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Measurement of Neutron Capture Cross Section of ^{99}Tc Between 0.007 eV and 47 keV

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Abstract

The neutron capture cross section of ^{99}Tc has been measured relative to the ^{10}B(n,α) standard cross section by the neutron time-of-flight (TOF) method in the energy range of 0.007 eV to 47 keV using a 46 MeV electron linear accelerator (linac) at the Research Reactor Institute, Kyoto University (KURRI). In order to experimentally prove the result obtained, the supplementary cross section measurement has been made from 0.3 eV to 1 keV using the Kyoto University Lead slowing-down Spectrometer (KULS) coupling to the linac. The relative measurement by the TOF method has been normalized to the reference value (20.01 b) at 0.0253 eV and the KULS measurement to that by the TOF method.

The existing experimental data and the evaluated capture cross sections in ENDF/B-VI, JENDL-3.2, and JEF-2.2 have been compared with the current measurements by the linac TOF and the KULS experiments. The energy dependency of the KULS data is close to that of the TOF data which are energy-broadened by the resolution function of the KULS.

1. Introduction

The nuclear cross section data of long-lived fission products (LLFPs) are of great importance for the assessment of reactor safety and the investigation of fuel-burn-up characteristics. In recent years, a great interest has been taken in the capture cross sections of LLFPs from the points of research and development of the nuclear transmutation technology.[1)]

Although the cross section of the ^{99}Tc(n,γ) reaction has been measured by a few experimental groups[2-5)], no data has been reported below 3 eV, except for the data at the thermal neutron energy.[6-8] Chou and Werle have measured the data from 3 eV to 50 keV by using a lead slowing-down spectrometer.[2] Resonance peaks of the cross section are energy-broadened due to the poor energy resolution of the spectrometer. In the keV energy region, Little and Block[3] and Macklin[4] measured the cross section by the neutron time-of-flight (TOF) method using an electron linear accelerator (linac). An experimental group of Igashira has also obtained the data with a Pelletron accelerator.[5] Very recently, the capture cross section has been measured from 3 to 400 eV at the Institute for Reference Materials and Measurements (IRMM) using the Geel linac GELINA and the resonance parameters obtained have been analized.[9,10] The evaluated cross section of the $^{99}Tc(n,\gamma)$ reaction has been compiled in ENDF/B-VI[11], JENDL-3.2[12] and JEF-2.2[13].

Since a lead slowing-down spectrometer can give us an intense neutron flux/spectrum in the eV or resonance energy region, a good signal-to-noise ratio can be achieved in the cross section measurement using even radioactive materials.[14] However, the energy resolution of the spectrometer is poor and about 30 to 35% at full width at half maximum (FWHM).[14,15] Recently, the lead slowing-down spectrometer has been successfully applied to the cross section measurement.[16]

2. Experiment and Measurement

The capture cross section measurement has been carried out by the neutron TOF method using a 46 MeV linac at the Research Reactor Institute, Kyoto University (KURRI). The experimental arrangement is shown in **Fig. 1**. The neutron collimation system was mainly composed of B_4C, Li_2CO_3 and Pb materials and tapered from 12 cm in diameter at the entrance of the flight tube to about 2 cm in diameter at the capture sample. The experimental arrangement is similar to the previous one.[16]

Technetium sample was made of oxide powder (TcO_2, chemical purity of Tc: 95%) of 72.8 mg, which was packed in an aluminum disk-container of 30 mm in diameter and 1.9 mm in thickness. No other γ-ray peak has been observed from the sample with a high purity Ge detector. A boron (B) plug of 1.8×1.8 cm^2, 0.5 cm in thickness (1.102 g/cm^2) was used for the neutron flux/spectrum measurement.

A pair of the C_6D_6 liquid scintillators (11 cm in diameter and 5 cm in thickness), which was placed at a distance of 12.0 ± 0.02 m from the pulsed neutron source of Ta, was employed for the capture cross section measurement of ^{99}Tc, and the sample was put in the neutron beam between the scintillators. The scintillators are less sensitive to neutrons inscattered from the sample/surroundings. The background measurement has been made with an empty disk-container for the sample. The background level has been also confirmed by the block-off method with a thick ^{10}B plug (4.54 g/cm^2) putting into the neutron beam.

