Measurement of Thermal Neutron Cross Section and Resonance Integral of the Reaction $^{99}$Tc$(n,\gamma)^{100}$Tc

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To obtain fundamental data for research on the transmutation of nuclear waste, the thermal neutron cross section and the resonance integral of the reaction $^{99}$Tc$(n,\gamma)^{100}$Tc have been measured using an activation method.

Four ammonium pertechnetate targets containing 371~375 kBq of $^{99}$Tc were irradiated for 2 min with reactor neutrons. Activation detectors of Co/Al and Au/Al alloy wires were irradiated for 10 min to monitor the neutron flux and the fraction of the epithermal part (Westcott's epithermal index). The Tc samples and flux monitors were irradiated with and without a Cd capsule. The $\gamma$-ray spectra from the irradiated samples were measured using a high purity Ge detector.

The thermal neutron cross section (2,200 m/s neutron cross section) and the resonance integral of the $^{99}$Tc$(n,\gamma)^{100}$Tc reaction were found to be $22.9 \pm 1.3$ b and $398 \pm 38$ b, respectively. The thermal neutron cross section obtained agrees with the previously reported values (20~2 b by Lucas, 24.8 by Pattenden, 24~4 by Ovechkin) within the limits of error. On the other hand, the resonance integral is twice the value reported by Lucas (186~16 b).

KEYWORDS: nuclear transmutation, incineration, radioactive waste, neutron capture, technetium 99, technetium 100, resonance integrals, thermal neutrons, cross sections, half-life, gamma spectra, activation method, fission products, experimental data

I. INTRODUCTION

Accurate neutron cross sections of radioactive fission products and transuranics are required for the research on nuclear transmutation methods. Especially, thermal neutron cross sections ($\sigma_\theta$) and resonance integrals ($I_\theta$) are important for the study of the transmutation methods(1)~(5) using a high flux fission reactor, a high intensity accelerator, and also a high flux fusion reactor.

The present authors have recently reported(6)~(11) the thermal neutron capture cross sections of $^{137}$Cs and $^{96}$Sr. The cross section of $^{137}$Cs obtained was twice the value measured in 1960 by Stupegia(12). The value of $^{96}$Sr was only 1/50 of the value measured in 1966 by Zeisel(13).

The present paper describes the experimental results on $^{99}$Tc, which is another important nuclide in the nuclear waste management because of its large fission yield(14), 6.1%, and its extremely long half-life $2 \times 10^5$ yr(15).

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The thermal neutron capture cross section of $^{99}\text{Tc}$ has been measured by four groups. The values varies from 20 to 100 b. On the other hand, there is only one experimental data on the resonance integral of $^{99}\text{Tc}$ reported by Lucas et al. (16) ($186 \pm 16$ b). However, the evaluated cross section libraries (17) (18) adopted the values of 19.5 ~ 19.6 b as $\sigma_0$ and 312 ~ 351 b as $I_0$.

The present experiment was designed to obtain a reliable value of the cross section of the $^{99}\text{Tc}(n,\gamma)^{100}\text{Tc}$ reaction. Four $^{99}\text{Tc}$ samples were irradiated by reactor neutrons with and without a Cd capsule. Activation detectors of Co/Al and Au/Al alloy wires were also irradiated to monitor the neutron flux and Westcott's epithermal index (19) $r(T/T_0)^{1/2}$. Taking advantage of a high efficiency Ge detector and fast data acquisition electronics, the decay $\gamma$-rays from the short-lived $^{100}\text{Tc}$ nucleus were measured.

The thermal neutron capture cross section and the resonance integral of the $^{99}\text{Tc}(n,\gamma)^{100}\text{Tc}$ reaction obtained were compared with the values measured by Lucas et al. (1977) and those in the evaluated nuclear data libraries.

