Determination of the ^{229}Th half-life

Zsolt Varga,^{*} Adrian Nicholl, and Klaus Mayer

European Commission, Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany

(Received 12 April 2014; revised manuscript received 19 May 2014; published 16 June 2014)

The paper describes the remeasurement of the half-life of ^{229}Th (ground state). The half-life was obtained by the measurement of amount content of ^{229}Th in the certified ^{229}Th radioactivity standard (SRM 4328C) from the National Institute of Standards and Technology. The measurement was performed by isotope dilution inductively coupled plasma mass spectrometry by using two independent standards for quantification. From the results the ^{229}Th half-life is determined to be 7917 years with an expanded uncertainty of 48 years using a coverage factor of $k = 2$. The methodology is traceable to the International System of Units, and the calculation complies with the recommendations of the Bureau International des Poids et Mesures for nuclear data evaluation.

DOI: [10.1103/PhysRevC.89.064310](https://doi.org/10.1103/PhysRevC.89.064310)

PACS number(s): 21.10.Tg, 23.60.+e, 27.90.+b

I. INTRODUCTION

^{229}Th is an α -decay nuclide of the neptunium radioactive decay series, which was recognized and was characterized in 1947 simultaneously by Hagemann *et al.* [1] and English *et al.* [2]. Although already extinct in nature, ^{229}Th occurs in some 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 isotope dilution mass spectrometry due to the relatively long half-life and the fact that it does not occur in nature. Recently, ^{229}Th was used to produce ^{225}Ac or ^{213}Bi for targeted α therapy [3,4]. ^{229}Th as an isotopic tracer plays a special role in nuclear forensic investigations since the tracer concentration uncertainty is one of the most dominant sources of uncertainty for the production date determination of illicit nuclear materials [5,6]. As the ^{229}Th standards are currently certified only as activity concentration (e.g., Bq g^{-1}), the exact knowledge of the ^{229}Th half-life is of utmost importance to convert it to amount content (e.g., $\text{mol } ^{229}\text{Th g}^{-1}$ standard) as required for mass spectrometry. The first precise half-life measurement of ^{229}Th was determined to be 7340 ± 160 years by Hagemann *et al.* [7] in 1950, which is still adapted in several nuclear databases, such as the Joint Evaluated Fission and Fusion File (JEFF-3.1.1). However, a later measurement by Goldstein *et al.* [8] by isotope dilution mass spectrometry reported a half-life of 7880 ± 120 years (2σ), which is approximately 7% higher than the earlier published value. This value is also adapted in several nuclear databases, such as the Nuclear Data Sheets [9] or Evaluated Nuclear Structure Data File [10]. Recently, Kikunaga *et al.* [11] measured the ^{229}Th half-life by high-resolution α spectrometry. Their half-life value of 7932 ± 55 years (at a confidence level of 2σ) is consistent with the result obtained by Goldstein *et al.*, and the authors suggest that the inaccuracy of the early half-life value by Hagemann *et al.* is attributed to incomplete separation of ^{233}U from its impurities.

The aim of the present study is to remeasure the ^{229}Th half-life accurately and traceable to the International System

of Units (SI). The SRM 4328C ^{229}Th radioactivity standard was used for the measurement, which was recently released by the National Institute of Standards and Technology (NIST) [12] and has a significantly lower relative combined standard uncertainty (0.30%) than the one used in the earlier study by Goldstein *et al.* (0.5%) [8]. The amount content of ^{229}Th in the certified ^{229}Th radioactivity standard was determined against two independent thorium tracers using inductively coupled plasma mass spectrometry (ICP-MS). In the first method (Method A), the ^{229}Th amount content was measured by the isotope dilution ICP-MS technique, which used a custom-made (certified as mass ratio) natural thorium (dominantly ^{232}Th) certified reference material as a tracer. The ^{232}Th standard used is fully traceable to SI. In the second method (Method B), the ^{229}Th amount content was quantified against ^{230}Th , which was obtained from a highly enriched uranium (HEU) sample. The HEU had been completely separated from ^{230}Th (progeny of ^{234}U) at a well-known time. Thus, ^{230}Th (daughter product of ^{234}U), which had grown in after the separation, serves actually as the tracer for the ^{229}Th determination, can be very precisely calculated by knowing the ^{234}U content and the elapsed time between the uranium separation and ^{229}Th determination [5]. Combining the two ^{229}Th amount content measurement results with the certified ^{229}Th activity concentration, the half-life can be calculated.

