Activity measurements of the radionuclide ^{124}Sb by the LNE-LNHB, France as a pilot study for the ongoing comparison BIPM.RI(II)-K1.Sb-124

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Abstract

In 2005, the Laboratoire national de métrologie et d'essais - Laboratoire national Henri Becquerel (LNE-LNHB) submitted a sample of known activity of ^{124}Sb to the International Reference System (SIR) in the frame of a pilot study. The value of the activity submitted was about 5.9 MBq. The comparison result shows agreement within the standard uncertainty with the published result for France in the key comparison identified as BIPM.RI(II)-K1.Sb-124.

1. Introduction

The SIR for activity measurements of γ-ray-emitting radionuclides was established in 1976. Each national metrology institute (NMI) may request a standard ampoule from the BIPM that is then filled (3.6 g) with the radionuclide in liquid (or gaseous) form. The NMI completes a submission form that details the standardization method used to determine the absolute activity of the radionuclide and the full uncertainty budget for the evaluation. The ampoules are sent to the BIPM where they are compared with standard sources of ^{226}Ra using pressurized ionization chambers (IC). Details of the SIR method, experimental set-up and the determination of the equivalent activity, A_e, are all given in [1].

From its inception until 31 December 2005, the SIR has measured 891 ampoules to give 651 independent results for 63 different radionuclides. The SIR makes it possible for national laboratories to check the reliability of their activity measurements at any time. This is achieved by the determination of the equivalent activity of the radionuclide and by comparison of the result with the key comparison reference value determined from the results of primary realizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the BIPM key comparison database (KCDB) that was set up under the CIPM Mutual Recognition Arrangement (MRA) [2]. The comparison referred to in this report is known as the BIPM.RI(II)-K1.Sb-124 key comparison.

In 2005, the LNE-LNHB submitted a ^{124}Sb ampoule in the frame of a pilot study and not for the purpose of producing degrees of equivalence. This submission enables the LNE-LNHB to check its SIR result in preparation for a possible future EUROMET exercise. In this report, the pilot result is compared to previous ^{124}Sb SIR results and to predicted equivalent activity values obtained from the SIR efficiency curves.

2. Participants

Apart from the present solution, the LNE-LNHB and two other NMIs have submitted four ampoules for the comparison of ^{124}Sb activity measurements since 1992. The LNE-LNHB details are given in Table 1 and the details of the other participants are given in [3].

The dates of measurement in the SIR are given in Table 1 and used in all references in this report.

Table 1. Details of the LNE-LNHB participation in the comparison BIPM.RI(II)-K1.Sb-124

NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM YYYY-MM-DD
LNE-LNHB*	Laboratoire national de métrologie et d'essais - Laboratoire national Henri Becquerel	France	EUROMET	1995-12-19 2005-04-26 #

***** previously known as the LPRI and the BNM-LNHB
# pilot study submission

3. NMI standardization methods

Each NMI that submits ampoules to the SIR has measured the activity either by a primary standardization method or by using a secondary method, for example a calibrated ionization chamber. In the latter case, the traceability of the calibration needs to be clearly identified to ensure that any correlations are taken into account.

A brief description of the standardization methods for the laboratory, the activities submitted and the relative standard uncertainties (k = 1) are given in Table 2. More detail can be found in [4]. The uncertainty budget for the 2005 SIR submission is given in Appendix 1. The acronyms used for the measurement methods are given in Appendix 2.

Details regarding the solution submitted are shown in Table 3, including any impurities, when present, as identified by the laboratories. When given, the standard uncertainties on the evaluations are shown.

The half-life used by the BIPM for the SIR measurement is 60.20 (3) days [5, 6].

Table 2. Standardization methods of the LNE-LNHB for ^{124}Sb

NMI	Method used and acronym (see Appendix 2)	Half-life / d	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty / 10^{-2} by method of evaluation A	B
LNE-LNHB	4πγ counting 4P-NA-GR-00-00-00	–	5729 5745†	95-11-21 12 h UTC	0.3	0.4
		[6]	5938	04-11-22 12 h UTC	0.10	0.17

† two ampoules submitted

Table 3. Details of the solution of ^{124}Sb submitted

NMI SIR year	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. / (μg g^{-1})	Density / (g cm^{-3})	Relative activity of any impurity†
LNE-LNHB 1995	$SbCl_3$ in HCl	2	$SbCl_3$: 10	1.034	^{125}Sb : 0.250 (5) %
2005					^{125}Sb : 0.069 (2) % * ^{125}Te^{m} : 0.44 (9) %

† the ratio of the activity of the impurity to the activity of ^{124}Sb at the reference date
* the influence of its ^{125}Te^{m} daughter is negligible in the SIR measurement.