### II. Experiments

#### 1. Target

The target nucleus $^{99}\text{Tc}$ does not emit any observable $\gamma$-rays. Therefore, the specific activity of the 0.01 M ammonium hydroxide containing $^{99}\text{Tc}$ as ammonium pertechnetate was measured in advance with a liquid scintillation counter. The value measured was $461 \pm 7$ kBq per gram of the solution. This solution was put into 4 polyethylene tubes (10 mm in inner diameter, 45 mm in outer length, and 1 mm in thickness) which served as the irradiation targets. The number of $^{99}\text{Tc}$ atoms contained in each target, represented by the activity, was determined from the weight of each sample times the specific activity and had a value ranging from 371 to 375 kBq. The weight of each sample was determined to an accuracy of 0.1 %. These polyethylene tubes were located in an outer polyethylene bottle. A Cd shield (1 mm in thickness, 22 mm in outer diameter and 63 mm in outer length) was inserted between the polyethylene tube and the outer bottle when it was required to reduce the intensity of thermal neutrons in the target tube.

Two kinds of flux monitor wires, Co/Al and Au/Al alloys, were used as activation detectors of the neutron flux at the irradiation position. Because Co and Au differ in sensitivities to thermal and epithermal neutrons, these monitors could be used to determine the thermal and the epithermal neutron fluxes. The metal contents and diameters of the monitor wires are listed in Table 1. The monitor wire was shaped into a spiral and inserted in the polyethylene tube. About 0.8 g of water was poured into the tube to imitate the liquid target of $^{99}\text{Tc}$.

<table>
<thead>
<tr>
<th>Material of wire</th>
<th>Diameter of wire (mm)</th>
<th>Radionuclide of interest</th>
<th>Half-life (yr)</th>
<th>$\sigma_0$ (b)</th>
<th>$I_0$</th>
<th>Detected $\gamma$-ray</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co/Al 0.46 wt% in Co</td>
<td>0.381</td>
<td>$^{60}\text{Co}$</td>
<td>5.271</td>
<td>37.2</td>
<td>1.83</td>
<td>1,173</td>
</tr>
<tr>
<td>Au/Al 0.112 wt% in Au</td>
<td>0.510</td>
<td>$^{198}\text{Au}$</td>
<td>2.696 d</td>
<td>98.8</td>
<td>17.02</td>
<td>412</td>
</tr>
</tbody>
</table>

$^\dagger$ The quantities $G_\gamma$ and $s_0$ are defined by Eqs. (1) and (4) in the text, respectively.

#### 2. Irradiation

The target was placed in a rotary specimen rack (RSR), which was located in the reflector surrounding the core of the TRIGA MARK II reactor at Rikkyo University. The irradiation position was characterized as having a thermal neutron flux of $5 \times 10^{11}$ $n/(cm^2\cdot s)$ and an epithermal index in the Westcott's convection (19) of 0.033.

The four targets of $^{99}\text{Tc}$ were each irradiated for 2 min one at a time. Each target was irradiated with the Cd capsule, and after
40 min it was irradiated without the Cd capsule. After 40 min, the activity of $^{100}$Tc produced by the previous irradiation is negligible because the lifetime of $^{100}$Tc is only 15.5 s. This set of irradiations of the $^{99}$Tc sample was repeated twice for each target. Using a stopwatch, the time interval between the stop time of the irradiation and the start time of the activity measurement was determined. That was typically 1 min. The Co and Au flux monitors were irradiated for 10 min with the Cd capsule and without it. This set of the irradiations of the flux monitors was repeated three times during the irradiations of the $^{99}$Tc targets.

3. Measurement of Radioactivity

The 540- and 591-keV $\gamma$-rays associated with the decay of $^{100}$Tc were measured with a high purity Ge-detector of 90% relative efficiency and 2.1 keV FWHM at 1.33 MeV. The irradiated polyethylene tube containing $^{99}$Tc and $^{100}$Tc was mounted on an acrylic box where the distance between the center of the front surface of the endcap of the Ge detector and the center of the tube was 100 mm.

