Update of the BIPM comparison BIPM.RI(II)-K1.Lu-177 of activity measurements of the radionuclide ¹⁷⁷Lu to include the 2013 result of the IFIN-HH (Romania), the 2014 result of the LNE-LNHB (France) and the 2022 result of the IRA (Switzerland)

R. Coulon¹, C. Michotte¹, S. Courte¹, M. Nonis¹, G. Ratel¹, S. Judge¹, M. Sahagia², A. Luca², A. Antohe², M.R. Ioan², F. Juget³, Y. Nedjadi³, T. Duran³, C. Bobin⁴, C. Fréchou⁴

E-mail: cmichotte@bipm.org

Abstract Since 2000, 7 laboratories have submitted 7 samples of ¹⁷⁷Lu to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM), with comparison identifier BIPM.RI(II)-K1.Lu-177. Recently, the IFIN-HH (Romania), the LNE-LNHB (France) and the IRA (Switzerland) participated in the comparison. The degrees of equivalence between each equivalent activity measured in the SIR or linked to the SIR from the CCRI(II)-K2.Lu-177 comparison have been calculated. The results are given in the form of a table and a graphical representation is also displayed.

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 with 3.6 g of the radioactive solution. For radioactive gases, a different standard ampoule is used. Each 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

¹ Bureau International des Poids et Mesures, Pavillon de Breteuil, F-92312 Sèvres Cedex, France.

² Institutul National de Cercetare - Dezvoltare pentru Fizica si Inginerie Nucleara "Horia Hulubei" (IFIN-HH), Bucarest - Magurele, Romania.

³ Institut de Radiophysique (IRA), Lausanne, Switzerland.

⁴ Université Paris-Saclay, CEA, List, Laboratoire National Henri Becquerel (LNE-LNHB), F-91120 Palaiseau, France.

ionization chambers. Details of the SIR method, experimental set-up and the determination of the equivalent activity $A_{\rm e}$, are all given in [1].

From its inception until 31 December 2022, the SIR has been used to measure 1045 ampoules to give 799 independent results for 72 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 standardizations. These comparisons are described as BIPM continuous comparisons and the results form the basis of the BIPM key comparison database (KCDB) of the Comité International des Poids et Mesures Mutual Recognition Arrangement (CIPM MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Lu-177 key comparison. The results of earlier participations in this key comparison were published previously [3,4].

2. Participants

Laboratory details are given in Table 1, with the earlier submissions being taken from [3, 4]. The dates of measurement in the SIR given in Table 1 are used in the KCDB and all references in this report.

Table 1: Details of the participants in the BIPM.RI(II)	-K1.Lu-177.
---	-------------

NMI or labora- tory	Previous acronyms or other insti- tutes	Full name	Country	Regional Metrology Organi- zation (RMO)	Date of SIR measurement
IFIN-HH	-	Institutul National de Cercetare - Dezvoltare pentru Fizica si Inginerie Nucleara "Horia Hulubei"	Romania	EURAMET	2013-07-11
IRA	IER	Institut de Radiophysique	Switzerland	EURAMET	2022-06-23
JRC	IRMM,	EC-JRC Institute for Ref-	European	EURAMET	2009-04-30
	CBNM	erence Materials and Measurements	Union		
LNE-	LMRI,	Université Paris-Saclay,	France	EURAMET	2014-06-25
LNHB	LPRI,	CEA, List, Laboratoire			
	BNM-	National Henri Becquerel			
	LNHB				
NIST	NBS	National Institute of Stan-	United	SIM	2000-02-15
		dards and Technology	States		
NPL	-	National Physical Labora-	United King-	EURAMET	2009-04-28
		tory	dom		
PTB	-	Physikalisch-Technische	Germany	EURAMET	2000-01-26
		Bundesanstalt			

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 appropriate correlations are taken into account.

A brief description of the standardization methods used by the laboratories, the activities submitted, the relative standard uncertainties and the half-life used by the participants are given in Table 2. The uncertainty budget for the new submission is given in Appendix D attached to this report; previous uncertainty budgets are given in the earlier K1 reports [3,4]. The list of acronyms used to summarize the methods is given in Appendix E.

The half-life used by the BIPM is 6.647(4) days as published in BIPM Monographie 5 vol. 2 [5].