II. EXPERIMENTAL PROCEDURES

A. Standards and reagents

Suprapur grade hydrofluoric and nitric acids (Merck, Darmstadt, Germany) were used for the sample preparation. HNO_3 was further purified by sub-boiled distillation (AHF analysentechnik AG, Germany). For dilutions ultrapure water was used (Elga LabWater, Celle, Germany). TEVATM extraction chromatographic resin (50–100- μm particle size, active component: aliphatic quaternary amine) supplied by Triskem International (Bruz, France) was used for the thorium separations in Method B. Some 0.4 ml of the TEVATM resin was placed in plastic Bio-Rad holders (diameter: 6 mm, length: 14 mm), and a porous Teflon frit was placed on the top to avoid mixing (Reichelt Chemietechnik Heidelberg, Germany). Before use, the resin was cleaned with 0.8 ml of 0.02M

^{*}zsolt.varga@ec.europa.eu

HF/0.02M HNO₃ followed by conditioning with 3 ml 2M HNO₃. All labware was thoroughly cleaned with ethanol and dilute HNO₃ before use. All weighings were carried out gravimetrically on calibrated balances, and the accuracy of the balances was checked daily against certified weights.

The SRM 4328C ²²⁹Th radioactivity standard was purchased from the National Institute of Standards and Technology (Gaithersburg, USA) and was opened directly before the study to minimize possible evaporation or adsorption. The activity of the SRM 4328C standard was decay corrected to the time of the measurement using the ²²⁹Th half-life value from Kikunaga *et al.* [11]. The decay correction was practically independent from the half-life value chosen and amounts to -0.056% .

The ²³³U (New Brunswick Laboratory, USA) isotopic standard was used to spike the samples for the uranium concentration measurements. A custom-made natural Th solution from Spex Certiprep, Inc. (Metuchen, USA) was used for the ²³²Th isotope dilution measurements as a spike for Method A. The thorium concentration of $999 \pm 5 \mu\text{g g}^{-1}$ is certified as a mass fraction with a relative expanded uncertainty of 0.5% ($k = 2$), and it contains almost exclusively ²³²Th [the $n(^{230}\text{Th})/n(^{232}\text{Th})$ amount ratio is $(2.1 \pm 0.1) \times 10^{-6}$]. Nominally 1% enriched uranium U-010 standard reference material from the National Bureau of Standards (USA) was used to correct for instrumental mass discrimination. IRMM-035 [certified $n(^{230}\text{Th})/n(^{232}\text{Th})$ is $(1.1481 \pm 0.0039) \times 10^{-5}$] and IRMM-185 [certified $n(^{235}\text{U})/n(^{238}\text{U})$ is $(2.00552 \pm 0.00030) \times 10^{-2}$] isotopic reference materials were used to check the accuracy of the thorium and uranium isotope ratio measurements, respectively.

The completely purified HEU material, which is used to obtain the ²³⁰Th for Method B, was prepared from nuclear-grade purity uranium solution with approximately 90% ²³⁵U enrichment. The preparation of the material is described in detail elsewhere [5]. The material is an archive uranium-oxide sample dissolved in nitric acid. The HEU sample was purified with three consecutive extraction chromatographic separations to remove the ²³⁰Th decay product. The final thorium purification of the material took place on July 19, 2011 at 11:40 a.m., and the material was evaporated to dryness right after its preparation to avoid loss of Th by adsorption. The separation efficiency of Th was determined by γ spectrometric measurement and by the addition of ²³²Th to the starting material and its remeasurement following the chemical separations. A total separation factor of $(3.5 \pm 0.4) \times 10^7$ was achieved, which corresponds to a ²³⁰Th/²³⁴U ratio in the final reference material of $(6.7 \pm 0.8) \times 10^{-13}$ at the time of preparation, thus the residual ²³⁰Th is negligible compared to the ingrowth thereafter. Thus, the amount of ²³⁰Th in the material is then solely the function of the ²³⁴U amount, and its amount can be derived from the decay equations,

