

# A new evaluation of the half-life of $^{241}\text{Pu}$ †

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A new evaluation of the half-life of  $^{241}\text{Pu}$  has been made by combining previous results from earlier methods and techniques with further measurements of the isotopic ratio of an initially enriched  $^{241}\text{Pu}$  material. A correction to the previous published value has had to be made. The half-life of  $^{241}\text{Pu}$  is an important parameter for the measurement and control of amounts of plutonium as the decay of this isotope is usually the largest contributor to the uncertainty of estimations of the amount of total plutonium after storage. The value of the half-life together with its associated uncertainty needs to be known with a high degree of confidence. However, the half-life value of *ca.* 14.3 a (years) means the decay of the isotope is difficult to measure over one or more half-lives using a single method or instrument. The method employed in this evaluation is designed to conform to the Guide to the Expression of Uncertainty in Measurements (GUM) (International Organisation for Standardisation, Guide to the Expression of Uncertainty in Measurement, ©ISO, ISBN 92-67-10188-9, Geneva, Switzerland, 1993)<sup>1</sup> and care was taken to ensure that all measured values over the 30 year measurement time-span have been included. The value recommended from this set of measurements is  $14.325 \text{ a} \pm 0.024 \text{ a}$  ( $k = 2$ ). This agrees with the value from the first measurements of the same material in 1983 of  $14.33 \text{ a} \pm 0.02 \text{ a}$  (P. De Bièvre, P. Gallet and M. Wertz, *Int. J. Mass Spectrom., Ion Phys.*, **51**, 1983, 111).<sup>2</sup>

## Introduction

Because  $^{241}\text{Pu}$  has the shortest half-life of the plutonium isotopes commonly encountered in material recovered from irradiated nuclear fuel, it affects the measurement of the amount of plutonium for accountability purposes to a much greater extent than other isotopes. Corrections for the decay of this isotope make a considerable contribution to the overall uncertainty in the total amount of plutonium, especially after storage for several years.

Many laboratories had previously measured the half-life of this isotope; an overview was given in a previous paper.<sup>2</sup> The values lay between 13.0 years to 15.0 years. This spread was considered unacceptable and in 1976, in an attempt to measure the half-life using the best technique available, a series of isotope ratio measurements on a plutonium sample enriched in  $^{241}\text{Pu}$  was started at the reference materials institute of the European Commission, originally called the 'Central Bureau for Nuclear

Measurements, CBNM' which is now the 'Institute for Reference Materials and Measurements, IRMM'.<sup>2</sup>

The value of these measurements lay not only in being able to apply the power of thermal ionisation mass-spectrometry (TIMS) to achieve very accurate measurements of the isotope ratios, but also in the design of the measurements for which a ratio-of-ratios technique was used to eliminate first order mass bias effects.

The results of measurements carried out over the first 6 years and the entire period of 20 years to 1996 were published previously.<sup>2,3</sup> The measurements from the 20 year period gave an excellent fit to a decay with a half-life of  $14.290 \text{ a} \pm 0.006 \text{ a}$  (years), a value lower than previously measured in 1982 but also with an uncertainty lower than previously achieved.

The original material has been carefully stored at IRMM. It has an enrichment of approximately 74% in  $^{241}\text{Pu}$  at the present time and is available for further measurements. The initial measurements were made (in 1976 and later) using a single-collector TIMS and the measurements made in 1996 were also made using this technique, although on a different instrument. Nowadays, measurements of the isotope ratios of Pu are preferentially carried out on mass spectrometers equipped with multi-collector Faraday detector arrays, using the technique of total evaporation.<sup>4-6</sup> In this technique a small sized sample (typically ranging from a few nanograms up to 100 ng Pu) is evaporated to exhaustion. This method has the general advantage of considerably reducing the influence of mass-fractionation ('mass bias') effects on the isotope ratio measurements as the signal is integrated over the entire ablation period of the sample.

