NIST Technical Note 2192

Bilateral comparison of the massic activity for a standardized ^{137}Cs solution between the National Institute of Standards Technology (NIST) and the National Institute of Nuclear Research (ININ)

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National Institute of Standards and Technology
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Abstract

National metrology institutes (NMIs) around the world keep standards for activity measurements for a large number of radionuclides. An important part of this work is to compare measurement capabilities between these institutes. The National Institute of Standards and Technology (NIST) in the United States of America and the National Institute of Nuclear Research (ININ) in Mexico performed a bilateral comparison for the determination of the massic activity of a ^{137}Cs solution. This report summarizes the results of this comparison.

Key words

Bilateral comparison, Cs-137 solution, NIST, ININ.

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1. Introduction

National metrology institutes (NMIs) around the world keep standards for activity measurements for a large number of radionuclides. An important part of this work is to compare measurement capabilities between these institutes. The National Institute of Standards and Technology (NIST) in the United States of America and the National Institute of Nuclear Research (ININ) in Mexico performed a bilateral comparison for the determination of the massic activity of a ^{137}Cs solution. This report summarizes the results of this comparison.

2. Source preparation at NIST

The ^{137}Cs source (number 2519-8) was provided by the NIST Radioisotope Measurement Assurance Program from a bulk solution (ID 2519), prepared on 2 May 2019 and having a chemical composition of approximately 108 micrograms of CsCl per gram, with a density of (1.018 ± 0.002) g/mL at 20 ºC in a 5 mL flame sealed borosilicate ampoule. The massic activity of the solution was 498.5 kBq·g^{-1}, with a relative expanded uncertainty ($k = 2$) of 0.90 % as of 2 May 2019, 1200 EST. The bulk solution was dispended into ten (10) 5 mL ampoules. Ampoule 2519-8 was used for this comparison, having a nominal mass of (5.103 ± 0.003) g. The half-life value used for the determination of the massic activity was (30.05 ± 0.08) years [Ref. 1]. The massic activity was measured using a NIST 4π-γ pressurized ionization chamber. The NIST 4π-γ pressurized ionization chamber had been calibrated in 1982 using a ^{137}Cs solution standardized by 4π beta-gamma anti-coincidence. [Ref. 4]

Source 2419-8 was shipped from NIST to ININ in June 2020.

3. Source preparation at ININ

The NIST ampoule source 2519-8 was measured as received using a re-entrant ionization chamber ("dose calibrator") at the ININ that was calibrated in terms of activity. The dose calibrator was calibrated using a ^{137}Cs reference source with traceability to NIST with 7.385 MBq as of 1 December 2006, 12:00 PST. This "as received" value for the activity of the ampoule was (2516 ± 13.1) kBq ($k = 2$) as of 21 October 2020, 12:00 PM EST. Part of this intercomparison included the production and measurement of point sources at ININ. Therefore, after the dose calibrator measurements, the ampoule was opened and a quantity of the ^{137}Cs solution was extracted from the NIST 2519-8 ampoule using a pycnometer to prepare five point sources. For the mass measurements when preparing each point source, the masses of the deposited solution were determined by mass difference get with a microbalance Mettler-Toledo, model XP-26. The masses for each of the point sources are listed in **Table 1**.

Table 1: List of masses and combined standard uncertainty (k = 2) for the point sources made at ININ

Source identification number	Deposited mass (g)
1CsNIST	(0.046 ± 0.001)
2CsNIST	(0.046 ± 0.001)

3CsNIST	(0.046 ± 0.001)
4CsNIST	(0.052 ± 0.001)
5CsNIST	(0.047 ± 0.001)

