

Standardization of ^{237}Np

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HIGHLIGHTS

- Standardization of ^{237}Np by $4\pi\alpha\beta$ liquid scintillation counting and confirmatory methods.
- All results agree within the respective method's uncertainties.
- $^{237}\text{Np}/^{233}\text{Pa}$ radioactive equilibrium disturbed when making dilutions and/or removing aliquots.

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ABSTRACT

The standardization of ^{237}Np was investigated. The certified massic activity for ^{237}Np was obtained by $4\pi\alpha\beta$ liquid scintillation (LS) counting with correction for the ^{233}Pa daughter using the CIEMAT/NIST efficiency tracing method using a ^3H standard. Confirmatory measurements were also performed by high-resolution HPGe gamma-ray spectrometry, and by $4\pi\alpha\beta(\text{LS})-\gamma(\text{NaI})$ anticoincidence counting. All results agree within the respective method's uncertainties. It was confirmed that the $^{237}\text{Np}/^{233}\text{Pa}$ radioactive equilibrium is disturbed when making dilutions and/or removing aliquots.

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1. Introduction

Neptunium is a byproduct of plutonium production activities and results from the neutron capture by uranium isotopes. Neptunium-237 is the most stable neptunium isotope and is also the product of alpha decay of ^{241}Am . It is produced as a long-lived waste product in nuclear reactors. Hence, accurate standardization of ^{237}Np is necessary for environmental monitoring of nuclear waste.

Neptunium-237, with a half-life of $(2.144 \pm 0.007) \times 10^6$ a (Chechev and Kuzmenko, 2010a), decays by alpha emission to the ground state and excited states of Protactinium-233. Protactinium-233, with a half-life of (26.98 ± 0.02) d (Chechev and Kuzmenko, 2010b), decays by beta emission to levels of Uranium-233. During both decays numerous gamma transitions occur.

In order to determine international measurement compatibility for ^{237}Np , and to produce a reliable and consistent decay scheme, a EUROMET project was conducted during 1998 (Smith et al., 2001). A high-purity solution of ^{237}Np was prepared and disseminated to the participating National Metrology Institutes (NMIs), including NIST. The ^{237}Np exercise EUROMET action 416, which was hosted by the National

Physical Laboratory (NPL; Middlesex, UK), was completed in 1999. However, for unforeseeable circumstances, NIST did not contribute to these results. During this exercise ^{237}Np – ^{233}Pa equilibrium/stability issues were identified by numerous participants.

In this work the standardization of ^{237}Np by different methods was investigated and the equilibrium/stability issues were re-examined.

2. Experimental methods, results and discussion

2.1. Overview

The scheme used to prepare the ^{237}Np standard and the counting sources that were used for the LS analysis and confirmatory measurements performed at NIST is shown in Fig. 1. The certified ^{237}Np massic activity for the standards was obtained from a primary standardization based on $4\pi\alpha\beta$ liquid scintillation (LS) spectrometry, with corrections for the ^{233}Pa daughter using the CIEMAT/NIST efficiency tracing method (CNET).² Confirmatory standardizations of the ^{237}Np massic activity were performed by live-timed anticoincidence counting $4\pi\alpha\beta$

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² The acronym CNET refers to CIEMAT/NIST Efficiency Tracing method. CIEMAT/NIST refers to the two laboratories that collaborated in developing the protocol for this LS tracing methodology; viz., the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) and the National Institute of Standards and Technology (NIST).

