

Improved chemical separation scheme of Pa isotopes from a ^{232}Th target toward observing the radiative decay of $^{229\text{m}}\text{Th}$ using ^{229}Pa

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The first excited state of the ^{229}Th nucleus ($^{229\text{m}}\text{Th}$) has an excitation energy of ~ 8.3 eV (150 nm),¹⁾ potentially leading to an ultraprecise nuclear clock. We aim to observe the radiative decay (γ rays) of $^{229\text{m}}\text{Th}$ and determine its radiative half-life; this is an important parameter to develop the nuclear clock by doping a CaF_2 crystal with ^{229}Pa , which decays to $^{229\text{m}}\text{Th}$ with negligibly small recoil energy.²⁻⁴⁾ In a previous study,⁴⁾ we ionized ^{233}Pa and implanted it into a CaF_2 crystal with a total efficiency of 0.53(1)% using the surface ionization technique. However, when we tried the ionization and implantation of ^{229}Pa that was produced in the $^{232}\text{Th}(p, 4n)^{229}\text{Pa}$ reaction and chemically purified,³⁾ the efficiency was limited to $\sim 0.1\%$. A probable reason for the low efficiency is that some amount of ^{232}Th and impurities remain in the purified ^{229}Pa solution and inhibit the ionization of Pa. Therefore, we improved the chemical separation scheme in this study by adding a separation step using the CL resin (Triskem).⁵⁾ Moreover, for faster separation, we increased the flow rate of eluents during column chromatography by a factor of ~ 10 , compared with our previous separation experiments.^{2,3)}

The chemical separation scheme was developed using ^{230}Pa ($T_{1/2} = 17.4$ d) and ^{233}Pa ($T_{1/2} = 27.0$ d) produced in the $^{232}\text{Th}(d, 4n)^{230}\text{Pa}$ and $^{232}\text{Th}(d, n)^{233}\text{Pa}$ reactions at the RIKEN AVF cyclotron. Two ^{232}Th foils ($70 \text{ mg/cm}^2 \times 2$, ~ 140 mg) were irradiated with $9.1 \mu\text{A}$ of a 24-MeV deuteron beam for 70 min. The foils were dissolved with concentrated HCl and 0.05 M $(\text{NH}_4)_2\text{SiF}_6$ ⁶⁾ 62 days after the irradiation. The chemical separation of the Pa isotopes was performed following the scheme shown in Fig. 1, which consists of three column separations using anion-exchange resin (Column A and C) and the CL resin (Column B). The flow rate of every eluent was set to 1–2 mL/min by pushing the air in the column using a peristaltic pump. The radioactivity of Pa isotopes and fission products (FPs) for each fraction was measured via γ -ray spectroscopy using a Ge detector to check chemical yields and radioactive impurities. The final Pa fraction was subjected to inductively coupled plasma mass spectrometry (ICP-MS) to check the chemical purity.

The separation by Column A removed the majority of ^{232}Th and FPs, and the Pa fraction that eluted from Column A included radioactive impurities of ^{95}Zr ($T_{1/2} = 64.0$ d), ^{95}Nb ($T_{1/2} = 35.0$ d), and ^{103}Ru ($T_{1/2} = 39.2$ d). The separation with Column B reduced

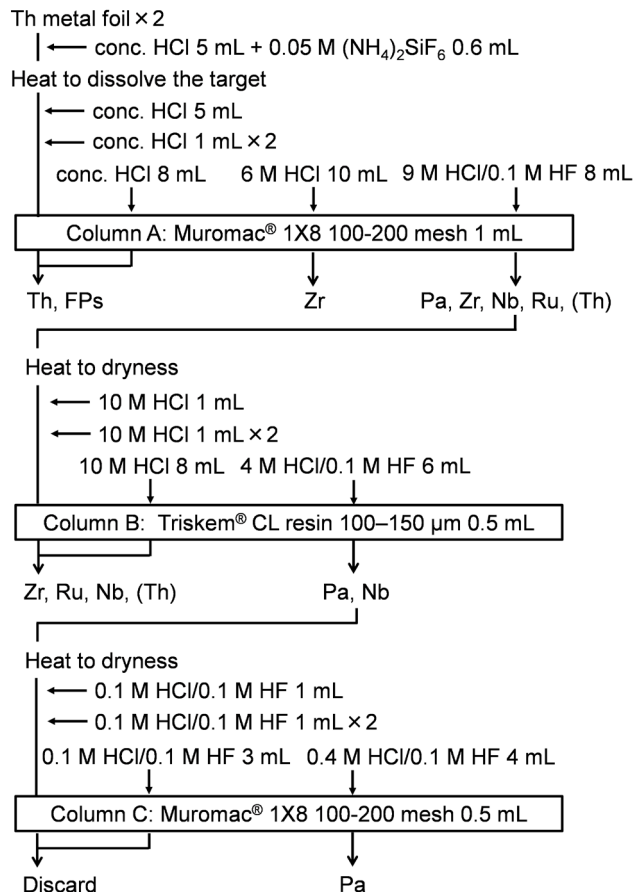


Fig. 1. Chemical separation scheme of Pa.

^{95}Zr and ^{103}Ru to levels less than the detection limits of the γ -ray measurement. ^{95}Nb , which behaves similarly to Pa, was separated with Column C by a decontamination factor of 220, which is high enough for the experiment of ^{229}Pa . It was reported that Nb can be separated with the Eichrom DGA resin;⁵⁾ thus, we checked the performance of this method. However, the decontamination factor was measured to be only 12, and thus, we selected the method in Fig. 1. The chemical yields of Pa isotopes for Columns A, B, and C were 95(4)%, 96(3)%, and 95(3)%, respectively (total yield 86(5)%), meaning that the separation at the high flow rate does not lead to the loss of Pa. We expect that the whole separation will be finished in ~ 4 h, which is ~ 3 times shorter than that of the previous scheme, minimizing the decay loss of ^{229}Pa . For the ICP-MS measurement of the purified Pa solution, the impurities of >100 ng were Mg (627 ng), Al (284 ng), Zn (141 ng), and Th (167 ng). Therefore, the amount

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of ^{232}Th was reduced to the same level as other impurities.

We are now developing a new Pa implantation apparatus that enables in varying the implantation energy of ^{229}Pa in the range of 0.5–30 keV, not limited to 15 keV.⁴⁾ In a preliminary experiment using the purified $^{230,233}\text{Pa}$ solution and the new apparatus, the total ion implantation efficiency was measured to be $\sim 0.2\%$, which is higher than the previous value using ^{229}Pa ($\sim 0.1\%$), probably due to the improved chemical purification scheme. Higher efficiency may be further obtained by improving and optimizing the new apparatus.

References

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