The signals from the detector were transferred to a fast data acquisition system consisting of a transistor reset preamplifier (ORTEC model 232 P), an amplifier (CANBERRA model 2024) and an ADC (CANBERRA 450 MHz model 8077). The digital pulse height signals of the ADC were accumulated through an adapter (TOYO T3500) in a histogramming memory (LeCroy model 3588). The adapter was modified to read the ADC dead time by two clocks of 1 MHz; one measures the ADC live time and the other the real operating time. These clocks were controllable using CAMAC functions. The pulse height data with 4 k channels were transferred through a crate-controller (TOYO CC/7000) into a hard disk of a personal computer (NEC PC98) at an interval of 22 s; the real operating time was 20 s and the data transfer time was 2 s. The data acquisition program controlling this system could monitor, analyze, and save the pulse height data automatically. To correct the pulse pile-up loss, the counting loss of the pulser’s pulse was measured in advance as a function of the ratio between ADC live time and real operating time; this ratio could be varied by changing the distance between the detector and a standard source ($^{137}$Cs). The correction factor of the pile-up loss for $\gamma$-rays from $^{100}$Tc was obtained by inserting the ratio between the live time and the real time for each measurement into this function.

The radioactivities of the irradiated flux monitors, $^{60}$Co and $^{198}$Au, were measured with the same Ge detector. The monitor was sealed with a vinyl tape and mounted on the acrylic box. The $\gamma$-ray from $^{198}$Au was measured for about one hour and those from $^{60}$Co were about one day.

The peak efficiency of the detector was calibrated with a mixed standard source containing $^{113}$Sn, $^{85}$Sr, $^{137}$Cs, $^{88}$Y and $^{60}$Co supplied by Saclay. The summing loss of the two $\gamma$-rays in cascade was corrected for the $\gamma$-rays of the standard source and the irradiated flux monitors as well as for those associated with the decay of $^{100}$Tc, where the angular correlation effect was taken into account as described in Chap. III.

Self-absorption of $\gamma$-rays was calculated for the flux monitors using the next equation (see APPENDIX):

$$ G_{\gamma} = 1 - \frac{8\mu R}{3\pi}, \quad (\mu R \ll 1), $$

where $R$ is the radius of the wire and $\mu$ the linear absorption coefficient. The resulting self-absorption coefficient, $G_{\gamma}$, as well as nuclear data are listed in Table 1 for each $\gamma$-ray of interest. The absorption of $\gamma$-rays in the polyethylene tube was also corrected; the absorption probability in the tube wall of thickness 1 mm is 0.88% for a $\gamma$-ray energy of 540 keV.

III. ANALYSES AND RESULTS

Figure 1 shows $\gamma$-ray spectra obtained from 20 s measurements of $\gamma$-rays from a $^{99}$Tc sample irradiated with and without the Cd capsule. The $\gamma$-rays of $^{100}$Tc produced by the $^{99}$Tc(n,$\gamma$)$^{100}$Tc reaction can be clearly seen at 540, 591 and 1,512 keV in both the spectra. The two intense $\gamma$-rays, 540 and 591 keV,
were used for the determination of the radioactivity of $^{100}$Tc.

Figure 2 shows the decay curves of these $\gamma$-rays in run 1 for target No. 3, and Table 2 summarizes the half-life value of $^{100}$Tc determined from the analysis of these decay curves. Each value in Table 2 corresponds to the averaged value of the half-lives obtained by analyzing four independent decay curves, which were obtained by repeating twice the irradiations of a $^{99}$Tc sample with and without the Cd capsule. The error of the half-life includes not only the statistical error but also the systematic error of the pulse pile-up cor-

<table>
<thead>
<tr>
<th>Target No.</th>
<th>Half-life (s)</th>
<th>540 keV</th>
<th>591 keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>15.5 ± 0.1</td>
<td>15.4 ± 0.1</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>15.5 ± 0.1</td>
<td>15.4 ± 0.1</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>15.5 ± 0.1</td>
<td>15.5 ± 0.1</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>15.5 ± 0.1</td>
<td>15.5 ± 0.1</td>
<td></td>
</tr>
</tbody>
</table>

Weighted average: 15.5 ± 0.1
rection factor, that is, ±0.09 s. The weighted average is 15.5±0.1 s. This value is 2% smaller than the previously reported one (15.8±0.1 s)\(^{(22)}\). We use the half-life of \(^{100}\)Tc measured here in the analysis of the activity of \(^{100}\)Tc. Nuclear data used\(^{(15)}\)\(^{(22)}\) in the present analysis are shown in Table 3.

