STANDARDIZATION OF ^{241}Pu: ISSUES AND PROBLEMS

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ABSTRACT. A number of issues and problems in the standardization of ^{241}Pu by liquid scintillation (LS) methods have been encountered in work that has been performed over several years at the National Institute of Standards and Technology (NIST). The methods used for this work include: $4\pi\beta$-LS CIEMAT/NIST efficiency tracing (CNET) using ^{3}H as an efficiency monitor; $4\pi\alpha$(LS)-γ(NaI) live-timed anticoincidence (LTAC) measurements of ^{241}Am in aged solutions to follow the ingrowth from ^{241}Pu; and $4\pi\beta$-LS-based triple-to-double coincidence ratio (TDCR) determinations. Standardizations were performed on several linked solutions, which included a well-characterized master solution, identified as A1, that has been followed at NIST for the past 30+ years, two NIST-disseminated Standard Reference Materials (SRM 4340A and 4340B), and a solution prepared and distributed by the National Physical Laboratory (NPL) as part of an international measurement comparison. Features of our findings include: (i) A serious initial discrepancy between the CNET and TDCR methods was eventually resolved; (ii) Some concerns about cocktail composition/stability still exist because the CNET results show greater variability between LS cocktails than between counters with different operating characteristics (e.g. detection threshold), which is usually not the case for low-energy beta emitters; (iii) The CNET determinations show a distinct efficiency (quench) dependency irrespective of beta spectrum shape assumptions; (iv) It is apparent that the most accurate determination of the massic activity for the NPL comparison solution could be achieved by relative LS rate measurements with the aged solution A1, whose activity in turn had been determined by LTAC ^{241}Am ingrowth; and (v) The CNET and TDCR methods were shown to be in reasonable to somewhat good agreement with the LTAC ^{241}Am ingrowth determinations, particularly when considering the larger uncertainties associated with the former methods.

INTRODUCTION

Plutonium-241 (^{241}Pu) is a fissile material that accounts for a significant proportion of fissions in thermal reactor fuel that has been used for some time. In the environment, it is found in areas where nuclear weapons testing have occurred and/or nuclear fallouts. Quantitation of ^{241}Pu in low-level waste and environmental samples is of interest because ^{241}Pu is a precursor of other transuranium nuclides that have longer half-lives, greater environmental mobility, and greater toxicity (L'Annunziata 2003). ^{241}Pu is a very low-energy beta emitter with $E_{\beta(max)} = 20.8 \pm 0.2$ keV; $E_{\beta(ave)} = 5.23 \pm 0.5$ keV; and a half-life of 5234 ± 15 days (Chechev and Kuzmenko 2009). It decays primarily (99.998%) to the ground state of ^{241}Am, an emitter of alpha particles, X- and gamma rays with a half-life of 432.6 ± 0.6 yr (Chechev and Kuzmenko 2010) and is much more radiotoxic than its parent. ^{241}Pu also decays (0.00244%) to levels of ^{237}U (6.749 d) (Kuzmenko and Chechev 2010).

The low beta energy makes the measurement of ^{241}Pu somewhat difficult because of the low liquid scintillation (LS) detection efficiency. However, the LS efficiencies for the ^{241}Pu determinations have been determined by a tracing-based method (CIEMAT/NIST)[2] using composition-matched LS cocktails of a ^{3}H standard as the efficiency detection monitor (CNET). The ^{241}Pu activity can also be determined indirectly by measurements of its daughter nuclide ^{241}Am, but measurements based on the ingrowth of the daughter radionuclide ^{241}Am can be done only after a reasonable ingrowth period.

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[2] The acronym CIEMAT/NIST refers to the 2 laboratories that collaborated in developing the protocol for this LS tracing methodology: the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) and the National Institute of Standards and Technology (NIST).

