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FORMATION OF ^{99m}Tc BY THE (γ, γ') NUCLEAR EXCITATION

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Abstract: A new activation process to obtain ^{99m}Tc has been studied by (γ, γ') excitation of its ground state isomer ^{99}Tc (2.1×10^5y). Bremsstrahlung irradiation of the target ^{99}Tc using an electron linear accelerator gave radioactivities of ^{99m}Tc (6.0hr), ^{96}Tc (4.3d) and ^{95}Tc (20hr). The integrated cross section of ^{99}Tc for the ^{99}Tc (γ, γ') ^{99m}Tc process was found to be $(0.9\sim2.0)\times10^{-26}$MeV·cm^2 for incident electron energy 15~50MeV. Based on the above results, the (γ, γ') reaction was applied to radioactivation analysis of ^{99}Tc for the first time. The production rate of ^{99m}Tc and the detection limit of ^{99}Tc were 0.048Bq ^{99m}Tc/Bq ^{99}Tc and 3ng ^{99}Tc (50pCi) at 50MeV incident electron energy for 3hr irradiation, respectively.

(^{99}Tc (γ, γ') ^{99m}Tc, integrated cross section, radioactivation analysis of ^{99}Tc)

Introduction

No report has been published on nuclear excitation of ^{99}Tc (half life: 2.1×10^5y) through (γ, γ') reaction. As one of the most abundant fission products of uranium in many nuclear reactors operated in the world this nuclide is accumlated year by year. And it is also detected in radioactive fallout in the environment. Importance of the study of this nuclide is increasing remarkably from various points of view including waste management and effective utilization of byproducts of reactor operation.

We have succeeded nuclear excitation of ^{99}Tc to its isomeric state ^{99m}Tc (half life: 6.0hr) by (γ, γ') reaction using an electron linear accelerator of Tohoku University. The magnitude of the integrated cross section for the reaction is similar to that for ^{115}In (γ, γ') ^{115m}In reaction.

Application of this (γ, γ') reaction to radioactivation analysis of ^{99}Tc has been studied. This process was found to be useful when the amount of ^{99}Tc was more than nanogram, proving that easy and specific detection of ^{99}Tc is possible by our method.

Experimental

^{99}Tc in the chemical form of ammonium pertechnetate was purchased from the Amersham International Company and was dispersed in silica which was chosen as a supporting material. Irradiation of technetium was performed using an electron linear accelerator of Tohoku University for about 3-8 hours. Electron energy varied in the range of 15-50MeV. The beam intensity was 100μA. A platinum converter was used to get bremsstrahlung for irradiation. Radioactivities were measured with Ge(Li) detectors connected to 4k pulse-height analyzers.

Results and Discussion

Basic nuclear data

A typical γ-ray spectrum of a technetium target irradiated with bremsstrahlung of 50MeV electrons is shown in Fig. 1. Photopeaks of ^{99m}Tc (140keV), ^{96}Tc (778, 813, 850, 1127keV etc.) and ^{95}Tc (766, 1074keV) appear in the figure. ^{99m}Tc was produced by nuclear excitation ^{99}Tc (γ, γ') ^{99m}Tc, ^{96}Tc by ^{99}Tc (γ, 3n) ^{96}Tc reaction, and ^{95}Tc by ^{99}Tc (γ, 4n) ^{95}Tc reaction.

Table 1. Integrated cross sections of ^{99}Tc (γ, γ') ^{99m}Tc and ^{115}In (γ, γ') ^{115m}In reactions

Incident electron energy (MeV)	^{99}Tc (γ, γ') ^{99m}Tc (10^{-26}MeV·cm^2)		^{115}In (γ, γ') ^{115m}In (10^{-26}MeV·cm^2)	
15	1.02 ± 0.10 0.80 ± 0.08	Av. 0.91 ± 0.07	1.46 ± 0.15 1.55 ± 0.16	Av. 1.50 ± 0.11
20	1.51 ± 0.15 1.48 ± 0.15	Av. 1.50 ± 0.11	1.95 ± 0.20 2.06 ± 0.21	Av. 2.01 ± 0.14
50	2.07 ± 0.20 2.04 ± 0.20 1.92 ± 0.19	Av. 2.01 ± 0.12	2.11 ± 0.21 2.30 ± 0.23	Av. 2.21 ± 0.16

