International comparison of activity measurements of a solution of ^{51}Cr

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1. Introduction

A bilateral comparison between the Laboratoire National Henri Becquerel, belonging to CEA (France) and named LNE-LNHB, and the National Centre for Nuclear Research Radioisotope Centre POLATOM (Poland) on activity measurements of a solution of ^{51}Cr was organized in the second half of the year 2022 in order to link both the LNE-LNHB and POLATOM to the BIPM International Reference System (SIR). Table 1 lists the participating laboratories and indicates also the persons who carried out the measurements. The project was declared as a EURAMET.RI(II)-K2.Cr-51 key comparison and was registered in the EURAMET database under the number 1554. An extra ampoule, prepared by the POLATOM from the same batch used for the bilateral comparison between the POLATOM and the LNE-LNHB for measurements, was sent to the BIPM.

The exercise started in the third week of May 2022 when a master ^{51}Cr solution was prepared at POLATOM and measurements ended 21 November 2022. A schedule of the inter-comparison was given in the technical protocol for an EURAMET key comparison of ^{51}Cr accepted by the BIPM. The measurand for this exercise was the massic activity in the diluted ^{51}Cr solution.

2. Decay scheme

Chromium-51 decays by electron capture to the ground state of ^{51}V (90.09%) and to the 320.0835 keV excited level (9.91%). The decay scheme can be seen in Fig. 1, which has been taken from the Table de Radionucléides [1]. The recommended half-life value $T_{1/2}$ = 27.704 (4) d was taken by participants. The less intense electron-capture transition is followed by the γ transition with energy of 320.0835 keV to the ground state of ^{51}V.

3. Characteristics of the solution

The radioactive material was prepared at POLATOM in the form of $^{51}CrCl_2$ in carrier solution containing 25 μg Cr as Na_2CrO_4 in 1 mL 0.1 M HCl. The master solution BW/09/22 with an approximate massic activity of 313 $MBq{\cdot}g^{-1}$ on 1 June 2022 was obtained. The master solution was diluted (solution BW/09/22/R_1) and the following glass ampoules were prepared: two type BIPM SIR and two type P6, filled in order to obtain a volume of about 3.6 mL of liquid solution. The dilution factor was R_1 = 45.37628. The type BIPM SIR glass ampoules were measured at POLATOM in an ionization chamber over two weeks.

One P6 glass ampoule and one glass ampoule type BIPM SIR containing 3.62244 g of diluted solution were sent to LNE-LNHB by 1 June 2022. The massic activity in the diluted solution contained in the BIPM SIR glass ampoule was measured in an ionization chamber. Six liquid scintillation sources were prepared at LNE-LNHB in sand-blasted vials filled with 10 mL Ultima Gold scintillator mixed with aliquots (~ 27 mg) of the solution taken from the P6 glass ampoule. The above sources were measured at LNE-LNHB using the liquid scintillation counting (LSC) technique.

The second glass ampoule type BIPM SIR contained 3.64199 g of diluted solution and was sent to the BIPM on 1 June 2022 in order to link the results achieved in the bilateral comparison to the BIPM SIR.

The second P6 ampoule was used at POLATOM to prepare a set of six sources in 20 mL high-performance PerkinElmer sand-blasted glass vials filled-in with 10 mL of Ultima Gold liquid scintillator, containing a mass of radioactive material ranging from 39.2 mg to 40.7 mg. The glass vials were measured at POLATOM using the LSC technique over a period of one week.

No impurities were detected by γ-ray spectrometry. An upper limit for a potential impurity of ^{60}Co was determined to be $A(^{60}\text{Co})/A(^{51}\text{Cr}) \leq 7.9\ 10^{-7}$, which should not influence the results of the intercomparison.

4. Measurement methods used

The ^{51}Cr diluted solution had been standardized at both LNE-LNHB and POLATOM by using primary activity measurements techniques, based on 4π(LS)-γ coincidence and anti-coincidence counting methods. The LNE-LNHB also made measurements with the 4πγ method using a large well-type NaI(Tl) detector. Sources used for the measurements were prepared by drop deposition of aliquots of the solution using the pycnometer technique (difference weighing) with masses between 10 and 30 mg and subsequent drying of the samples. The detection efficiency was calculated using the Geant4 code on the basis of a comprehensive modelling of the detection system (CPG1). Correction for the zero-energy extrapolation was applied to compensate for counting losses under the detection threshold. This measurement of LNE-LNHB activity was only indicative, as the calculated detection efficiency was very low for ^{51}Cr (about 9%).