The length of time needed to measure the half-life poses problems for any laboratory or group of laboratories: methods and instrumentation change every few years. However the decay

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**Table 1** Isotopic composition of the enriched  $^{241}\text{Pu}$  material

Isotope	100 × amount fraction		
	1976-Jan-15	1996-Sep-15	2006-Jul-01
$^{238}\text{Pu}$	0.01	0.02	0.03
$^{239}\text{Pu}$	1.15	2.79	4.04
$^{240}\text{Pu}$	4.05	9.79	14.21
$^{241}\text{Pu}$	92.73	82.41	74.47
$^{242}\text{Pu}$	2.06	4.99	7.25

should be followed for longer than a single half-life to ensure no isobaric interferences are present in the measurements. This considerably enhances the confidence we can place in the value of the derived half-life. The experimental set-up used in this work approaches this problem by measuring a single material that was initially highly enriched in  $^{241}\text{Pu}$ . The isotopic ratios of the material could then be measured over a time period corresponding to more than a single half-life period (two half-lives are covered in the work reported here). The complete isotopic composition of the material is given in Table 1 at the start of the experiment in 1976, in 1996 when the second set of measurements were carried out and in 2006 for the measurements reported here. The methodology employed (ratio-of-ratios isotopic measurements) is the least sensitive to changes in measurement methods and instruments and allows a consistent long-term approach to measuring the decay of the isotope.

Over the last 10 years the Guide to the Expression of Uncertainty in Measurements ('GUM') published by BIPM/ISO<sup>1</sup> has had an increasing impact on the calculation of uncertainties in the nuclear measurement field. Improvements in mass spectrometry and a time difference of about ten years since the last evaluation made it feasible to re-measure the isotope ratios of the material originally used in 1976 with the latest measurement techniques and to use the opportunity to apply the concepts of the GUM as rigorously as possible to arrive at a final value of the half-life and associated uncertainty for the use by the nuclear measurement community for calculations of plutonium isotopic composition.

During the course of the present measurements, the calculations in<sup>3</sup> were found to have inadvertently included a double correction for the decay of the isotopes  $^{240}\text{Pu}$  and  $^{239}\text{Pu}$ , which lowered the published value of the half-life of  $^{241}\text{Pu}$ . The opportunity was therefore taken to calculate a new value of the half-life and at the same time to critically look at its uncertainty estimation and re-evaluate this as well. It was recognised that the goal was an uncertainty that would be "fit for intended use", not necessarily that having the lowest value.

The present measurements therefore allowed us to extend the length of decay over which the isotope measurements have been made. The new measurement technique of total evaporation can also be expected to reduce the uncertainty associated with the ratio measurements themselves, although this does not have a great effect on the final uncertainty of the calculated half-life. The difference between this work and that done previously is the rigorous application of the GUM in calculating the final uncertainty. Therefore, laboratories calculating plutonium decays can include the value arrived at in this work as a standard 'type B' GUM-defined uncertainty.

## New measurements

The original material was plutonium oxide with a 92.7% abundance of  $^{241}\text{Pu}$  (1976) supplied from Oak Ridge. The material was dissolved in nitric acid and has been carefully stored in a glass bottle behind lead shielding because of the heavy in-growth of  $^{241}\text{Am}$ .

It is critical in these measurements to remove  $^{241}\text{Am}$  completely and measure the isotope ratios within a short time after separation to ensure there is no isobaric interference at mass 241. The separation has been described previously.<sup>3</sup> An aliquot of the original solution is evaporated to dryness and taken up in 8M nitric acid; subsequently two separations on anion-exchange columns are carried out. The second separation is made so that measurement of the isotope ratios can be done within 48 hours after separation. Under these conditions there is no significant interference from the small amount of  $^{241}\text{Am}$  that grows in over this short time period. This point was previously investigated in detail<sup>3</sup> and any possible interference from  $^{241}\text{Am}$  was shown to be negligible in the half-life calculations.