$$N_{230\text{Th}} = N_{234\text{U}} \frac{\lambda_{234\text{U}}}{\lambda_{230\text{Th}} - \lambda_{234\text{U}}} (e^{-\lambda_{234\text{U}}t} - e^{-\lambda_{230\text{Th}}t}), \quad (1)$$

where $N_{230\text{Th}}$ and $N_{234\text{U}}$ are the amount content of ²³⁰Th and ²³⁴U in the sample, respectively, $\lambda_{230\text{Th}}$ and $\lambda_{234\text{U}}$ are the decay constants of ²³⁰Th and ²³⁴U, respectively, and t is the elapsed time

since the separation of the material. The uranium content and isotopic composition in the HEU material were determined by both thermal ionization mass spectrometry (TIMS) and ICP-MS. The ²³⁴U concentrations in the sample measured by TIMS and ICP-MS are (94.97 ± 0.09) and $(95.23 \pm 0.20) \mu\text{g g}^{-1}$, respectively, and they are in good agreement. Based on the ²³⁴U concentration and Eq. (1), the ²³⁰Th content in the HEU sample can be derived if the elapsed time since the separation is known. For the calculation the weighted average of the TIMS and ICP-MS ²³⁴U concentrations were used $[(95.01 \pm 0.08) \mu\text{g g}^{-1}]$, whereas for the half-lives of ²³⁴U and ²³⁰Th the recommended values by the Decay Data Evaluation Project (DDEP) were used (245500 ± 600 and 75380 ± 300 years, respectively). Note that much lower (approximately one-fifth) uncertainties for the half-lives have already been reported for these nuclides [13], although the DDEP values were chosen for traceability reasons. The ²³⁰Th concentration in the HEU sample added to the ²²⁹Th standard is $(6.653 \pm 0.017) \times 10^{-11} \text{ g } (^{230}\text{Th})$ per gram solution at the time of separation (June 14, 2013, 11:30 a.m.), which was performed 696.0 ± 0.1 days after the preparation of the material. The uncertainty of the elapsed time is calculated taking into account the length of separation (both the preparation of the purified HEU and the separation for the ²²⁹Th half-life measurement).

All standards used in the study were kept under weight control to correct for potential evaporation losses until use. All spiking and sample measurements were performed gravimetrically, and all weights and weight differences were higher than 0.5 g to minimize weighing uncertainties.

III. INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRIC MEASUREMENTS

The mass spectrometric analysis of the thorium samples was carried out by using a double-focusing magnetic sector ICP-MS equipped with a single electron multiplier (ELEMENT2, Thermo Electron Corp., Bremen, Germany). All measurements were carried out in low-resolution mode ($R = 300$) by using a low-flow microconcentric nebulizer operated in a self-aspirating mode (flow rate was approximately $100 \mu\text{l min}^{-1}$) in combination with a Teflon Scott-type spray chamber. Optimized operating parameters are summarized in Table I. Prior to the analysis of the samples the instrument was tuned by using a 1-ng g^{-1} multielement solution (Alfa Aesar, Karlsruhe, Germany). The optimization was carried out with respect to maximum uranium sensitivity and low UO^+/U^+ ratio. The sensitivity was approximately 1.0×10^6 counts/s for 1-ng g^{-1} ²³⁸U, UH^+/U^+ and UO^+/U^+ ratios were less than 7×10^{-5} and 2×10^{-2} , respectively. The dead time of the detector was measured prior to the measurement. For the measurements all signals were measured in counting mode to avoid the use of the conversion factor between the analog and the digital modes. The mass bias was estimated by the measurement of U-010 standard reference material, measured after three samples. The obtained bracketing values of U-010 were used to calculate the mass bias. The mass bias correction was approximately -0.027% per mass unit. Among the three samples an IRMM-185 standard was used to check the accuracy. The replicate $n(^{235}\text{U})/n(^{238}\text{U})$ measurement of

TABLE I. Optimized ELEMENT ICP-MS operating conditions and data-acquisition parameters.

