted from the RRC. Three metallic $^{23}$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.

After the irradiation, the second foil of each set of three $^{232}$Th foils was subjected to $\gamma$-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 model5) 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 $\gamma$-line of $^{213}$Bi. The two-body successive decay equation ($^{222}$Ra $\rightarrow$ $^{225}$Ac $\rightarrow$ $\cdots$) was applied to fit the decay curve after subtracting the small contribution of the $^{440.4}$keV $\gamma$-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 $\beta^-$ 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).5) 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/µA-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 μA; target thickness: 4.5 g/cm²; 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