Up to 1996, 12 sets of measurements had been made and these constituted the basis of the previously reported half-life over a time span of measurements of 20 years. Three new sets of measurements were carried out in 2006, two of which were performed on a multi-collector thermal-ionisation mass spectrometer and the 15 independent sets of measurements have now been combined in the present report. All these measurements were made using a ratio-of-ratios approach. The advantages of this were shown previously<sup>2</sup> to eliminate mass-fractionation effects during the measurements and to render the process independent of instrumentation or measurement technique. This made it particularly suitable for the present case where measurements using a multi-collector mass-spectrometer are combined with the earlier, single-collector instrument measurements.

## Principle of the measurements

Following the original design a ratio-of-ratios is constructed: the intensity ratios at masses 241/240 and 240/239 are measured and the ratio-of-ratios calculated:

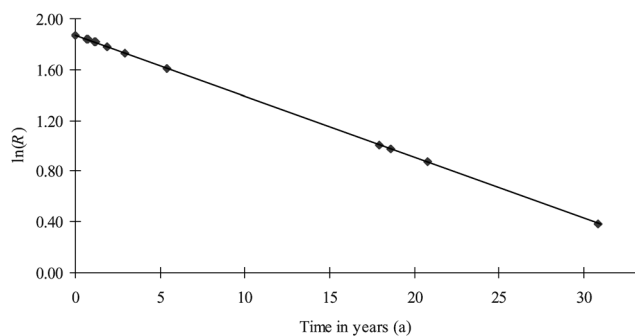
$$R = \frac{\frac{n(^{241}\text{Pu})}{n(^{240}\text{Pu})}}{\frac{n(^{240}\text{Pu})}{n(^{239}\text{Pu})}} \quad (1)$$

The mass fractionation in each measurement is essentially eliminated when applying this equation, an important factor when comparing measurements carried out on different instruments with different mass responses.

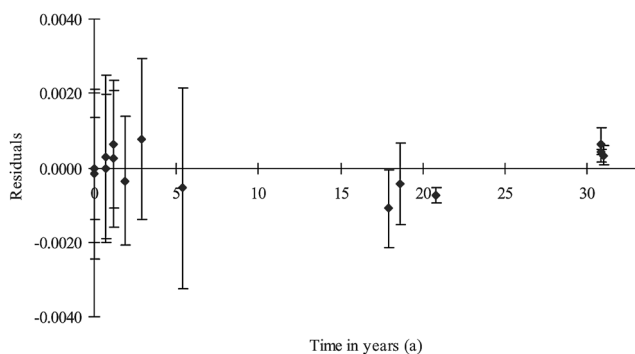
A plot of the natural logarithm of the ratio-of-ratios with time for all 15 measurements is shown in Fig. 1 and the residuals after an unweighted regression fit in Fig. 2.

## Measurement by thermal ionisation mass-spectrometry

The first points shown in Fig. 1, with decay times of 5 years or less, are the means of group measurements made using a Tele-dyne thermal-ionisation mass spectrometer.<sup>2</sup> From each separation, six individual filaments were prepared and measured.



**Fig. 1** Natural log of the isotopic ratio-of-ratios  $R = (n(^{241}\text{Pu})/n(^{240}\text{Pu}))/ (n(^{240}\text{Pu})/n(^{239}\text{Pu}))$  vs. decay time in years. The three measured points around 20 years decay were used in the consistency test of the calculated half-life. The uncertainties of the points are covered by the size of the plotted squares.



**Fig. 2** Residuals of the measured data points fitted with a unweighted least squares. The dispersion indicated for each point is  $2 \times$  standard deviation of repeated measurements and does not include any contribution from the fitting process.

The three main isotope intensities were measured:  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$ . An isotope reference material, IRMM-290 with a certified  $n(^{242}\text{Pu})/n(^{239}\text{Pu})$  ratio, was measured at the same time to allow a mass-fractionation factor for each measurement set to be calculated.

The value of the mass-fractionation factor is a valuable measure of the behaviour of the mass-spectrometer. Although not applied as a correction to the measured ratios in eqn. 1, single ratios *e.g.*  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$  must be corrected for this effect and it can be a major contributor to the overall uncertainty of the measurements of the values of single isotopic ratios. The isotopic ratio  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$ , corrected for mass-fractionation as described above has been used as an independent check of the consistency of the measurements over the complete time period and is described more fully below.