Measurement parameters	
Forward power (W)	1350
Cooling gas flow rate (l min^{-1})	16
Auxiliary gas flow rate (l min^{-1})	0.91
Nebulizer gas flow rate (l min^{-1})	1.305
Solution uptake rate ($\mu\text{l min}^{-1}$)	~ 100
Data-acquisition parameters	
Resolution	300 (low)
Runs and passes	3×25
Mass window (%)	Th analysis: 25 U analysis: 25
Samples per peak	Th analysis: 50 (^{229}Th , ^{230}Th), 25 (^{232}Th) U analysis: 100 (^{233}U , ^{234}U , ^{235}U , ^{236}U , ^{238}U)
Sampling time (s)	Th analysis: 0.080 (^{229}Th), 0.110 (^{230}Th), 0.025 (^{232}Th) U analysis: 0.100 (^{233}U , ^{234}U , ^{236}U), 0.030 (^{235}U), 0.010 (^{238}U)
Search window (%)	Th analysis: 15 U analysis: 20
Integration window (%)	Th analysis: 25 U analysis: 25
Integration type	Average
Scan type	E scan

IRMM-185 gave $0.020\,038 \pm 0.000\,071$ ($n = 13$), which is in agreement with the certified value of $0.020\,0552 \pm 0.000\,0030$. The accuracy was also checked by the measurement of IRMM-035 thorium isotope reference material. The procedure was identical to that of uranium with the exception that the abundance sensitivity at $m/z = 2$ of the instrument, which is crucial for the measurement of the low $^{230}\text{Th}/^{232}\text{Th}$ ratio, was measured each day by scanning the $m/z = 233\text{--}240$ region by using a natural uranium sample with ^{236}U abundance less than 10^{-9} . The low concentration level of ^{236}U in the natural uranium sample was verified by TIMS with a modified total evaporation [14]. The measured abundance sensitivity of the ICP-MS instrument used was 1.6×10^{-6} at $m/z = 2$. The replicate $n(^{230}\text{Th})/n(^{232}\text{Th})$ measurement of the IRMM-035 sample gave $(1.13 \pm 0.07) \times 10^{-5}$, which is in agreement with the certified value of $(1.1481 \pm 0.0039) \times 10^{-5}$. Note that the $n(^{230}\text{Th})/n(^{232}\text{Th})$ ratios in the analyzed samples are far higher than in this standard. The absolute detection limit of ^{230}Th is about 20 fg.

IV. DATA EVALUATION

All uncertainties in the present paper are given as combined standard uncertainties. The half-life calculation is based on the relationship between the ^{229}Th activity and the amount content [Eq. (2)],

$$T_{1/2} = \frac{\ln(2)N}{A}, \quad (2)$$

where N is the amount of ^{229}Th in the sample and A is the activity in the same sample aliquot. The half-life is obtained in seconds, and it is converted to years by using the fact that $31\,556\,925$ s is equivalent to one tropical year. Concentrations of isotopes of interest necessary for the half-life calculations were experimentally determined as a function of $^{229}\text{Th}/^{230}\text{Th}$ or $^{229}\text{Th}/^{232}\text{Th}$ ratios according to the isotope dilution method. All raw data were corrected by taking into account instrumental mass bias that uses linear correction [15]. As the concentrations of the ^{232}Th and ^{233}U were certified as

mass fractions (i.e., gram analyte per gram standard solution), the amount content was calculated by using the atomic weights of ^{232}Th and ^{233}U [232.038 055(21) and 233.039 6352(29), respectively] [16]. The overall uncertainty of the results was calculated by taking into account the uncertainty of the weight measurements, spike and standard concentrations, measured isotope ratios, relative atomic masses, half-lives, and elapsed times according to the Bureau International des Poids et Mesures (BIPM) guide [17]. The calculations were carried out by the use of the commercially available software GUM WORKBENCH [18]. All uncertainties in the present paper are given as combined standard uncertainties if not indicated otherwise.