DESCRIPTIVE BACKGROUND AND METHODS

The National Institute of Standards and Technology (NIST) has made various ^{241}Pu determinations over many years. Below is a short background of the solutions origin and the links to other solutions for the past 30+ years presented chronologically by the measurement time. In addition, a more detailed look at the methods and evaluations performed in the last 3 years is presented. Refer to Figure 1 for the scheme of preparation and calibration of the ^{241}Pu solutions and standards. Solid lines are mass connections while dashed lines are measurement connections.
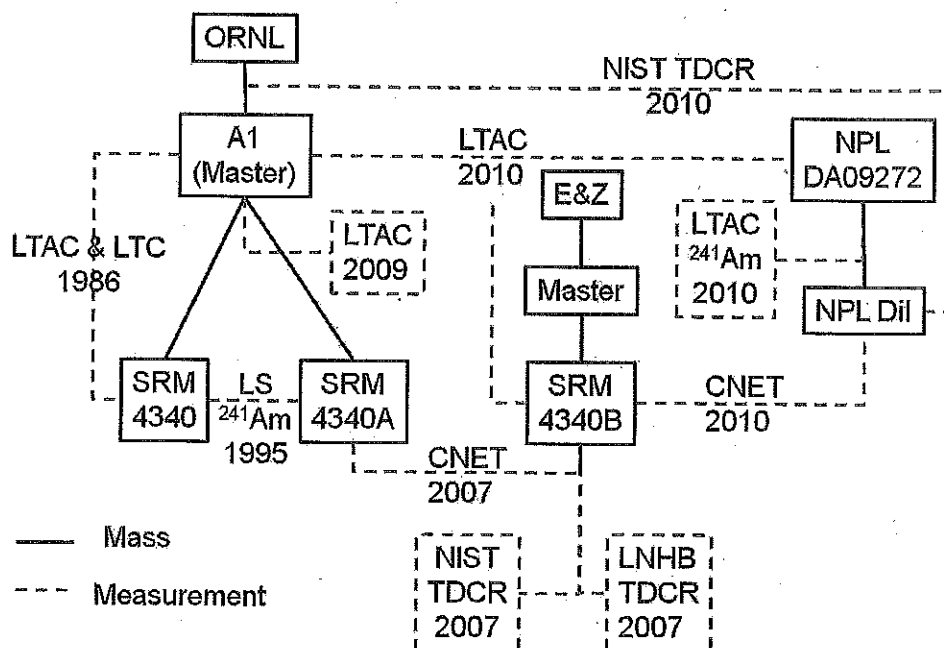

Figure 1 Scheme for preparation and calibration of the ^{241}Pu solutions and standards. Refer to text for acronyms.

A ^{241}Pu solution was provided by Oak Ridge National Laboratory (ORNL) in 1977 right after separation. It was standardized at NIST by both liquid scintillation and 4π(LS)-γ(NaI) coincidence and anticoincidence counting (Coursey et al. 1989). At the time of standardization, it was examined for radionuclidic impurities and all the emissions observed could be attributed to the ^{241}Pu plus progeny. This solution was diluted and used as the master solution (A1) for NIST ^{241}Pu Standard Reference Materials (SRM 4340 and 4340A) that were subsequently disseminated. SRM 4340 (NBS 1986) was calibrated by the 4π(LS)-γ coincidence counting of ^{241}Am daughter ingrowth, while a new dilution, SRM 4340A (NIST 1996), was standardized by LS following the ingrowth of ^{241}Am over the course of 18 years.

In 2007, a ^{241}Pu solution was obtained from Eckert & Ziegler (E&Z) Isotope Products (California, USA)[3], diluted and standardized by CNET using CN2003 (E Günther, personal communication, 2003) code calculations. This solution was disseminated as SRM 4340B (NIST 2008). The LS

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[3]Certain commercial equipment, instruments, and materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by NIST, nor does it imply that the materials and/or equipment are the best available for the purpose.