Between 1993 and 1996, three further separations were made from the source material and each time a group of 6 filaments was prepared and measured on a Finnigan MAT 261 single Faraday collector thermal-ionisation mass-spectrometer.<sup>3</sup>

In 2006, measurements were made using a Finnigan MAT 262 TIMS equipped with a Faraday multi-collector. (One measurement using the technique described in<sup>3</sup> on the MAT 261 was also carried out for comparison between the techniques). A total evaporation technique was applied.<sup>6</sup> The mass fractionation is

exceedingly small using this technique and is approximated, as for the previous methods, by the measurement of an isotope reference material, IRMM-290. A single loading and measurement sequence included 7 filaments loaded with Pu from the enriched  $^{241}\text{Pu}$  material after separation and an equal or greater number loaded with IRMM-290.

### Control of the measurements using the $n(^{240}\text{Pu})/n(^{239}\text{Pu})$ ratio

The ratio of the measured  $^{240}\text{Pu}$  and  $^{239}\text{Pu}$  intensities can be used as a check of the consistency of the measurements over the 30 year time period. The  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  intensities are corrected for mass fractionation and for radioactive decay using the recommended values of the half-lives for  $^{239}\text{Pu}$  ( $24110 \text{ a} \pm 60 \text{ a}$ ) and  $^{240}\text{Pu}$  ( $6563 \text{ a} \pm 14 \text{ a}$ ).<sup>7</sup>

The corrected  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$  ratio is plotted against time for the 15 groups of measurements in Fig. 3.

The points show an overall consistency of measurement values over the long time span. The final two points, measured by the total evaporation technique, have a smaller associated uncertainty than the previous measurements.

### Basis of the method to calculate the half-life

Two ratios are measured:  $n(^{241}\text{Pu})/n(^{240}\text{Pu})$  and  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$  and the natural logarithm of the ratio of these is plotted with time (eqn. 1). The critical value to be determined is the slope of this ratio of ratios with time.

The slope  $p_2$  (following previously published papers<sup>2,3</sup>) and the decay constants of the 3 isotopes  $\lambda_{239}$ ,  $\lambda_{240}$  and  $\lambda_{241}$  are related:

$$p_2 = \lambda_{241} + \lambda_{239} - 2\lambda_{240} \quad (2)$$

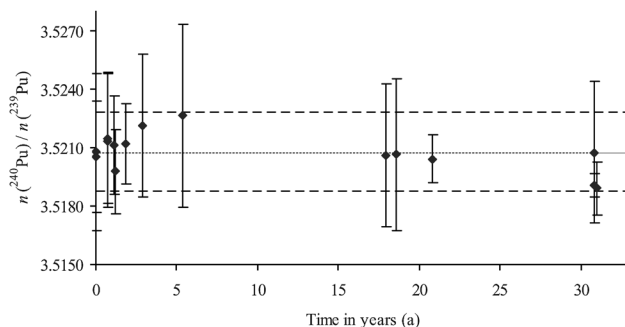
and therefore

$$\lambda_{241} = p_2 + 2\lambda_{240} - \lambda_{239} \quad (3)$$

The half-life  $\tau_{1/2}(^{241}\text{Pu})$  is

$$\tau_{1/2}(^{241}\text{Pu}) = \frac{\ln(2)}{\lambda_{241}} \quad (4)$$

In the previous paper,<sup>3</sup> the value of the slope was found by carefully applying a conventional regression analysis and considering two extra terms, one for possible influence of  $^{241}\text{Am}$



**Fig. 3** Measured ratio  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$  corrected for mass-fractionation and decay of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ . The dotted lines are  $\pm 2s$  of the non-weighted mean of all points.

and another to introduce a factor for the change in techniques, mass-spectrometer operators and chemical methods between the various measurements. Both of these factors were shown to be non-significant.

