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Re-determination of the half-life of ^{229}Th

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^{229}Th is an alpha-decay nuclide of the neptunium decay series. Though already extinct in nature, ^{229}Th occurs in high amount in spent fuel as the progeny of the long-lived ^{237}Np . ^{229}Th is one of the most widely used tracers in geology, environmental sciences or nuclear chemistry to determine ^{230}Th and ^{232}Th concentrations by mass spectrometry due to the relatively long half-life and to the fact that it does not occur in nature. ^{229}Th as an isotopic tracer plays a special role in nuclear forensics, where its uncertainty associated with its concentration is one of the most dominant components in the uncertainty budget of radiochronometry of illicit nuclear materials¹. As the ^{229}Th standards are certified by activity concentration (e.g. Bq g⁻¹), the accurate knowledge of ^{229}Th half-life value is of utmost importance to convert it to amount content (e.g. mol g⁻¹) needed for mass spectrometry. The first precise half-life measurement of ^{229}Th was accomplished by Hagemann et al.² and gave a value of 7340 ± 160 years. However, a later measurement by Goldstein et al.³ by isotope dilution mass spectrometry reported an approximately 7% higher half-life of 7880 ± 120 years. Recently, Kikunaga et al.⁴ measured the ^{229}Th half-life by alpha spectrometry. Their half-life value of 7932 ± 55 years is consistent with the result obtained by Goldstein et al., and the authors suggest that the inaccuracy of the early half-life value is attributed to incomplete separation of ^{233}U from its impurities.

The aim of the present study is to re-measure the ^{229}Th half-life using inductively coupled plasma mass spectrometry (ICP-MS) at lowest possible uncertainty. By the measurement of the ^{229}Th amount content in the certified ^{229}Th radioactivity standard (SRM 4328C), the half-life of ^{229}Th can be calculated. The amount content of ^{229}Th was determined by two independent methods, both traceable to SI. In the first method, the ^{229}Th amount content was measured by isotope dilution ICP-MS technique, using a natural thorium certified reference material as a tracer. In the second method, the ^{229}Th amount content was measured against a completely separated highly-enriched uranium solution. In this case, the applied uranium sample was completely purified from its thorium decay products beforehand at a well-known time. Thus the ^{230}Th daughter product of the ^{234}U , which serves actually as the tracer for the ^{229}Th determination, can be very precisely calculated knowing the ^{234}U content and the elapsed time between the uranium separation and ^{229}Th determination¹. Our measured ^{229}Th half-life of 7921 years with an expanded uncertainty of 56 years ($k=2$) is in agreement with the latest values. Uncertainty was calculated according to the GUM (Guide for the Expression of Uncertainty in Measurement). The obtained value can help resolve the problem with ^{229}Th half-life discrepancy, and can result in more precise age dating measurements both for geological and nuclear samples.

References

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