CNET measurements for this standardization comprised 3 cocktail compositions (Table 1, series I–III), each with 6 samples, measured for 2 to 10 cycles on 1 or 2 measurement occasions, in 3 counters for a total of $n = 12$ data sets ($\nu = 312$ effective degrees of freedom), normally distributed. At the same time, a set of LS sources was prepared as shown in Table 1, series III, for the remeasurement of SRM 4340A. This data (SRM 4340A) was analyzed identically to SRM 4340B (by CNET). The CNET method has been described in detail elsewhere along with the specific analysis procedure used for this work (Collé and Zimmerman 1997 and references therein). The ^3H nuclear data used in the calculation was obtained from Chechev (2006). SRM 4340B was examined for alpha-particle-emitting impurities by alpha spectrometry with surface barrier detectors and for photon-emitting impurities by high-purity intrinsic germanium detectors. The solution showed the presence of ^{242}Pu, $^{240+239}Pu$, ^{238}Pu, and ^{241}Am and no photon-emitting impurities were observed.

Table 1 LS cocktail compositions used in the standardization of ^{241}Pu. Series I through III were used in the CNET measurements during the standardization of SRM 4340B in 2007; series IV was used in the TDCR measurements during the standardization of SRM 4340B in 2007. Series V was used in the CNET measurements during the standardization of NPL Dil. Series VI–VII were used in a LS comparison between NPL Dil, A1, and SRM 4340B in 2010. All the sources were prepared in nominal 20-mL scintillation vials.

Cocktail series	Scintillant[a]	Nr of samples	M (g)[b]	H₂O mass fraction	C_a (mol L⁻¹)[c]	Solution(s)
I	UGAB	6	10.5	0.07	1.2	4340B
II	UGAB	6	10	0.05	0.2	4340B
III	PCS	12	11.2	0.10	1.0	4340B/4340A
IV	UGAB	3	9.2	0.10	1.8	4340B
V	UGAB	18	11.8	0.06	2.8	4340B/NPL Dil
VI	UGAB	15	9.9	0.06	2.8	4340B/NPL Dil/A1
VII	UGAB	15	9.8	0.06	3.0	4340B/NPL Dil/A1
VIII	UGAB	6	9.9	0.06	3.0	NPL Dil/A1

[a]UGAB = Ultima Gold™ AB (PerkinElmer, USA); PCS = Phase Combining Systems (Amersham Biosciences, Sweden).
[b]Mass of scintillation fluid in each cocktail.
[c]HNO₃ concentration in aqueous fraction of cocktail.

In order to obtain an independent confirmation of the ^{241}Pu activity (SRM 4340B), measurements using the triple-to-double coincidence ratio (TDCR) were performed. The sources' composition is as presented in Table 1, series IV. However, the TDCR results showed a large discrepancy with CNET (7.7%). This discrepancy was puzzling; thus, an ampoule of the solution was sent to Laboratoire National Henri Becquerel (LNHB) in order to obtain a separately independent and impartial measurement of the ^{241}Pu by TDCR. The TDCR method for SRM 4340B and measurements are explained in detail in Bergeron and Zimmerman (these proceedings).

In 2009, as part of an internal verification, a new determination of the ^{241}Am content of solution A1 was performed by $4\pi(LS)-\gamma(NaI)$ live-timed anticoincidence (LTAC) counting with efficiency extrapolation. The ^{241}Pu activity of the solution was derived from the ^{241}Am ingrowth using known separation dates and literature values for the relevant half-lives. This result was in very good agreement with the one obtained in 1986.