4. Measurement setup using the HPGe detectors at ININ

Therefore, the mass activity concentration in units of $kBq \cdot g^{-1}$ of 5 different point sources was measured using two HPGe detectors. For the activity measurements at ININ two high purity germanium (HPGe) detectors were used. One HPGe detector was a CANBERRA Industries detector (the detector characteristics are listed in **Table 2**)[1] that was calibrated for efficiency using as reference a point standard source (Areva, Cerca LEA ^{137}Cs, Serial No. 50488, traceable to the Laboratoire National Henri Becquerel (LNHB) with Comité Français d'accréditation (COFRAC) certificate Accreditation No 2-1529) at a source to detector distance of (10.74 ± 0.02) cm. The source activity reported in the Calibration Certificate as of 11 September 2011 was (3.25 ± 0.05) × 10^4 Bq (k = 2). The full energy peak efficiency at 661.66 keV was determined to be equal to 1.389 x 10^{-3} (± 1.67 %) (k = 2). The point sources prepared from the NIST solution were measured using this HPGe detector. Measurements were performed on 21 October 2020 with a typical measurement time of 900 s, the dead time was approximately 0.35 % and the source to detector distance was (10.74 ± 0.02) cm. Five point sources were prepared with the NIST solution and one measurement for each point source was performed.

The nuclear data used for the measurements were obtained from the LNHB, see Ref. 1.

Table 2: ININ Canberra HPGe detector characteristics

Detector Class: HPGe	**Manufacturer: CANBERRA**
Model: 7229P	Serial: 88513223
Number of detectors: 1	Solid angle: 0.12 sr.
Type: P	Coaxial: YES
Diameter: 42 mm	Volume: 19180 mm^3
Material of window: Aluminum	
Window thickness: 5 mm	
Distance between source and photon counter (mm): 107.35 ± 0.02	
Original value: Resolution at: 1.33 MeV	FWHM: 1.71 keV
Actual value: Resolution at: 1.33 MeV	FWHM: 2.8 keV

[1] Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

The second HPGe detector was an ORTEC detector (the HPGe detector characteristics are listed in **Table 3**) that was calibrated for efficiency using the same reference point standard source (Areva, Cerca LEA ^{137}Cs, Serial No. 50488) at a source to detector distance of (21.73 ± 0.09) cm. The source activity reported in the Calibration Certificate as of 11 September 2011 was (3.25 ± 0.05) × 10^4 Bq (k = 2). The full energy peak efficiency at 661.66 keV was determined to be equal to 8.83 x 10^{-4} ± 1.66 % (k = 2). The point sources prepared from the NIST ampoule source 2519-8 solution were measured using this HPGe detector. Measurements were performed on 21 October 2020 with a typical measurement time of 1200 s, the dead time was approximately 0.35 % and the source to detector distance was 21.73 cm. Five point sources were prepared with the NIST solution and one measurement for each point source was performed.

Table 3: ININ ORTEC HPGe detector characteristics

Detector Class: HPGe	**Manufacturer: ORTEC**
Model: GMX30-70-LB-B	Serial: 53-N13407A
Number of detectors: 1	Solid angle: 0.06 sr.
Type: N	Coaxial: YES
Diameter: 57.1 mm	Volume: 1618.4 mm^3
Material of window: Aluminum	
Window thickness: 3 mm	
Distance between source and photon counter (mm): 217.26 ± 0.09	
Resolution at: 1.33 MeV	FWHM: 1.90 keV

5. Results of the measurements at ININ

Preliminary measurements of the NIST ampoule source 2519-8 were performed prior to opening it using a dose calibrator. The activity of the solution was measured to be equal to (2516 ± 13) kBq (k = 2) as of 21 October 2020, 12:00 PM EST.

The measurements performed of the 5-point sources in each HPGe detector were averaged to obtain the massic activity for the source provided by NIST. The results are summarized in

Table **4**.

Table 4: Results of measured massic activity performed at ININ

HPGe Detector	**Massic activity ($kBq \cdot g^{-1}$)**	**Reference time**
Canberra detector	473.7 ± 12.9 (k = 2)	22 October 2020, 12:00 PM EST
ORTEC detector	472.7 ± 14.2 (k = 2)	21 October 2020, 12:00 PM EST

The uncertainty analysis for the measurements performed using both of the HPGe detectors are listed in **Table 5** and **Table 6**.

