(LNS Experiment: #2389, #2408, #2424, #2443, #2458, #2460, #2475, #2490, #2492, #2502, #2503, #2513, #2514, #2525, #2526, #2541, #2542)

Search for the Decay of ^{229m}Th by Photon Detection

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§1. Introduction

The first excited state of thorium-229, ^{229m}Th, is expected to show an intriguing decay property because of its extremely low-lying excitation energy. The excitation energy has been reported to be either 3.5 ± 1.0 eV or 3.4 ± 1.8 eV from the results of precise γ-ray spectroscopy conducted at the decay of ^{233}U.

In the disintegration process from ^{229m}Th to ^{229g}Th, the emission of internal conversion electrons is forbidden because the disintegration energy is lower than even the first ionization energy of thorium atoms. Thus the de-excitation from ^{229m}Th to the ground state is expected to take place through a direct γ-ray transition or decay via an electron bridge (EB) mechanism in which part of the transition energy is deposited to excite the valence-shell electrons of ^{229m}Th. This implies that the half-life of ^{229m}Th is directly affected by the chemical state of ^{229m}Th.

Several experimental results have been reported by different groups about the detection of photons emitted from ^{233}U samples that contain a considerable amount of ^{229m}Th produced through the α decay from ^{233}U with a branching ratio of about 2 %. These attempts were not successful, however, owing to interference of radiation-induced fluorescence from the materials around the radioactive ^{233}U sample.

In our experiments, Th atoms were chemically separated by means of an anion exchange method for removing the influence of intense radiation from ^{233}U. As for the photon detection, the detection efficiency was raised as high as possible by developing a novel detection system. In this report, the limits of the half-life of ^{229m}Th in HNO_3 solution are deduced based on the experimental observations.

§2. Experiment

Thorium-229m samples were prepared by a chemical separation from about 93mg of ^{233}U containing 4.4 ppm of ^{232}U by the following procedure. Uranium-233 was first sorbed in 2 cm^3 of an anion exchange column (Dowex 1X8, 200-400 mesh) in 9M HCl solution and its descendants except for Bi and Tl were eluted. After a given growth time, accumulated ^{229}Th and ^{229m}Th were eluted in 5 ml of 9M HCl solution and separated from ^{233}U. In order to thoroughly remove ^{233}U, the eluate was again passed through

another anion exchange column of the same resin in 9M HCl solution. We were able to accomplish the whole chemical separation within only about 3 minutes. Following the separation, 1 mg of Al and approximately 10 ml of concentrated ammonia water were added into the eluate. Thorium atoms were coprecipitated with Al in the form of hydroxide. After the decantation of the supernatant liquid, the precipitate was dissolved in 3 to 4 droplets of 8 M HNO_3 in order to minimize the sample volume to prepare a quasi point source. The solution sample was transferred into a small quartz bowl of a size of 6 mm in internal diameter and 10 mm in height, and photon measurements were performed.

Taking a wide range of the expected half-life of ^{229m}Th into account, two different experiments were carried out varying the growth time: 2 and 17 hours for the first and second experiments, respectively. It took 18 minutes to accomplish the chemical separation and treatment in the first experiment, and 30 minutes in the second.

In this work, we consider that the detection efficiency precedes the energy information. Although the output signal of a photomultiplier (PM) has no information concerning the energies of incident photons, therefore, we adopted the PM designed for single photon counting. In order to improve the detection efficiency, an oval reflector was employed to focus as many photons as possible on the photocathode of the PM.

The detection efficiency was estimated taking into account several parameters: the sample geometry, the reflection efficiency of the reflector, the effect of a finite volume of the ^{229m}Th solution sample, the quantum efficiency of the PM, and the absorbance by the HNO_3 solution. The whole detection efficiency was accordingly estimated to be at least 4×10^{-4} for the photons in the range between 340 and 600 nm (3.6 - 2.1 eV).

§3. Result and Discussion

It should be noted that ^{229m}Th accumulates, during the growth time, depending on T^{HCl}, and ^{229m}Th disintegrates, during the measurement, at the rate T^{HNO3}. Here, T^{HCl} and T^{HNO3} denote the half-lives of ^{229m}Th as a solute in HCl and HNO_3 solutions, respectively. Since the chemical condition of ^{229m}Th turns during the preparation of the sample, the following discussion on T^{HNO3} is based on the number of ^{229m}Th atoms grown in the HCl solution.

In Fig. 1 and 2 are shown the time variation of the photon counting rate for the first experiment (2-hour growth time) and that for the second experiment (17-hour growth time), respectively. The horizontal dashed lines represent the constant background level obtained by a blank measurement. It is obvious from the figures that additional photons deriving from the ^{229m}Th source as well as from the other descendants were not evidently detected.

Although the half-life cannot be determined, the limits of the half-life can be estimated from the results. The estimation of the limit of T^{HNO3} requires 1) the lower limit of the number of ^{229m}Th atoms at the end of the growth in the HCl solution and 2) the detection efficiency which has already been estimated. The number of ^{229m}Th atoms can be figured from the activity of ^{229m}Th at the end of the growth time. If T^{HCl} is 3 minutes, which was taken for the chemical separation, or shorter, it would be difficult to estimate the limit of T^{HNO3} because of their decay loss during the separation. From these

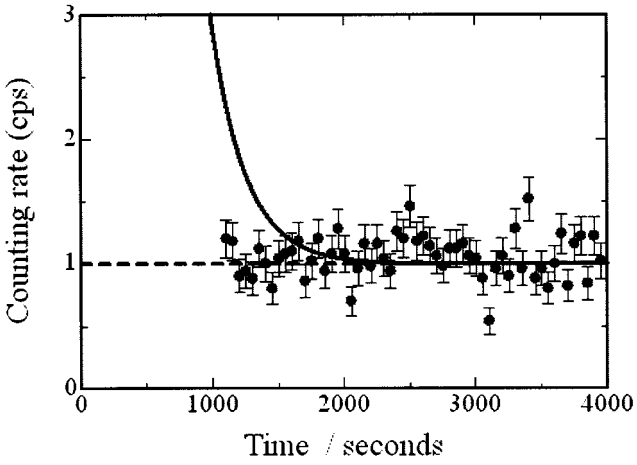

Fig.1. Time variation of the counting rate for photons emitted from ^{229m}Th HNO_3 solution sample grown for 2 hours in the 9M HCl.

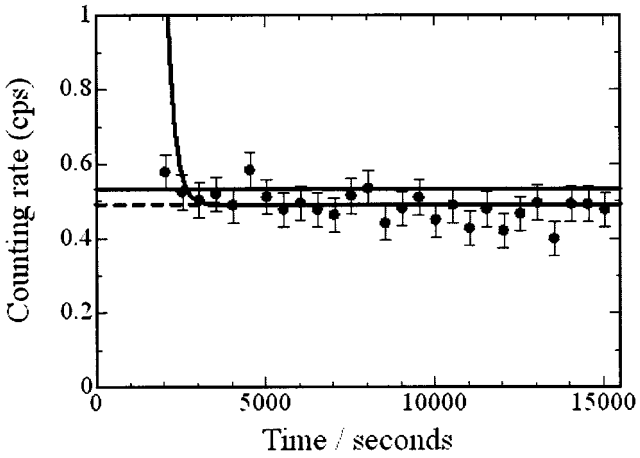

Fig.2. Time variation of the counting rate for photons emitted from ^{229m}Th HNO_3 solution sample grown for 17 hours in the 9M HCl.

conditions, the limits were estimated as follows.

1) If T^{HCl} is shorter than 3 minutes, it is difficult by the present experimental method to observe evident photon events for any half-life of ^{229m}Th in HNO_3 solution.
2) If T^{HCl} is longer than 3 minutes, T^{HNO3} can be limited as $<$ 3 minutes or $>$ 60 hours as indicated by solid line in Fig. 1. Especially referring to the case that T^{HCl} is longer than 3 hours, the number of ^{229m}Th atoms accumulated during the growth time is almost constant in both experiments and

T^{HNO3} should be < 2 minutes or > 400 days as indicated by the solid lines in Fig. 2.

For further discussion about T^{HNO3}, more detailed information on T^{HCl} is necessary. A new device for the preparation of a point source of ^{229m}Th is now under development.