Although the value from the regression is assumed to be reliable, there are more doubts about the validity of the uncertainty associated with this value, which arises purely from the fit of the points to the straight line regression. The GUM prescribes that the uncertainties of the measurements should be propagated into the uncertainty of the final result. We have doubts that a regression analysis is the right tool to do this. Other half-life investigations have pointed out the dangers of accepting the uncertainty from least-squares methods, particularly in regard to measurements of half-lives.<sup>8,9</sup> These considerations have considerable consequences if applied to all cases where least squares analysis is applied as a tool and where the uncertainties derived need to be GUM compatible.

Because of the limitations we have developed a method that overcomes the inconsistencies and which results in a value and, importantly, an associated uncertainty for the half-life of <sup>241</sup>Pu that is fit-for-the-intended-use—in this case for the calculation of plutonium inventories where the effect of the decay of <sup>241</sup>Pu is significant. The method is based on defining the starting point and the end point of the line and calculating the slope of the line between these two points. In this method the intermediate measured values are not used in the calculation of the value of the slope but are used to check for consistency. The basis for the consistency check is given in Appendix 1. All time differences used and applied in this method were calculated in days (86400 seconds per day) and converted to years using the factor 365.24219878.<sup>10</sup>

The method to calculate the half-life is as follows:

1 A set of measured points is selected to represent a starting point of the decays. The measured values of this first set of ratios are then individually corrected for the decay of <sup>241</sup>Pu to a single, arbitrary point in time which is then the starting point for the slope calculation. These decay corrections are small as the points are close together in time and therefore the uncertainty in the assumed <sup>241</sup>Pu half-life for this correction contributes very little to the uncertainty of the starting point. A consistency check is applied to this set of points and an extra uncertainty contribution is added to each measured value as necessary.<sup>11</sup>

2 The last group of measured values is treated in the same way and adjusted for <sup>241</sup>Pu decay to a time marking the end of the slope.

3 The slope,  $p_2$ , is calculated simply as the difference of the logarithm of the measured ratios divided by the difference in time between the measurements. The uncertainties of the ratios at the start and end points are calculated from the individual measurements ('type A') plus an eventual extra type B component derived from the consistency check. The start and end times are defined values and therefore have no associated uncertainty.

4 The corrections for the half-lives of <sup>240</sup>Pu and <sup>239</sup>Pu are made following eqn. 2 and the half-life of <sup>241</sup>Pu calculated from eqns. 3 and 4.

5 Points 1 through 4 are repeated using the updated value of the <sup>241</sup>Pu half-life. Further iterations are carried out until the calculated half-life does not change.

6 All measured ratios not included in the definition of the starting or ending point are checked for consistency with the half-life

value and uncertainty. If necessary the uncertainty of the half-life is increased until the measured ratios agree with the predicted value either from the starting point plus decay corrections or analogously from the end point.

## Justification of the method

The half-life measurements discussed here are based on the knowledge that the decay follows strictly an exponential law. Under this condition we know that the difference in the natural logarithm of two ratio of ratios  $R_0$  and  $R_1$  (eqn. 1) between two points in time  $t_0$  and  $t_1$  is proportional to the time difference between the time points.

$$\ln(R_1) - \ln(R_0) \sim t_1 - t_0 \quad (5)$$

From this principle the calculation for  $p_2$  follows:

$$p_2 = \frac{\ln(R_1) - \ln(R_0)}{t_1 - t_0} \quad (6)$$

If we assume that the uncertainty of the time measurements is not significant and if we further assume that the uncertainty for the ratio of ratios measurement is almost independent of time difference, we can see from the partial derivatives that the time difference is a scaling factor for the uncertainty.

$$\frac{\partial p_2}{\partial R_1} = \frac{1}{R_1 \times (t_1 - t_0)} \quad (7)$$

$$\frac{\partial p_2}{\partial R_0} = -\frac{1}{R_0 \times (t_1 - t_0)} \quad (8)$$

This means that the uncertainty of the half-life will be decreased if the time difference is increased. As a consequence, regardless of whatever method we choose to evaluate  $p_2$ , the value and its basic uncertainty will be determined by the two points where the time difference is at maximum.

Based on these considerations we select two measurement points to calculate  $p_2$ . The points are defined as times  $t_0$  and  $t_1$  with no uncertainty.

In addition, different instruments and methods were used to measure the ratio of ratios throughout this sequence of measurements. Our approach allows us to easily group similar results together and consolidate them for the ratio of ratios at the reference time points.

To consolidate the measurements we adjust them to the reference time point by correcting for decay. The adjustment of the measurements to a reference point in time allows us to compare the different measurement results and to check if they are metrologically consistent. This approach allows us to include all knowledge which is available from the measurements close to the start and endpoints. The effect of the uncertainty of the half-life which was used in the adjustment is small because the adjustment factors are small.

The influence of the measured results close to the centre of the decay diagram in Fig. 1 (points 10, 11, 12 at a decay time around 20 years) is low because of the time difference to the starting and end points. It was decided, therefore, not to include these data points in the evaluation of the final value but to use them as part of an additional quality assurance measure. In essence the

uncertainty of the half-life was increased until the estimation of the half-life was consistent with all measurement values.

## Calculations and discussion

The measured data set consists of 15 individual points starting in 1976 and including the last measured values in 2006. Each point is a ratio-of-ratios,  $[n(^{241}\text{Pu})/n(^{240}\text{Pu})]/[n(^{240}\text{Pu})/n(^{239}\text{Pu})]$  as measured, uncorrected for any decays and is the mean of the measured ratios from a set of filaments prepared from a single separation of the plutonium isotope mixture (Table 2).

The raw data are the values of the intensities of the two mass ratios, (eqn. 1) and the standard uncertainty of each measured ratio is derived from the repeated measurements: a type A uncertainty as defined by the GUM.

The three groups of measured points can clearly be distinguished in Fig. 2 in which the residuals between the measured values and those calculated for convenience from an unweighted regression analysis are shown. (The uncertainties shown in Fig. 2 are twice the standard deviation from the repeat measurements at each point.)

### Selection of the start and end points

To define the start point we have an option to select measurements carried out relatively close to the beginning of the whole

series, in which case the normalisation for the decay of  $^{241}\text{Pu}$  will be small, or to use all the measurements done with the first Teledyne machine up to and including the value shown above at 5 years decay. The second option has the advantage that the group is then quite homogeneous from the point of view of measurement technique. With the first option the ratio of ratios at the starting point (date: 13 January 1976) has a value of  $6.5066 \pm 0.0018$ ,  $k = 2$  using the first 6 measured points and for the second option using the first 9 points a value of  $6.5073 \pm 0.0020$ ,  $k = 2$  is obtained. Both of these values are the result of 3 iterations of the half-life and in each case a sizeable contribution of more than 50% to the uncertainty of each measured point had to be added to obtain a consistent solution involving all the points, despite the relatively large standard deviations derived from individual measurements. For the purpose of this exercise, the value of  $6.5066 \pm 0.0018$ ,  $k = 2$  was chosen for the first point.

The grouping of the last 3 measurements is clear and a value for the ratio of ratios of  $1.47188 \pm 0.00027$ ,  $k = 2$ , (date: 13 November 2006) is obtained after the third iteration for the half-life.

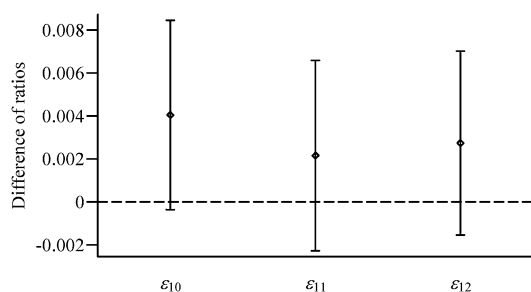
The value of the half-life calculated from the slope of the ratio-of-ratios decay before checking the remaining points for consistency is  $14.3258 \text{ a} \pm 0.0031 \text{ a}$ ,  $k = 2$ . An uncertainty budget is given in Table 3 showing that the greatest relative contributions to the uncertainty of the half-life come from the measurements defining the start point (69.2%) and the end point (30.6%).