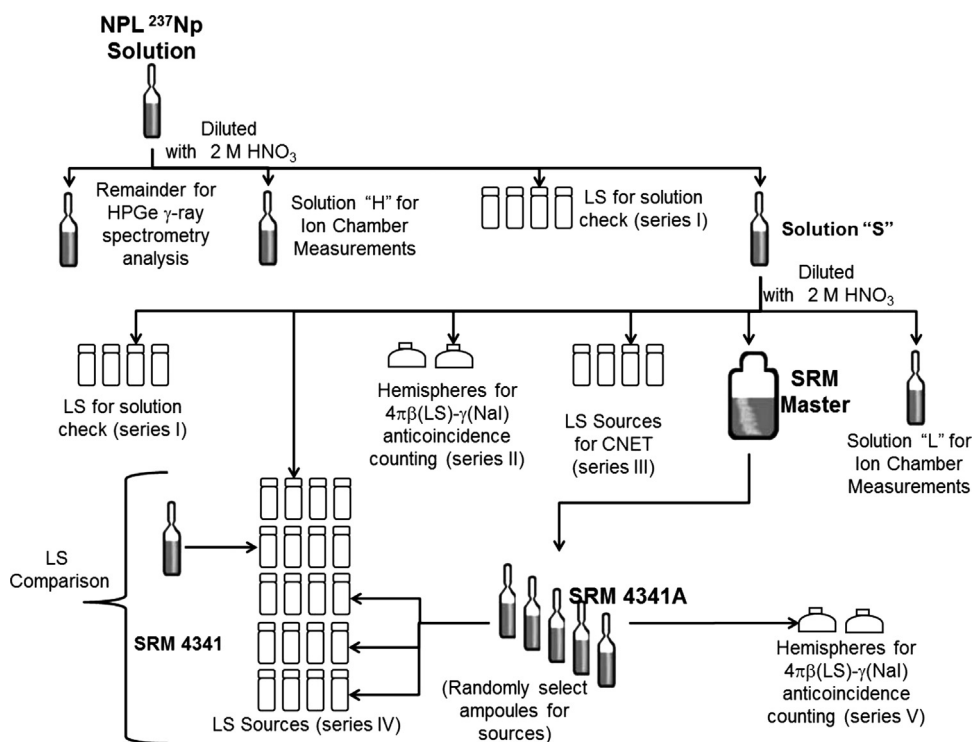


Fig. 1. Scheme for the solution's dilutions and source preparation for ^{237}Np as used for calibration and confirmatory measurements.

(LS)- γ (NaI) (LTAC) and gamma-ray spectrometry (γ -spec). LTAC measurements were performed on both SRM 4341a (NIST, 2013) and high level solution "S". High-resolution HPGe gamma-ray spectrometry (γ -spec) was used to determine photon emitting impurities, as well as to measure the activity on the initial NPL solution. A direct LS comparison of the SRM 4341a standardization was made with that for previous ^{237}Np issue (NIST, 1993), which was first disseminated in 1993 as SRM 4341, and with the higher level solution "S" that was a dilution of the solution used for the EUROMET comparison.

2.2. Preparation of solutions and counting sources

The ^{237}Np stock solution used to prepare this SRM was obtained from NPL as part of the EUROMET action 416 (^{237}Np exercise) measurement comparison amongst NMIs (Smith et al., 2001). The stock solution was chemically purified on approximately 19 to 22 August 1997 by the Institute for Reference Materials and Measurements (IRMM; Geel, BE). It consisted of approximately 4 g of a 5.8 mol L^{-1} hydrochloric acid and $33 \mu\text{g g}^{-1}$ oxalic acid solution in which ^{237}Np was dissolved. Protactinium-233, the daughter product that results from ^{237}Np decay, has been growing in since that separation time. Since there was concern of solution instability in this study, the NPL solution was diluted in steps and stability tested at every step by ion chamber (IC "A"), Auto chamber (IC "B"), LS, NaI(Tl) detectors and $4\pi\alpha\beta$ (LS)- γ (NaI) live-timed anticoincidence counting (LTAC).

The cocktail compositions of all the counting sources used in the standardization are presented in Table 1. Series I sources were used for a preliminary solution check. Series II and V sources were used for the LTAC determinations. Series III sources were used for the CNET. Series IV sources were used to perform the LS comparison between SRM 4341, high level solution "S" (linked to the EUROMET comparison solution), and three randomly selected ampoules from the newly prepared SRM 4341a.

Timing for this work was with respect to the SRM 4341a Reference Time of 1 September 2012 ($t=0$). The initial dilutions of the NPL solution and preparation of the SRM ampoules were

performed on 5 to 6 September 2012 ($t=4 \text{ d}$ to $t=5 \text{ d}$). The LS source preparation and initiation of counting was done on 3–4 October 2012 ($t=32 \text{ d}$ to $t=33 \text{ d}$).

2.3. Measurement methods

2.3.1. Liquid scintillation measurements

Three different instruments were used for the LS measurements; (i) Packard TriCarb A2500 TR (Perkin-Elmer, Wesley, MA)³; (ii) Wallac 1414 Winspectral (Perkin-Elmer, Wesley, MA) and (iii) Beckman LS 6500 (Beckman Coulter, Fullerton, CA). They have considerably different operating condition 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).

