Measurement of the neutron capture cross section of ^{99}Tc using ANNRI at J-PARC

Tatsuya Katabuchi[1,a], Motoharu Mizumoto[1], Masayuki Igashira[1], Kazushi Terada[2], Atsushi Kimura[2], Shoji Nakamura[2], Taro Nakao[2], Osamu Iwamoto[2], Nobuyuki Iwamoto[2], Kazuhito Mizuyama[2], Hideo Harada[2], Jun-ich Hori[3], and Koichi Kino[4]

[1] Laboratory for Advanced Nuclear Energy, Tokyo Institute of Technology, Ookayama, Meguro, Tokyo 152-8550, Japan
[2] Nuclear Science and Engineering Center, Japan Atomic Energy Agency, Tokai, Naka, Ibaraki 319-1195, Japan
[3] Research Reactor Institute, Kyoto University, Kumatori, Sennan, Osaka 590-0494, Japan
[4] Faculty of Engineering, Hokkaido University, Kita-ku, Sapporo 060-8628, Japan

Abstract. The neutron capture cross section of ^{99}Tc was measured using NaI(Tl) detectors of the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) at the Japan Proton Accelerator Research Complex (J-PARC) in the energy range from thermal to the keV energy region. Preliminary results were presented and compared with previous measurements and evaluations.

1. Introduction

Long-lived fission products (LLFP) in nuclear spent fuel have been an issue to deal with in the nuclear power industry. Currently-planned geological disposal of nuclear waste has been disputed in public. To solve this long-standing issue, nuclear transmutation, in which LLFPs are transmuted into shorter-life or stable isotopes through neutron-induced reactions, has been proposed. Technetium-99 is one of the LLFPs and has the highest priority to transmute due to its high fission yield and radiotoxicity. In order to design a nuclear transmutation system for ^{99}Tc, reliable neutron cross section data of ^{99}Tc in a wide range of neutron energy are necessary.

In the present work, we carried out time-of-flight (TOF) experiments to measure the neutron capture cross section of ^{99}Tc with the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) [1,2] at the Japan Proton Accelerator Research Complex (J-PARC). ANNRI has been built for nuclear data measurement utilizing a high-intensity pulsed neutron beam from a spallation neutron source [3] in the Materials and Life Science Facility of J-PARC. The neutron capture cross section of ^{99}Tc was measured from thermal to keV neutron energy region. In particular, an effort was made to extend the high energy limit of measurement to a few hundred keV.

2. Experiments and data analysis

NaI(Tl) detectors of ANNRI were used for measurements. Two NaI(Tl) detectors were placed at detection angles of 90° and 125° with respect to the beam axis. The sizes of the NaI(Tl) crystals were 330 mm diameter ×203 mm length and 203 mm diameter ×203 mm length, respectively. A fast data acquisition system based on pulse-width analysis method [4] was implemented to achieve measurement in the high energy region.

A ^{99}Tc metal sample with a radioactivity of 52 MBq was used in measurement. The sample was sealed in an Al container. The size of the sample was 6.3 mm in diameter and 0.28 mm in thickness. The sample was placed at a flight distance of 27.9 m from the neutron source. In addition to the ^{99}Tc sample, runs for blank, dummy Al container and carbon sample were carried out for background measurements. The incident neutron spectrum was determined from detected counts of 478 keV γ-rays from the $^{10}B(n,\alpha)^{7}Li$ reaction. Measurement of ^{197}Au was also made as standard measurement.

The TOF method was employed to determine the incident neutron energy. The repetition rate of the pulsed neutron beam was 25 Hz. The incident proton beam power was approximately 200 kW. The pulsed proton beam was operated in the single-bunch mode. The single-bunch mode operation was significantly beneficial in the present work. This allows us to analyze resolved resonances with a fine time resolution of 100 ns in a simple way. Details of data analysis can be found elsewhere [4,5]. Only a brief description is given here. After dead time correction, backgrounds from frame overlap neutrons, blank, the container and scattering neutrons were subtracted. The frame overlap background, which was caused by slow neutrons generated in prior TOF frames, was estimated using the special J-PARC accelerator operational pattern. The incident proton beam pulses from a 3-GeV synchrotron were switched between the spallation neutron target and a larger 50-GeV synchrotron. A small fraction of the beam pulses (1–2%) was not transmitted to the spallation target. This beam absent period was considerably useful because background by overlapping neutrons can be directly measured during the period [6]. Background of scattering neutrons was estimated from measurements with a graphite sample. The net TOF

[a] e-mail: buchi@nr.titech.ac.jp

DOI: 10.1051/epjconf/201714611050

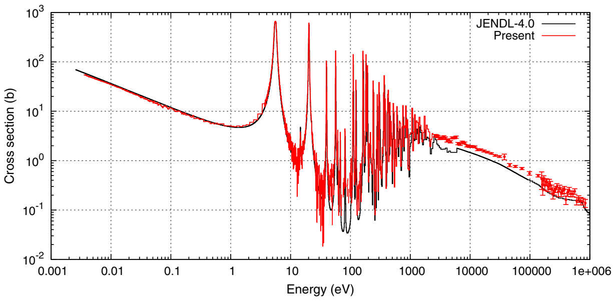

Figure 1. Neutron capture cross section of ^{99}Tc. The present results (red) are compared with the evaluated cross section of JENDL-4.0 (black).