### Table 3 Nuclear data used in present work

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>(T_{1/2}) (s)</th>
<th>(E)</th>
<th>(\gamma)-intensity(^{(12)})(^{(22)}) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{100})Tc</td>
<td>15.5±0.1</td>
<td>540 keV</td>
<td>7.00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>591 keV</td>
<td>5.74</td>
</tr>
</tbody>
</table>

The reaction rate \((R)\) of the \(^{99}\)Tc\((n,\gamma)\)\(^{100}\)Tc was evaluated using the following equation:

\[
R = A_2/N_1\left[1 - \exp\left(-\lambda_2 T_{IR}\right)\right], \quad (2)
\]

where \(A_2\) is the radioactivity of \(^{100}\)Tc, \(\lambda_2\) the decay constant of \(^{100}\)Tc \((\lambda_2 = \ln 2/T_{1/2})\), \(N_1\) the number of \(^{99}\)Tc nuclei and \(T_{IR}\) the irradiation period.

The reaction rate divided by the neutron capture cross section \(\sigma_{\gamma}\) at thermal neutron energy (25 meV) can be expressed for a well-moderated thermal neutron spectrum as follows\(^{(19)}\):

\[
R/\sigma_{\gamma} = n v_0 [g G_{th} + r(T/T_0)^{1/2} s_0 G_{epi}], \quad (3)
\]

where \(n v_0\) is the “neutron flux” in the Westcott convention with the neutron density \(n\) and the neutron velocity \(v_0 = 2,200\) m/s. In Eq. (3), \(g\) is the non-1/\(v\) factor, that is, the measure of the cross section deviation from the 1/\(v\) law in a thermal energy region. The quantity \(r(T/T_0)^{1/2}\) gives the fraction of epithermal neutrons in the neutron spectrum, and \(s_0\) is defined by

\[
s_0 = \frac{2}{\sqrt{\pi}} \frac{I_0'}{\sigma_{\gamma}}, \quad (4)
\]

where \(I_0'\) means the resonance integral after subtracting the 1/\(v\) component. The \(G_{th}\) and \(G_{epi}\) denote self-shielding coefficients for thermal and epithermal neutrons, respectively. The \(g\) factor of \(^{99}\)Tc is 1.008 according to the evaluation by Pattenden\(^{(23)}\). Noting that \(g, G_{th}\) and \(G_{epi}\) are almost unity for \(^{99}\)Tc targets and flux monitors\(^{(24)}\)\(^{(25)}\), we assumed unity for \(g G_{th}\) and \(G_{epi}\) in the following analysis.

Equation (3) can be rewritten\(^{(16)}\)\(^{(11)}\), using simplified flux notations, as

\[
R/\sigma_{\gamma} = \phi_1 + \phi_2 s_0, \quad (5)
\]

for irradiation without a Cd capsule,

\[
R'/\sigma_{\gamma} = \phi_1' + \phi_2' s_0, \quad (6)
\]

for irradiation with a Cd capsule.

The simplified flux factors, \(\phi_{1,2}\) and \(\phi_{1,2}'\), were determined using the \(R\) and \(R'\) values of the flux monitors, Au and Co. The \(\sigma_{\gamma}\) and \(s_0\) of Au and Co are shown in Table 1. Figure 3 shows the experimental relation between \(R/\sigma_{\gamma}\) (or \(R'/\sigma_{\gamma}\)) and \(s_0\). The irradiations of the flux monitors were repeated three times and three sets of the \(R\) and \(R'\) agreed each other within the limits of the statistical errors. The averaged values of the three \(R\) and \(R'\) sets were then used to determine \(\phi_{1,2}\) and \(\phi_{1,2}'\). The obtained values of the simplified flux are shown in Table 4.

