*Journal of Nuclear and Radiochemical Sciences, Vol. 6, No. 3, pp. 283-286, 2005*

# Cross Section of Thermal-Neutron Capture Reaction by $^{99}$Tc

**Kazuyoshi Furutaka,*[a] Hideo Harada,[a] Shoji Nakamura,[a] Toshio Katoh,[a] Toshiyuki Fujii,[b] Hajimu Yamana,[b] and Subramanian Raman[c]**

[a]*Tokai works, Japan Nuclear Cycle Development Institute, 4-33 Muramatsu, Tokai-mura, Naka-gun, Ibaraki 319-1194, Japan*

[b]*Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan*

[c]*Physics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, U.S.A.*

***Received: July 31, 2005; In Final Form: September 30, 2005***

An account is given of a series of study to accurately determine the cross section of the $^{99}$Tc($n$,γ)$^{100}$Tc reaction for thermal neutrons. Methods used are an activation analysis in which γ rays in $^{100}$Ru are examined, and analyses of ground-state transitions in $^{100}$Tc. To reduce uncertainty of the cross section obtained in the activation measurement, an attempt was made to accurately determine emission probabilities of γ rays in $^{100}$Ru.

## 1. Introduction

Technetium-99 (half-life $T_{1/2} = 2.1 \times 10^5$ years)[1] is one of the long-lived fission products (LLFP) that are produced in commercial nuclear power plants with relatively large yields (cumulative yield for $^{235}$U: 6.11%).[2] Technetium-100, which is produced in neutron-capture reaction by $^{99}$Tc, β-decays to its stable daughter nuclide $^{100}$Ru, with a short half-life of 15.3 s.[3] Also, cross section of $^{99}$Tc($n$,γ) reaction is reported to be relatively large. These facts make the nuclide $^{99}$Tc among possible candidates for transmutation using neutron.

In order to design transmutation systems for $^{99}$Tc using neutron-capture reaction and to study their feasibilities, accurate cross section of the $^{99}$Tc($n$,γ) reaction is in need (in Reference 4, the cross section is requested to be determined as accurate as 5% in an energy range between thermal and 20 MeV), and therefore in recent years many efforts have been put to measure the cross section experimentally (see Reference 5 and the references therein). Among cross section of the ($n$,γ) reaction, the one at thermal energy (2200 m/s), $\sigma_0$, is important, because not only is it used by itself to calculate quantities such as reaction rate for thermal neutrons, but also it is often used to process experimental data obtained in the so-called resolved resonance region, namely to normalize, to check consistencies of experimentally obtained resonance parameters and so on.

Several authors have reported the cross section of the $^{99}$Tc($n$,γ) reaction for thermal neutrons. Pomerance[6] and Tattersall[7] reported the cross sections obtained by using the pile oscillator method in which a sample is oscillated between a center of a reactor and a position outside the reactor and the resulting modulation of the pile power is observed as a measure of the cross section. Pattenden[8] and Watanabe et al.[9] measured total neutron cross sections as functions of neutron energy, and deduced capture cross sections at thermal energy by subtracting scattering cross sections. Some researchers employed activation methods to deduce the cross section. Ovechkin et al.[10] detected γ rays in $^{100}$Ru emitted in the capture reaction to deduce rate of the reaction and the cross section. Lucas et al.[11] examined changes in isotopic composition of Ruthenium in the irradiated samples by a mass-spectroscopic method after neutron irradiations. The leftmost quarter of Figure 1 shows the reported values of $\sigma_0$ of the $^{99}$Tc($n$,γ) reactions. As can be seen in the figure, the reported values are not accurate enough (the uncertainty of the most precise one being as large as 8%)[8] and at the same time scattering around about 20 barns.