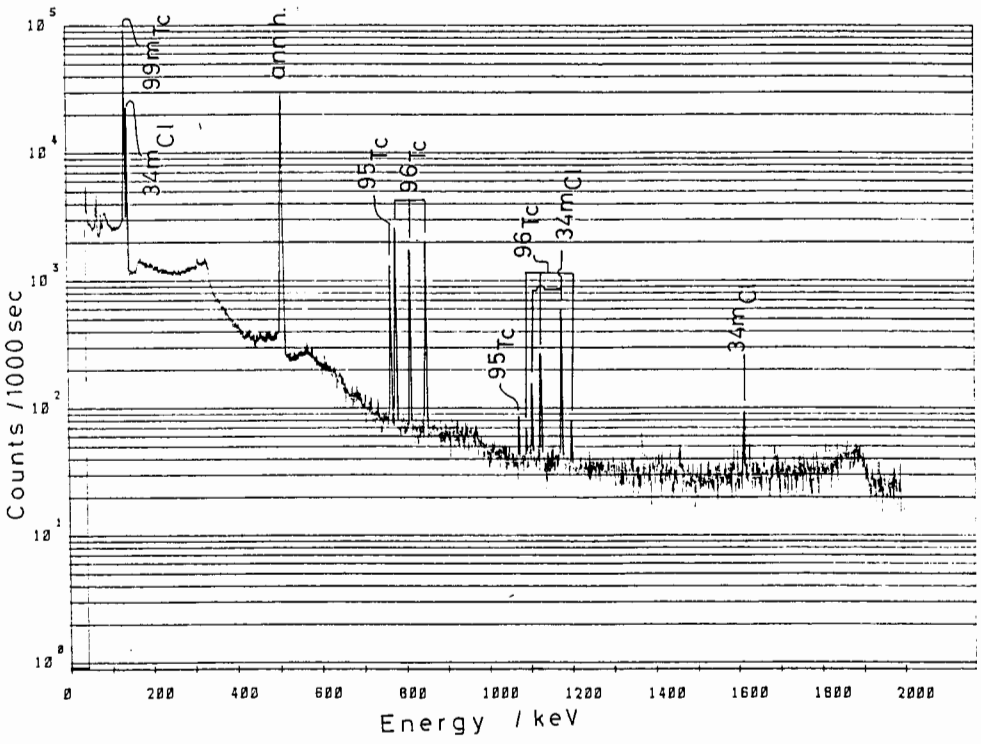

Fig.1. γ-Ray spectrum of a technetium sample irradiated with bremsstrahlung

Integrated cross sections of the $^{99}Tc(\gamma, \gamma')^{99m}Tc$ and $^{115}In(\gamma, \gamma')^{115m}In$ were determined at incident electron energies 15, 20 and 50MeV. They are listed in Table 1. Each value is calculated on the assumption that a giant resonance peak lies between 4 and 12MeV in both cases. These determinations were referred to gold, copper and yttrium monitors, of which cross sections are relatively well known.[1] We have got reliable and self-consistent results within 5% errors using these cross section data.

The integrated cross sections of ^{115}In for the process $^{115}In(\gamma, \gamma')^{115m}In$ are $(1.5\sim2.2)\times10^{-26}$MeV·cm^2. These values are larger than the reported values[2-5] $(3\sim10)\times10^{-27}$MeV·cm^2 in the 4-12MeV region. Probably tailing of the excitation function of the $^{115}In(\gamma, \gamma')^{115m}In$ process may be concerned with the observed differences.

Activation analysis

Although neutron activation analysis of technetium have been attempted by some authors,[6-9] it was not a convenient method because the product nuclide ^{100}Tc has a short half-life (15.8 s) and emits weak γ-rays only.

Instead of the (n, γ) activation, we have studied radioactivation analysis using the $^{99}Tc(\gamma, \gamma')^{99m}Tc$ reaction described in the previous section. ^{99m}Tc has a half-life long enough to allow ordinary chemical separation procedure, and its γ-ray energy is suitable for measurement by an ordinary γ-ray spectrometer. Nondestructive analysis is also possible with this (γ, γ') process.

Considering that the radioactivity of ^{99m}Tc produced by the (γ, γ') process depends on the flux of photons and on the distance from the platinum converter, we used monitors (usually Cu) in front of and behind the technetium samples in order to make corrections concerned with their positions.

The relation between the amount of technetium and counts of ^{99m}Tc is shown in Fig. 2. Radioactivity increases linearly with increasing amount of technetium up to μg level. This relation is useful in radioactivation analysis as a calibration line. In this condition the production rate is not optimum because a sweep magnet was used in irradiation. Therefore, the maximum production rate was sought under different irradiation conditions as follows. (1) Condition 1. Maximum electron energy was 50MeV, a platinum converter of 1mm thickness was used, and a sweep magnet was inserted in order to remove unconverted electrons. Irradiation time was 8 hrs. (2) Condition 2. Maximum electron energy was 30MeV, a platinum converter of 2mm thickness was used without a sweep magnet. Irradiation time was 3 hrs. (3) Condition 3. Maximum electron energy was 50MeV, a platinum converter of 2mm thickness was used without a sweep magnet. Irradiation time was 3 hrs. The beam current is about 100μA in all the cases. The maximum production rate of ^{99m}Tc was obtained in the condition 3. A linear relationship between the production rate of ^{99m}Tc and of ^{64}Cu (in the copper monitor) shown in Fig. 3 was found.