Table 2: Standardization methods of the participants for ¹⁷⁷ Lu	u.
--	----

NMI or	Method used and the	Activity	Relati	ve	Reference	Half-life
labora- tory	acronym	$A_i/{f kBq}$	$oxed{standa} \ egin{array}{c} ext{standa} \ ext{uncert} \ /10^{-2} \ \end{array}$		date	/ d
			A	В	yyyy-mm- dd	
IFIN-HH	$4\pi(PC)\beta$ -γ coincidence (4P-	8155.72	0.55	0.7	2013-07-10	6.647(4) [5]
	PC-BP-NA-GR-CO)				12:00 UT	
IRA	$4\pi\beta(PS)-4\pi\gamma$ coincidence	$20924.5^{\rm a}$	0.41	0.17	2022-06-15	6.6463(15)
	(4P-PS-BP-NA-GR-CO)				12:00 UT	[6]
JRC	CIEMAT/NIST (4P-LS-	2462 ^b		1.64	2009-05-01	6.647(4) [5]
	MX-00-00-CN)				12:00 UT	
	4π (PC)β- γ coincidence (4P-					
	PC-BP-NA-GR-CO)					
LNE-	$4\pi(PC)\beta$ - γ anti-coincidence	5829.05	0.22	0.12	2014-06-23	6.647(4) [5]
LNHB	(4P-PC-BP-NA-GR-AC)				12:00 UT	
NIST	CIEMAT/NIST (4P-LS-	133 850	0.19	0.28	2000-02-01	6.60(1)
	MX-00-00-CN)				17:00 UT	
NPL	$4\pi(PC)\beta$ - γ coincidence (4P-	11 790°	0.03	0.3	2009-05-01	6.647(4) [5]
	PC-BP-NA-GR-CO)				12:00 UT	
	CIEMAT/NIST (4P-LS-					
	MX-00-00-CN)					
	$4\pi(\mathrm{LS})\beta$ - γ coincidence (4P-					
	LS-BP-NA-GR-CO)					
PTB	CIEMAT/NIST (4P-LS-	138 790 ^d	0.2	0.4	2000-01-19	6.646(5) [7]
	MX-00-00-CN)				00:00 UT	
	4π (PC)β- γ coincidence (4P-					
	PC-BP-NA-GR-CO)					

NMI or	Method	\mathbf{used}	and	\mathbf{the}	Activity	Relativ	e	Reference	Half-life
labora-	acronym				$A_i/{f kBq}$	standar	$^{\mathrm{d}}$	date	$ $ $/\mathbf{d}$
tory						uncerta	inty		
						$/10^{-2}$			
						A	В	уууу-тт-	
								dd	

... Continuation of Table 2.

Details regarding the solutions 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.

NMI or labo-	Chemical	Solvent	Carrier	Density	Relative activity of
ratory	composi-	conc.	conc.		any impurity ^d
	tion				
/ SIR year		/(mol dm ⁻³)	$/(\mu g g^{-1})$	$/(g \text{cm}^{-3})$	
IFIN-HH 2013	LuCl ₃ in HCl	0.1	LuCl ₃ :2	1	177 mLu:12(6)x10 ⁻³ %
IRA 2022	nonactive	0.1	nonactive	1.000(5)	177 mLu<2.8x10 ⁻³ %
	Lu^{3+} ions in		Lu^{3+} ions:20		
	HCl				
JRC 2009 ^a	LuCl ₃ in HCl	1	LuCl ₃ :20	1.037	177 mLu: $40(10)$ x 10^{-3} %
					$33.5(7)x10^{-3}$ % a2
LNE-LNHB	HCl	0.1	LuCl ₃ :10	1	177 mLu:10(1)x10 $^{-3}$ %
2014					
NIST 2000	Lu in HCl	0.01	-	1	^{177m} Lu:
					$27.1(5) \times 10^{-3} \%^{b}$
NPL 2009 ^c	LuCl ₃ in HCl	1	LuCl ₃ :20	1	^{177m} Lu:
					$33(3)x10^{-3}$ %
PTB 2000	LuCl ₃ in HCl	0.1	LuCl ₃ :20	0.999	177 mLu: $6.9(6)$ x
					$10^{-3} \%$

Table 3: Details of each solution of 177 Lu submitted.

^a The activity is the arithmetic mean of 12 efficiency extrapolated activities obtained with 4 plastic scintillation sources, from 2 dilutions, and 3 γ settings. The degrees of freedom of each of the twelve efficiency extrapolations ranged between 31 and 38.