**Table 2** Measured ratio-of-ratios  $(n(^{241}\text{Pu})/n(^{240}\text{Pu}))/(n(^{240}\text{Pu})/n(^{239}\text{Pu}))$ . The value of the start ratio-of-ratios is the mean of the first 9 measured values corrected for decay to the start date: the uncertainty includes a component for consistency. The value of the end ratio-of-ratios is the mean of points 13, 14 and 15 corrected for decay to the end date

No.	Date	Time in years (a)	$U_c (k = 2)$	Ratio-of-ratios $R$	$U_c (k = 2)$
<i>start</i>	1976-01-13	0.0		6.5066	0.0018
1	1976-01-13	0.0077	0.0090	6.5023	0.0082
2	1976-01-19	0.0233	0.0053	6.4965	0.0086
3	1976-09-27	0.7104	0.0054	6.2857	0.0085
4	1976-10-04	0.7291	0.0066	6.2820	0.0086
5	1977-03-08	1.1600	0.0121	6.1526	0.0084
6	1977-03-22	1.1988	0.0121	6.1435	0.0083
7	1977-11-23	1.8749	0.0126	5.9406	0.0024
8	1978-12-05	2.9025	0.0142	5.6599	0.0024
9	1981-06-02	5.3916	0.0036	5.0133	0.0034
10	1993-12-13	17.9216	0.0042	2.7387	0.0012
11	1994-08-25	18.6198	0.0023	2.6498	0.0012
12	1996-10-28	20.7959	0.0027	2.38517	0.00024
13	2006-11-13	30.8413	0.0025	1.47161	0.00066
14	2006-11-13	30.8402	0.0005	1.47139	0.00010
15	2007-01-16	31.0109	0.0003	1.45921	0.00042
<i>end</i>	2006-11-13			1.47188	0.00027

**Table 3** Uncertainty budget of  $^{241}\text{Pu}$  half-life  $T_{241}$  before adding uncertainty required for the consistency checking.  $t_0$  and  $t_1$  are the times at start and end of the observed decay;  $R_{1976}$  and  $R_{2006}$  the measured values of the ratio-of-ratios

Quantity	Value	Standard Uncertainty	Sensitivity Coefficient	Uncertainty Contribution	Index
$t_1$	30.8343 a				
$t_0$	0.0 a				
$R_{2006}$	1.471885	$133 \times 10^{-6}$	6.5	$870 \times 10^{-6} \text{ a}$	30.6%
$R_{1976}$	6.506619	$885 \times 10^{-6}$	-1.5	$-1.3 \times 10^{-3} \text{ a}$	69.2%
$\tau_{1/2}(^{239}\text{Pu})$	24110.0 a	30.0 a	$-350 \times 10^{-9}$	$-11 \times 10^{-6} \text{ a}$	0.0%
$\tau_{1/2}(^{240}\text{Pu})$	6563.00 a	7.00 a	$9.5 \times 10^{-6}$	$67 \times 10^{-6} \text{ a}$	0.2%
$\tau_{1/2}(^{241}\text{Pu})$	14.32580 a	$1.57 \times 10^{-3} \text{ a}$			



**Fig. 4** The differences (epsilon values) for the central three points with expanded uncertainties ( $k = 2$ ). Calculated or predicted values are based on a half-life of  $14.325 \pm 0.024$  a for  $^{241}\text{Pu}$ .

However, we include the measurements carried out in 1996 as a consistency check and as a result the uncertainty of the  $^{241}\text{Pu}$  half-life has to be considerably increased. This was done until the differences between predicted and measured values for the points numbered 10, 11 and 12 were zero within the uncertainty for each point. The difference for each of the points is shown in Fig. 4, where the expanded uncertainty,  $k = 2$ , includes the uncertainty of the half-life and the uncertainty for each measured point. The half-life and uncertainty to achieve this is  $14.325 \pm 0.024$  a,  $k = 2$ .

Because we have no reason to arbitrarily eliminate measured points, we are constrained to accept a half-life value with a considerably larger uncertainty than found by regressing all the points to determine the slope  $p_2$ . We believe, however, that this value is realistic and fit for intended use.