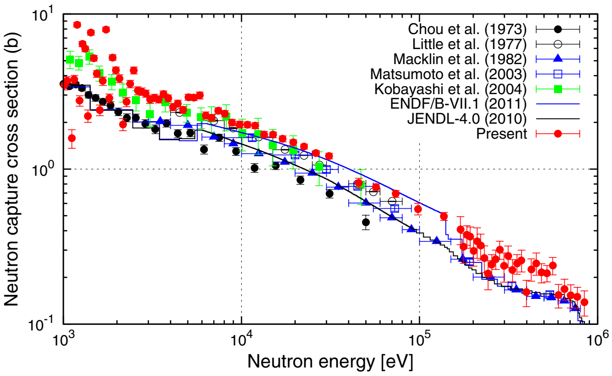

Figure 2. Neutron capture cross section of ^{99}Tc in the high energy region. The present points (red solid) are compared with previous measurements and evaluations. Only statistical errors of the present data are shown.

spectrum of ^{99}Tc was converted to the cross section by dividing by the incident neutron spectrum determined from the boron measurement. The absolute cross section was determined from a saturated resonance of ^{99}Tc at 5.6 eV.

3. Results

Results are shown in Fig. 1. The results are still preliminary because the pulse-height weighting technique [7] was not applied yet. Corrections for self-shielding and multiple scattering have not been made. Evaluated cross section of JENDL-4.0 [8] is shown for comparison. The self-shielding factors calculated from the resonance parameters were multiplied to the evaluated cross section for comparison. With a single-bunch mode time resolution (∼100 ns), the count statistics was enough for resolved resonance analysis up to around 500 eV. Statistics can be improved in planned future upgrade of the neutron source increasing the proton beam power up to 1 MW.

Figure 2 shows a close-up of the high energy continuum region. The present results are compared with previous measurements [9–13] and evaluations of JENDL-4.0 and ENDF/B-VII.1 [14]. The ENDF/B-VII.1 evaluation is closer to the present data than the JENDL-4.0 evaluation. However finalizing data analysis is necessary for more discussion.

4. Summary

We measured the neutron capture cross section of ^{99}Tc using the NaI(Tl) detectors of ANNRI at J-PARC in the energy range from thermal to the keV energy region. The present results were compared with previous measurements and evaluations. Data analysis is still ongoing. The pulse-height weighting technique will be applied. Corrections such as self-shielding and multiple scattering will be made. In the future analysis, resolved resonance parameters will be extracted.

The present study includes the result of “Research and Development for accuracy improvement of neutron nuclear data on minor actinides” entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

References

[1] M. Igashira, et al., Nuclear Instruments and Methods in Physics Research Section A **600**, 332 (2009)
[2] Y. Kiyanagi, et al., Journal of Korean Physical Society **59**, 1781 (2011)
[3] F. Maekawa, et al., Nuclear Instruments and Methods in Physics Research Section A **620**, 159 (2010)
[4] T. Katabuchi, et al., Nuclear Instruments and Methods in Physics Research Section A **764**, 369 (2014)
[5] K. Terada, et al., Progress in Nuclear Energy **82**, 1 (2014)
[6] K. Hirose, et al., Journal of Nuclear Science and Technology **50**, 188 (2013)
[7] R.L. Macklin, et al., Nuclear Instruments and Methods in Physics Research **91**, 565 (1971)
[8] K. Shibata et al., Journal of Nuclear Science and Technology **48**, 1 (2011)
[9] Jen-Chang Chou and H. Werle, Journal of Nuclear Energy **27**, 811 (1973)
[10] R.C. Little and R.C. Block, Transactions of the American Nuclear Society **26**, 574 (1977)
[11] R.L. Macklin, Nuclear Science and Engineering **81**, 520 (1982)
[12] T. Matsumot et al., Journal of Nuclear Science and Technology **146**, 61 (2004)
[13] K. Kobayashi, Nuclear Science and Engineering **81**, 209 (2004)
[14] M.B. Chadwick et al., Nuclear Data Sheets **112**, 2887 (2011)