^b Partially weighted mean (power=1) of the rather discrepant results obtained with the two methods indicated.

 $^{^{\}rm c}$ Corresponds to an activity concentration of 3286 kBq·g⁻¹ evaluated as the arithmetic mean of the three results obtained with the three methods

^d The result the weighted mean of the two methods.

^a Same solution as for the CCRI(II)-K2 Lu-177 comparison but diluted by a factor of 4.955317

^{a2} Weighted mean value from the CCRI(II)-K2.Lu-177 comparison, used for the evaluation of the KCRV and for the link of the K2 comparison

^b Measured at the BIPM

 $^{^{\}rm c}$ Same solution as for the CCRI(II)-K2 Lu-177 comparison

^d The ratio of the activity of the impurity to the activity of ¹⁷⁷Lu at the reference date

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a dedicated database based on CSV formatted files controlled by the Git version control system [8]. The latest submission has added 3 ampoules for the activity measurements for ¹⁷⁷Lu giving rise to 7 ampoules in total.

The SIR equivalent activity, A_{ei} , for each ampoule received from each NMI, i, including both previous and new results, is given in Table 4. The relative standard uncertainties arising from the measurements in the SIR are also shown. This uncertainty is additional to that declared by the NMI $(u(A_i))$ for the activity measurement shown in Table 2. Although submitted activities are compared with a given source of 226 Ra, all the SIR results are normalized to the radium source number 5 [1]. Table 4 also shows the comparison results selected for the KCRV as explained in section 4.1.

The IRA ampoule was measured in the SIR for one week, giving results in agreement within two combined standard uncertainties. No trend was observed.

NMI or labo-	Mass m_i	A_i	$^{226}\mathrm{Ra}$	$A_{\mathbf{e}i}$	Relative	$u_{\mathbf{c}i}$	$A_{\mathbf{e}i}$	for
ratory			source		uncert.		KCRV	
					from			
					SIR			
/ SIR year	$/\mathbf{g}$	/kBq		$/\mathbf{MBq}$	$/10^{-4}$	/MBq	$/\mathbf{MBq}$	
IFIN-HH 2013	3.597 584	8155.72	1	550.3	25	5.1	е	
IRA 2022	3.569	20 924.5	1	539.7 ^f	16	2.5	е	
	06(20)							
JRC 2009	3.674 99	2462	1	566.3	33 ^a	9.5		
				$565.2^{\rm cb}$	13	9.3	565.2(93)	
LNE-LNHB	3.65	5829.05	1	560.2	16	1.7	е	
2014								
NIST 2000	3.627 4	133 850	3	551.5	14	2	d	
NPL 2009	3.589 37	11 790	2	559.5^{c}	13	1.8	559.5(18)	
PTB 2000	3.675 5	138 790	3	559.0	11	2.7	559.0(27)	

Table 4: Results of SIR measurement of ¹⁷⁷Lu.

^a Dominated by the uncertainty in the impurity correction in the SIR measurement due to the large uncertainty on the 177m Lu activity stated by the participant.

^b Result when the weighted mean 177m Lu activity value from the CCRI(II)-K2 comparison is used in the impurity correction of the SIR measurement.

^c Result used to link to CCRI(II)-K2 comparison

^d Result not considered for the KCRV calculation, see details in [4]

^e This result is not included in the KCRV calculation, in accordance with a decision taken at the CCRI(II) meeting in June 2023.

^f After a year, two samples from the fully decayed solution were prepared and then measured by IRA for 40 h each on a Wallac 1220 QuantulusTM spectrometer. No long-lived impurity was found [9].

The CCRI(II)-K2.Lu-177 comparison was held in 2009 [10]. The results were linked to the BIPM.RI(II)-K1.Lu-177 comparison through the measurement in the SIR of at least one ampoule of the CCRI(II)-K2 comparison, as explained in [4].