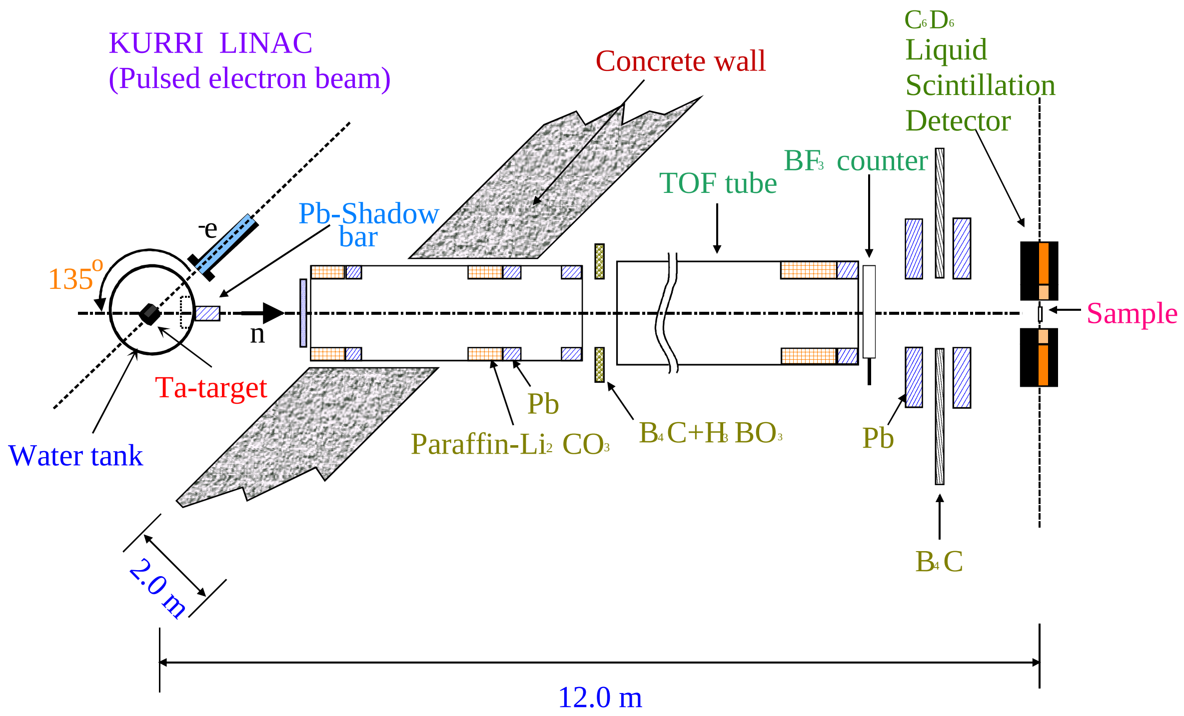

Fig. 1. Experimental arrangement for the linac time-of-flight method.

We have employed a coincidence method between the C_6D_6 detectors to reduce the background counts, although an anti-coincidence method has been applied to the B measurement. Through the amplifiers and the discriminators, signals from the detectors were fed into a time digitizer, which was started by the linac electron burst. The discrimination level was set at about 200 keV to detect the γ-rays from the ^{99}Tc and the B samples. Two sets of 4096 channels with a 0.5 μs channel-width were allotted to the C_6D_6 detectors and a BF_3 proportional counter as a neutron intensity monitor during the experiment. These signals were stored in a data-acquisition system for each measurement.

The linac was operated in two different modes: one was for the measurement below about 20 eV with a repetition rate of 50 Hz, a pulse width of 3 μs, a peak current of 0.4 A and an electron energy of 30 MeV, and the other is for the measurement above about 2 eV with a repetition rate of 200 Hz, a pulse width of 100 ns, a peak current of 6 A and an electron energy of 30 MeV.

The relation of capture yields for the ^{99}Tc(n,γ) reaction and the ^{10}B(n,α) reaction is obtained by the following relations: [16,17]

$$Y_X(E) = \frac{C_X(E)}{C_B(E)} \cdot Y_B(E) \quad , \tag{1}$$

where the subscripts X and B are for the ^{99}Tc and the B samples, respectively. $C_X(E)$ and $C_B(E)$ are the counting rates at energy E for each sample. $Y_B(E)$ is the energy dependent capture yield for the ^{10}B(n,α) standard cross section. There is a following relation between the capture cross section $\sigma_C(E)$ and the yield $Y(E)$:[16,17)]

$$Y(E) = \left\{ 1 - \exp(-N \cdot \sigma_T(E) \cdot t) \right\} \frac{\sigma_C(E)}{\sigma_T(E) \cdot Fc(E)} \quad , \tag{2}$$

where $\sigma_C(E)$ and $\sigma_T(E)$ are the neutron capture and the total cross sections, N and t are the atomic density and the thickness of the sample, and $Fc(E)$ is a correction function for the neutron scattering and/or self-shielding in the sample. In the current data analysis, we have employed the Monte Carlo code MCNP[18)] to derive the correction function $Fc(E)$.