In late 2009, as part of an international comparison piloted by NPL, a ^{241}Pu solution was sent to various laboratories for calibration. The ^{241}Pu solution received by NIST was diluted (referred to as

NPL Dil), measured and analyzed by CNET using CN2003 (E Günther, personal communication, 2003) code calculations with ^{3}H standard as the efficiency detection monitor, TDCR and LTAC. The LS CNET measurements consisted of 1 cocktail composition (Table 1, series V), with 6 samples, measured for 3 to 5 cycles on 1 or 2 measurement occasions, in 3 counters for a total of $n = 4$ data sets ($\nu = 84$ effective degrees of freedom), normally distributed. At the same time, SRM 4340B was remeasured (Table 1, series V) and analyzed identically (by CNET).

For the ^{241}Pu determination in the NPL solution, 2 more sets of LS vials were prepared (Table 1, series VI and VII) with identical compositions in order to obtain a relative LS comparison by the ratio of both the SRM solution to the well-known A1 and of NPL Dil solution to A1. The measurement comprise 5 samples, measured for 3 cycles on 1 to 3 measurement occasions, in 3 counters for a total of $n = 5$ data sets ($\nu = 75$ effective degrees of freedom), for each ratio, normally distributed.

For the ^{241}Pu determinations of both SRM 4340B (2007) and NPL Dil (2010), 3 different instruments were used for the LS measurements: (i) Packard Tri-Carb A2500 TR® (PerkinElmer, USA); (ii) Wallac 1414 Winspectral® (PerkinElmer, USA); and (iii) Beckman LS 6500® (Beckman Coulter, USA). They have considerably different operating conditions characteristics (e.g. low-energy threshold, deadtime, linear vs. logarithmic energy binning, etc.), which helps to ensure variation in measurement conditions during standardization. A more detailed description is presented elsewhere (Laureano-Pérez et al. 2007).

For the TDCR determinations during the NPL solution standardization, 2 sets of 3 LS sources with identical composition were prepared. The first set was for the determination of ^{241}Pu in the NPL Dil solution and the second set for the determination of ^{241}Pu in solution A1. The sources composition is as presented in Table 1, series VIII. The TDCR method and measurements for these solutions are explained in detail in Bergeron and Zimmerman (these proceedings).

The LTAC counting sources for NPL solution and A1 were glass hemispheres sealed at the top with epoxy. Each hemisphere was filled with about 4 g of Ultima Gold™ AB (PerkinElmer, USA) LS cocktail, and enough water and HNO_3 such that the aqueous fraction was 0.15 and the composition of that aqueous fraction was 2M HNO_3. The high aqueous fraction was used because the α emission rate of the NPL solution was low; hence, large sample sizes were needed to obtain a high-enough net count rate. The LTAC instrument contains a single-phototube LS detector surrounded by a NaI well detector. The instrument and method are described in Fitzgerald and Forney (these proceedings) and Fitzgerald and Schultz (2008). The ^{241}Pu activity of solution A1 was determined by measuring the ^{241}Am activity using LTAC. This measured ^{241}Am activity was used to calculate the ^{241}Pu activity with the known separation date.

RESULT AND DISCUSSION

The ^{241}Am ingrowth in A1 solution has been followed at NIST for the last 30+ years. As explained above, this solution is directly linked to SRM 4340 and 4340A (see Figure 1). From the ^{241}Pu activity determinations of these solutions, the following results were obtained: (i) the ^{241}Pu value obtained for SRM 4340A by following ^{241}Am ingrowth by LS determined in 1995 agreed to within +0.06% with the results for SRM 4340 (1986); (ii) the results for the measurement of SRM 4340A by LS CNET in 2007 were in agreement with the ^{241}Am ingrowth (1995) to within +1%; (iii) the ^{241}Pu determination in A1 by LTAC in 2009 agreed with the one performed in 1986 to within −0.1%. These results show that the ^{241}Am content of the solution is very well known and therefore so is the ^{241}Pu content.

