Preparation of a small high-density ^{229}Th target for the X-ray pumping of the ^{229}Th nuclear clock isomer

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The first excited state of the ^{229}Th nucleus (^{229m}Th) has an excitation energy of ∼8.3 eV,[1] which allows laser excitation and spectroscopy of the nucleus. One application is ultraprecise nuclear clocks with unprecedented uncertainty.[2] Our group aims to observe the γ-rays emitted from ^{229m}Th, which is an important step for realizing nuclear clocks. In 2019, we succeeded in actively producing ^{229m}Th by populating the 29.2-keV second excited state of ^{229}Th using X-rays at SPring-8.[3] With this technique, we are now trying to detect γ-rays of ^{229m}Th produced in a ^{229}Th-doped CaF_2 crystal, in which the internal conversion process of ^{229m}Th would be inhibited. For every beamtime at Spring-8, we have to precisely adjust the energy of X-rays for the resonant excitation of ^{229}Th to the 29.2-keV state. For the energy adjustment, we have used a source containing 0.24 μg of ^{229}Th placed in a hole with a diameter of 0.4 mm on a graphite plate. Owing to the small amount of ^{229}Th, searching for the resonant X-ray energy has required more than 14 h. The above ^{229}Th source was prepared by performing the ∼7000 sets of dropping a small amount of ^{229}Th solution (5 nL per drop) into the hole and evaporating it by heating. The amount of ^{229}Th that can be poured into the small hole was limited because the nanoscale drop was not stable and the density of the ^{229}Th sample was lower than expected, probably owing to the repeated rapid evaporation processes. In this study, we developed a new simple method and successfully prepared a small high-density ^{229}Th source, which is used to search for the resonant X-ray energy for the 29.2-keV state.

The new source-preparation method is drying one drop (∼1 μL) of nitric acid containing ^{229}Th on a Teflon plate. Owing to the high water repellency of Teflon, the shape of the drop remains spherical during the evaporation process, and the drop size becomes much less than 1 mm just before it is completely dried. We prepared two types of Teflon plates shown in Fig. 1. The type A plate has a small taper hole made by drilling, where the ^{229}Th solution is dropped. The hole in the type B plate was manually prepared by heating the Teflon plate and deforming it with a conical die. We found that a drop on the type B plate was evaporated and retained a spherical shape. In contrast, a drop on the type A plate slightly expanded, and the size of the residues became larger than 1 mm. The images obtained by scanning electron microscopy (SEM) for the type A plate (Fig. 2) showed scratches made by drilling, which would be the reason for the lower water repellency. The type B plate showed a smooth surface (Fig. 2); thus, we adopted the type B plate to prepare the ^{229}Th source.

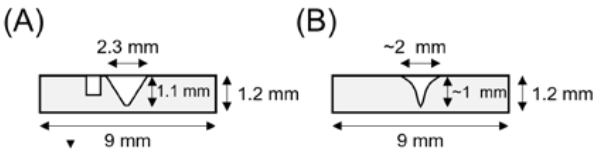

Fig. 1. Schematics of the Teflon plates of types A (left) and B (right).

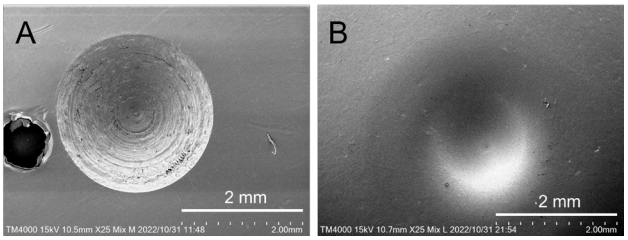

Fig. 2. SEM images of the Teflon plates of types A (left) and B (right).

^{229}Th was first purified via anion exchange chromatography using high-purity acids (metal impurities <100 ppt). The purified ^{229}Th was dissolved with 10 μL of 1-M HNO_3. The solution was dropped on a Teflon sheet and evaporated. Then, the residue was dissolved with 1 μL of 1-M HNO_3 and dropped on the type B plate placed on a hot plate. We washed the Teflon sheet with 1 μL of 1-M HNO_3, which was then added to the drop on the type B plate. The temperature of the hot plate was first maintained at ∼45°C for 20 min to slowly evaporate the majority of the solution. It was then increased to ∼120°C over 11 min and maintained at that temperature for 9 min. The temperature was then raised to 200°C over 13 min and maintained at that temperature for 19 min for complete dryness.

The ^{229}Th source shown in Fig. 3 had a residue with a diameter of ∼0.7 mm. The amount of ^{229}Th in the source was measured to be 1.61(2) μg by γ-ray spectroscopy, which is 6.7 times larger than the amount of the previous ^{229}Th source that we have used.[3] X-ray fluorescent measurements performed at SPring-8 showed that a large amount of ^{229}Th was condensed within a diameter of 0.65 mm. By using this new ^{229}Th source, we were able to finish the X-ray en-

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ergy search for the excitation to the 29.2-keV state four times faster than that for the previous source.

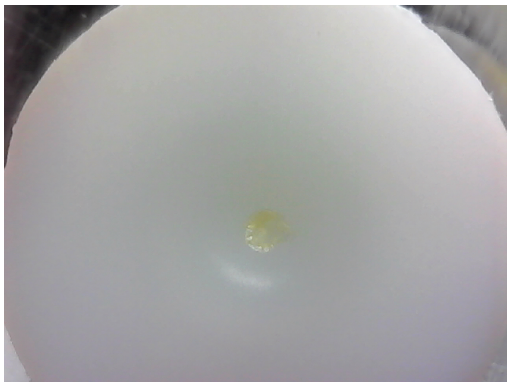

Fig. 3. Photograph of the prepared ^{229}Th target.

References
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3) T. Masuda *et al.*, Nature **573**, 238 (2019).