The certified massic activity for ^{237}Np was obtained by $4\pi\alpha\beta$ liquid scintillation (LS) counting. The LS detection efficiency for ^{233}Pa was calculated using the CN2003 code (Günther, 2003) for the CNET method with composition matched LS cocktails of a ^3H standard as the efficiency detection monitor. The ^3H nuclear data used in the calculation were obtained from Chechev (2006). This method has been described in detail elsewhere (Collé and Zimmerman, 1996a, b, 1997; Zimmerman and Collé, 1997a, b).

Series III and IV were measured concurrently and sequentially in three counters for a period of 80 days (from $t=33 \text{ d}$ to $t=113 \text{ d}$). The sources were preliminarily measured for a period of 20 days and an increase in counting rates with time was observed, showing an in-growth of ^{233}Pa in the traced activity (Fig. 2). An identical trend was observed in all LS sources, excepting those prepared from SRM 4341. In as much as the solution composition of SRM 4341 and the new SRM 4341a are nearly identical, this was

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 imply that the materials and/or equipment are the best available for the purpose.

Table 1

LS cocktail compositions used for the standardization of ^{237}Np . All the sources were prepared with Ultima Gold AB (Series I, III and IV, 9.9 g and series II and V, 4 g). Nitromethane was added to series III as a quenching agent. Series I and IV were used for LS measurements; series III was used for the $4\pi\alpha\beta$ LS measurements with ^3H -standard efficiency tracing and; series II and V was used for the $4\pi\alpha\beta(\text{LS})-\gamma(\text{NaI})$ anticoincidence counting experiments.

Cocktail series	Radionuclide solution	N_s	f_w	C_{HCl} (mol L $^{-1}$)	C_{HNO_3} (mol L $^{-1}$)	Aliquant (mg)
I	NPL and "S"	8	0.06	0.9	0–1.9	87–102
II	"S"	3	0.07	1.1–1.9	0.6–1.9	51–103
III	"S"	6	0.06	0.07	0.12	51–55
IV	"S", 4341, 4341a	20	0.06	0.06	0.55–0.76	160–200
V	4341a	2	0.11	0.80	1.3	400

N_s =number of samples in each series.

f_w =Aqueous mass fraction in cocktail.

C_{HNO_3} =HNO $_3$ concentration (in mol L $^{-1}$) in aqueous fraction of cocktail.

C_{HCl} =HCl concentration (in mol L $^{-1}$) in aqueous fraction of cocktail.

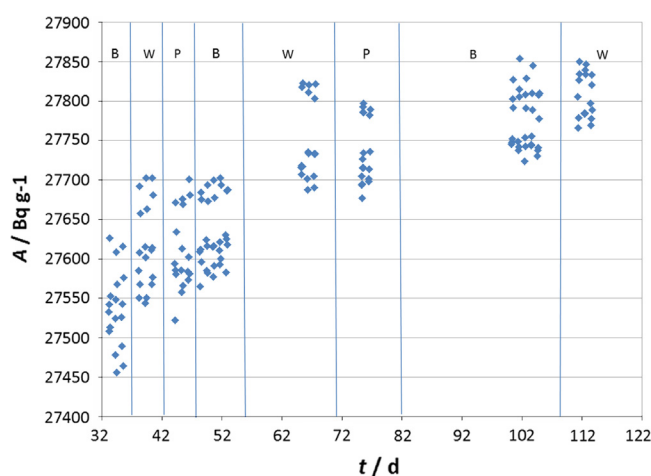


Fig. 2. LS results of traced ^{237}Np massic activity as a function of time. The B, P, and W refer to the LS counter used for the measurement: P=Packard TriCarb A2500 TR; W=Wallac 1414 Winspectral and B=Beckman LS 6500. The time scale is with respect to the SRM Reference Time of 1 September 2012 ($t=0$). The sources for this data were prepared on 3 October 2012 ($t=32$ d).

a surprising finding. It most likely arose because SRM 4341 solution was in radioactive equilibrium, and was not noticeably disturbed when removing aliquants to prepare the LS sources. The sources were then measured in different counters for 60 additional days. The counting time was typically 40 min per sample for three to five cycles per measurement occasion. As seen in Fig. 2, the data showed some stabilization (leveling off) after about $t=65$ d. Since the traced activity of the solution seemed to remain relatively constant, the four measurements performed after $t=65$ d were analyzed and used for the reported certified activity.