As mentioned, during the 2007 standardization it was found that the NIST TDCR differed from the CNET value by −7.7%. The TDCR result obtained from LNHB (P Cassette, personal communication, 2007) agreed with NIST TDCR to within +0.04%, which in turn differs from the CNET result by −7.6%. It was proposed that this difference might be due to the code used in the determination of the theoretical efficiency for CNET. Based on previous experience (Collé et al. 2008), it is believed that CN2003 is the adequate code since it uses a quench function applicable for a di-isopropyl-napthalene-based scintillation fluid, like Ultima Gold (PerkinElmer). However, in order to test this hypothesis the data was reanalyzed using EFFY4 (E Garcia-Toraño personal communication, 1993). The use of EFFY4 showed a lower result by 0.7%. The TRACER code (P Cassette, personal communication, 2006) was compared to CN2003 to determine the effect of the code in the activity. It was found that for $kB = 0.12$ cm MeV^{-1}, which is the number used in TDCR calculations, at 30% ^{3}H efficiency, it agrees with CN2003 to about +0.04%. Hence, this possible explanation was discarded.

The uncertainty on the SRM 4340B ^{241}Pu determination by CNET was 3.8% ($k = 2$). The main contributors were the uncertainty in measurement precision (scatter in data) and the uncertainty due to the model dependencies and computed beta spectra. It should be noted that the measurements showed greater variability in the traced activity between samples than between counters. This is rare for low-energy beta emitters where differences in counter characteristics (detection threshold) usually make significant differences.

At the time of the SRM 4340B standardization, the discrepancy found between TDCR and CNET could not be explained, and it was assumed that there was a systematic error during the TDCR analysis that could not be determined. This discrepancy between CNET and TDCR was found in the preliminary determinations of the NPL Dil solution, but further investigation during this calibration showed some of the reasons for the discrepancy. Certain modifications to the TDCR hardware, the use of a different model, a change in vial type, and a more accurate accounting for impurities corrections and ^{241}Am ingrowth caused the difference between CNET and TDCR to change from −7.7% to −2.6%. More details of these changes are presented in Bergeron and Zimmerman (these proceedings). At the same time of the NPL Dil solution calibration by TDCR, when the modifications were performed, measurements of the solution A1 were performed. The result obtained for ^{241}Pu in A1 solution agrees with the LTAC determination to within −2.3%, which is within the TDCR method uncertainty listed (~2.3%, $k = 2$).

The CNET performed for both ^{241}Pu standardization/calibration occasions (SRM 4340B and NPL) used CN2003 as a code and ^{3}H as the efficiency detection monitor. There was, however, a difference in the data analysis. The difference consisted in the LS counter data window selection during the data analysis. In the 2007 standardization, the whole spectra (wide open window) was taken into account when doing the CNET analysis. The traced activity was then corrected based on the determination of the ^{241}Am ingrowth performed at NIST by alpha spectrometry. At the time of the determination on the NPL solution (2010), any alpha emitters from impurities or ^{241}Am daughter were unknown. Therefore, a different window was set where only the ^{241}Pu beta region was considered. SRM 4340B was remeasured and analyzed exactly as the NPL solution (beta region window) in 2010. It was found that the results in 2010 agreed to within −0.18% to the determination for SRM 4340B in 2007, proving the consistency of the determination and confirming that the window setting was adequate. It also suggests that the uncertainty in the ^{241}Pu certified massic activity might have been overestimated.

The beta spectrum shape factor was also analyzed during this standardization. Figure 2 shows the effect of the beta spectrum shape factors on the efficiency calculation of CN2003 code. Efficiencies were calculated with the data as obtained from the literature assuming an allowed beta transition ($C(W) = 1$) and an endpoint energy $E_{\beta,max} = 20.8 \pm 2$ keV (Chechev 2006; Chechev and Kuzmenko