Table 5: Uncertainty analysis for measurements performed using the ININ Canberra HPGe detector

Sources of Uncertainty	**Relative standard Uncertainty expressed as a percentage**
Due to the measurement of the sources and the method of measurement	
Counting statistics	0.58 %
Photopeak area	0.27 %
Efficiency calibration	1.67 %
Source to Detector Distance	0.14 %
Gross Area	0.21 %
Thickness of source supporting material	0.01 %
Dead time	0.35 %
Pile-up	0.067 %
Emission intensity	0.2 %
Counting time	0.01 %
Repeatability	0.39 %
Impurities	UNDETECTED
Due to the Weighing	
Balance Certificate of Calibration	
Eccentricity	1.73E-06
Resolution	2.89E-07
Air Buoyancy Corrections	1.28E-06
Maximum Permissible Errors (EMT or EMP, characteristic of the balance)	5.77E-04
Repeatability	3.46E-06
Combined standard uncertainty, μ_c	***1.35 %***
Relative expanded uncertainty (k = 2), U	***2.7 %***

Table 6: Uncertainty analysis for measurements performed using the ININ ORTEC HPGe detector

Sources of Uncertainty	**Relative standard Uncertainty expressed as a percentage**
Due to the measurement of the sources and the method of measurement	
Counting statistics	1.24%
Photopeak area	0.73%
Efficiency calibration	1.66 %

Source to Detector Distance	0.40%
Gross Area	0.28%
Thickness of source supporting material	0.01 %
Dead time	0.35 %
Pile-up	0.0056 %
Emission intensity	0.20 %
Counting time	0.01 %
Repeatability	0.55 %
Impurities	UNDETECTED
Due to the Weighing	
Balance Certificate of Calibration	
Eccentricity	1.73E-06
Resolution	2.89E-07
Air Buoyancy Corrections	1.28E-06
Maximum Permissible Errors (EMT or EMP)	5.77E-04
Repeatability	1.47E-05
Combined standard uncertainty, μ_c	***1.50 %***
Relative expanded uncertainty (k = 2), U	***3.0 %***

6. Measurements at NIST

Each of the ten (10) samples prepared from the bulk solution were measured in the NIST 4π-γ reentrant ionization chamber "A" that is maintained as a secondary standard for γ-ray emitting radioisotopes [Ref. 4]. The standard procedure for using a pressurized ion chamber such as the NIST 4π-γ reentrant ionization chamber "A", are described by Calhoun [Ref. 2]. The measured quantity is the ratio, R, of the detector response for a given radionuclide of activity, A, to that of a NIST radium-226 reference source. A calibration factor (K-value) for a given radionuclide is defined as $K = A/R$. The K value for ^{137}Cs was determined in 1982 using a ^{137}Cs solution that had been standardized by the primary measurement method of 4π β-γ anti-coincidence counting with efficiency tracing [Ref. 5 and 6]. That K value, after small ^{226}Ra decay correction, was used to calibration solution 2519 in 2019. Each of the ten (10) ampoules was measured on five (5) separate occasions (insertions), resulting in 50 measurements of the original solution 2519. Additionally, NIST radium reference source number 50 (RRS50) was counted 12 times on six (6) different occasions (insertions) throughout the measurement interval, resulting in 72 measurements. After applying the decay corrected K-value for ^{137}Cs a resultant average activity of 498.5 kBq·g^{-1} and a counting precision, including the within and in-between insertion components of variance, of 0.11% (k = 1) was obtained. The uncertainty budget for these measurements is listed in Table 7. No gamma-ray emitting impurities were present in solution 2519; the limits of detection for photon-emitting impurities, as of 7 May 2019, are listed in Table 8.

Table 7: Uncertainty analysis for the NIST ^{137}Cs massic activity measurement of solution 2419.