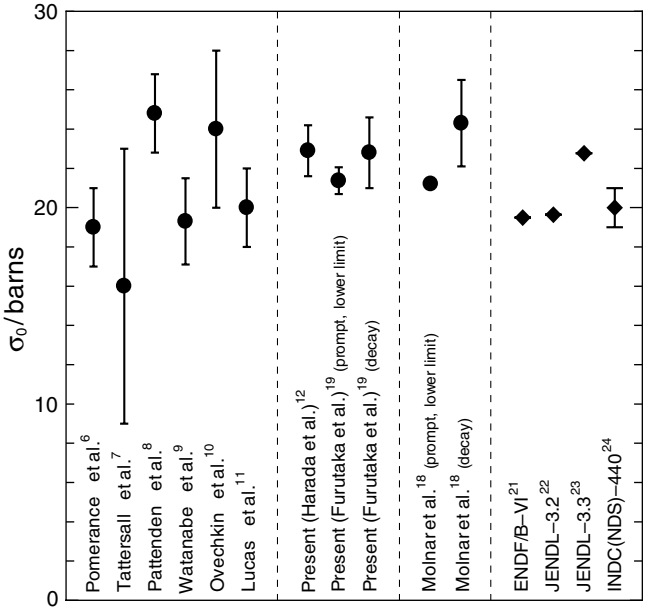


**Figure 1.** The cross sections of the $^{99}$Tc($n$,γ) reaction for thermal neutrons reported in References 12 and 18 are compared with other published experimental data as well as evaluated ones. The filled circles (●) represent experimental data, whereas the filled diamonds (◆) indicate evaluated ones.

In order to accurately determine the cross section of thermal-neutron capture reaction by $^{99}$Tc experimentally, a series of study has been performed:

- Measurement of $\sigma_0$ using an activation method in which γ rays in $^{100}$Ru were observed,
- Measurement of emission probabilities of γ rays in $^{100}$Ru, and
- Determination of $\sigma_0$ from analyses of the prompt γ rays emitted in the capture reaction.

In the following, the research is described, and the obtained results are compared to others. Possible improvements of the experimental methods are also presented.

## 2. Measurement of the $\sigma_0$ Using an Activation Method

The product of the $^{99}$Tc($n$,γ) reaction, $^{100}$Tc, β-decays to $^{100}$Ru

*Corresponding author. E-mail: furutaka.kazuyoshi@jaea.go.jp. FAX: +81-29-282-5458.

© 2005 The Japan Society of Nuclear and Radiochemical Sciences
Published on Web 12/28/2005

with $T_{1/2}$ = 15.3 s; most of the decays (93%) directly go to the ground state of $^{100}$Ru, but a part of the intensity feeds its excited states. The excited states eventually deexcite by emitting some γ rays, the most intense ones being 540-keV and 591-keV ones, and produce stable $^{100}$Ru nuclei in its ground state. Therefore, by counting these γ rays in $^{100}$Ru, one can deduce the number of $^{100}$Ru nuclei produced, and consequently reaction cross section, if the numbers of target nuclei and irradiated neutrons as well as the emission probabilities of the γ rays are known.

The details of the experiment have been presented elsewhere,[12] and so here it's described only briefly. Four irradiation samples of ammonium pertechnetate containing ~370 kBq of $^{99}$Tc were used in the measurement; the number of $^{99}$Tc nuclei in each sample was determined through β-ray counting using a liquid scintillation detector and weighing. Each sample was irradiated by neutrons for 2 minutes using a rotary specimen rack of the TRIGA MARK II reactor of Rikkyo University. At the irradiation position, the thermal neutron flux was $5 \times 10^{11}$ $n$/cm$^2$/s and the epithermal index in the Westcott's convention was 0.033. Each sample was irradiated four times, two with a surrounding Cd capsule and two without, with 40 minutes of cooling in between. The neutron flux at the irradiation position was measured separately between the Tc irradiations by using Co/Al and Au/Al alloys as activation detectors. After each irradiation, γ rays emitted from the sample were measured using a high purity Ge detector with 90% relative efficiency and 2.1 keV FWHM at 1.33 MeV. The distance between the sample and the front surface of the detector endcap was 100 mm.