2009). As part of the ^{241}Pu comparison, LNHB obtained an experimental beta spectrum measured in a bolometer from which the shape factors were deduced ($C(W) = 1 - 1.2w + 1.4/w - 0.4w^2$) (P Cassette, personal communication, 2010) with an endpoint energy $E_{\beta,max} = 20.8 \pm 2$ keV. The spectrum with these shape factors was used in the code, referred to as LNHB on the plot, and activity recalculated. In addition, extreme, largely unrealistic shape factors ($C(W) = 1-372.6w + 129.33/w - 243.4w^2$, referred to as NIST on plot) with an endpoint energy $E_{\beta,max} = 20.8 \pm 2$ keV were used to test the sensitivity on the calculated activity. In the ε_{H-3} range used for tracing (0.17–0.38), corresponding to 0.15–0.33 for ε_{Pu-241}, the change in beta spectrum shape factors could not produce a significant enough change in the theoretical efficiencies. There was an increase in traced activity, for both SRM 4340B and NPL Dil, of 2.0% when using the shape factor numbers provided by LNHB and 4.0% when using the NIST more unrealistic numbers. This increase is only in the magnitude of the value; it has no effect in the slope of the plot of traced activity vs. ε_{H-3}. These results show that the activities obtained from the CNET determinations demonstrate an efficiency (quench) dependency irrespective of beta spectrum shape assumptions. Since there was no practical reason for not using the literature values, the allowed beta spectrum shape was used.
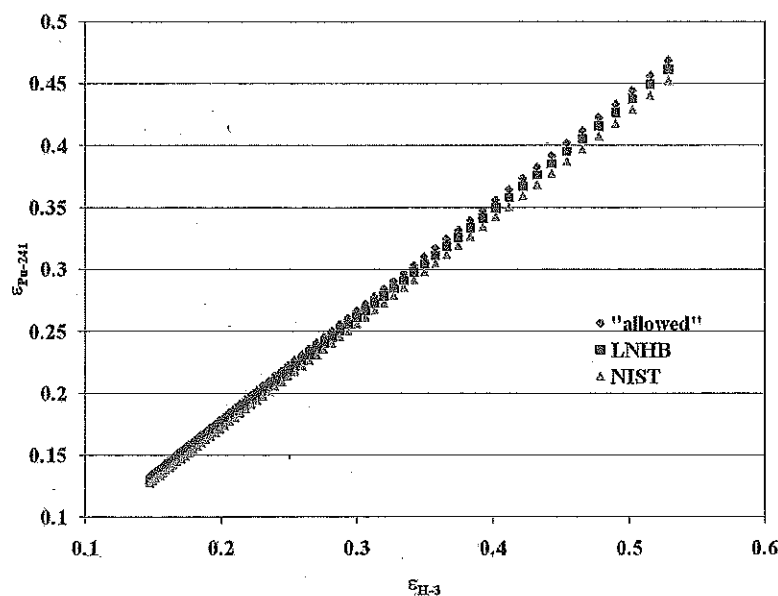

Figure 2 Effect of beta spectrum shape factors in CN2003 code efficiency determination. LNHB data provided by P Cassette (personal communication, 2010).

Due to the original discrepancy between CNET and TDCR and the fact that the solution A1 is very well known, it was decided to standardize the NPL solution by linking it to the A1 solution through a relative LS measurement. The A1 solution was then restandardized by LTAC. The LTAC method had the advantage over pure $4\pi\alpha$ LS counting in that the β/α energy discriminator could be set well above the β-endpoint energy while accounting for any α-counting losses (~0.3%). Those small α-counting losses caused by the discriminator were extrapolated to zero using the anticoincidence method. Also, a hardware discriminator was used, so that the higher β-decay rate did not increase the deadtime of the counter for alphas. The new ^{241}Pu activity value for the solution A1 differed from the 1986 SRM 4340 certificate by only −0.01%. The uncertainty on the new activity value is 0.38% ($k = 1$), where most of that is from to the uncertainties due to the ^{241}Pu half-life (0.28%) and the efficiency extrapolation (0.18%).

