Measurement of γ -ray Emission Probabilities of ¹⁰⁰Tc

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Abstract

In order to precisely determine thermal neutron capture cross section of ⁹⁹Tc nuclide, γ -ray emission probabilities of ¹⁰⁰Tc were measured using a newly developed β - γ coincidence system, which utilizes a plastic scintillator as a β -ray detector.

About 5 kBq of ⁹⁹Tc samples were irradiated for 15 seconds, and then β and γ rays were measured for three minutes. The measurements were repeated 99 times.

From the obtained data, ratios of coincidence counting rates to β singles counting rates, n_c/n_β , was determined for 540 keV γ ray. To determine γ -ray emission probabilities, a method has been developed which uses a Monte Carlo simulation and takes into account β feedings to excited levels and detection threshold of β rays.

1 Introduction

The ⁹⁹Tc is a long-lived $(T_{1/2} = 2.1 \times 10^5 \text{ y})$ fission product nuclide with a large fission yield. Therefore, it is suggested as a candidate for neutron transmutation, and many experimental works have been done in order to determine the thermal neutron capture cross sections of ⁹⁹Tc, in particular by using an activation method[1].

In order to precisely determine the cross section in the activation method, precise values of γ -ray emission probabilities of the reaction product, ¹⁰⁰Tc, are required. These values are, however, determined with large errors (7.0 ± 1.2 % for 540 keV γ ray [2]) mainly because of its short half-life ($T_{1/2} = 15.5$ s).

To determine γ -ray emission probabilities of the ¹⁰⁰Tc nuclide, a β - γ coincidence measurement has been done by using a newly developed β - γ coincidence system which utilizes a plastic scintillator as a β -ray detector [3].

2 The experiment and data analysis

The experiment has been carried out at the Research Reactor Institute of Kyoto University. About 10 μl of ammonium hydroxide solution which contained 5 kBq of ⁹⁹Tc as ammonium pertechnetate, was dried on an acrylic plate and irradiated by neutrons for 15 seconds using a pneumatic tube Pn-3 at the institute, and then β - and γ -rays emitted from the sample were measured.

Beta rays were measured using a plastic scintillation detector placed at 10 cm distance from the sample, whose thickness was 4 mm and which has a trapezoidal shape with an area of 33 cm². For γ -ray detection, a HPGe detector was used, whose relative efficiency is 90 % of that of 3"×3" NaI detector. The distance between the irradiated sample and the Ge detector was 3 cm. To follow the decay of the ¹⁰⁰Tc nuclei, the obtained data were partitioned every 5 seconds. The irradiations and the measurements were repeated 99 times to improve the statistical accuracy. A total of 2×10^6 counts were obtained for a peak area of the 540 keV γ -ray, which is emitted after β decay of the ¹⁰⁰Tc nuclei.

In **figure 1**, an energy spectrum of γ rays observed by the Ge detector is shown, which is obtained by summing singles (projection) spectra over all runs. From the spectrum, γ rays emitted following the decay of ¹⁰⁰Tc, namely 540, 591 and 1512 keV, are clearly identified. The figure also shows a



Figure 1: (upper) Singles spectrum of γ rays summed over all runs. (lower) Decay curve of 540 keV peak yields. The dashed line represents contribution from ¹⁰⁰Tc, while dotted lines are that from backgrounds.

decay curve of peak-areal counts of the 540 keV γ ray in a typical run. The decay data for each run were fitted by a function with the following form:

$$Y(t) = a \exp\left(-\frac{\ln(2)}{\tau}t\right) + C,$$
(1)

where τ , a and C were varied as free parameters. The τ corresponds to the half-life of ¹⁰⁰Tc. By averaging the obtained values for τ over all runs, τ for ¹⁰⁰Tc was determined to be 15.27 ± 0.02 (s), thus the 540 keV γ rays were confirmed to be emitted following the decay of ¹⁰⁰Tc.

Shown in **figure 2** is a β -ray energy spectrum summed over all runs. A bump around 50 channel bin was caused by noises, and therefore counts above 90 channel (inclusive) are summed as a β counts. **Figure 3** shows a decay curve for the β -ray counting rate in a typical run. To extract a contribution from ¹⁰⁰Tc to the singles β counts, the decay curves were fitted by a function with the following form:

$$Y'(t) = \sum_{i=1}^{3} a_i \exp\left(-\frac{\ln(2)}{\tau_i}t\right) + C',$$
(2)

In the fit, a_i s and C' were varied as free parameters while τ_i s were fixed to half-lives of ¹⁰⁰Tc (15.27 s), ¹⁹O (26.91 s [2]) and ²⁷Al (2.248 min. [3]). The solid line in the figure shows a result of a fit, while contribution from the decay of ¹⁰⁰Tc was shown by the dashed line. Coincidence counting rates of the β rays were determined in the same manner.



Figure 2: Singles spectrum of β rays summed over all runs.



Figure 3: Decay curves of singles β ray counts above 90 ch. The solid line represents the result of the fit and dashed line contribution from ¹⁰⁰Tc. The dotted lines express contributions from ¹⁹O, ²⁸Al and backgrounds.

From the result of the above mentioned fits, ratios of coincidence counting rates to β singles counting rates, n_c/n_β , were calculated for each time bin in each run. The final values of n_c/n_β were obtained after averaging over all time bins and all runs. For 540 keV γ ray, $n_c(540 \ keV)/n_\beta$ was determined to be 0.00168 ± 0.00001 .

To deduce γ -ray emission probabilities from the obtained n_c/n_β values, a method has been developed using the Monte Carlo simulation code EGS4[4]. It simulates the decay of the ¹⁰⁰Tc nuclei and the interaction between the detectors and the emitted particles (β and γ rays), and calculates n_c/n_β values as a function of β branching ratio to the ground state of ¹⁰⁰Ru. Four excited states of ¹⁰⁰Ru with large β branching ratios were taken into account in the simulation, namely the ground state, 540, 1131, 1362 and 2052 keV levels. With the five levels, more than 99.8 % of β decay feedings are exhausted[2]. Ratios between the feeding probabilities to the excited states were determined from the obtained singles γ -ray spectrum, after sum-coincidence corrections. By varying the β feeding probability to the ground state and searching for the value which reproduce the observed n_c/n_β values, the γ -ray emission probabilities can be determined. The analysis is now under way.

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