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "mother-file". The previous activity measurements for ^{124}Sb arise from four ampoules and the SIR equivalent activity, $A_{e,i}$, for each ampoule is given in [3] for each NMI, *i*. The SIR equivalent activities for the previous and new LNE-LNHB ampoule are given in Table 4. The relative standard uncertainty arising from the measurements in the SIR is also shown. This uncertainty is additional to that declared by the NMI for the activity measurement shown in Table 2. Although activities submitted are compared with a given source of ^{226}Ra, all the SIR results are normalized to the radium source number 5 [1].

The SIR correction for impurities is lower than 10^{-3} in each case. The contribution of ^{125}Te^{m} is negligible.

The present LNE-LNHB SIR result agrees within a standard uncertainty with their previous result based on the same measurement method. The deviation from the other participants in the SIR shown in Figure 1 is confirmed and may be related to the different standardization methods used (see [3, 4]). Further investigations on this aspect will be carried out by the LNE-LNHB, possibly in the frame of a EUROMET project.

As this latest SIR result was submitted as a pilot study, it is not eligible for inclusion in the BIPM.RI(II)-K1.Sb-124 key comparison reference value, that currently has the value x_R = 9517 (76) kBq which includes the previous LNE-LNHB result. Neither is the pilot result eligible for publication with degrees of equivalence in the KCDB.

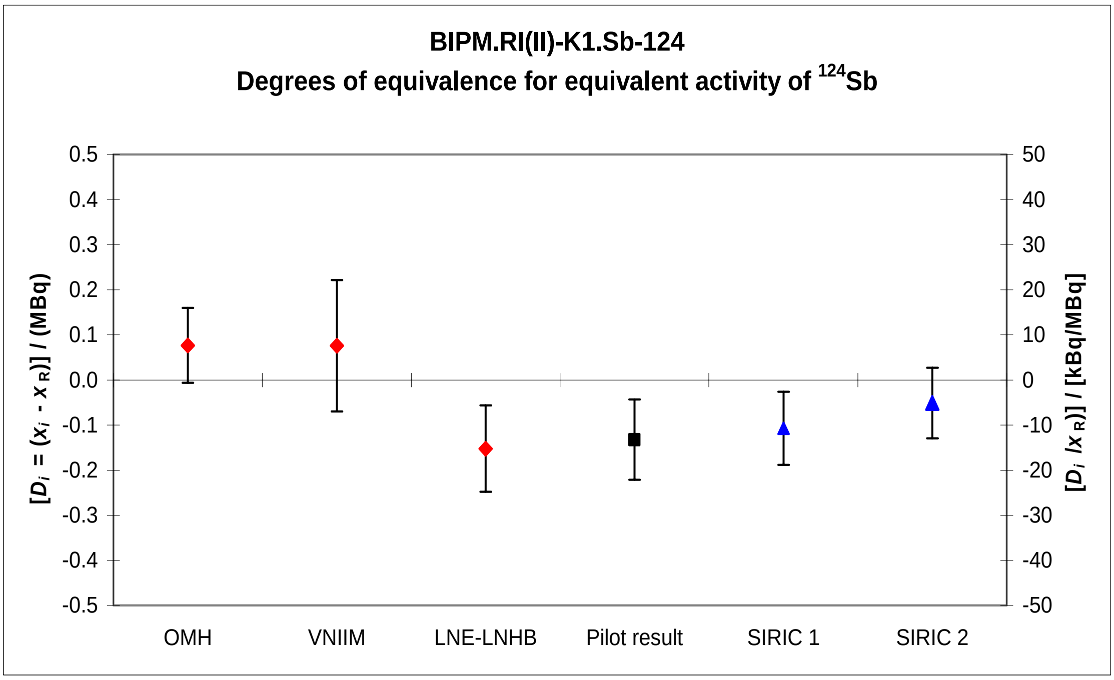
Table 4. Results of SIR measurements of ^{124}Sb for the LNE-LNHB

NMI SIR year	Mass of solution / g	Activity submitted / kBq	N° of Ra source used	SIR $A_{e,i}$ / kBq	Relative uncertainty from SIR / 10^{-2}	Total uncertainty $u_{c,i}$ ($A_{e,i}$) / kBq
LNE-LNHB 1995	3.744 2 3.755 2	5729 5745	5	9362 9367†	0.04	47 47†
2005	3.697 6	5938	3	9385	0.11 #	21

† the mean of the two A_e values shown for the same measurement date is used with an averaged uncertainty, as attributed to an individual entry [7].
# mainly due to the ^{124}Sb half-life uncertainty together with the delay of the submission for SIR measurement with respect to the reference date.