4.1. The key comparison reference value

In May 2013, the CCRI(II) decided to calculate the key comparison reference value (KCRV) by using the power-moderated weighted mean [11] rather than an unweighted mean, as had been the policy. This type of weighted mean is similar to a Mandel-Paule mean in that the NMIs' uncertainties may be increased until the reduced chi-squared value is one. In addition, it allows for a power α smaller than two in the weighting factor. As proposed in [11], α is taken as 2-3/N where N is the number of results selected for the KCRV. Therefore, all SIR key comparison results can be selected for the KCRV with the following provisions:

- (a) results for solutions standardized by only primary techniques are accepted, with the exception of radioactive gas standards (for which results from transfer instrument measurements that are directly traceable to a primary measurement in the laboratory may be included);
- (b) each NMI or other laboratory may use only one result (normally the most recent result or the mean if more than one ampoule is submitted);
- (c) results more than 20 years old are included in the calculation of the KCRV but are not included in data shown in the KCDB or in the plots in this report, as they have expired;
- (d) possible outliers can be identified on a mathematical basis and excluded from the KCRV using the normalized error test with a test value of 2.5 and using the modified uncertainties;
- (e) results can also be excluded for technical reasons; and
- (f) the CCRI(II) is always the final arbiter regarding excluding any data from the calculation of the KCRV.

Although the KCRV may be modified when other NMIs participate, on the advice of the Key Comparison Working Group of the CCRI(II), such modifications are made only by the CCRI(II) during one of its biennial meetings, or by consensus through electronic means (e.g., email) as discussed at the CCRI(II) meeting in 2013.

In view that at least one participant found discrepant results depending on the method used without finding the reason, the CCRI(II) agreed in June 2023 not to update the KCRV and invited NMIs to carry out investigations on the standardization of Lu-177 and participate in the further SIR comparisons. Consequently the KCRV is identical to the value of 559.9(18) MBq published in 2003 [3]. This is in conformance

with the SIRIC estimation of 560.1(31) MBq [12].

4.2. Degrees of equivalence

Every participant in a comparison is entitled to have one result included in the KCDB as long as the NMI is a signatory or designated institute listed in the CIPM MRA and the result is valid (i.e., not older than 20 years). No recent submission has been identified as a pilot study so the most recent result of each NMI is normally eligible for inclusion on the KCDB platform of the CIPM MRA [2]. An NMI may withdraw its result only if all other participants agree.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the KCRV [2]. The degree of equivalence is expressed quantitatively in terms of the deviation from the key comparison reference value and the expanded uncertainty of this deviation (k = 2). The degree of equivalence between any pair of national measurement standards is expressed in terms of their difference and the expanded uncertainty of this difference and is independent of the choice of key comparison reference value.

4.2.1. Comparison of a given NMI result with the KCRV

The degree of equivalence of the result of a particular NMI, i, with the key comparison reference value is expressed as the difference D_i between the values

$$D_i = A_{ei} - KCRV \tag{1}$$

and the expanded uncertainty (k = 2) of this difference, U_i , known as the equivalence uncertainty; hence

$$U_i = 2u(D_i) \tag{2}$$

When the result of the NMI i is included in the KCRV with a weight w_i , then

$$u^{2}(D_{i}) = (1 - 2w_{i})u_{i}^{2} + u^{2}(KCRV)$$
(3)

However, when the result of the NMI i is not included in the KCRV, then

$$u^2(D_i) = u_i^2 + u^2(KCRV)$$
(4)

The introductory text in Appendix A is the one agreed by the CCRI(II) for all the K1 comparisons.

4.2.2. Comparison between pairs of NMI results

The degree of equivalence between the results of any pair of NMIs, i and j, is expressed as the difference D_{ij} in the values

$$D_{ij} = D_i - D_j = A_{ei} - A_{ej} \tag{5}$$

and the expanded uncertainty (k=2) of this difference, $U_{ij}=2u(D_{ij})$, where

$$u^{2}(D_{ij}) = u_{i}^{2} + u_{j}^{2} - 2u(A_{ei}, A_{ej})$$
(6)

where any obvious correlations between the NMIs (such as a traceable calibration, correlations normally coming from the SIR, or from the linking factor in the case of linked comparison) are subtracted using the covariance $u(A_{ei}, A_{ej})$ (see [13] for more detail). However, the CCRI decided in 2011 that these pair-wise degrees of equivalence no longer need to be published as long as the methodology is explained.

Tables B1 and B2 show the matrix of all the degrees of equivalence as they will appear in the KCDB. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with A_{ei} replaced by x_i . The introductory text is that agreed for the comparison. The graph of the results in Table 5, corresponding to the degrees of equivalence with respect to the KCRV (identified as x_R in the KCDB), is shown in Figure C1. This graphical representation indicates in part the degree of equivalence between the NMIs but obviously does not take into account the correlations between the different NMIs. It should be noted that the final data in this paper, while correct at the time of publication, will become out-of-date as NMIs make new comparisons. The formal results under the CIPM MRA [2] are those available in the KCDB.