## Conclusions

The intention of this study was firstly to correct the previous value for the half-life of  $^{241}\text{Pu}$ <sup>3</sup> and to introduce a new method of calculating it, including new measured values and with a method we believe is fully compatible with the GUM. Uncertainty contributions were rigorously separated into type ‘A’—arising from the repeated measurements of the ratios of a single purified material—and type ‘B’ which includes the published half-lives of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  as well as a major uncertainty contribution to achieve consistency between all measured points.

Previous methods were based on more or less complicated applications of linear regression analysis. We are not convinced that the resulting uncertainties for the half-life are compatible with the GUM methodology and therefore have applied a method based on defining measured values of the isotopic ratio-of-ratios at the start and end point of the decay under observation. This method has an extra advantage of fitting the measurement history as different machines and methods of data acquisition were used for the measurement groups around the starting point, after about 20 years and at the end point, about 30 years.

In this work we considered each point as a valid measurement that was carried out with great care and under the best metrological understanding at that time. One consequence of this is that the uncertainty of the half-life, as derived from the start and end points, has been increased considerably in order for the points measured around 20 years decay to be included. This technique has previously been applied when combining multiple

measurements.<sup>11</sup> In the present paper the technique is referred to as a consistency test (see Appendix 1). For the measurements leading to the half-life of  $^{241}\text{Pu}$  it is the half-life itself whose uncertainty has to be increased so as to be consistent with all the measured values, as we have at present no reason to increase the uncertainty of the measured points.

The resulting half-life value for  $^{241}\text{Pu}$  is  $14.325 \pm 0.024$  a. The uncertainty, given here with  $k = 2$ , is certainly a conservative value which we believe will be reduced after further measurements have been carried out. These measurements should allow us to judge whether certain of the previous measurements can be eliminated from the half-life determination.

## Appendix: Metrological consistency

The interpretation of measurement uncertainty following the GUM is a metrological or state-of-knowledge based interpretation. In this interpretation uncertainty characterises the spread of values which is compatible with our knowledge of the natural system being measured.

The applied consistency check follows the concept suggested by Kessel et. al.<sup>11</sup> For example, the ratio-of-ratios ( $R$  in equation 1) was measured several times close to the starting time point. If the time difference between the measurements is known, they can be corrected to a common point in time. If we further assume that the material is homogeneous, we would expect that we would measure the same ratio of ratios. From this expectation we can derive a useful consistency check. Since we are measuring the same measurand in a group of measurements with a very similar uncertainty, we can use an arithmetic mean to combine the individual measurement results of the group.

$$R_p = \frac{1}{n} \sum_{i=1}^n R_i \quad (9)$$

with  $R_p$  being the arithmetic mean of the ratio-of-ratios of the group of measurements and  $R_i$  the  $n$  individual measurements.

If we have indeed measured the same quantity with procedures reliable within their uncertainty, we would expect that the difference between the mean  $R_p$  and the contributing  $R_i$  is small compared to the uncertainty of this difference.

The difference between contributing  $R_i$  and  $R_p$  is  $\varepsilon_i$ .

$$\varepsilon_i = R_i - R_p \quad (10)$$

To be metrologically consistent all  $\varepsilon_i$  must meet the following condition.

$$|\varepsilon_i| < k u(\varepsilon_i) \quad (11)$$

The coverage factor  $k$  is chosen to be 2 which represent 95% coverage for the individual  $\varepsilon_i$  if the distribution of the  $\varepsilon_i$  is assumed to be normal. An equal amount of uncertainty is added to each measurement if necessary for this condition to be met.

The same consistency criterion was applied to ensure that the measured points at around 20 years of decay were covered by the calculated half-life. The difference was calculated between the measured values and the calculated values based on the end points and the new half-life. The differences together with their expanded uncertainty ( $k = 2$ ) are shown in Fig. 4 after additional

uncertainty was added to the value of the half-life. The value zero is always included in the uncertainty intervals. Therefore the values are consistent.

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