For this analysis ^{237}Np was assumed to have 100% efficiency due to alpha emissions and highly converted gammas. Its daughter, ^{233}Pa , efficiency was calculated using a simplified decay scheme of 21 decay paths (cascades) with 6 betas and 13 gammas with a probability of 0.03% or higher. The simplified decay scheme was input into CN2003. For the ^3H efficiency range used (17% to 40%), the total efficiency ranged from 194% to 197%.

Even though some disequilibrium was observed at the beginning of the measurements, stability was accomplished after $t=65$ d (or one ^{233}Pa half-life since the start of counting) and equilibrium was assumed in the final calculations. The main contributor to the uncertainty of the certified value was the uncertainty due to the model dependencies and computed beta spectra including uncertainties in the decay scheme data. The uncertainty budget for the ^{237}Np is presented in Table 2.

A fit of the data indicated a ^{237}Np – ^{233}Pa disequilibrium of about 1% at the time of source preparation. For comparison, an LS source prepared from an aliquot of solution "S" on 5 September ($t=4$ d) did not exhibit any disequilibrium over a period of 7 days to within measurement precision ($<0.1\%$). We could not discern the cause of this difference and cannot explain it.

The solution "S" to NPL solution dilution factor obtained by LS measurements from the series I sources with two counters agreed with the gravimetric dilution to $+0.30\%$ and $+0.14\%$. The SRM to solution "S" dilution factor obtained by comparing the series III and IV LS sources was $+0.16\%$ higher than the gravimetrically determined dilution factor.

2.3.2. $4\pi\beta(\text{LS})-\gamma(\text{NaI})$ live-timed anticoincidence counting (LTAC)

Confirmatory massic activity determinations were carried out by LTAC of the SRM 4341a solution and of the higher-level solution, "S". For details of the method, see the report (Fitzgerald et al., 2010) of the standardization of ^{229}Th , another mixed alpha-beta chain, and references therein. In the present case, the efficiencies for both the alpha decay of ^{237}Np and the beta decay of ^{233}Pa were monitored using three γ -ray gates in the NaI spectrum, and extrapolated to 100% efficiency to reveal the total ^{237}Np and ^{233}Pa activity of the solution. The total activity was measured in this way over a period 90 days.

Based on ingrowth fits of the data, the ratio of ^{233}Pa activity to ^{237}Np activity at the time of source preparation was (0.9955 ± 0.0003) for solution "S" and (0.930 ± 0.003) for SRM 4341a. That is, 7% of the ^{233}Pa in the SRM solution was apparently left behind when the LTAC sources were prepared, indicating that disequilibrium occurs on transfers. The ingrowth curves were then used to extract the respective ^{237}Np massic activities for the two solutions. The ratio of the resultant ^{237}Np massic activities in the two solutions leads to a dilution factor of (182.13 ± 0.28) , which is $(0.11 \pm 0.16)\%$ lower than the gravimetric dilution factor. This good agreement implies that the disequilibrium was caused by loss of only ^{233}Pa , not ^{237}Np , from the solution during processing.

Since the disequilibrium was smaller for solution "S" than for SRM 4341a, the LTAC massic activity for the former, divided by the gravimetric dilution factor, was used as the confirmatory value for the SRM. This massic activity was $(151.8 \pm 0.3) \text{ Bq g}^{-1}$. Most of the uncertainty is due to the fits of the ingrowth curves and also the efficiency extrapolation, which was estimated based on the spread of extrapolations using one or more NaI gates and on Monte Carlo simulations.

2.3.3. $4\pi\gamma$ Ion chamber measurements

Two ampoules with different ^{237}Np activity levels were prepared for $4\pi\gamma$ ionization chamber measurements. Two ion chambers were used: NIST pressurized IC "A"; and Auto chamber IC "B". Details of each chamber can be found in Unterwieser and Fitzgerald (2012), and Fitzgerald (2010), respectively. Solutions "H" and "L" (see Fig. 1) with dilution factors of approximately 4 and 10 were measured every week for a period of 90 days in IC "A". Each ampoule and the Ra reference were manually inserted five times in IC "A". Hence each measurement point is the average of five measurements. The same solutions were measured in IC "B" nine times in a period of 100 days. Each ampoule and the Ra reference were automatically inserted four times in IC "B", so each measurement point is the average of four measurements. The results of both chambers showed a slight increase in massic counting rates for the first 2 weeks of measurements. The increase was approximately 0.5%. However, after 30 days and up to 90 days the data showed no discernible trend, which made us conclude that the solution had reached equilibrium.