V. RESULTS AND DISCUSSION

A. Method A: ^{229}Th half-life measurement by isotope dilution with ^{232}Th

The methodology applied for the ^{229}Th half-life measurement against the natural ^{232}Th standard is similar to that used by Goldstein *et al.* [8]. Both the natural Th standard used for spiking and the ^{229}Th are in liquid form, and they were mixed gravimetrically in precleaned polyethylene vials to yield solutions with an approximate total thorium concentration of 40 ng g^{-1} . The $^{229}\text{Th}/^{232}\text{Th}$ ratio is approximately 1:40. For the gravimetric mixing the minimum sample size was 0.5 g to decrease the uncertainties due to weighings. For the dilutions 4% (by weight) HNO_3 solution was used. Note that the ^{232}Th and the ^{229}Th standards have similar chemical compositions (both are supplied in about 5% HNO_3 solution). The mixed samples were thoroughly homogenized and were heated to 50°C for 48 h to ensure isotopic equilibrium. Before measurements the sample aliquots were diluted to about 1.5 ng g^{-1} ^{232}Th concentration. The gravimetric mixing was performed in three replicates, and each replicate was analyzed twice on different days.

The amount content of the diluted SRM 4328C ^{229}Th radioactivity standard with an activity concentration of $(3.628 \pm 0.013)\text{ Bq g}^{-1}$ was measured to be

TABLE II. Uncertainty budget of Method A.

Uncertainty component ^a	Relative standard uncertainty (%)	Sensitivity factor	Uncertainty component (%)
Activity of SRM 4328C standard	0.30	263	78.9
Concentration of natural thorium standard	0.26	13.1	3.4
Mass bias	0.17	8.4	1.4
²²⁹ Th/ ²³² Th ratio of the mixed sample	0.54	27.9	15.1
Gravimetric mixing of diluted ²²⁹ Th and natural thorium	0.023	17.8	0.4
IRMM-010 certified ²³⁵ U/ ²³⁸ U ratio	0.05	2.5	0.12
SRM 4328C gravimetric dilution	0.028	14.3	0.4

^aOther components included in the model, with an uncertainty component below 0.1%: elapsed time and half-life for ²²⁹Th decay correction, atomic weights of ²²⁹Th, ²³²Th, ²³⁵U, and ²³⁸U, abundance sensitivity, dead time, ²²⁹Th/²³²Th ratio in the natural Th, ²²⁹Th/²³²Th ratio in the SRM 4328C, and gravimetric dilution of the natural thorium standard.

$(1.3030 \pm 0.0024) \times 10^{12} \text{ g}^{-1}$ [equivalent to $(2.1637 \pm 0.0040) \times 10^{-12} \text{ mol } (^{229}\text{Th}) \text{ g}^{-1}$ solution]. The derived ²²⁹Th half-life from these values is 7889 ± 32 years (uncertainty is expressed as combined standard uncertainty). The uncertainty budget of the calculation (Method A) is shown in Table II. The most important contributors to the half-life uncertainty are the activity concentration of the SRM 4328C standard, the measured ²²⁹Th/²³²Th ratio by ICP-MS, and the concentration of the natural Th standard.

