Production of ^{93}Zr sample in the ^{93}Nb(n, p) reaction towards accurate determination of ^{93}Zr half-life

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The half-live of a long-lived fission product (LLFP) is not reliable, even though it is one of the important basic data to evaluate the long-term radiological risk of nuclear fuel waste. For example, although ^{93}Zr will be the second significant contributor to the fission product activity of nuclear wastes in 1000 years[1] due to its long half-life, the half-life of ^{93}Zr has been reported as 1.53 Myr,[2] 1.13(11) Myr,[3] and 1.64(6) Myr,[1] and the discrepancy between these values is as large as 30%.

Because it is impossible to measure the decay of radioactivity directly by observing it, the half-lives of LLFPs have been deduced by the formula $T_{1/2} = (\ln 2)N/A$ where N is the number of the atoms and A the radioactivity. In the measurements of A and N, impurities in samples cause unpredictable errors, such as isobar impurities in a mass spectrometry. To reduce such errors and to obtain reliable half-lives, we plan to establish a new method using the collinear laser spectroscopy technique for the accurate determination of half-lives of LLFPs, starting with ^{93}Zr. The ^{93}Zr samples in the previous measurements were chemically separated from a spent nuclear fuel and a zircaloy cladding, but both of them contain many nuclides which may lead to unexpected error. We instead prepared a ^{93}Zr sample using the ^{93}Nb(n, p) ^{93}Zr reaction. Because other radioactive nuclides produced simultaneously by fast neutron beams on this target are almost limited to the Nb isotopes from the (n, xn) reaction, pure radioactivity of ^{93}Zr can be obtained by the chemical separation. The number of ^{93}Zr will be determined from the resonance peak intensities in spectra measured by a collinear laser spectroscopy using the ^{93}Zr sample added with a quantified neutral Zr standard. This can avoid the errors from isobar impurities that could occur in a mass spectrometry.

We conducted a 1-day beam time test in October 2021. Neutrons with an energy of approximately 10 MeV were produced in the ^{9}Be(d, n) reaction by irradiating a 30-MeV d beam from the AVF cyclotron onto a beryllium target of 1.85 g/cm^2. The ^{93}Zr isotopes were then produced in the ^{93}Nb target behind the ^{9}Be target by the ^{93}Nb$(n, p)^{93}$Zr reaction. We prepared a ^{93}Nb target of 102.8 g/cm^2, which was composed of 120 layers of 1-mm thick and ϕ15 mm ^{93}Nb disks. During the beam time, the d beam intensity was measured using a ^{9}Be target holder as a Faraday cup. We evaluated that the ^{9}Be target was irradiated with 5.8×10^{18} deutrons in total. Based on the production cross section data from JENDL-4.0, ^{92m}Nb ($T_{1/2} = 10.15$ d) was expected to be the most abundant radioactive source in the target.

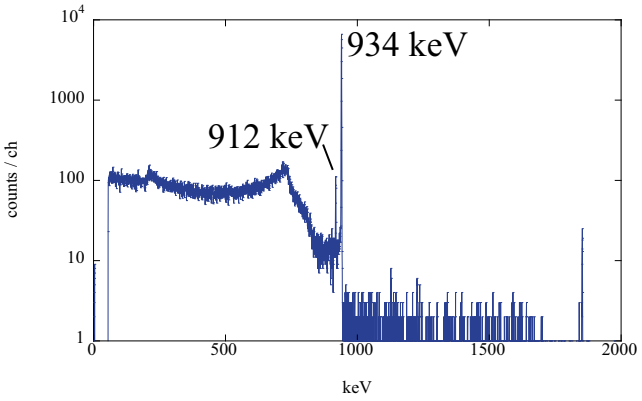

Fig. 1. Example γ-ray spectrum for the neutron-irradiated ^{93}Nb target.

We started γ-ray measurements for the ^{93}Nb targets 5 days after the irradiation. The 120 ^{93}Nb targets were measured one by one using a Ge detector. Figure 1 shows the γ-ray spectrum from the most downstream ^{93}Nb target, as an example, which presents intense peaks of ^{92m}Nb at 934 keV and 912 keV. Using the γ-ray yields of 934 keV from ^{92m}Nb, we evaluated the ^{92m}Nb radioactivity of each target at the end of the beam time, taking into account the measurement date, peak count rates, half-life, γ-ray intensity, and detector efficiency. The total radioactivity was found to be 27.39(9) MBq. A further detailed analysis is in progress.

The target will be chemically separated to extract ^{93}Zr after cooling down to reduce its radioactivity. Then, β-particles from the ^{93}Zr sample will be measured by liquid scintillation counting to measure the ^{93}Zr radioactivity. If we find that the amount of ^{93}Zr is sufficient, we will proceed to the number measurement using our collinear laser spectroscopy setup.[4,5]

The production yields and their dependences on the target thickness obtained in this experiment for ^{92m}Nb and other nuclides can be used to check the validity of the evaluated fast neutron cross section data. We will compare the present results with the predictions using, for example, PHITS[6] with the JENDL data library.

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