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Observation of α-decay of ^{229m}Th produced from ^{229}Ac

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We produced ^{229}Th in the nuclear reaction ^{232}Th(γ, p2n)^{229} Ac, followed by disintegration to ^{229m,g}Th. The α-decay signals from ^{229m}Th were searched for and the alpha-particle events of the energy region between 4.93 MeV and 5.05 MeV were observed in the separated thorium fraction from an actinium source highly purified from the ^{232}Th + γ reaction products. The energy values of the α-particles coincide with those expected for ^{229m}Th.

§1. Introduction

Recent investigations of ^{229}Th have reported energy difference between the ground state ^{229g}Th and the first excited state ^{229m}Th to be (3.5 ± 1.0) eV [1, 2] or (3.4 ± 1.8) eV [3]. In those studies, ^{229m,g}Th was produced from the α-decay of ^{233}U. Additional evidence for the existence of ^{229m}Th was given by Burke *et al*. [4] using ^{230}Th (d,t) particle reaction spectroscopy. The internal conversion process is prohibited in ^{229}Th because the energy of the isomeric transition from ^{229m}Th to ^{229g}Th is lower than the first ionization potential of thorium. Therefore, the investigation of the ^{229m}Th decay is expected to provide a strong verification of the electron bridge mechanism (EBM) [5], which is a de-excitation process competing with isomeric transition and α-decay. Since the probability of the EBM depends on the energy state of the outer-shell electrons, the half-life of ^{229m}Th can vary with its chemical state; hence the isomer is also interesting from a chemical viewpoint.

It is believed that no one has thus far succeeded in detecting a decay signal from ^{229m}Th. In a few studies, the observation of photons due to isomeric or EBM transitions was attempted by ultraviolet and visible spectroscopy [6, 7], but there is still some doubt regarding the results [8, 9]. Browne *et al*. [10] searched for ^{229m}Th from a sample prepared from approximately 25g of ^{233}U. They tried to observe the growth of γ-rays associated with the α-decay of ^{229g}Th due to the feeding from ^{229m}Th, but observed no evidence of such growth. The failure to observe these events might be attributable to two reasons: the difficulty in detecting extremely low energy photons and the small branching ratio from ^{233}U to ^{229m}Th,

which is about 2%.

In order to address the first difficulty, it seems reasonable to devise the identification of ^{229m}Th by the detection of its α-decay. The most favorable decay-mode of ^{229m}Th should be the direct γ-transition or the EBM [5]. The observation of photon emission from the isomeric state requires detecting ultraviolet-visible photons under considerable interference, such as α-particle induced fluorescence. Such a measurement is very difficult. On the other hand, the α-decay of ^{229m}Th is undoubtedly nuclear disintegration phenomenon, though as a minor decay-mode. The technique for α-particle detection is well established and its detection is subjected to little interference from the background. Therefore, it is expected that if α-particles from ^{229m}Th exist, they are observable. The α-decay of ^{229m}Th has been discussed by Mitsugashira *et al*. [11] and Dykhne *et al*. [12]. Both expected the favored α-particle decays from ^{229m}Th had energies higher than those from ^{229g}Th, i.e. 4.93-5.05 MeV. This implies that the partial half-life of the α-decay of ^{229m}Th is shorter than that of ^{229g}Th.

To address the difficulty of the small branching ratio from ^{233}U to ^{229m}Th, we produced ^{229m}Th from β-decay of ^{229}Ac following a nuclear reaction process, whereas in all previous investigations on the decay properties of ^{229m}Th, the nuclide was produced from the α-decay of ^{233}U. By using a suitable nuclear reaction, it was expected that the ^{229}Th sample would have a m/g ratio large enough to observe the minor α-decay from ^{229m}Th, although complicated chemical procedures for eliminating by-products would be required.

Based on these ideas, we previously reported the detection of α-decays of ^{229m}Th produced by the ^{230}Th(γ, n) reaction [11]. Unfortunately, it appeared that the results contained interfering effects due to pile-up of the α-particles from ^{230}Th decay and β-rays of the other nuclear reaction products. In this reaction system, the interference was unavoidable because it is impossible to chemically separate the ^{229m}Th from the target ^{230}Th and other thorium isotopes produced in the reaction.

In this study, we attempted to detect the α-decay signals from ^{229m}Th produced using another reaction system. We obtained ^{229m}Th from β-decay ^{229}Ac that was produced through the reaction ^{232}Th(γ, p2n). A sample of ^{229m,g}Th was prepared using several chemical separation methods and was measured with an α-spectrometer.

§2. Experimental

2.1 Preparation of the thorium dioxide target

A ^{232}Th reagent, allowed to stand unprocessed for more than 40 years, contains daughter nuclei in radioactive equilibrium mixture, such as ^{228}Ra and ^{228}Ac. The ^{228}Ac feeds ^{228}Th that significantly interferes with the observation of the α-particles of ^{229m}Th. Therefore, ^{228}Ra and ^{228}Ac should be removed from thorium in radioactive equilibrium with its decay series nuclei just before bremsstrahlung irradiation. To perform such chemical separation, about 1 g of thorium dioxide was dissolved in a mixture of 10 mL of 13M (mol/dm^3) nitric acid (HNO_3) and a drop of 27M hydrofluoric acid (HF) under heating, and the solution was passed through a column packed with anion exchange resin (Dowex 1X8, 100-200 mesh, 30 mmϕ × 70 mm) to adsorb thorium isotopes. The resin was washed with 150 mL of 8M HNO_3 to remove radium and actinium, and then thorium isotopes were eluted from the column with

150 mL of 2M HCl. A greater than 3-fold excess of saturated oxalic acid was added to the eluate under heating, then the resulting precipitate, $Th(C_2O_4)_2$, was aged for one night. The precipitate was washed with distilled water and ignited at 800 ºC for 8 hours to produce thorium dioxide.

2.2 Bremsstrahlung irradiation

A sample of about 1.65 g of $^{232}ThO_2$ was enclosed in a quartz tube for irradiation with bremsstrahlung radiation. The irradiation was carried out using the Electron Linear Accelerator at Tohoku University. The accelerator was operated at an electron energy of 30 MeV with a mean current of around 0.1 mA. The $^{232}ThO_2$ target was placed in close contact with the back of a platinum converter and cooled with running tap water during the 1-hour irradiation. After the irradiation, the target was chemically treated according to the procedure outlined in Fig.1 and the details given below.

2.3 Isolation of actinium from the irradiated target

The target material was dissolved in a mixture of 10 mL of 13M HNO_3 and one drop of 27M HF under heating, and diluted with water to 40 mL (diluted to about 4M HNO_3). In order to remove thorium isotopes from the solution, including aimed actinium, solvent extraction with 40 mL of 1M HDEHP-benzene was carried out three times in series. After washing the aqueous phase with 40 mL benzene, actinium isotopes were co-precipitated with iron hydroxide by adding 1 mg of iron and 15M ammonia water (NH_{3aq}). The precipitate was then dissolved in 1M HNO_3 and the solution passed through a cation exchange resin column (Dowex 50X8, 200-400 mesh, 5.5 mmϕ × 40 mm), which adsorbs actinium isotopes. The resin was washed with 8 mL of 3M HNO_3 to remove radium and iron. Then actinium isotopes were eluted from the column with 12 mL of 8M HNO_3. The solution was passed through an anion exchange resin column (Dowex 1X8, 200-400 mesh, 5.5 mmϕ × 40 mm) to completely eliminate thorium from the solution. The actinium fraction containing rare earth fission products was adjusted to a 9M HCl solution and passed twice in series through an anion- exchange resin column (Dowex 1X8, 200-400 mesh, 5.5 mmϕ × 40 mm) to adsorb completely ^{231}Pa produced from ^{231}Th. These purification procedures for actinium were carried out within about one hour after the end of bremsstrahlung irradiation.

2.4 Preparation of $^{229m,g}Th$ source for α-particle counting and spectrometry

To allow the decay of ^{229}Ac ($T_{1/2}$ = 62.7 min) and the growth of $^{229m,g}Th$ isotopes, the purified actinium fraction was left to stand for 3 hours after the end of the purification of actinium. The solution containing thorium isotopes was adjusted to 8M HNO_3 solution during these 3 hours and passed through an anion- exchange resin column (Dowex 1X8, 200-400 mesh, 5.5 mmϕ × 40 mm) to adsorb the thorium isotopes. The resin was washed with 20 mL of 8M HNO_3 to remove rare earth fission products and actinium isotopes. The thorium isotopes were eluted from the column with 5 mL of 2M HCl and co-precipitated with samarium hydroxide by adding 30 μg of samarium and 15M NH_{3aq}. The precipitate was collected on a 0.02-μm alumina filter (Whatman, ANODISC membrane) of 18 mm diameter to prepare a counting source. The filter was fixed on a stainless-steel supporting ring and dried at 130 ºC. All the chemical procedures were finished about 5 hours after the end of irradiation. The sample was measured by α-ray spectrometry with a 450 mm^2 silicon detector and a 2k-channel PC-PHA system. The background count rate for this detector was about 20 counts/day in the energy range of 3 to 8 MeV

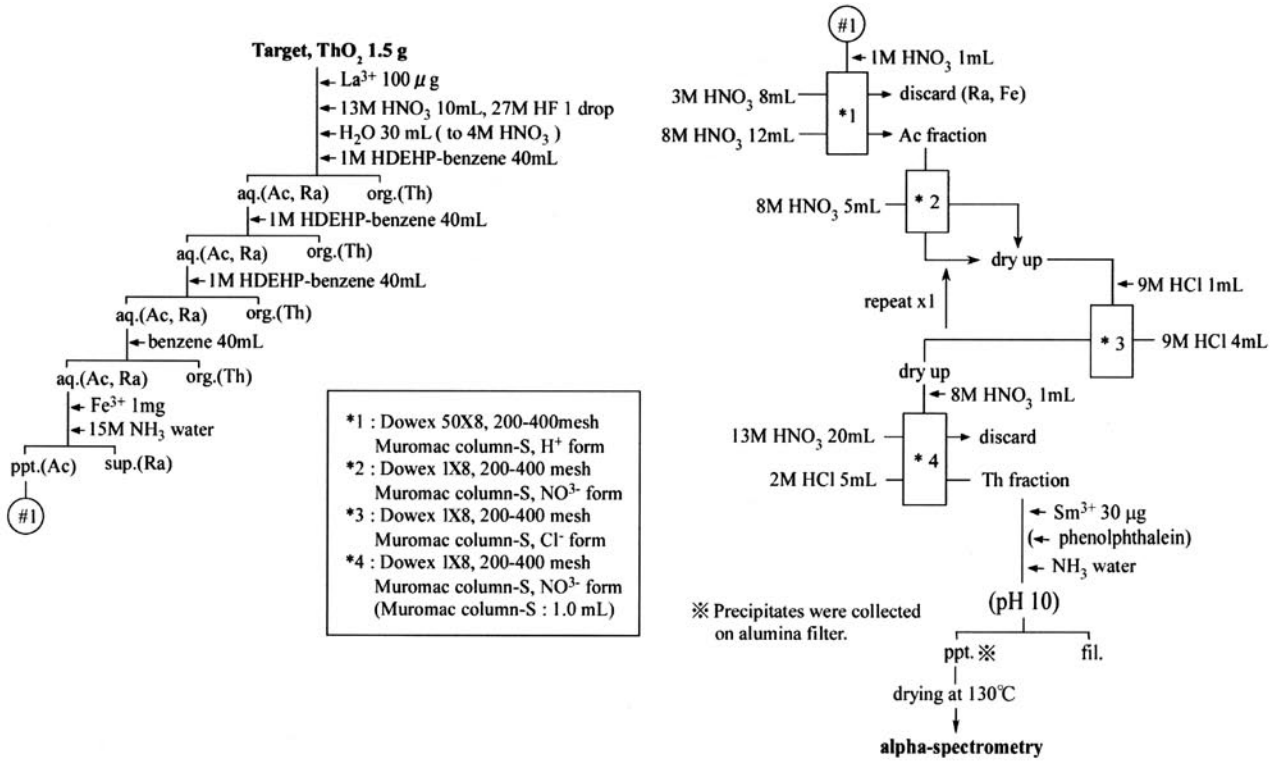

Fig.1. Chemical procedure for preparation of the ^{229m,g}Th sample.

and less than 1 count/day in the energy range of 4.5 to 5.1 MeV. Collection of α-events was started within 10 minutes of completing the counting source preparation and the accumulation of an α-ray spectrum for 10^4 -sec was repeated 90 times.

§3. Results and discussion

An α-ray spectrum obtained as a sum of the first 30 spectra is shown in Fig.2. The α-peaks of ^{147}Sm, ^{232}Th, ^{230}Th, and ^{228}Th and its daughters can be observed in the spectra. Some dozen α-counts are observed between the ^{230}Th peaks and the ^{228}Th peaks, at an energy consistent with that expected for ^{229m,g}Th. It was found that the ratio of the α-counts of ^{230}Th/^{229}Th/^{228}Th was about 3/1/70, while it was about 5500/1/10 for the thorium sample isolated from ^{230}Th(γ, n) reaction products [11]. In this experiment, the count number of ^{230}Th decays was relatively small and had no influence on the identification of ^{229m,g}Th. However, interference from ^{228}Th on the observation of ^{229}Th may have occurred, as discussed below, because the count number of ^{228}Th decays was greater by a factor of seven in this experiment.

A magnification of the α-spectrum for the region related to ^{229}Th is shown in Fig.3 (a). The spectrum of the energy region differs from the spectrum expected for only ^{229g}Th. The causes of this difference are believed to be the peak tail from the peaks of ^{228}Th and its daughters, α-counts from impurities, and/or the existence of ^{229m}Th.

^{228}Th was formed simultaneously with ^{229m,g}Th by the β-decay of ^{228}Ac produced in the ^{232}Th(γ, p3n) reaction in the source. It has the largest number of counts in the spectra and the energy region is

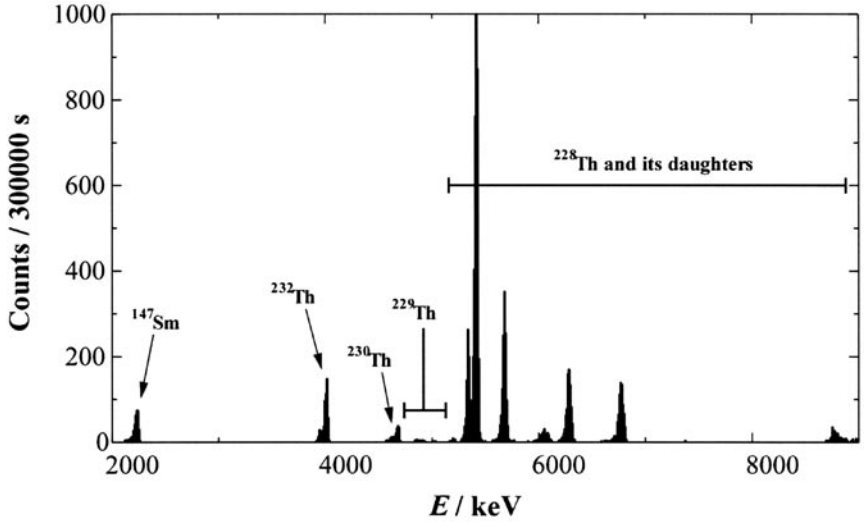

Fig.2. α-ray spectrum acquired over a counting duration of 3×10^5 s, starting 10 minutes after the chemical separation.

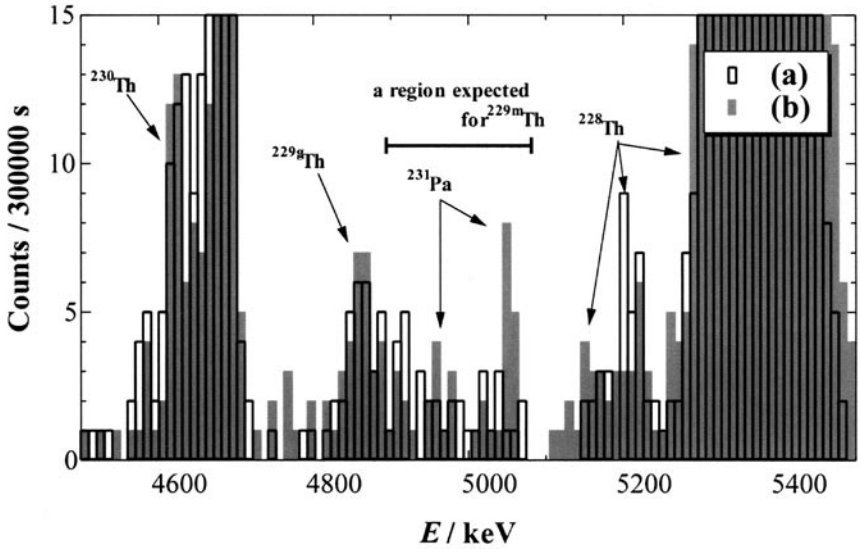

Fig.3. Expanded α-ray spectra acquired over a counting duration of 3×10^5 s, starting 10 minutes after the chemical separation (a), and acquired 1 week after the chemical separation (b).

higher than that of ^{229m,g}Th. It was found, however, that the high resolution spectrum is little influenced by the peak tail of ^{228}Th. In Fig.3 (a) the ^{228}Th peak has a FWHM of about 29 keV measured at 5423 keV.

A growth in time of the α-count rate was observed. These counts were identified by the energy value as the α-peaks of ^{231}Pa. The growth of ^{231}Pa interfered with the identification of ^{229m}Th. The spectrum obtained one week after the chemical separation is shown in Fig.3 (b), overlaying Fig.3 (a), demonstrating the growth of ^{231}Pa. It is thought that ^{231}Pa was completely eliminated by the repetitive anion exchange at the end of the chemical purification of the actinium fraction and, therefore, the observed ^{231}Pa may be the decay product of ^{231}Th, produced in the ^{232}Th(γ, n) reaction. The counting source included ^{231}Th because elimination of thorium was not complete. Although the decontamination

factor of thorium isotopes in the purified actinium fraction was estimated to be about 5×10^{-5}, sufficient ^{231}Pa was produced to interfere with the identification of ^{229m}Th because of the large cross-section of the ^{232}Th(γ, n) reaction.

The spectrum in Fig.3 (b) can be reproduced by sum of the ^{229g}Th spectrum and the ^{231}Pa spectrum. It indicates that the half-life of ^{229m}Th is not as long as one week. On the other hand, the early spectrum in Fig.3 (a), before the growth of ^{231}Pa, differs from the spectrum expected for only ^{229g}Th. In particular the observed α-counts between 4.93 MeV and 5.05 MeV, which coincides with the expected energy of ^{229m}Th, were larger than those expected for ^{229g}Th. It may be considered that the new α-emitter ^{229m}Th was observed in this work, although the α-peaks are not clearly defined, probably due to poor counting statistics.

§4. Conclusion

We attempted to produce ^{229m}Th by the nuclear reaction ^{232}Th(γ, p2n)^{229}Ac, followed by disintegration to ^{229m,g}Th. The alpha-events in the expected energy region for ^{229m,g}Th were observed with less interference than has been achieved in previous experiments. The spectral shape differs from that expected for only ^{229g}Th, which may be due to presence of ^{229m}Th. A better chemical separation of actinium from thorium would eliminate the interference of ^{231}Pa and allow more definitive results to be obtained.

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References

[1] C.W. Reich and R.G. Helmer: Phys. Rev. Lett. **64** (1990) 271.

[2] R.G. Helmer and C.W. Reich: Phys. Rev. C**49** (1994) 1845.

[3] V. Barci, G. Ardisson, G. Barci-Funel, B. Weiss, O. El Samad, and R.K. Sheline: Phys. Rev. C**68** (2003) 034329.

[4] D.G. Burke, P.E. Garrett, T. Qu, and R.A. Naumann: Phys. Rev. C**42** (1990) R499.

[5] E.V. Tkalya, A.N. Zherikhin, and V.I Zhudov: Phys. Rev. C**61** (2000) 064308.

[6] G.M. Irwin and K.H. Kim: Phys. Rev. Lett. **79** (1997) 990.

[7] D.S. Richardson, D.M.. Benton, D.E. Evans, J.A.R. Griffith, and G Tungate: Phys. Rev. Lett. **80** (1998) 3206.

[8] S.B. Utter, P. Beiersdorfer, A. Barnes, R.W. Lougheed, J.R. Crespo López-Urrutia, J.A. Becker, and M.S. Weiss: Phys. Rev. Lett. **82** (1999) 505.

[9] R.W. Shaw, J.P. Young, S.P. Cooper, and O.F. Webb: Phys. Rev. Lett. **82** (1999) 1109.

[10] E. Browne, E.B. Norman, R.D. Canaan, D.C. Glasgow, J.M. Keller, and J.P. Young: Phys. Rev. C**64** (2001) 014311.

[11] T. Mitsugashira, M. Hara, T. Ohtsuki, H. Yuki, K. Takamiya, Y. Kasamatsu, A. Shinohara, H. Kikunaga, and T. Nakanishi: J. Radioanal. Nucl. Chem. **255** (2003) 63.

[12] A.M. Dykhne, N.V. Eremin, and E.V. Tkalya: JETP Lett. **64** (1996) 345.