In the observed spectra, peaks due to the 540- and 591-keV γ rays were clearly identified. From the peak counts of these γ rays, reaction rates of the capture reaction were deduced and analyzed based on a modified version of Westcott's convention. With the emission probabilities for the two γ rays listed in Reference 13, after a correction of summing-coincidence effect, the cross section was deduced. We have adopted the cross section obtained from the analysis of the 540-keV γ ray. The value obtained is 22.9 ± 1.3 b.[12] The result agrees well with the values reported in References 8 and 10, but about 3 barns higher than that listed in References 6, 7, 9, and 11. Note that the uncertainty of the emission probability is not included in the error. By including the uncertainty, the result would be 22.9 ± 2.6 b.[12] As is easily seen, the overall accuracy can be largely improved by reducing the uncertainty of the emission probability.

## 3. Measurement of Emission Probabilities of γ rays in $^{100}$Ru

The emission probabilities listed in Reference 13 are based on the study done by Berzins et al.[14] In the study, the number of (β, 591-keV γ) coincidences per emitted γ ray were measured by using a NaI(Tl) and a plastic scintillator as a γ- and β-ray detector, respectively. Neglecting indirect β feeding to the 1131-keV state, the authors have deduced the β feeding to the 1131-keV level as 5.7%. The authors did not quote the uncertainty of the value; in Reference 15 the uncertainty was estimated to be about 16%. The uncertainties of the absolute intensity of the 540-keV γ ray quoted in References 1 and 13 are 17.14% and 10%, respectively, which are rather large. In order to determine emission probabilities of the γ rays in $^{100}$Ru and to estimate their uncertainties accurately, a β–γ coincidence system has been developed which consist of a plastic scintillation detector as a β detector and a large-volume high-purity germanium detector as a γ detector,[16] and applied to an experiment.[3]

The experiment has been performed at the Kyoto University Research Reactor Institute. Samples of technetium containing 5 kBq of $^{99}$Tc were irradiated for 15 s using one of the pneumatic irradiation tubes of the research reactor in the institute. The neutron flux at the irradiation position is $2.3 \times 10^{13}$ $n$/cm$^2$/s for thermal and $8.4 \times 10^{11}$ $n$/cm$^2$/s for epithermal neutrons. After each irradiation, the sample was placed between the detectors and coincidence measurement was done for at least 120 s. The distances between the sample and the front surfaces of the detectors were 30 mm and 100 mm for γ- and β-ray detectors, respectively. In some of the measurements an acrylic plate (16 mm in thickness) was inserted between the sample and the β-ray detector to prevent the β-ray from entering the detector and to estimate the sensitivity of the detector to γ rays. A total of 99 measurements were done.

From the observed β and γ spectra, ratios of the number of β–γ coincidences to the number of β rays are deduced for 540- and 591-keV γ rays as functions of applied β-ray detection thresholds. By extrapolating the functions to vanishing threshold value, the effect of the β detection threshold to the ratio was eliminated. After making corrections of summing-coincidence effect and γ-ray sensitivity in the β detector, the emission probabilities of the 540- and 591-keV γ rays were obtained as 6.6 ± 0.5% and 5.5 ± 0.3%, respectively. The obtained results agree with that reported in Reference 1 within limits of errors, but are a little bit smaller. The estimated errors are much smaller than those quoted in Reference 1.