### Table 4 Results of simplified neutron flux measurement in RSR of Rikkyo Reactor monitored by Co/Al and Au/Al flux wires

<table>
<thead>
<tr>
<th>Irradiation type</th>
<th>Irradiation period</th>
<th>(\phi_1) or (\phi_1')</th>
<th>(\phi_2) or (\phi_2')</th>
</tr>
</thead>
<tbody>
<tr>
<td>Without Cd</td>
<td>10 min</td>
<td>4.615±0.076</td>
<td>0.152±0.009</td>
</tr>
<tr>
<td>With Cd</td>
<td>10 min</td>
<td>0.196±0.014</td>
<td>0.168±0.004</td>
</tr>
</tbody>
</table>

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Fig. 3 Plots of \(R/\sigma_{\gamma}\) or \(R'/\sigma_{\gamma}\) against \(s_0\) for neutron flux monitors irradiated without and with a Cd capsule.
Because Eqs. (5) and (6) give the relation

\[ s_o = \frac{\phi_1 - \phi_2(R/R')}{\phi_2 - \phi_2(R/R')} \tag{7} \]

the values of \( s_o \) of the \(^{99}\text{Tc}(n,\gamma)\) reaction is deduced from the ratio, \( R/R' \) of \(^{99}\text{Tc}\). The value of the \( \sigma_0 \) of this reaction is then obtained by inserting the \( s_o \) into Eq. (5).

The resonance integral \( I_0 \) can be calculated by assuming the Cd cut-off energy to be 0.5 eV. Here, \( I_0 \) is given by

\[ I_0 = I'_0 + 0.45 \tilde{\alpha}_0 \tag{8} \]

where 0.45 \( \alpha_0 \) is the \( 1/\nu \) contribution.

In the analysis of \( \sigma_0 \) and \( I_0 \), the value of 15.5 s was used as the half-life of \(^{100}\text{Tc}\). The adoption of this half-life instead of the value of 15.8 s reported by Berzins(22) enlarges the reaction rate of \(^{99}\text{Tc}\) about 5% in the analysis.

The angular correlation(26) of the two cascade \( \gamma \)-rays, 540 and 591 keV, was also taken into account in the evaluation of the summing-loss factor, because the \( 0^+ \rightarrow 2^+ \rightarrow 0^+ \) cascade transitions have a strong angular correlation as:

\[ W(\theta) = 1 + 0.36P_2(\cos \theta) + 1.14P_4(\cos \theta) \tag{9} \]

where \( \theta \) is the angle between the two cascading \( \gamma \)-ray directions and \( P_i \) the \( i \)-th Legendre polynomial. The decay schemes of \(^{100}\text{Tc}\) and \( W(\theta) \) are shown in Figs. 4 and 5, respectively.

Here, we define the 591 and 540 keV \( \gamma \)-rays as \( \gamma_1 \) and \( \gamma_2 \), respectively. The true \( \gamma \)-ray intensity emitted from the \(^{100}\text{Tc}\) target can then be expressed as

\[ I_{\gamma} = I_{\gamma}^{\text{obs}} \times F_{\text{SC}}, \tag{10} \]

where \( I_{\gamma}^{\text{obs}} \) is the observed \( \gamma \)-ray intensity and \( F_{\text{SC}} \) the correction factor of sum-coincidence. \( F_{\text{SC}} \) is expressed as

\[ F_{\text{SC}}^{(1)} = (1 - \varepsilon_{\gamma_1} f_w)^{-1}, \tag{11} \]

for \( \gamma_1 \) and

\[ F_{\text{SC}}^{(2)} = (1 - \frac{b_i}{b_s} \varepsilon_{\gamma_2} f_w)^{-1}, \tag{12} \]

for \( \gamma_2 \), where \( b_i \) is the \( i \)-th \( \gamma \)-ray branch and \( \varepsilon_{\gamma_i} \) the total efficiency for \( i \)-th \( \gamma \)-ray in the Ge detector. The \( f_w \) is the average of \( W(\theta) \) over the detector solid angle weighted by the \( \gamma \)-ray attenuation probability in the Ge crystal calculated for each direction. The calculated value of \( f_w \) is 2.2 for the present case.