Uncertainty Component	Uncertainty value (%)
4π-γ pressurized ionization chamber calibration factor	0.42
50 ionization-chamber measurements on this solution	0.11
photon-emitting impurities in this solution	< 0.001
radium-226 reference source ratios (N = 72)	0.01
radium reference source positioning	0.05
radium reference source decay correction	0.01
electrometer response linearity	0.10
decay correction	<0.001
gravimetric measurements	0.05
detection limits for photon-emitting impurities	0.01
Combined standard uncertainty, μ_c	***0.45***
Relative expanded uncertainty (k = 2), U	***0.90***

Table 8: Detection limits for photon-emitting impurities as of 7 May 2019

Limits of detection ($\gamma \bullet s^{-1} \bullet g^{-1}$)	Energy region (keV)
170	25 – 40
97	45 – 130
170	135 – 640
720	650 – 670
14	680 – 2000

In addition to these measurements, the massic activity of the 2519-8 ampoule source was also measured (as a check) using 3 different HPGe detectors (1 measurement in each detector). The massic activity of the solution was determined to be equal to 496.1 $kBq \cdot g^{-1}$, with a relative expanded uncertainty ($k = 2$) of 0.90 % as of 2 May 2019, 1200 EST.

7. Comparison of measurement results

The measurements performed at ININ were compared to the NIST certified value obtained from the measurements made using the NIST 4π-γ reentrant ionization chamber "A", see Table 9. In addition, the NIST measurements performed as a check in the HPGe detectors are also listed.

Table 9: Summary of massic activity measurements at a reference time of 2 May 2019 12:00 PM EST. The uncertainties on massic activity are combined standard uncertainty.

Detector description	Massic activity ($kBq \cdot g^{-1}$)	Difference with NIST value (%)	U_D (%) (k=2)
NIST 4π-γ reentrant ionization chamber*	498.5 ± 2.2	-	-
NIST HPGe detectors	496.1 ± 3.8	-0.5	1.8
ININ Dose calibrator	510.1 ± 4.2	2.3	1.9
ININ Canberra detector**	490.1 ± 6.7	-1.7	2.9
ININ ORTEC detector	489.1 ± 7.3	-1.9	3.1
* NIST value used for the comparison ** ININ value used for the comparison			

The final reported value by the ININ is the one obtained using the ININ Canberra detector (marked with two asterisks in Table 9), the other measurements were performed as secondary measurements to support the reported value. For this comparison the ININ Canberra detector value is compared with the NIST 4π-γ reentrant ionization chamber. Therefore, the ININ value is -1.7 % different from the NIST reference value.

8. Conclusions

A solution of 137Cs was prepared at NIST and calibrated using a 4π-γ reentrant ionization chamber (source certificate in Annex A). Prior to shipping a gravimetrically-prepared ampoule of the solution to the ININ, the ampoule was measured in multiple NIST HPGe detectors to confirm the activity. Upon arrival to the ININ the source was measured in a dose calibrator, after which several point sources were prepared and measured in 2 HPGe detectors. The measured value at ININ agrees with the NIST reference value to (1.7 ± 2.9) % (k = 2).

References

[1] Laboratoire National Henri Becquerel, C.E.A. Saclay - Gif-sur-Yvette Cedex - France. Table of Radionuclides, Recommended Data (on-line), updated January 14, 2018. Available at http://www.nucleide.org/DDEP_WG/DDEPdata.htm. Last accessed on February 2019.

[2] B.N. Taylor and C.E. Kuyatt (1994). Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results. NIST Technical Note 1297.

[3] Laboratoire National Henri Becquerel, C.E.A. Saclay - Gif-sur-Yvette Cedex - France. Table of Radionuclides, Recommended Data (on-line), updated January 14, 2018. Available at http://www.nucleide.org/DDEP_WG/DDEPdata.htm. Last accessed on February 2019.

[4] J. M. Calhoun (1986). Radioactivity Calibrations with the NBS "4π"γ Ionization Chamber, and Other NBS Radioactivity Calibration Capabilities, NBS SP 250-10, Gaithersburg, MD, USA.