For the relative LS measurements, SRM 4340B, NPL Dil, and A1 solution sources where prepared with identical cocktail compositions and measured in 3 different counters. The ratio was determined by selecting the counts from the first window or the "beta region window," which was set after obtaining the total spectra for each source. The ratio of NPL/A1 was found to be 0.2401 with a relative standard uncertainty of 0.17%. Using the activity determined by LTAC we obtained the ^{241}Pu activity in the NPL solution that will be reported by NIST. The result is not presented here because the comparison is not yet complete.

The ^{241}Pu content of the NPL solution was also determined by following the ^{241}Am ingrowth over 3 months using LTAC (as described above) and then fitting that ingrowth curve, leaving the α-emitting impurities as a free parameter. The initial ^{241}Am activity was allowed to vary within the range indicated by NPL. This method works because the initial ^{241}Am and also the Pu impurities all have long half-lives relative to the ingrowth of ^{241}Am, so they are nearly constant during the 3-month span of the measurement. Our fitting model also accounts for the small ^{241}Pu alpha branch. So far, the fit uses 3 data points covering a timespan from 210 days after separation to 285 days after separation. We will continue making measurements for a few more months, to reduce the uncertainty in the deduced ^{241}Pu activity.

SRM 4340B was measured and analyzed identically to the NPL solution (LS ratio with LTAC measurements of A1). The result obtained using the combination of methods showed a −3.1% difference from the value standardized in 2007 by CNET and −2.9% with the measurement performed in 2010 by CNET. Although it is within the uncertainty of the certified massic activity (3.8% at $k = 2$), it is disconcerting that there is a disagreement with the SRM when a relatively good agreement was found with the NPL solution. In addition, there was a very good agreement when remeasuring SRM 4340B. This might indicate that the beta spectrum shape could be more important than it was assumed and/or an unknown issue that has not been addressed in this determination.

The ^{241}Pu activity obtained by the combination of LTAC and LS (to be reported) has a relative expanded uncertainty of 0.86% ($k = 2$). The uncertainty evaluation is presented in Table 2. The ^{241}Pu content in the NPL solution obtained by the combination of methods agrees with CNET to within +1.9%, which is within the uncertainty of the CNET method (3.8% at $k = 2$) for this radionuclide.

Table 2 Uncertainty evaluation for the ^{241}Pu massic activity in the NPL solution.

	Uncertainty component	Assessment type	Relative standard uncertainty contribution on massic activity of ^{241}Pu (%)
1	Uncertainty for LTAC determination of ^{241}Pu in solution A1	A	0.38
2	LS measurement precision; reproducibility in activity ratio for 1 cocktail composition, with 5 samples, measured in 3 counters on 1 or 2 measurement occasions; standard deviation of the mean for $n = 5$ data sets ($v = 75$ effective degrees of freedom), normally distributed	A	0.17
3	Background; wholly embodied in 2	A	—
4	LS counters dependencies; wholly embodied in component 2	A	—
5	Live time determinations for LS counting time intervals, includes uncorrected deadtime effects	B	0.06
6	Gravimetric (mass) measurements for LS sources	B	0.05
7	Gravimetric (mass) measurements for dilution of NPL solution	B	0.05
8	^{241}Pu decay corrections for half-life uncertainty of 0.28%	B	0.004
Relative combined standard uncertainty			**0.43**
Relative expanded uncertainty ($k = 2$)			**0.86**

This LS uncertainty was determined using very conservative numbers due to our lack of knowledge of the shape of the beta spectrum. The value to be reported also agrees to within −0.67% with the improved NIST TDCR method. The value for the LTAC ^{241}Am ingrowth determination is +2.5% from the value to be reported. This is within the method uncertainty listed, 2.5% at $k = 1$, and is dominated by the uncertainty from the ingrowth fit, which itself contains most of the uncertainty from the anticoincidence measurements. The NPL solution was examined for alpha-particle-emitting impurities by alpha spectrometry with surface barrier detectors and for photon-emitting impurities and ^{241}Am ingrowth by high-purity intrinsic germanium detectors. The solution showed the presence of ^{242}Pu, $^{240+239}$Pu, ^{238}Pu, and ^{241}Am. All the photon emissions observed could be attributed to the ^{241}Pu progeny.