The \( \varepsilon_T \) was calculated by using a Monte Carlo simulation code EGS4(27), which simulates \( \gamma \)-ray and electron transport in materials, e.g. a Ge crystal. In the simulation, the single open ended structure of the Ge crystal used in this experiment was taken into account. The \( \varepsilon_T \) was calculated for the energy range of 0.2~1.4 MeV and the absolute value of \( \varepsilon_T \) was normalized at 1.25 MeV using the experimental value of \( \varepsilon_T \) determined using \( \gamma \)-rays from a \(^{60}\text{Co}\) standard source. The obtained correction factor \( F_{\text{SC}} \) is shown in Table 5 together with the value of \( \varepsilon_{\gamma_i} \). The correction of sum-coincidence also enlarges the reaction rate about 5%.

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The experimental results \( (R/R', s_0, \sigma_0 \text{ and } I_0) \) are summarized in Table 6. The errors of the values in Table 6 include not only statistical errors but also systematic errors listed below with their uncertainties:

1. Error in the \( \gamma \)-ray detection efficiency resulting from the ambiguity of the source position \( 1.5\% \)
2. Error in the \( \gamma \)-ray detection efficiency resulting from the ambiguity of the calibration source intensity \( 3.0\% \)
3. Error in the measurement of the specific activity \( 1.5\% \)
4. Error in the measurement of the neutron flux \( 3.5\% \)
5. Error in the measurement of the half-life of \( ^{100}\text{Tc} \) \( 2.7\% \)
6. Error in the measurement of the target weight \( 0.1\% \)
7. Error in the reaction rate that comes from the error in the measurement of time \( (\pm 0.5 \text{ s}) \) \( 2.2\% \)
8. Error in the correction of the angular correlation \( 0.5\% \)

### Table 6

<table>
<thead>
<tr>
<th>Target No.</th>
<th>( R/R' )</th>
<th>( s_0 )</th>
<th>( \sigma_0 )</th>
<th>( I_0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.15±0.04</td>
<td>19.8±1.9</td>
<td>22.6±1.5</td>
<td>407±46</td>
</tr>
<tr>
<td>2</td>
<td>2.21±0.04</td>
<td>18.9±1.8</td>
<td>23.0±1.5</td>
<td>395±45</td>
</tr>
<tr>
<td>3</td>
<td>2.24±0.04</td>
<td>18.8±1.8</td>
<td>23.1±1.5</td>
<td>396±45</td>
</tr>
<tr>
<td>4</td>
<td>2.20±0.04</td>
<td>19.2±1.8</td>
<td>22.8±1.5</td>
<td>397±45</td>
</tr>
</tbody>
</table>

Weighted average: \( \sigma_0 = 22.9\pm1.3 \text{ b} \) and \( I_0 = 398\pm38 \text{ b} \)

In the calculation of the weighted average, only the statistical errors and the systematic errors of (7) in each run and each target were treated as independent errors. The \( \sigma_0 \) and \( I_0 \) in Table 6 were obtained by the analysis of the 540-keV \( \gamma \)-ray. The values obtained were 22.9±1.3 and 398±38 b, respectively. The \( \sigma_0 \) and \( I_0 \) of the \( ^{99}\text{Tc}(n,\gamma)^{100}\text{Tc} \) reaction obtained by analyzing the 591-keV \( \gamma \)-ray were 23.9±1.4 and 414±40 b, respectively. These values are about 5\% larger than those in Table 6. We adopted the values obtained by the analysis of the 540-keV \( \gamma \)-ray because the intensity of the 591-keV \( \gamma \)-ray involved not only the error in absolute intensity (10\%) but also the error in relative intensity (7\%) but the intensity of the 540-keV \( \gamma \)-ray involved only the error in the absolute intensity. This error in the absolute \( \gamma \) intensity is not included in the errors of Table 6. Including this error, the errors in \( \sigma_0 \) and \( I_0 \) are ±2.6 and ±55 b, respectively.

### IV. DISCUSSION

The \( \sigma_0 \) in this work agrees with those in evaluated nuclear data libraries JENDL-3 and ENDF/B-VI, as shown in Table 7. The \( \sigma_0 \) values in the data libraries were evaluated with the experimental data: 100±25 b in BNL-325 (1955 and 1957)(28), 24.8 b by Pattenden (1958)(25), 24±4 b by Ovechkin et al. (1973)(29), and 20±2 b by Lucas et al. (1977)(16). The \( I_0 \) in this work (388±38 b) is about twice that reported by Lucas et al. \( (I_0'=186±16 \text{ b}) \), which is only one experimental data before our measurement. Their irradiations were carried out in two different locations of a swimming-pool reactor with different spectra; the spectral indices \( r \) were 0.03 and 0.15. The \( r \) of 0.15 is about 5 times the value of \( r \) of the neutron spectrum in our irradiation position. The expression of Eq. (3) is valid only for a well moderated neutron spectrum and for a small \( r \). The difference in \( r \) between Lucas’s irradiation and our measurement could be the origin of the discrepancy in the \( I_0 \) value.