5. Conclusion

The BIPM continuous key comparison for ¹⁷⁷Lu, BIPM.RI(II)-K1.Lu-177, currently comprises 5 results. The SIR results, together with the previously published CCRI(II)-K2.Lu-177 results, have been analyzed, providing degrees of equivalence for 12 national metrology institutes. Other results may be added when other NMIs contribute with ¹⁷⁷Lu activity measurements to this comparison or take part in other linked comparisons.

6. References

- [1] Ratel, G. The Système International de Référence and its application in key comparisons, *Metrologia*, 2007, 44(4), S7-S16.
- [2] CIPM MRA: Mutual recognition of national measurement standards and of calibration and measurement certificates issued by national metrology institutes, International Committee for Weights and Measures, 1999, pp. 45, Technical Supplement revised in October 2003 (pages 38-41).
- [3] Ratel G., Michotte C., BIPM comparison BIPM.RI(II)-K1.Lu-177 of activity measurements of the radionuclide ¹⁷⁷Lu, *Metrologia*, 2003, 40, 06028.

- [4] Michotte C., Ratel G., Courte S., Johansson L., Keightley J., Arinc A., Bakhshandeiar E., Pommé S., Altzitzoglou T., Paepen P. and Van Ammel R., BIPM comparison BIPM.RI(II)-K1.Lu-177 of activity measurements of the radionuclide ¹⁷⁷Lu for the NPL (UK) and the IRMM (EU), with linked results for the comparison CCRI(II)-K2.Lu-177, *Metrologia*, 2014, 51, 06002.
- [5] Bé M.-M., Chisté V., Dulieu C., Browne E., Chechev V., Kuzmenko N., Helmer R., Nichols A., Schönfeld E., Dersch R., *Table of radionuclides*, Monographie BIPM-5, 2004, Vol 2.
- [6] Kellett M.A., ¹⁷⁷Lu: DDEP Evaluation of the decay scheme for an emerging radiopharmaceutical, Applied Radiation and Isotopes, 109, 2016, 129-132.
- [7] Schötzig U. et al., Standardisation and decay data of ¹⁷⁷Lu and ¹⁸⁸Re, Applied Radiation and Isotopes, 55, 2001, 89-96.
- [8] Coulon R., Courte S., Judge S., Michotte C. and Nonis M., Digitalization of the reporting of key comparisons for radionuclide metrology, *Measurement Science and Technology*, 2021, **33** 024003.
- [9] Nedjadi Y., Juget F., Durán M.T., Desorgher L., Bochud F. and Bailat C., Activity standardisation of ¹⁷⁷Lu, *Applied Radiation and Isotopes*, 2023, **200**, 110986.
- [10] Zimmerman B.E., Report of the CCRI Comparison CCRI(II)-K2.Lu-177, Metrologia, 2013, 50, Tech. Suppl. 06020.
- [11] Pommé S. and Keightley J., Determination of a reference value and its uncertainty through a power-moderated mean, *Metrologia*, 2015, **52**(3), S200.
- [12] Cox M.G., Michotte C., Pearce A.K., Measurement modelling of the International Reference System (SIR) for gamma-emitting radionuclides, 2007, Monographie BIPM-7, 48 pp.
- [13] Michotte C. and Ratel G., Correlations taken into account in the KCDB, CCRI(II) working document, 2003, CCRI(II)/03-29.

Appendix A. Introductory text for ¹⁷⁷Lu degrees of equivalence

Key comparison BIPM.RI(II)-K1.Lu-177

MEASURAND: Equivalent activity of ¹⁷⁷Lu

Key comparison reference value: the SIR reference value $x_{\rm R}$ for this radionuclide is 559.9 MBq, with a standard uncertainty, $u_{\rm R}$ equal to 1.8 MBq (see Section 4.1 of the Final Report). The value x_i is taken as the equivalent activity for a laboratory i.

and U_i , its expanded uncertainty (k=2), both expressed in MBq, and $U_i=2((1-2w_i)u_i^2+u_{\rm R}^2)^{1/2}$, where w_i is the weight of The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms: $D_i = (x_i - x_R)$ laboratory i contributing to the calculation of $x_{\rm R}$.