The list of the methods used by both laboratories is given in Table 2. The methods used are also indicated in Tables 3a, 3b, 4 and 5 as well as in Fig. 2.

The POLATOM system was equipped with the CAMAC electronic modules for amplification, analysis and recording of pulses. The LNE-LNHB anti-coincidence system was equipped with CAMAC and NIM modules for amplification and for the acquisition of γ-spectra. Home-made modules based on extendable dead-time combined to a live time circuitry were used for counting [3, 4].

The measuring system used at POLATOM were:

- 4π(LS)-γ coincidence and anti-coincidence measuring system with two photomultipliers type EMI 9899 B working in coincidence and two scintillation detectors with NaI(Tl) crystals working in the sum system [2];

The measuring systems used at LNE-LNHB were:

- 4π(LS)-γ anti-coincidence counting system based on a TDCR-3PMTs apparatus in the beta channel and a scintillation detector NaI(Tl) 3”x3” crystal in the gamma channel. The TDCR detector is equipped with 3 XP2020Q PMTs.
- Large 152 mm x 127 mm well-type NaI(Tl) detector with a central well of 21 mm diameter and 47.5 mm height used for 4πγ counting [5]. A MTR2 module was used for counting [3].

5. Uncertainties

All contributions to the total uncertainty for each individual method used in both laboratories are detailed in Table 3a for POLATOM and Table 3b for LNE-LNHB. The uncertainty evaluation type is also given as provided by the laboratories.

6. Results

Four values of massic activity given at the reference date of 1 June 2022, 12:00 h UTC and obtained using all the methods implemented by both laboratories, are given in Table 4 and shown in Fig. 2.

Following a provision of the CIPM Mutual Recognition Arrangement [6], the outcome of an international comparison should not give several results for one laboratory. Both laboratories selected for this reason one of its values obtained using the most robust among the methods they used: LNE-LNHB - the value obtained using the 4π(LS)-γ anti-coincidence method and POLATOM - the weighted mean value of the 4π(LS)-γ coincidence and 4π(LS)-γ anti-coincidence method results. Table 5 has been constructed following the preferences of the laboratories.

The power-moderated mean of the values given in Table 5 yields a massic activity equal to $\overline{A}$ = 6828 kBq·g^{-1}; u = 20 kBq·g^{-1}, 0.30%. All the results in Table 4 are comprised in a range of (+0.12%, –0.47%) of the weighted mean.

Employing the convention used in CCRI(II) [7], the degree of equivalence between the participants, *Dij*, was calculated by :

$$D_{ij} = x_i - x_j$$

where in this case x_i and x_j are the ^{51}Cr massic activity obtained for the solution at LNE-LNHB and at POLATOM, respectively, given in Table 5. The expanded uncertainty, U_{ij}, of D_{ij} is given by

$$U_{ij} = 2\sqrt{(u_i)^2 + (u_j)^2}$$

assuming that x_i and x_j are not correlated and where u_i and u_j are the standard uncertainty (k=1) of the massic activity estimated at LNE-LNHB and at POLATOM, respectively.

The performances of the two laboratories were evaluated by computing the quantity D_{ij}/U_{ij}. The results x_i and x_j are considered to be in agreement if the absolute value of D_{ij}/U_{ij} is lower than 1. The degree of equivalence was calculated to be D_{ij} = 41 kBq·g^{-1}; U_{ij} = 93 kBq·g^{-1} (k = 2); the results of both laboratories are in excellent agreement. The D_{ij}/U_{ij} values of any pair of results are computed and listed in Table 6. Because all absolute values of D_{ij}/U_{ij} are significantly lower than 1, this confirms that the results obtained by both laboratories are in good agreement.

References

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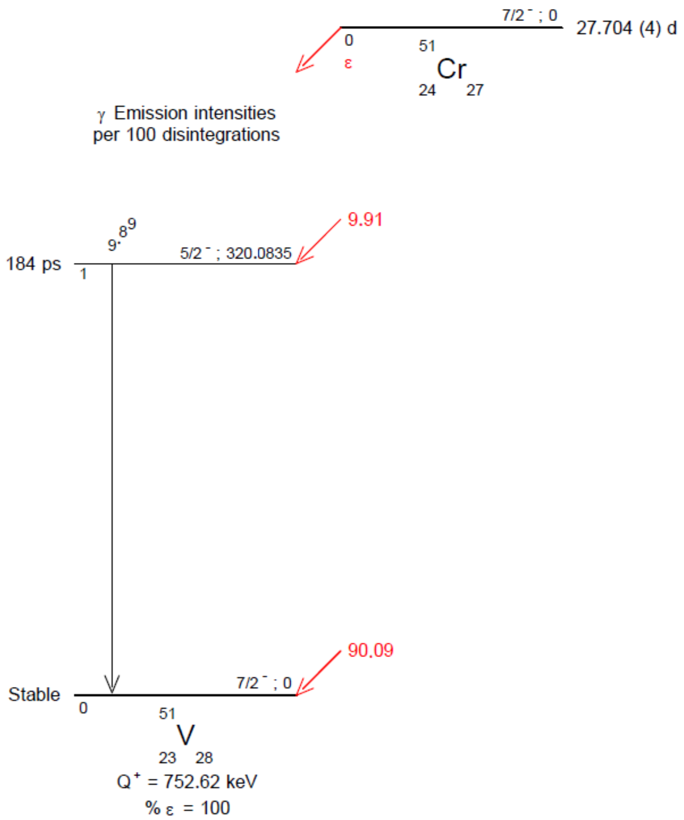

Figure 1 Decay scheme of ^{51}Cr taken from V.P. Chechev, N.K. Kuzmenko. *Table de Radionucléides*, LNE-LNHB/CEA, Saclay 02/04/2014 – 06/05/2014.

Table 1 List of participants

POLATOM	National Centre for Nuclear Research Radioisotope Centre POLATOM, Otwock, Poland (T. Ziemek, J. Marganiec-Gałązka, P. Saganowski and Z. Tymiński, measurements, A. Listkowska and E. Lech, source preparation)
LNE-LNHB	Université Paris-Saclay, CEA, List, Laboratoire National Henri Becquerel (LNE-LNHB), F-91120 Palaiseau, France (Ch. Bobin and Ch. Thiam for measurements and L. Chambon and V. Lourenço for source preparation)

Table 2 List of the methods used

Name of the method and its acronym	Description of the method	Description of the detector	Laboratories
4π(LS)-γ coincidence 4P-LS-MX-NA-GR-CO	4π liquid-scintillation coincidence counting	4π liquid-scintillation coincidence detector with two NaI(Tl) detectors in the γ channel	POLATOM
4π(LS)-γ anti-coincidence 4P-LS-MX-NA-GR-AC	4π liquid-scintillation anti-coincidence counting	4π liquid-scintillation coincidence detector with two NaI(Tl) detectors in the γ channel	POLATOM
4π(LS)-γ anti-coincidence 4P-LS-MX-NA-GR-AC	4π(LS)-γ anti-coincidence counting	TDCG detector (a liquid-scintillation triple detector and a NaI(Tl) detector in the γ channel)	LNE-LNHB
4πγ high efficiency 4P-NA-GR-00-00-HE	4πγ high efficiency counting	A large well-type NaI(Tl) detector	LNE-LNHB

Table 3a Uncertainty assessment for the ^{51}Cr massic activity where *u* is a relative standard uncertainty (*k* = 1) measured at POLATOM

Laboratory		POLATOM	
Method		4π(LS)-γ coincidence	4π(LS)-γ anti-coincidence
Uncertainty component	Assessment type	*u* (%)	*u* (%)
Counting statistics	A	0.76	0.7
Weighing	A	0.05	0.05
Dead time	B	0.001	0.001
Resolving time	B	0.023	0.023
Decay correction	A	0.06	0.06
Extrapolation of efficiency curve	A	0.45	0.047
Anti-coincidence window	B	-	0.031
Combined uncertainty (as quadratic sum of all uncertainty components)	A	0.887	0.847
	B	0.023	0.039
	total	0.887	0.848

Table 3b Uncertainty assessment for the ^{51}Cr massic activity where *u* is a relative standard uncertainty (*k* = 1) measured at LNE-LNHB

Laboratory		LNE-LNHB	
Method		4π(LS)-γ anti-coincidence	4πγ high efficiency
Uncertainty component	Assessment type	*u* (%)	*u* (%)
Counting statistics	A	0.04	0.2
Background	B	0.16	0.1
Weighing	B	0.1	0.05
Dead time	B	0.02	0.01
Decay correction	B	0.06	0.013
Decay data	B	0.04	0.3
Extrapolation of efficiency curve	A	0.23	-
Zero-energy extrapolation	B	-	0.2
Detection efficiency	B	-	0.7
Combined uncertainty (as quadratic sum of all uncertainty components)	A	0.233	0.224
	B	0.203	0.789
	total	0.309	0.820

Table 4 Results

Laboratory Method	Massic activity ($kBq \cdot g^{-1}$)	Combined standard uncertainty ($kBq \cdot g^{-1}$)	Relative standard uncertainty (%)
POLATOM			
4π(LS)-γ coincidence	6799	60	0.887
4π(LS)-γ anti-coincidence	6809	58	0.848
LNE-LNHB			
4π(LS)-γ anti-coincidence	6845	21	0.309
4πγ high efficiency	6800	56	0.820

Table 5 Results; one value per designated institute

Laboratory	Massic activity ($kBq \cdot g^{-1}$)	Combined standard uncertainty ($kBq \cdot g^{-1}$)	Relative standard uncertainty (%)
POLATOM [1)]	6804	42	0.613
LNE-LNHB [2)]	6845	21	0.309

[1)] result obtained as a weighted mean of 4π(LS)-γ coincidence and 4π(LS)-γ anti-coincidence method results.
[2)] final result from the 4π(LS)-γ anti-coincidence method only.

Table 6 D_{ij}/U_{ij}. values of any pair of results for the ^{51}Cr massic activity

LNE-LNHB / POLATOM	4π(LS)-γ anti-coincidence	4πγ high efficiency
4π(LS)-γ coincidence	0.36	0.01
4π(LS)-γ anti-coincidence	0.29	0.06

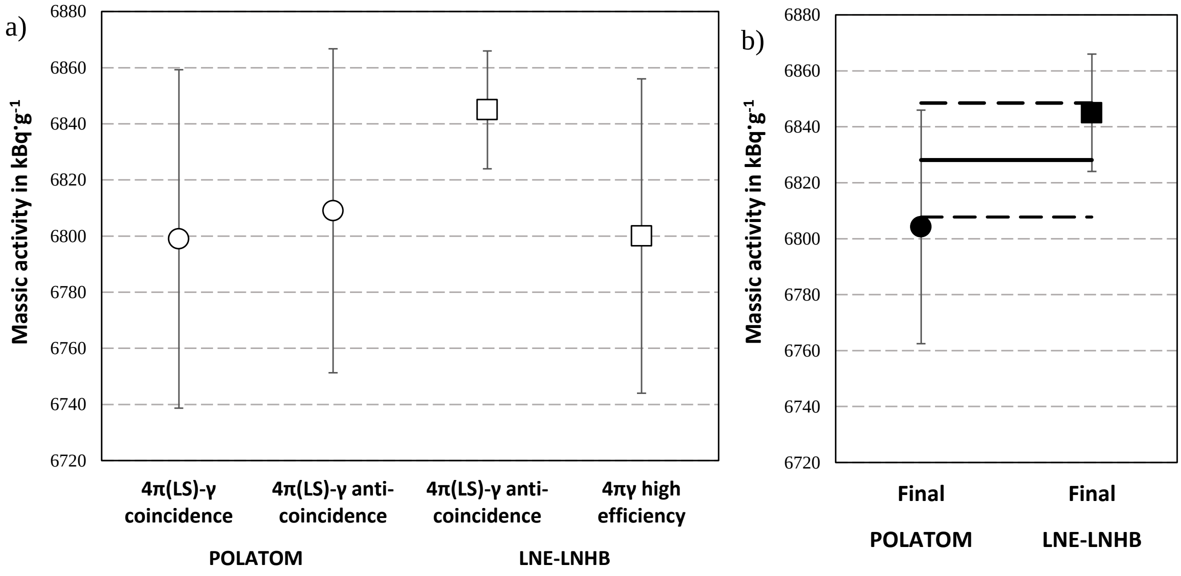

Fig. 2 Final results of the ^{51}Cr bilateral comparison of massic activity. a) All results reported by laboratories. b) The final results reported by POLATOM and LNE-LNHB. The power-moderated mean value (solid line) is shown with its uncertainty (dotted lines).