B. Method B: ²²⁹Th half-life measurement by isotope dilution with ²³⁰Th

In Method B, a known amount of the HEU standard was mixed with an aliquot of the SRM 4328C standard gravimetrically. By knowing the ²³⁴U amount and the elapsed time, the ²³⁰Th present in the sample can be calculated, which enables the quantification of the ²²⁹Th amount content in the NIST standard after the measurement of the ²²⁹Th/²³²Th amount ratio. The solid-form HEU material was dissolved immediately before analysis in concentrated nitric acid. For the dissolution, 3-ml ultrapure nitric acid was added to the sample in a PFA container, the cap was screwed on, and the vial was placed on a hot plate. Dissolution was performed at about 120 °C for 1 h. After cooling, accurately weighed 500 μl aliquots of this stock solution (which correspond to about 5-mg uranium) were used for each thorium and uranium

measurement. The thorium and uranium content and isotopic composition determinations are described in detail elsewhere [19]. In short, the ²³⁴U content of the sample was determined by using isotope dilution ICP-MS and TIMS measurements by applying ²³³U tracer after successive gravimetric dilution of the sample aliquot. For the ²²⁹Th analysis, an aliquot of the HEU stock solution was gravimetrically mixed with the SRM 4328C standard in a PFA vial and was placed on a hot plate for 1 h to ensure isotopic equilibrium. For the gravimetric mixing the minimum sample size was 0.5 g to decrease the uncertainties due to weighing. Due to the lower thorium content, the ²²⁹Th/²³⁰Th ratio was determined afterwards by using extraction chromatographic separation with a TEVATM column. After cooling, the mixed HEU stock/²²⁹Th aliquot was diluted to threefold with high-purity water to adjust the HNO₃ concentration to about 5M. To follow loading the aliquot on the TEVATM column, uranium was removed by washing five times with 2-ml 2M HNO₃. The thorium content of the sample retained on the resin was stripped with 0.8 ml of 0.02M HF/0.02M HNO₃ in a precleaned polyethylene vial. Before the ICP-MS measurement the sample was diluted to about 1.2 ml with 5% ultrapure HNO₃. The analysis was performed in three replicates that involved the gravimetric mixing, and method blanks prepared parallel to the samples were used for background correction.

The separation of the HEU aliquot mixed with the ²²⁹Th standard took place on June 14, 2013 at 11:30 a.m., which

TABLE III. Uncertainty budget of Method B.

Uncertainty component ^a	Relative standard uncertainty (%)	Sensitivity factor	Uncertainty component (%)
Activity of SRM 4328C standard	0.30	220	66.1
Mass bias	0.11	50.0	5.5
²²⁹ Th/ ²³⁰ Th ratio of the mixed sample	0.62	32.7	20.3
Gravimetric mixing of diluted ²²⁹ Th and natural thorium	0.023	20.4	0.5
IRMM-010 certified ²³⁵ U/ ²³⁸ U ratio	0.05	4.0	0.2
SRM 4328C gravimetric dilution	0.028	14.3	0.4
²³⁴ U half-life	0.24	25.2	6.1
Elapsed time between HEU preparation and separation for half-life measurement	0.014	1.5	0.02
²³⁴ U amount concentration in HEU	0.083	8.5	0.7

^aOther components included in the model with an uncertainty component below 0.1%: elapsed time and half-life for ²²⁹Th decay correction, atomic weights of ²²⁹Th, ²³⁰Th, ²³⁵U and ²³⁸U, abundance sensitivity, dead time, ²²⁹Th/²³⁰Th ratio in the HEU, ²²⁹Th/²³⁰Th ratio in the SRM 4328C, Avogadro number, half-life of ²³⁰Th, and gravimetric dilution of the HEU sample.

corresponds to 696.0 ± 0.1 days after the preparation of the HEU material. The elapsed time is calculated as the difference of the uranium-thorium separation of the HEU stock/ ^{229}Th mixture (not the time of the spiking but when the $^{230}\text{Th}/^{234}\text{U}$ daughter-parent is separated) and the preparation of the HEU material. By using the ^{234}U results and Eq. (1), the ^{230}Th tracer concentration in the HEU sample added to the ^{229}Th standard is $(6.653 \pm 0.017) \times 10^{-11}$ g (^{230}Th) per gram solution. The ICP-MS measurements were carried out within a week after the preparation of the samples to avoid significant ingrowth of ^{230}Th from the residual HEU after separation, although the ^{230}Th ingrowth is negligible as the U/Th separation factor is typically higher than 10^3 . The $n(^{229}\text{Th})/n(^{230}\text{Th})$ amount ratio was measured from the separated samples. Details for the ICP-MS analysis are described in detail elsewhere [19].

The amount content of the diluted SRM 4328C ^{229}Th radioactivity standard with an activity concentration of (3.628 ± 0.013) Bq g $^{-1}$ was measured to be $(1.3134 \pm 0.0034) \times 10^{12}$ g $^{-1}$ [equivalent to $(2.1810 \pm 0.0057) \times 10^{-12}$ mol (^{229}Th) g $^{-1}$ solution]. The derived ^{229}Th half-life from these values is 7952 ± 36 years (uncertainty is expressed as combined standard uncertainty). The uncertainty budget of the calculation (Method B) is shown in Table III. The most important contributors to the half-life uncertainty, similar to Method A, are the activity concentration of the SRM 4328C standard, the measured $^{229}\text{Th}/^{232}\text{Th}$ ratio by ICP-MS, and the concentration of the natural Th standard.

C. Calculation of the combined half-life value

The half-life values obtained by Methods A and B are in good agreement, and no significant deviation is observed even at a 1σ confidence level. By using the calculated half-lives by Methods A and B, a weighted average of 7917 years can be obtained for the ^{229}Th half-life that uses the combined uncertainties of the individual values as weights. The combined standard uncertainty, calculated by uncertainty propagation, is equal to 24 years. This combined standard uncertainty corresponds to an expanded uncertainty of 48 years at a coverage factor of $k = 2$ (approximately 95% level of

confidence). The obtained ^{229}Th half-life is in agreement with the values from Goldstein *et al.* [8] and Kikunaga *et al.* [11] and is significantly different than that of Hagemann *et al.*, which verifies their possibly biased value (Fig. 1). The achieved relative combined standard uncertainty of 0.30% is slightly lower than that achieved by Kikunaga *et al.* (0.35%) [11].

VI. CONCLUSIONS

In the present paper, the half-life of ^{229}Th (ground state) was remeasured by using two different approaches for quantification. The obtained half-life and its associated combined standard uncertainty are 7917 ± 24 years. Both methods are fully traceable to SI, and the results are calculated in accordance with the BIPM methodology.

By applying the evaluation rules of the DDEP, a recommended ^{229}Th half-life value, which uses the previously reported results can also be calculated [20]. For the calculation the values reported by Hagemann *et al.* [7], Goldstein *et al.* [8], Kikunaga *et al.* [11], and the combined result of the present study (7340 ± 80 , 7880 ± 60 , 7932 ± 28 , and 7917 ± 24 years, respectively) were applied, although the Hagemann *et al.* value appears to be a clear outlier. By using the DDEP evaluation rules, the half-life reported by Hagemann *et al.* cannot be clearly identified as an outlier based on Chavenet's criterion as the probability to obtain such values is 53.9%, slightly higher than the critical probability of 50% for the outlier rejection. However, by using the four reported half-lives, the internal variance (i.e., the expected uncertainty of the mean, based on the individual *a priori* variances) and the external variance (includes the scatter of the data and is based on the amount by each individual half-life value deviates from the mean) can also be calculated. A measure of the consistency of the data is the ratio of external and internal uncertainty [20]. The calculated ratio is 4.1, significantly higher than a unit, which implies that either at least one of the data has an underestimated uncertainty or an outlier is present among the data. By assuming the Hagemann *et al.* value as an outlier, we can obtain a more consistent dataset with a weighted average half-life of 7920 years with internal and external uncertainties of 17.3 and 20.8 years, respectively. The calculated ratio of the external and internal uncertainties is 1.2, close to the unit that indicates the consistency of the data. The $\chi^2/(n - 1)$ (reduced χ^2) can be calculated for the dataset as the square of the ratio of the calculated internal and external uncertainties as a practical test for discrepant data. The obtained $\chi^2/(n - 1)$ is 1.45, significantly smaller than the critical value of 4.6 for $n = 3$ (degree of freedom is 2). Therefore, based on the DDEP methodology, the recommended ^{229}Th half-life obtained is 7920 ± 17 years where the uncertainty indicated is the combined standard uncertainty, equal to the internal uncertainty. The often used, equivalent uncertainty expressed as expanded uncertainty, is 34 years at a coverage factor of $k = 2$ (approximately 95% level of confidence). The three half-life results used for the calculation were reported by different authors by using different analytical techniques, therefore it can be considered as a reliable estimate for the ^{229}Th half-life.

