Production yield of ^{93}Zr by ^{93}Nb (n,p) reaction for collinear laser spectroscopy

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The half-lives of long-lived fission products (LLFP) serve as crucial nuclear data for managing nuclear waste.[1)] Among LLFPs, ^{93}Zr is the second strongest radioactive element in spent nuclear fuel 1000 years later. However, its half-life ($\sim 1.6\times10^6$ y) has not been measured so accurately, showing significant discrepancies in values obtained from previous mass spectroscopy measurements. To obtain a reliable reference value of the half-life, a new method using collinear laser spectroscopy is proposed.[2,3)] Previous measurements involved the chemical separation of ^{93}Zr samples from spent nuclear fuel. However, such samples may contain impurities from various elements, leading to systematic errors in half-life measurements. Contrastingly, in our method, ^{93}Zr is produced through ^{93}Nb(n,p) reaction using accelerator neutrons. As this method limits the formation of reaction products other than ^{93}Zr, high-purity samples can be obtained after chemical separation.

In October 2021, a one-day test irradiation of the ^{93}Nb target with accelerator neutrons was conducted. The neutrons, which had a continuous energy spectrum of up to approximately 30 MeV, were provided by ^{9}Be(d,n) reaction using a 30 MeV deuteron beam from the AVF cyclotron. The ^{9}Be target had a thickness of 1.85 g/cm^2, which was sufficient to stop the incident deuteron beam. The ^{93}Nb target, which comprised 120 layers of ^{93}Nb metal disks measuring 1 mm in thickness and 15 mm in diameter, had a thickness of 102.8 g/cm^2. The targets were arranged to test the validity of the evaluated energy and angular distributions of the accelerator neutrons. The ^{93}Nb target furthest upstream was set 3.5 mm behind the Be target. The deuteron beam current was monitored during the beam time using the ^{9}Be target holder as a Faraday cup. The beam intensity was approximately 10 μA.

To estimate the production yield of ^{93}Zr, we conducted gamma-ray measurements on the ^{93}Nb targets using an HPGe detector. The measurements were obtained 5 days and 10 months after the end of irradiation. In the first measurement, we observed strong γ-rays of ^{92m}Nb ($T_{1/2}$ = 10.15 d) produced by the ^{93}Nb$(n,2n)$ reaction. We determined the production yield of ^{92m}Nb by considering the half-life, branching ratio of observed γ-rays, and the gamma-ray detection efficiency of the HPGe detector. The total yield for 120 ^{93}Nb targets was determined to be $3.465\times10^{13}(13)$ atoms (27.39(9) MBq) at the end of irradiation. During a cooling period of several months, the radioactivity of ^{92m}Nb decreased. As a result, in the second measurement, 703- and 871-keV -rays of ^{94}Nb ($T_{1/2} = 2.03\times10^4$ y) produced by the ^{93}Nb(n,γ) reaction, were observed (see Fig. 1). Other γ-rays in the spectrum are from the decay of ^{91m}Nb, ^{182}Ta, which were produced from impurities in the target such as Mo and Ta. The ^{94}Nb yield in the most upstream target was determined to be $6.1(6)\times10^{11}$ atoms.

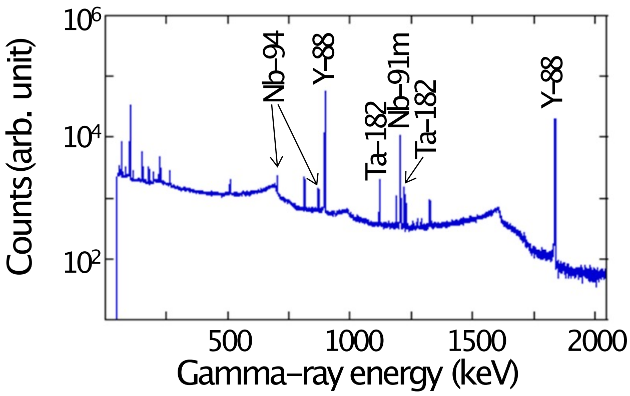

Fig. 1. Example γ-ray spectrum of ^{93}Nb target irradiated with accelerator neutrons measured 10 months after the end of irradiation.

To compare the measured yields with the estimated ones, we used the MCNP code[4)] to calculate the energy and angular distributions of the neutron flux. We assumed the same thickness of the ^{9}Be target and the same energy of the deuteron beam as the experiment. The yields were estimated by multiplying the neutron flux and reaction cross sections in a given ^{93}Nb target geometry from JENDL-5.[5)] The estimated yields for ^{92m}Nb and ^{94}Nb are higher than the measured values, with 1.6×10^{15} and 8.6×10^{12} atoms, respectively. The estimated dependence of yields on the ^{93}Nb target thickness does not agree well with the measured one. The reason for the discrepancies is still unknown. The production yield of ^{93}Zr is estimated to be 1.4×10^{14} atoms, but the actual value may be smaller. ^{93}Zr will be chemically separated from the ^{93}Nb targets and then placed in the ion source of the mass separator for collinear laser spectroscopy. We are currently improving the sensitivity of collinear spectroscopy based on the estimated yield of ^{93}Zr.

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