On the basis of the relation in Fig. 3 we calculated the maximum production rate of ^{99m}Tc in the technetium sample : 0.048Bq ^{99m}Tc/Bq ^{99}Tc. If we put the samples at the nearest position to the platinum converter and increase irradiation time, the production rate will be improved. The

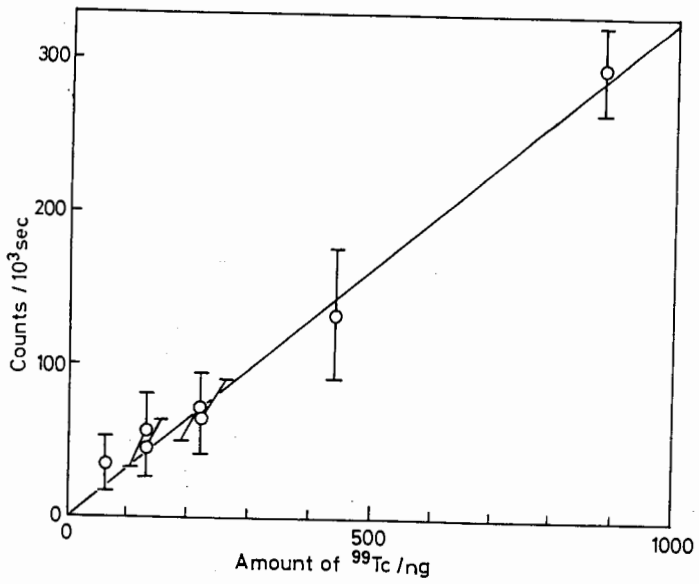

Fig.2. The relation between ^{99}Tc content and ^{99m}Tc radioactivity.

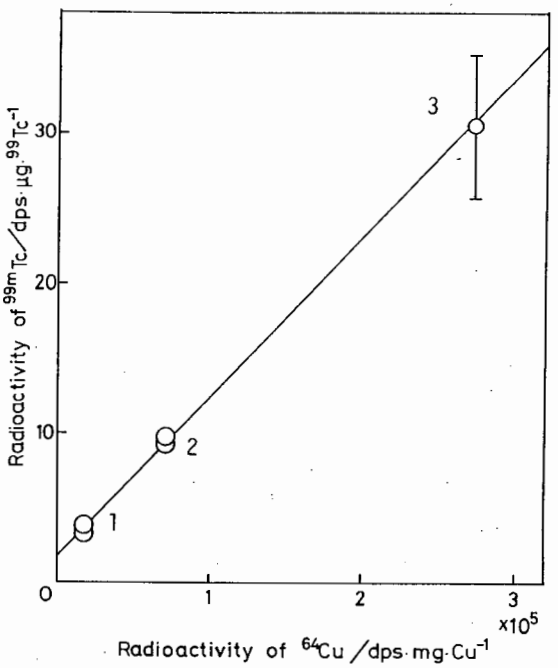

Fig.3. The relation between the production rate of ^{99m}Tc and specific activity of ^{64}Cu in the monitor. Numbers show the conditions indicated in the text.

detection limit in the condition 3 is calculated to be 3ng ^{99}Tc (50pCi). Although this detection limit is worse compared to that in liquid scintillation counting of ^{99}Tc, the photoactivation analysis is useful in the following point.

1. Detection of technetium is easy and specific using a γ-ray spectrometer.
2. Technetium can be analyzed in a nondestructive manner. No chemical separation procedure is requested.
3. If preconcentration procedure is adopted, the detectable concentration of technetium will be much improved.

Summary

^{99}Tc(γ, γ′)^{99m}Tc nuclear excitation was observed in irradiation of ^{99}Tc with bremsstrahlung of LINAC electron beam. The integrated cross section was determined in various conditions for the first time; the value $(0.9\sim2.0)\times10^{-26}$ MeV·cm^2 was comparable with that of ^{115}In(γ, γ′)^{115m}In.

Radioactivation analysis of ^{99}Tc using the ^{99}Tc(γ, γ′)^{99m}Tc reaction was studied with a γ-ray spectrometer. Detection of ^{99m}Tc was easy and the detection limit of ^{99}Tc was 3ng (50pCi) at present.

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