In addition to checking for the stability of the solution, the ion chamber measurements were used to confirm the gravimetric

Table 2
Standard uncertainty components for the ^{237}Np massic activity of SRM 4341a, standardized by $4\pi\alpha\beta$ LS counting with ^3H -standard efficiency tracing for the ^{233}Pa correction.

Uncertainty component	Assessment type ^a	Relative standard uncertainty contribution on massic activity of ^{237}Np (%)
1 LS measurement precision; standard deviation of the mean for 4 sets of measurements obtained with 3 different LS counters each measured 3 to 5 times in each counter on 1 or 2 occasions. The typical internal relative standard deviation of the mean within a measurement data set was typically 0.03 % for $n=18$ to $n=30$ measurements with 6 LS sources	A	0.12
2 Background LS measurement variability and cocktail stability; wholly embodied in component 1	B	–
3 Live time determinations for LS counting time intervals, includes uncorrected dead time effects; assumed from specified tolerance limits of counters' gated oscillators	B	0.10
4 LS α -detection inefficiency for ^{237}Np	B	< 0.01
5 Gravimetric (mass) determinations for LS sources, dilution factors and counting source preparations	B	0.17
6 Decay corrections for ^{237}Np and ^{233}Pa ; half-life uncertainties of 0.07% and 0.33%, respectively [6]	B	2×10^{-7}
7 Assumed radioactive equilibrium between ^{237}Np and ^{233}Pa in the LS sources after 33 days of measurements	B	0.1
8 Decay corrections for ^3H half-life uncertainty of 0.16% [6]	B	0.002
9 Calculated beta efficiency for ^{233}Pa , including uncertainties in decay scheme data	B	0.4
10 Impurities, report of alpha impurity activity ratio to ^{237}Np of 0.0015 (5) from the 1997 EUROMET measurement comparison [6] of the master solution. No photon-emitting impurities were found. No ^{241}Am was found, indicating that beta-emitting ^{241}Pu was not present	B	0.05
Relative combined standard uncertainty		0.46
Relative expanded uncertainty ($k=2$)		0.92

^a Letter A, denotes evaluation by statistical methods; B denotes evaluation by other methods.

dilution ratio. This was done by determining the ratio of the solutions (H/L) on each of the measurement occasions and comparing it to the gravimetric dilution factor. It must be emphasized that the ion currents for solution “L” are small and exhibit considerable variability. The currents were a factor of 10 lower than the lower-limit used for the routine measurements. The H/L for IC “B” differed from the gravimetric dilution factor by -0.12% . In general, the IC “B” massic counting rates were higher than those obtained in IC “A”. The average value for the twelve measurements points in IC “A” differed from the gravimetric dilution factor by -1.2% , leading us to suspect that there was instrumental (electrometer) problems with IC “A”.

2.3.4. Gamma-ray spectrometry

Analysis for photon-emitting impurities was performed using a high purity intrinsic germanium detector: X-detector (length, $L=36$ mm; diameter, $\phi=43$ mm). The detector is an n-type coaxial detector with a 0.5 mm Be window. The detector and spectrometry procedures used by NIST have been described by Pibida et al. (2006, 2007). The ^{237}Np activity was determined using gamma-ray lines: 143.25 keV, 151.41 keV and 155.24 keV.

No impurity photons were detected, although the estimated lower limits of detection for photon-emitting impurities, expressed as massic photon emission rate, were relatively large compared to the ^{237}Np massic activity, namely in the range of $100 \text{ s}^{-1} \text{ g}^{-1}$ to $200 \text{ s}^{-1} \text{ g}^{-1}$.

2.3.5. NaI measurements

Select ampoules of SRM 4341a solution were measured in two NaI(Tl) well detectors to examine for the possible presence of ^{237}Np – ^{233}Pa disequilibrium. Sixteen randomly selected ampoules were measured on two occasions and of those 5 ampoules were selected and measured on 2 more occasions. The principal factor initially taken into account was the ratio of the integrated photo-peaks for the 390-keV and 1230-keV gamma-rays, representing primarily the ^{237}Np and ^{233}Pa daughter, respectively. No substantial change in this ratio with time was observed and we mistakenly concluded that the SRM solution was in radioactive equilibrium. Further thought revealed that the 390 keV peak also has a contribution from ^{233}Pa . Re-analyzing the initial data, in light of this, showed a similar level of $^{237}\text{Np}/^{233}\text{Pa}$ disequilibrium right

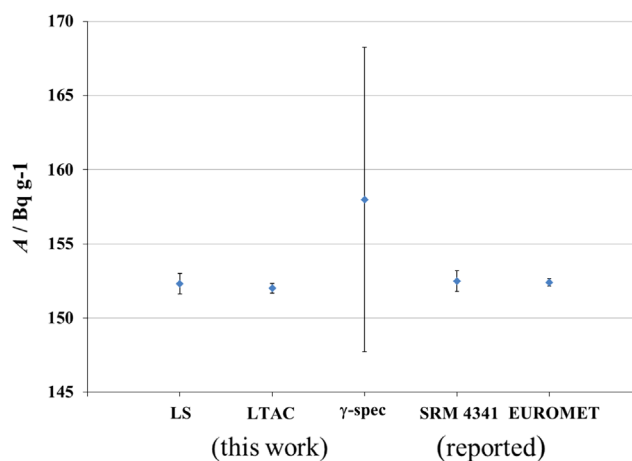


Fig. 3. A comparison of the results for the ^{237}Np massic activity. The uncertainty intervals on the data points represent the standard uncertainties ($k=1$) as determined by each method. The massic activity values reported for SRM 4341 (NIST, 1993) and EUROMET (Smith et al., 2001) were scaled relative to that from SRM 4341a for comparison here.

after the SRM was dispensed as observed by other methods. The principal finding obtained from the NaI measurements was that no difference was seen in the gamma-ray emission rates between the ampoules. This result implies that there were no solution instability problems to the extent that it would be discernible by the sensitivity of the NaI measurements.

3. Summary

The certified massic activity of ^{237}Np (SRM 4341a) was obtained from the $4\pi\alpha\beta$ liquid scintillation based standardization and it could be directly compared to the results obtained from the unweighted mean of nine primary standardizations by five laboratories that were performed in 1998–99 as part of the EUROMET ^{237}Np measurement comparison. Confirmatory standardizations of the ^{237}Np massic activity for SRM 4341a were obtained by LTAC and by γ -spec. A direct LS comparison of this SRM standardization was also made with a previous issue of ^{237}Np (SRM 4341), which was first disseminated in 1993. The results obtained by LTAC, LS

comparison of SRM 4341 and EUROMET report (applying the dilution factors used in this standardization), agreed to the LS CNET result to within -0.20% , $+0.13\%$ and $+0.07\%$, respectively. The γ -spec result differs from the LS CNET result by 3.7%, however, the results agree within the method's uncertainty. The results of these comparisons are presented in Fig. 3.

It was confirmed that the $^{237}\text{Np}/^{233}\text{Pa}$ equilibrium is disturbed when making dilutions and/or removing aliquots. It was observed that it is slightly disturbed during dilutions, as could be seen in IC data when equilibrium was reestablished two weeks after preparation. However, additional dilutions of the solutions (i.e., solution “L” as compared to “S”) showed a higher level of disequilibrium. A marked disequilibrium was observed in the LS sources. Equilibrium was not obtained until 33 days after source preparation (i.e., $t=65$ d). This effect, however, was taken into account in all the determinations.

The ^{237}Np solution standards that will be disseminated by NIST as SRM 4341a are contained in 5-mL, flame-sealed borosilicate glass ampoules, and consist of (5.3196 ± 0.0003) g of a carrier-free nominal 2 mol L^{-1} nitric acid solution, having a density of $(1.067 \pm 0.002) \text{ g mL}^{-1}$ at 16.3°C .

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Lizbeth Laureano-Pérez was responsible for coordinating all aspects of this work, assisted in the wet-lab work, and performed all aspects of the LS measurements and data analyses. R. Fitzgerald independently performed all of the LTAC measurements, and served as the project's devil's advocate. R. Collé did the solution manipulation and gravimetric work (dilutions, source preparations; and dispensing); performed all of the NaI measurements; and provided oversight, data reconciliation, and final analyses and interpretations leading to certification of the standardization. The authors would like to gratefully acknowledge Leticia Pibida for carrying out the photonic emission impurity analyses; and particularly thank Daniel Golas for cheerfully assisting us in flame sealing all of the SRM ampoules during their preparation.

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