We have also made a supplementary measurement of the ^{99}Tc(n,γ) cross section by the Kyoto University Lead slowing-down Spectrometer (KULS)[14)] using an Ar-gas proportional counter for the capture γ-ray measurement, as we have measured before.[16)] Using the lead slowing-down spectrometer, the capture cross section can be obtained in principle with the above Eqs.(1) and (2), although the energy resolution is broadened to be about 35 %.[14)]

3. Results and Discussion

Making use of the C_6D_6 detectors and the 46 MeV electron linac at KURRI, the capture cross section of ^{99}Tc has been measured relative to that of the ^{10}B(n,α) reaction from 0.007 eV to 47 keV and the result has been normalized to the reference cross section value of 20.01 b at 0.0253 eV in ENDF/B-VI. The measurement of the high energy run has been normalized to that between 3 eV and 10 eV in the low energy run. The major uncertainties are due to the statistical error, which is especially larger in the resonance region and the worst case is 208 %, deviation from the 1/v form in the ^{10}B(n,α) standard cross section, the data normalization with the reference value and the error related to the corrections by the neutron self-shielding and/or inscattered neutrons, as shown in **Table 1**. The correction for the neutron self-shielding and/or the neutron scattering in the capture sample was made by using the Monte Carlo code MCNP, as we did before.[16)] In the off-resonance energy region, we have summed up some of the channel-data to give better statistics. However, we have not added the measured data in the resonance peak region to keep the energy resolution better. As we have done before.[16)], we have investigated the detection efficiency of the C_6D_6 detection system, by measuring the pulse height spectra with neutrons (1) from 0.007 to 2 eV, (2) from 2 to 15 eV in the first resonance and (3) from 15 to 150 eV. The results processed by the weighting function method showed that the effect on the relative efficiency might be 3 % to 10 % at most in the resonance cross section region and that the factor was within the experimental uncertainty.

Table 1. Experimental Uncertainties for the Current Time-of-Flight Measurement.

Reasons for Uncertainties	Experimental Error (%)	
	E_n (min)	Error (%)
Statistical error	0.07 eV	5.8 ~ 6.7
	1.0 eV	3.8 ~ 25
	7.0 eV	8.0 ~ 208
	300 eV	17 ~ 47
	47 keV	———
Normalization to the reference value	3	
Deviation from the 1/*v* form in the ^{10}B(n,α) cross section	~ 2	
Background subtraction	~ 1	
Correction for the neutron self-shielding and/or neutron scattering effects	1.5 ~ 2.3	
Total uncertainty	5.6 ~ 208 (%)	

The capture cross section obtained by the linac TOF method is presented in **Fig. 2**. Since the data measured by Chou and Werle[2)] are energy-broadened by a lead slowing-down spectrometer, we cannot directly compare the data with the TOF measurement. However, their data are discrepant from the energy-broadened evaluation values and the current data at the resonance energy region. Above 200 eV, the cross sections measured by Chou and Werle[2)], Little and Block[3)], Macklin[4)] and the Igashira's group[5)] are close to the current measurement in general, although it seems that the data by Little and Block are a little higher at energies of 4 to 30 keV and that the data by Chou and Werle are a little lower above 10 keV. The evaluation data in ENDF/B-VI[11)], JENDL-3.2[12)] and JEF-2.2[13)] are almost similar to each other. They are in general agreement with the current measurement in the relevant energy range. In the cross section minimum region from 10 eV up to 200 eV, the measured values are close to the evaluations, although the statistical errors are pretty larger. The uncertainties may be caused by poor signal-to-background ratio due to the limited amount of ^{99}Tc sample.

The supplementary capture cross section of the ^{99}Tc(n,γ) reaction has been also measured relative to the ^{10}B(n,α) cross section, using the KULS[14)]. **Figure 3** shows the measured result from 0.3 eV to 1 keV. The experimental uncertainties, which are mainly due to the statistical ones, are from 0.3 to 23 %. The KULS data have been normalized to the energy-broadened