## 4. Measurement of the $\sigma_0$ from Yields of the Prompt γ rays

In a separate series of study, one of the present authors has demonstrated the great capabilities of determining the cross sections of the neutron capture reactions, by detecting γ rays in singles measurements with a sensitive detection system and constructing level schemes.[17] In the study, prompt γ rays emitted in neutron capture reactions by nuclides with masses up to 60 were measured, and almost complete decay schemes were constructed by analyzing the obtained data based on the Rydberg-Ritz combination principle. By summing up intensities of primary transitions from a capturing state or of secondary ones to the ground state of a nuclide, one can deduce cross section of the reaction. The method was applied to a measurement of $\sigma_0$ of the $^{99}$Tc($n$,γ) reaction. Results of a study along the same line have been published recently.[18]

The details of the study have already presented in Reference 19 and here only brief description is given. The measurement of γ rays was done at the Los Alamos National Laboratory Omega West Reactor. A metallic sample of $^{99}$Tc (84.6 mg) was placed in the thermal column of the internal target facility at the reactor. The target position was 1.5 m from the edge of the reactor core and the thermal neutron flux was ~$6 \times 10^{11}$ $n$/cm$^2$/s and the Cd(In) ratio was ≈2000. γ rays were measured with a spectrometer consisting of 30-cm$^3$ coaxial intrinsic Ge detector positioned inside a 20-cm-diameter by 30-cm-long NaI(Tl) annulus; The annulus is optically divided into two halves. The spectrometer was located 6.3 m from the target position and was operated either in the Compton-suppressed(CS) mode or in the pair-spectrometer(PS) mode. γ rays with energies of up to ~3 MeV (~11 MeV) were measured in the CS (PS) mode. Energies of the γ rays were calibrated with the prompt γ ray from the $^1$H($n$,γ) reaction plus the annihilation radiation in the CS mode; in PS mode, the calibration was done with the prompt γ rays emitted in the $^{14}$N($n$,γ) reaction using a melamine ($C_3H_3N_6$) sample. Calibration of detection efficiencies were done with a set of standard sources (CS) and relative intensities of the γ rays in $^{14}$N($n$,γ) reaction (PS). Absolute intensities were determined through measurements in which the target was irradiated together with a 100.0-mg $CH_2$ standard; the recommended value of $\sigma_0$ = 332.6 ± 0.7 mb was used for the $^1$H($n$,γ) reaction.

Figure 2 shows a selected portion of the observed spectrum in the CS mode. Many γ rays were observed in both modes, including the 540-and 591-keV γ rays in $^{100}$Ru. Gamma-ray peaks observed in both modes are analyzed and the energies as well as the intensities were determined. The obtained data in both modes are combined, and after elimination/correction of background, a total of 1086 γ rays were assigned to be emitted

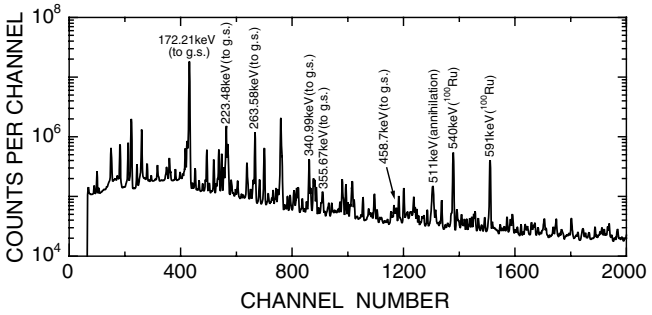


**Figure 2.** A selected portion of the observed spectrum in the prompt γ-ray measurement emitted in $^{99}$Tc($n$,γ) reaction, in the CS mode. The assigned ground-state transitions are indicated. The two intense γ rays in $^{100}$Ru, namely 540- and 591-keV γ rays, are also clearly seen.

in the capture reaction. Starting from a skeleton level scheme built from the available data, a decay scheme was constructed by analyzing the observed γ rays using the Rydberg-Ritz combination principle. A total of 88 γ rays have been incorporated into the level scheme consisting of 36 bound levels including eight new ones. Among these, six were assigned as transitions to the ground state, namely 172.21-, 223.48-, 263.58-, 340.99-, 355.67- and 458.7-keV ones. From the intensities of the ground-state transitions and their internal conversion coefficients calculated using the code HSICC,[20] $\sigma_0$ was obtained as 21.37 ± 0.68 b; the value should be regarded as a lower limit, because the constructed decay scheme is far from complete and there may be more ground-state transitions which were not incorporated into the scheme. From the yields of the γ rays in $^{100}$Ru observed simultaneously, the cross section was also deduced; the obtained value is 22.8 ± 1.8 b. In Figure 1 the obtained results are compared with other experimental/evaluated data.