Appendix B. Table of degrees of equivalence for BIPM.RI(II)-K1.Lu-177

Table B1: The table of degrees of equivalence for ${\rm BIPM.RI(II)\text{-}K1.Lu\text{-}177}$

NMI i	D_i / \mathbf{MBq}	U_i / \mathbf{MBq}
JRC	5	17
NPL	-0.4	3.5
IFIN-HH	-10	11
LNE-LNHB	0.2	5.0
IRA	-20.2	6.2

Table B2: The table of degrees of equivalence for the CCRI(II)-K2.Lu-177(2009) comparison

NMI i	D_i / \mathbf{MBq}	U_i / \mathbf{MBq}
ANSTO	-4.1	4.7
ENEA-INMRI	4.2	8.5
LNMRI-IRD	-2.1	7.8
NIST	-0.5	5.3
NMISA	0.7	4.8
POLATOM	-1.7	7.2
PTB	-1.3	4.4

Appendix C. Graph of degrees of equivalence with the KCRV for ¹⁷⁷Lu (as it appears in Appendix B of the MRA)

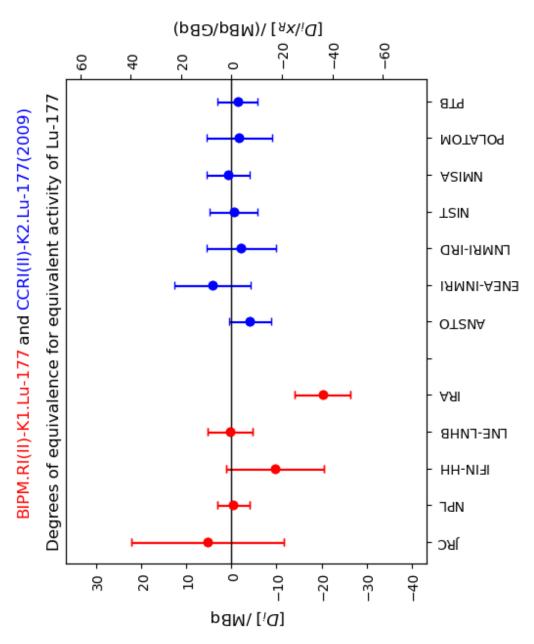


Figure C1. Degrees of equivalence for equivalent activity of ¹⁷⁷Lu.

Appendix D. Uncertainty budgets for the activity of $^{177}\mathrm{Lu}$ submitted to the SIR

Detailed Uncertainty Budget

Laboratory:IFIN-HH Radionuclide: ¹⁷⁷Lu; Ampoule number:SIR2.

Uncertainty components*, in % of the activity concentration, due to

		Remarks	Evaluation	Relative
			type (A or	B) sensitivity
				Factor
counting statistics	0.55	Standard deviation of the	Α	
		mean activity concentration	ı	
		calculated from 8 sources		
weighing	0.1	Balance reading	В	
dead time	0.3	Uncertainty propagation	В	0.06
background	0.04	Uncertainty propagation	В	0.06
pile-up				
counting time				
adsorption	0.1	Not measured, evaluated	В	
impurities	0.012	Impurity content	В	
tracer				
input parameters and statistical model				
quenching				
interpolation from calibration curve				
decay-scheme parameters				
half life $(T_{1/2} = 6.647 \text{ d};$ u = 0.004 d)	0.02	Uncertainty propagation	В	0.5
self absorption				
extrapolation of efficiency curve	0.12	Uncertainty propagation considering the slope uncertainty	ninty B	0.02
other effects (if relevant)				0.02
(explain) Secondary beta pulse	s 0.6	Beta plateau slope	В	
combined uncertainty (as quadratic sum of all uncertainty components)	0.89 L			

^{*} The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, ISO, corrected and reprinted 1995).

^{*}UTC = coo universal time March 8, 2012 Reference BIPM/RI-SIR-F-05(1.6)

SIR/SIRTI reporting form - radioactive solution

V2.2 page 3a

BIPM.RI(II)-K1 or BIPM.RI(II)-K4

Measurement method	4-pi-Beta(PS)-4pi-Gamma coincidence				
ACRONYM	4P-(PS)-BP-4P-NA-GR-CO	Comments:			
Activity concentration at					
reference date / kBq g ⁻¹	5862.75675				
Relative standard					
uncertainty / 10 ⁻²	0.44				
Date of measurement at					
the NMI (YYYY-MM-DD)	2022-06-15				

Uncertainty budget			
	Relative		
Uncertainty component	uncertainty /	Evaluation type (A or B)	Comment
Oncertainty component	10	туре (А ог в)	
			ΔΒγ/Rymin where ΔΒγ the maximum dispersion of
			the γ-background rate during the campaign, while Rymin is the smallest γ-countrate measured at the
Background	0.03	D	two gamma settings
Buckground	0.03	Ь	Maximum value of the propagation of the half-life
			uncertainty to the decay correction factors (latest
Half-life	0.06	В	measurements)
			$\Delta \tau \times \rho \beta$ where $\Delta \tau$ is the uncertainty of the deadtime
			and ρβ is a typical true beta countrate for the
Dead time	0.13	В	campaign
			(2.2/2) ())
			(ΔτR/τR)•(pacc/pcmax) where ΔτR/τR is the relative
			standard uncertainty of the resolving time and pacc is the accidental coincidence countrate, while pcmax is
			the largest measured true coincidence countrate
Resolving time	0.03	В	the largest measured true comeracine countrate
Timing	0.002	В	Worst case time base error
Weighing	0.08	В	Δm/m for a median source of the whole set used
Dilution factor	0.01	В	
			Statistical standard deviation of the mean of ρβ·ργ/ρς
			observed during repeated counting of sources
Counting statistics	0.1	А	
			Relative standard deviation of 12 efficiency
reporducilbility	0.3	^	extrapolated activities obtained from 4 sources from 2 dilutions, and three gamma settings
reportuciibility	0.3	A	anations, and three gaining settings
			Typical relative standard deviation of an intercept
			obtained by Monte Carlo fits in which $(1-\epsilon\beta)/\epsilon\beta$
Extra-/Inter-polation of efficiency			and ρβργ/ρc are varied stochastically 10 ⁴ times
curve	0.25	A	within their distributions assumed to be Gaussian
Combined standard			
uncertainty	0.44		

Detailed Uncertainty Budget

Laboratory: LNE-LNHB; Radionuclide: 177Lu; Ampoule number: 3

Uncertainty components*, in % of the activity concentration, due to

	W	Remarks	Evaluation	Relative
			type (A or B)	Sensitivity
				Factor
statistics	0,07	12 sources	A	
weighing	0,05	Pycnometer technique	В	
dead time	0,01	Live time technique	В	
background	0,05		A	
pile-up				
counting time				
adsorption				
impurities	0,05	Lu-177m	B	
tracer input parameters			Tel 100 100 100 300 100 100 100	
and statistical model				
quenching interpolation from calibration curve				
decay-scheme parameters				~~~~~
half life ($T_{1/2}$: 6,647 (4) d)	0,1		В	
self absorption				
extrapolation of efficiency curve	0,2	Extrapolation technique	A	
other effects (if relevant) (geometry factor, retrodiffusion)				
combined uncertainty (as quadratic sum of all uncertainty component	0,25			

^{*} The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, ISO, corrected and reprinted 1995).

Appendix E. 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
4 π	4P	proportional counter	PC
defined solid angle	SA	press. Prop. Counter	PP
2 π	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
		Cerenkov detector	CD
		calorimeter	CA
		solid plastic scintillator	SP
		PIPS detector	PS
		CeBr3	СВ

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	СО
bremsstrahlung	BS	anticoincidence	AC
gamma rays	GR	coincidence counting with	CT
		efficiency tracing	
x-rays	XR	anticoincidence counting	AT
		with efficiency tracing	
photons $(x + \gamma)$	PH	triple-to-double coincidence	TD
		ratio counting	
photons + electrons	PE	selective sampling	SS
alpha particle	AP	high efficiency	HE
mixture of various radi-	MX	digital coincidence counting	DC
ation			

Examples of methods	acronym
$4\pi(PC)\beta-\gamma$ coincidence counting	4P-PC-BP-NA-GR-CO
$4\pi(PPC)\beta-\gamma$ coincidence counting	4P-PP-MX-NA-GR-CT
eff. trac	
defined solid angle α -particle	SA-PS-AP-00-00-00
counting with a PIPS detector	
$4\pi(PPC)AX-\gamma(GeHP)-$	4P-PP-MX-GH-GR-AC
anticoincidence counting	
$4\pi \text{CsI-}\beta, AX, \gamma \text{ counting}$	4P-CS-MX-00-00-HE
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG