Production and photon measurement of ^{229}Pa toward the observation of radiative decay of ^{229m}Th

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The first excited state of ^{229}Th (^{229m}Th) has an extremely low excitation energy of ∼8.3 eV (150 nm),[1)] which may enable a nuclear clock with unprecedentedly low uncertainty. So far, the radiative half-life of ^{229m}Th, which is an important parameter to develop the nuclear clock, has not yet been determined. To directly observe the radiative decay (γ-ray emission) of ^{229m}Th, the internal conversion (IC) process with a half-life of ∼7 μs[2)] must be prohibited by placing ^{229m}Th in the chemical environments where the electron binding energy is higher than the excitation energy of ^{229m}Th. ^{229m}Th doped into fluoride crystals is a candidate for such chemical environments. We are aiming to dope a CaF_2 crystal with ^{229m}Th by doping with ^{229}Pa, which decays to ^{229m}Th by electron capture with a negligibly small recoil energy. Suitable doping can be realized by implanting high-energy ^{229}Pa ions into a crystal and then annealing it. In this study, we developed a method for producing ^{229}Pa ($T_{1/2}$ = 1.5 d) in the ^{232}Th$(p, 4n)^{229}$Pa reaction and separating it from the target. We also measured low-energy photons from Pa isotopes on a CaF_2 crystal to evaluate the background toward the future γ-ray measurement of ^{229m}Th.

Two ^{232}Th metallic foils (thickness: 69.07 mg/cm^2, purity: 99.5%) were irradiated with a 30-MeV proton beam having an intensity of 1 μA for 10 h at the RIKEN AVF cyclotron. After the irradiation, we measured γ-ray spectra for the ^{232}Th foils and fractions resulting from the subsequent chemical separation process using a Ge detector.

The chemical separation process for one of the foils was performed as follows. First, we dissolved the foil in 2 mL of 11.3 M HCl plus 300 μL of 1 M HF and heated the solution to dryness. The sample was dissolved in 2 mL of 11.3 M HCl, following which 1.1 g of $Al(NO_3)_3 \cdot 9H_2O$ was added as a masking agent for the remaining F^- ions. After the solution was dried up and dissolved in 2 mL of 11.3 M HCl, the solution was fed into an anion-exchange column (Muromac 1X8, 100–200 mesh, ∼1.0 mL). We poured 10 mL of 11.3 M HCl into the column to elute Th isotopes, Ac isotopes, and some fission products. Next, we added 10 mL of 6 M HCl to elute Zr, following which 5 mL of 8 M HNO_3 was added to elute Zr, Mo, Ru, Sb, and Te isotopes. After we added 1 mL of 11.3 M HCl, Pa isotopes were eluted with 8 mL of 9 M HCl/0.1 M HF. The chemical yield of Pa isotopes in the whole process was 94(2)% (residual Pa isotopes were observed in the 8 M HNO_3 eluate). The radioactivity of chemically separated ^{229}Pa at the end of bombardment was evaluated to be 30(1) MBq, while those of ^{232}Pa, ^{230}Pa ($T_{1/2}$ = 17.4 d), ^{95}Zr ($T_{1/2}$ = 64.03 d), and ^{97}Zr ($T_{1/2}$ = 16.75 h), included as impurities, were 2.18(3), 0.89(4), 0.0054(7), and 0.40(2) MBq, respectively.

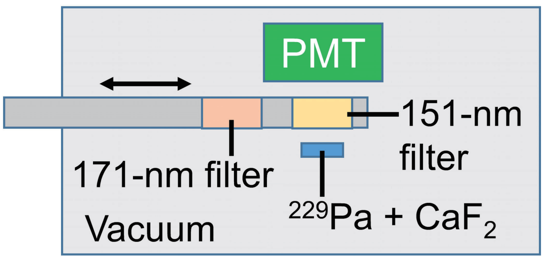

Fig. 1. Setup of the photon measurement of ^{229}Pa.

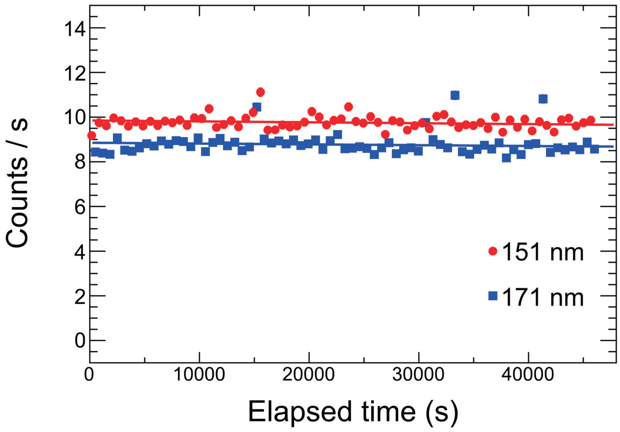

Fig. 2. Count rates of photons as a function of the elapsed time for the 151-nm (red circle) and 171-nm (blue square) filters. Red and blue lines show the decay curves of ^{230}Pa fitted to the data.

The ^{229}Pa sample dissolved with 27 M HF was dropped on a CaF_2 crystal, which was then annealed at 900°C for 1 h in a He gas flow (3 L/min). Photons from the crystal were measured with a photomultiplier (PMT, Hamamatsu R10454) in vacuum (Fig. 1). Band-pass filters for the photons of 151 ± 20 and 171 ± 20 nm (eSource Optics) placed between the crystal and the PMT were switched every 5 min. The radioactivities of ^{229}Pa, ^{230}Pa, and ^{232}Pa were 10.7(8), 35(2), and 0.381(8) kBq at the start of the measurement (11 days after the proton irradiation).

As shown in Fig. 2, the count rates of photons for both filters are ∼10 counts per second (cps), which

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are much higher than the dark count rate of the PMT (0.96 cps). The detected photons would originate from the Cherenkov radiation caused by the passage of beta particles though the CaF_2 crystal. The long decay time of photons (half-life >7 d) in Fig. 2 indicates that the Cherenkov photons dominantly originate from the beta decay of ^{230}Pa (0.004 photons per beta particle). If we implant 100 kBq of ^{229}Pa into a CaF_2 crystal, anneal it, and start the measurement one day after the proton irradiation, the background photons from ^{232}Pa and ^{230}Pa are estimated to be ∼30 cps, which is much higher than the estimated count rate of γ rays from ^{229m}Th (2 cps). Thus, we plan to perform the mass separation of ^{229}Pa to reduce the amount of ^{232}Pa and ^{230}Pa by a factor of >10 when we implant ^{229}Pa into a CaF2 crystal. Implanting ^{229}Pa into a crystal with an efficiency of 0.1–1% allows us to observe a growth and decay curve of photons of several cps only for the 151-nm filter, resulting in the unambiguous identification of the γ rays from ^{229m}Th.

References

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