## 5. Discussion

Figure 1 compares the values of cross section of the $^{99}$Tc($n$,γ) reaction for thermal neutrons reported in References 12 and 19 with other published experimental data as well as evaluated ones. At first glance, the results obtained from the yields of prompt γ rays in References 18 and 19 look very accurate; these values should be regarded as lower limits of the cross section, because there may be more ground-state transitions left unobserved/unassigned. Some of the evaluated values are smaller than these lower limits, reflecting the older values around 20 b; the present authors recommend to revise the values to larger ones. Although the results obtained from the prompt γ-ray measurements demonstrate a possibility to accurately determine cross sections from prompt γ-ray measurements, it seems very difficult and tedious to construct complete level schemes by incorporating thousands of observed γ rays. Therefore, subsidiary methods are highly desirable. A possible method would be the one that enables one to estimate unobserved portion of the intensities of the prompt γ rays from those of the observed ones.

The uncertainties of the cross section reported in References 18 and 19 obtained in the activation measurements are still larger than the requested one in Reference 4, because the uncertainties of the emission probabilities of the γ rays in $^{100}$Ru are not yet small enough. Figure 3 shows a comparison of the ratios of the intensity of the 591-keV γ ray to that of the 540-keV one calculated from the data in References 3, 14, 18, and 19. The ratios obtained from the newer and improved data in References 18 and 19 published after Reference 3, clearly indicate that the ratio that was obtained from the γ-ray emission probabilities in Reference 3, is a little bit too large. Thus, the accuracy of the emission probabilities should be improved further. To determine γ-ray emission probabilities accurately, 2 or 4π β–γ coincidence measurements are usually done; in the method, it is not difficult to determine emission probabilities

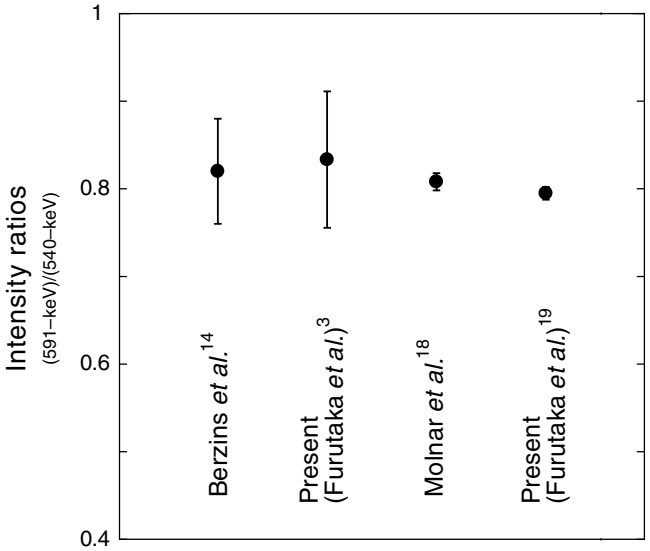


**Figure 3.** Ratios of the intensity of 591-keV γ ray in $^{100}$Ru to that of 540-keV one.

with uncertainties of less than a percent. However, the short half-life of $^{100}$Tc makes it difficult to apply the technique to the cross section measurements. One possible way would be to do the measurement with a neutron beam on-line.

## 6. Conclusion

Results are presented of a series of study to determine cross sections of $^{99}$Tc($n$,γ) for thermal neutrons. In spite of the efforts, the attained accuracy doesn't meet the requests, and further improved measurements are in need.

**Acknowledgements.** The present work has been carried out in part under the Visiting Researcher's Program of the Research Reactor Institute, Kyoto University. A part of the present work has been done as the collaboration between Japan Nuclear Cycle development institute (JNC) and Oak Ridge National Laboratory (ORNL) operated by UT-Battelle, LLC, under the contract No. FERD-01-2094. A part of the experimental work in the present study was sponsored by the U.S. Department of Energy, under Contract No. DE-AC05-00OR22725 with UT-Battelle, LLC.

## References

(1) R. B. Firestone, C. M. Baglin, and S. Y. Chu, *Table of Isotopes*, *8th ed. (1998 Update with CD-ROM)*, (John Wiley & Sons, New York, 1998).
(2) H. Ihara (Ed), JAERI-M 89-204, (1989).
(3) K. Furutaka, S. Nakamura, H. Harada, T. Katoh, T. Fujii, and H. Yamana, J. Nucl. Sci. Tech. **38**, 1035 (2001).
(4) NEA Nuclear Science Committee, Working Party on International Evaluation Co-operation Subgroup C, *The NEA High Priority Nuclear Data Request List*, (Status in March 2001).
(5) K. Kobayashi, S. L. Lee, S. Yamamoto, and T. Kawano, Nucl. Sci. and Eng. **146**, 209 (2004).
(6) H. Pomerance, ORNL-1975, p. 31 (1975).
(7) R. B. Tattersall, H. Rose, S. K. Pattenden, and D. Jowitt, J. Nucl. Energy, Part A **12**, 32 (1960).
(8) N. J. Pattenden, *Proc. 2nd Int. Conf. Peaceful Uses of Atomic Energy, Geneva*, 1958, vol. 16, p. 44. (1958).
(9) T. Watanabe and S. D. Reeder, Nucl. Sci. and Eng. **41**, 188 (1970).
(10) V. V. Ovechkin, D. F. Rau, and V. S. Rudenko, *Proc. 2nd Conf. on Neutron Physics, Kiev,* 1973, Vol.2, p. 131 (1974).
(11) M. Lucas, R. Hagemann, R. Naudet, C. Renson, and C.

Chevalier, IAEA-TC-119/14, p. 407 (1977).
(12) H. Harada, S. Nakamura, T. Katoh, and Y. Ogata, J. Nucl. Sci. Tech. **32**, 395 (1995).
(13) C. M. Lederer, V. S. Shirley, *Table of Isotopes, 7th ed.*, (John Wiley & Sons, New York, 1978).
(14) G. Berzins, M. E. Bunker, and J. W. Starner, Phys. Rev **187**, 1618 (1969).
(15) B. Singh, Nuclear Data Sheets **81**, 1 (1997).
(16) K. Furutaka, S. Nakamura, H. Harada, and T. Katoh, J. Nucl. Sci. Tech. **37**, 832 (2000).
(17) S. Raman, X. Ouyang, M. A. Islam, J. W. Starner, E. T. Jurney, J. E. Lynn, and G. Martinez-Pinedo, Phys. Rev. **C 70**, 044318 (2004).
(18) G. L. Molnár, T. Belgya, Zs. Révay, and S. M. Qaim, Radiochim. Acta **90**, 479 (2002).
(19) K. Furutaka, H. Harada, and S. Raman, J. Nucl. Sci. Tech. **41**, 1033 (2004).
(20) "ENSDF Analysis Program-HSICC", ver.11.13c, Feb. 9 2001, Energy sciences and Technology Department, National Nuclear Data Center, Brookhaven National Laboratory (2001).
(21) P. F. Rose, BNL-NCS-17541, 4th ed. Brookhaven National Laboratory (1991).
(22) T. Nakagawa et al., J. Nucl. Sci. Tech. **32**, 1259 (1995).
(23) K. Shibata et al., J. Nucl. Sci. Tech. **39**, 1125 (2002).
(24) S. F. Mughabghab, INDC(NDS)-440, INDC, Vienna (2003).