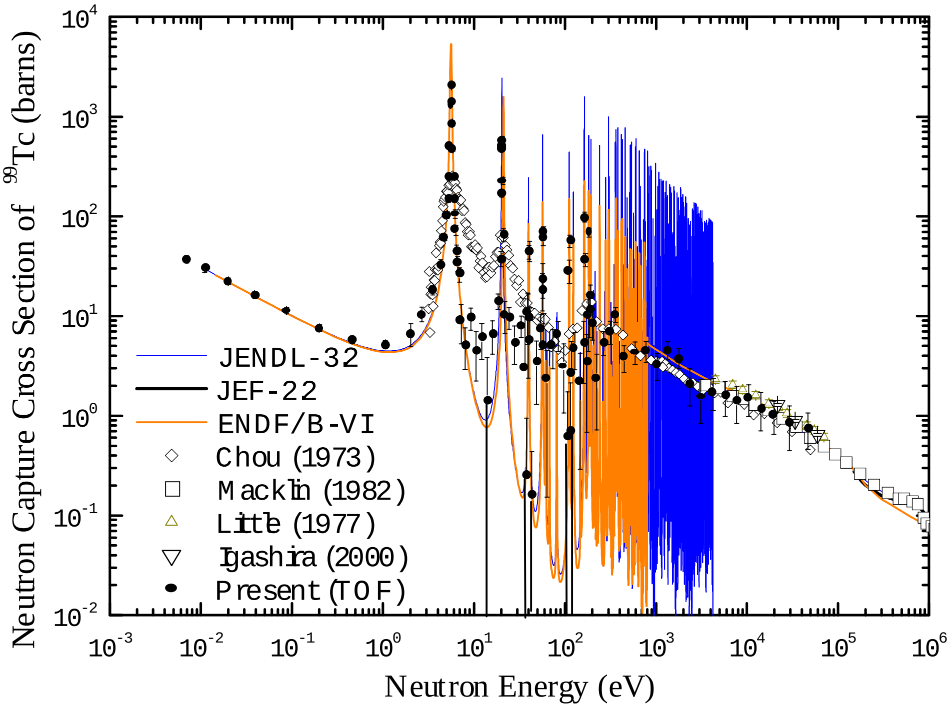

Fig. 2. Comparison of the current measurement and the experimental/evaluated cross sections of the ^{99}Tc(n,γ) reaction.

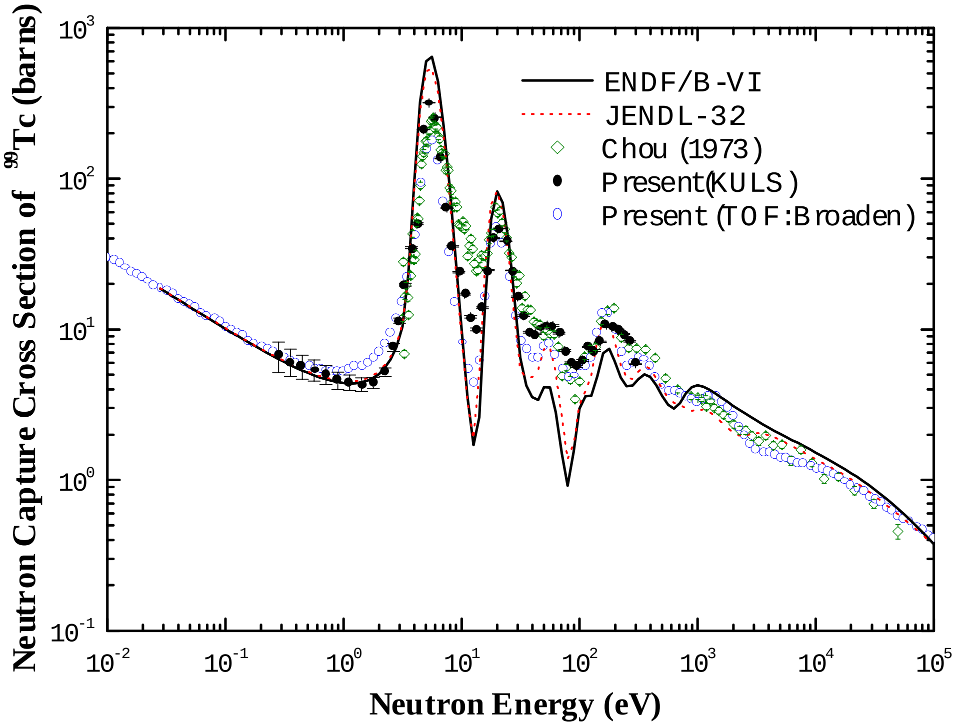

Fig. 3. Comparison of the measured data by the linac TOF method and the KULS and the evaluated cross sections in ENDF/B-VI and JENDL-3.2. The evaluated data and the linac TOF data are energy-broadened by the resolution function of the KULS.

TOF data by integrating them in the relevant energy region. The data by the KULS are practically close to the energy-broadened TOF data, although slight discrepancies are found between the data in the resonance structure region. As we have discussed before[16)], the discrepancies would be due to the fact that there may exist some problems in the resolution function to reproduce the KULS measurement. In the cross section minimum region, the data by Chou and Werle seem to be higher compared to the KULS data and the energy-broadened TOF data, which are closer to the energy-broadened evaluations in ENDF/B-VI and JENDL-3.2.

4. Conclusion

The existing experimental data by Little and Block, Macklin and the Igashira's group and the evaluation data in ENDF/B-VI and JENDL-3.2 are close in general above 200 eV to the current measurement from 0.007 eV to 47 keV by the linac TOF method. In the resonance energy region, the supplementary measurement has been also made from 0.3 eV to 1 keV with the lead slowing-down spectrometer, KULS, and the result is close to the data by Chou and Werle and to the energy-broadened TOF and the evaluation data, although some discrepancies among them are seen in the resonance cross section minimum region. We could provide the interesting data for the capture cross section, especially in the lower or resonance energy region.

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