FIG. 1. The obtained half-life values in comparison with the reported values. All uncertainties are expressed at a confidence level of 2σ or as expanded uncertainties with a coverage factor of $k = 2$.

- [1] F. Hagemann, L. I. Katzin, M. H. Studier, A. Ghiorso, and G. T. Seaborg, *Phys. Rev.* **72**, 252 (1947).
- [2] A. C. English, T. E. Cranshaw, P. Demers, J. A. Harvey, E. P. Hincks, J. V. Jelley, and A. N. May, *Phys. Rev.* **72**, 253 (1947).
- [3] A. Morgenstern, F. Bruchertseifer, and C. Apostolidis, *Curr. Radiopharm.* **4**, 295 (2011).
- [4] C. Apostolidis, R. Molinet, G. Rasmussen, and A. Mogenstern, *Anal. Chem.* **77**, 6288 (2005).
- [5] Z. Varga, A. Nicholl, M. Wallenius, and K. Mayer, *Anal. Chim. Acta* **718**, 25 (2012).
- [6] R. W. Williams and A. M. Gaffney, *Proc. Radiochim. Acta* **1**, 31 (2011).
- [7] F. Hagemann, L. I. Katzin, M. H. Studier, G. T. Seaborg, and A. Ghiorso, *Phys. Rev.* **79**, 435 (1950).
- [8] S. J. Goldstein, M. T. Murrell, and R. W. Williams, *Phys. Rev. C* **40**, 2793 (1989).
- [9] E. Browne and J. K. Tuli, *Nucl. Data Sheets* **109**, 2657 (2008).
- [10] Evaluated Nuclear Structure Data File (ENSDF), <http://www.nndc.bnl.gov/ensdf/>
- [11] H. Kikunaga, T. Suzuki, M. Nomura, T. Mitsugashira, and A. Shinohara, *Phys. Rev. C* **84**, 014316 (2011).
- [12] R. Fitzgerald, R. Collé, L. Laureano-Pérez, L. Pibida, M. M. Hammond, S. Nour, and B. E. Zimmerman, *Appl. Radiat. Isot.* **68**, 1303 (2010).
- [13] H. Cheng, H. L. Edwards, J. Hoff, C. D. Gallup, D. A. Richards, and Y. Asmerom, *Chem. Geol.* **169**, 17 (2000).
- [14] S. Richter and S. A. Goldberg, *Int. J. Mass Spectrom.* **229**, 181 (2003).
- [15] K. G. Heumann, S. M. Gallus, G. Rädlinger, and J. Vogl, *J. Anal. At. Spectrom.* **13**, 1001 (1998).
- [16] G. Audi, A. H. Wapstra, and C. Thibault, *Nucl. Phys. A* **729**, 337 (2003).
- [17] Joint Committee for Guides in Metrology, *Evaluation of Measurement Data—Guide to the Expression of Uncertainty in Measurement*, 1st ed. (Joint Committee for Guides in Metrology, Sèvres, France, 2008).
- [18] GUM WORKBENCH PRO, Version 2.4.1.375 (Metrodata GmbH, Weil am Rhein, Germany, 2009).
- [19] Z. Varga and G. Surányi, *Anal. Chim. Acta* **599**, 16 (2007).
- [20] Decay Data Evaluation Project, <http://www.nucleide.org/DDEP.htm>