### Table 7

<table>
<thead>
<tr>
<th>( \sigma_0 )</th>
<th>( I_0 )</th>
<th>( I_0' )</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present</td>
<td>22.9±1.3</td>
<td>398±38</td>
<td>388±38</td>
</tr>
<tr>
<td>Lucas et al.</td>
<td>20±2</td>
<td>186±16</td>
<td></td>
</tr>
<tr>
<td>JENDL-3</td>
<td>19.7</td>
<td>312</td>
<td></td>
</tr>
<tr>
<td>ENDF/B-VI</td>
<td>19.5</td>
<td>351</td>
<td></td>
</tr>
</tbody>
</table>

The present value of \( I_0 \) is even close to the values in JENDL-3 and ENDF/B-VI,
although the $I_0$ in our measurement is 13% or 28% larger than the values in ENDF/B-VI and JENDL-3, whose evaluation of $I_0$ in the data libraries is based on not only neutron capture data but also scattering data.

V. SUMMARY AND CONCLUSION

The thermal neutron cross section and the resonance integral of the $^{99}$Tc$(n,\gamma)^{100}$Tc reaction were measured using an activation method to provide basic data for research on the nuclear transmutation of radioactive waste.

The $^{99}$Tc targets were irradiated by thermal neutrons in a rotary specimen rack (RSR) of the Rikkyo University Reactor. The irradiations with and without a Cd capsule were repeated twice for each of four targets. Neutron fluxes and their epithermal neutron fractions were determined with flux monitor wires Co/Al and Au/Al, which possess different responses to epithermal neutrons. About 1 min after the irradiation, the measurement of $^{100}$Tc activity was started using an automatic data acquisition program controlling a Ge detector system. The analysis of the decay curves of $^{100}$Tc activity show that the half-life of $^{100}$Tc was $15.5\pm0.1$ s. This value is 2% shorter than the previous reported value.

The analyses of the reaction rates of $^{99}$Tc and flux monitors with and without a Cd capsule have yielded the thermal cross section $\sigma_\theta$ (for 2,200 m/s neutrons) as well as the resonance integral $I_0$ (including the 1/ν part) for the reaction $^{99}$Tc$(n,\gamma)^{100}$Tc:

$$\sigma_\theta = 22.9 \pm 1.3 \text{ b} \quad \text{and} \quad I_0 = 398 \pm 38 \text{ b}.$$ 

In this analysis, the observed $T_{1/2}$ of $^{100}$Tc was used and the angular correlation effect on the sum-coincidence was taken into account.

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(16) LEMMEL, H. D.: IAEA-NDS-100 Rev. 5, Nov.
[APPENDIX]

Self-absorption of γ-rays for the Flux Monitor Wires

Self-absorption of γ-rays in the flux wires is approximately given by the next equation because the wire is set perpendicular to the wire-to-detector direction in our setup:

\[ G_r = \frac{\int e^{-\mu I(x, y)} dx dy}{\int dx dy}, \quad (A1) \]

where \( \mu \) is the linear absorption coefficient of the wire material and \( I(x, y) \) the distance between a source point \((x, y)\) on the cross section of the wire and the wire's surface, that is,

\[ I(x, y) = \sqrt{R^2 - y^2 - x}. \quad (A2) \]

Here, the point on the cross section of the wire is expressed using the \( x \) and \( y \) coordinates, and \( R \) is the radius of the wire.

Replacing the numerator of Eq. (A1) by its Maclaurin series expansion to the second order, we obtain

\[ G_r \approx 1 - \frac{2 \mu R}{3\pi^2}. \quad (A3) \]