[5] A. Rytz (1982). International comparison of activity measurements of a solution of ^{137}Cs. Rapport BIPM-83/8. https://www.bipm.org/en/publications/rapports-bipm-1981-1990

[6] A. Rytz (1982). International comparison of activity measurements of a solution of ^{137}Cs Preliminary report Rapport BIPM-82/14. https://www.bipm.org/en/publications/rapports-bipm-1981-1990

Appendix A: Copy of Test report

U.S. DEPARTMENT OF COMMERCE
National Institute of Standards & Technology
Gaithersburg, MD 20899

REPORT OF TEST

for

Instituto Nacional De Investigaciones Nucleares
La Marquesa, Ocoyoacac, México

Radionuclide	Cesium-137
Source identification	2519-8
Source description	Liquid in a 5 mL flame-sealed borosilicate glass ampoule
Solution composition	Approximately 108 micrograms of CsCl per gram of approximately 1.1 mol•L^{-1} hydrochloric acid [1]*
Mass	5.103 ± 0.003 grams
Massic activity	498.5 kBq•g^{-1}
Reference time	1200 EST May 2, 2019
Relative expanded uncertainty	0.90 percent [2]
Photon-emitting impurities (at reference time)	None observed [3]
Half-life	30.05 ± 0.08 years [4]
Measurement method	NIST pressurized "4π"γ ionization chamber "B" calibrated by 4$\pi\beta$-γ anticoincidence efficiency-extrapolation technique

For the Director,

Gaithersburg, MD 20899
January 2020

Brian E. Zimmerman, Leader
Radioactivity Group
Physical Measurement Laboratory

*Notes on back

NOTES

[1] The solution density is 1.018 ± 0.002 g•mL^{-1} at 20°C.

[2] The uncertainty analysis methodology and nomenclature used for the reported uncertainties are based on uniform NIST guidelines and are compatible with those adopted by the principal international metrology standardization bodies [cf., B.N. Taylor and C.E. Kuyatt, *NIST Technical Note 1297* (1994)].

The relative **combined standard uncertainty**, $u_c = 0.45$ percent, is the quadratic combination of the standard deviations (or standard deviations of the mean where appropriate), or approximations thereof, for the following component uncertainties:

a) "4π"γ pressurized ionization chamber calibration factor	0.42 percent
b) 50 ionization-chamber measurements on this solution	0.11 percent
c) photon-emitting impurities in this solution	< 0.001 percent
d) radium-226 reference source ratios	0.01 percent
e) radium reference source positioning	0.05 percent
f) radium reference source decay correction	0.01 percent
g) electrometer response linearity	0.10 percent
h) decay correction	<0.001 percent
i) gravimetric measurements	0.05 percent
j) detection limits for photon-emitting impurities	0.01 percent

The **relative expanded uncertainty**, $U = 0.90$ percent, is obtained by multiplying u_c by a coverage factor of $k = 2$ and is assumed to provide an uncertainty interval of approximately 95 percent confidence.

[3] Estimated limits of detection for photon-emitting impurities, as of May 7, 2019, were:

1.7×10^2 γ•s^{-1}•g^{-1} for energies between 25 and 40 keV,
9.7×10^1 γ•s^{-1}•g^{-1} for energies between 45 and 130 keV,
1.7×10^2 γ•s^{-1}•g^{-1} for energies between 135 and 640 keV,
7.2×10^2 γ•s^{-1}•g^{-1} for energies between 650 and 670 keV,
1.4×10^1 γ•s^{-1}•g^{-1} for energies between 680 and 2000 keV,

provided that the impurity photons are separated in energy by four keV or more from photons emitted in the decay of cesium-137.

[4] Laboratoire National Henri Becquerel, C.E.A. Saclay - Gif-sur-Yvette Cedex - France. Table of Radionuclides, Recommended Data (on-line), updated January 14, 2018. Available at http://www.nucleide.org/DDEP_WG/DDEPdata.htm. Last accessed on February 2019.