CONCLUSIONS

Some issues and problems encountered in the standardization of ^{241}Pu by CNET, TDCR, and LTAC are discussed here. It was found that results from CNET and LTAC were in very good agreement for both the calibrations performed in 2007 as part of the standardization of SRM 4340B and the calibration of the NPL solution in 2010. On the other hand, TDCR agreement to the previous methods was only achieved after major modifications, which are now understood (Bergeron and Zimmerman, these proceedings).

The ^{241}Pu content of solution A1, by ^{241}Am ingrowth, has been followed by various determinations over 30+ years. Every determination was within 1% of the previous result irrespective of measurement method, i.e. 4π(LS)-γ(NaI) coincidence and anticoincidence counting of ^{241}Am daughter ingrowth, LS following the ingrowth of ^{241}Am, LTAC, and CNET. Even though the TDCR value obtained was −2.3% different, it was still within the method uncertainty; therefore, it was considered in reasonably good agreement.
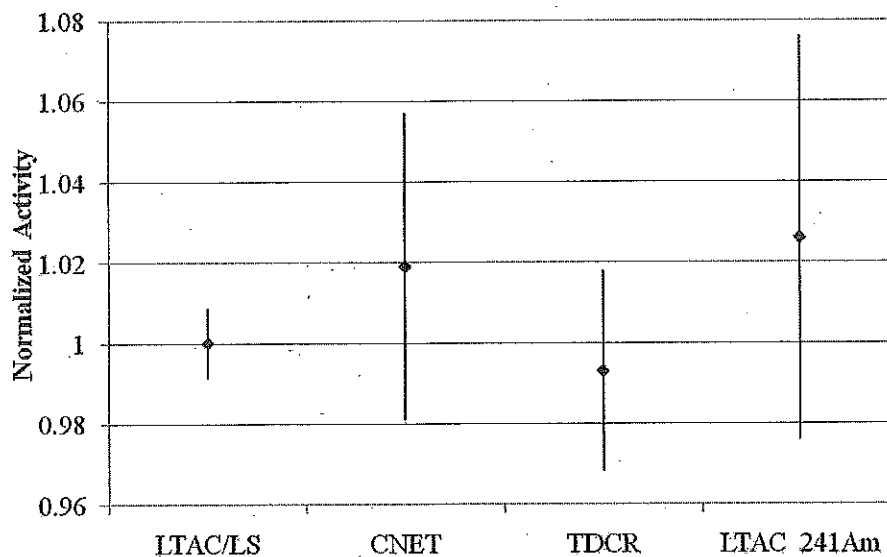

Figure 3 Normalized ^{241}Pu activity in the NPL solution and its respective uncertainty as determined by each method.

CNET analysis was performed on 2 standard solutions in addition to the NPL solution. From these determinations, it was found that the activities obtained from the CNET determinations demonstrate an efficiency dependency irrespective of beta spectrum shape assumptions. Also, similar results were obtained when using just a beta region of the LS spectrum for the data analysis compared to when the whole spectrum was used and corrected for the ^{241}Am content and alpha impurities.

Since the ^{241}Pu content in A1 was well established with determinations over many years, a relative LS measurement between the A1 and the NPL solution with a LTAC restandardization of A1 was selected as a more accurate way to determine the ^{241}Pu activity in the NPL solution. The result was confirmed by the measurements of the NPL solution by CNET, TDCR, and ^{241}Am ingrowth by LTAC. Figure 3 shows the normalized ^{241}Pu activity in the NPL solution and their respective uncertainty ($k = 2$) as determined by each method.

ACKNOWLEDGMENTS

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