As an equivalent activity is by definition the inverse of the detection efficiency of the SIR IC for a given radionuclide, predicted A_e values can be obtained from the SIR efficiency curves and using nuclear data for that radionuclide. The efficiency curves were determined with the SIRIC program [8, 9] by non-linear least-squares adjustment of the measurement model to selected equivalent activity measurement results for 38 different radionuclides (^{124}Sb being excluded in this case). A predicted A_e value of 9410 (27) kBq is deduced using ENSDF nuclear data for ^{124}Sb [10], while 9466 (19) kBq is obtained when *Nucléide 2000* [11] is used. It is interesting to note that both calculated values are in better agreement with the LNE-LNHB SIR results based on 4πNaI counting than with the results based on coincidence counting (see Figure 1). However, the coincidence measurement results are independent of the ^{124}Sb decay scheme, while this is not the case for 4πNaI counting and the SIRIC calculation.

Figure 1. Comparison of the SIR measurement results x_i with the KCRV x_R and the calculated SIRIC values (SIRIC 1 uses ENSDF and SIRIC 2 uses Nucléide 2000). Uncertainties are k = 2.



Conclusion

The BIPM ongoing key comparison for ^{124}Sb, BIPM.RI(II)-K1.Sb-124 currently comprises three results. The KCRV determined for this radionuclide, and the matrix of degrees of equivalence was approved by the CCRI(II) and published in the BIPM key comparison database in 2003.

As this latest SIR result was submitted as a pilot study, it is not eligible for inclusion in the matrix of degrees of equivalence in the KCDB for the BIPM.RI(II)-K1.Sb-124 key comparison. The present pilot result for the LNE-LNHB in this ongoing comparison agrees with their previous result of 1995 at the level of a standard uncertainty, while the difference with the other SIR participants remains. The result is, however, in agreement with the predicted equivalent activity values obtained using the SIRIC evaluation of the SIR efficiency curves, and different published nuclear data for ^{124}Sb.

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Appendix 1. Uncertainty budgets for the activity of ^{124}Sb submitted to the SIR

Uncertainty budget for the LNE-LNHB measurement (2005)

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
Counting statistics	10	–
Interpolation from calibration curve	–	11
Extrapolation below energy threshold	–	4
Counting time	–	< 1
Weighing	–	5
Impurities	–	9
Background	0.2	–
Half life	–	6
Quadratic summation	**10**	**17**
Relative combined standard uncertainty, u_c	**20**	

Appendix 2. Acronyms used to identify different measurement methods

Each acronym has six components, geometry-detector (1)-radiation (1)-detector (2)-radiation (2)-mode. When a component is unknown, ?? is used and when it is not applicable 00 is used.

Geometry	acronym	Detector	acronym
4π	4P	proportional counter	PC
defined solid angle	SA	press. prop. counter	PP
2π	2P	liquid scintillation counting	LS
undefined solid angle	UA	NaI(Tl)	NA
		Ge(HP)	GH
		Ge(Li)	GL
		Si(Li)	SL
		CsI(Tl)	CS
		ionization chamber	IC
		grid ionization chamber	GC
		bolometer	BO
		calorimeter	CA
		PIPS detector	PS
Radiation	**acronym**	**Mode**	**acronym**
positron	PO	efficiency tracing	ET
beta particle	BP	internal gas counting	IG
Auger electron	AE	CIEMAT/NIST	CN
conversion electron	CE	sum counting	SC
mixed electrons	ME	coincidence	CO
bremsstrahlung	BS	anti-coincidence	AC
gamma rays	GR	coincidence counting with efficiency tracing	CT
X - rays	XR	anti-coincidence counting with efficiency tracing	AT
photons (x + γ)	PH	triple-to-double coincidence ratio counting	TD
photons + electrons	PE	selective sampling	SS
alpha - particle	AP	high efficiency	HE
mixture of various radiations	MX	digital coincidence counting	DC

Examples method	acronym
4π(PC)β–γ-coincidence counting	4P-PC-BP-NA-GR-CO
4π(PPC)β–γ-coincidence counting eff. trac.	4P-PP-MX-NA-GR-CT
defined solid angle α-particle counting with a PIPS detector	SA-PS-AP-00-00-00
4π(PPC)AX-γ(Ge(HP))-anticoincidence counting	4P-PP-MX-GH-GR-AC
4π CsI-β,AX,γ counting	4P-CS-MX-00-00-HE
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG