

**BIPM comparison BIPM.RI(II)-K1.Cu-64
of the activity measurements of the radionuclide ^{64}Cu**

C. Michotte¹, S. Courte¹, G. Ratel¹, K. Kossert², O.J. Nähle²
¹BIPM, Sèvres, ²PTB, Germany

Abstract

In 2009, the Physikalisch-Technische Bundesanstalt (PTB), Germany, submitted a sample of known activity of ^{64}Cu to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM). The value of the activity submitted was about 9.3 MBq. The result of this new comparison has been approved for publication by Section II of the Consultative Committee for Ionizing Radiation (CCRI(II)), comparison identifier BIPM.RI(II)-K1.Cu-64.

1. Introduction

The SIR for activity measurements of γ -ray-emitting radionuclides was established in 1976. Each NMI may request a standard ampoule from the BIPM that is then filled (3.6 g) with the radionuclide in liquid form. For radioactive gases, a different standard ampoule is used. 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. 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 2008, the SIR has measured 914 ampoules to give 670 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 standardizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the BIPM key comparison database (KCDB) of the CIPM Mutual Recognition Arrangement (CIPM MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Cu-64 key comparison.

2. Participants

The PTB is the only participant to date for the comparison of ^{64}Cu activity measurements and one ampoule was submitted in 2009. The laboratory details are given in Table 1.

Table 1. Details of the participants in the BIPM.RI(II)-K1.Cu-64

NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
PTB	Physikalisch-Technische Bundesanstalt	Germany	EURAMET	2009-02-12 08 h 54 UT

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 each laboratory, the activities submitted and the relative standard uncertainties ($k = 1$) are given in Table 2. The uncertainty budget is given in Appendix 1.

The activity reported by the PTB is the weighted mean of the measurements using the two methods indicated. Correlation due to weighing uncertainties using the same balance has been taken into account.

Table 2. Standardization method of the participant for ^{64}Cu

NMI	Method used and code (see Appendix 2)	Half-life / d	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty / 10^{-2} by method of evaluation	
					A	B
PTB	LSC (CIEMAT/NIST) 4P-LS-MX-00-00-CN and $4\pi(e,x)-\gamma$ coincidence 4P-PC-MX-NA-GR-CO	0.529 17 (9)	9 311	09-02-13 0 h 00 UT	0.47	

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 BIPM standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer is described in [3]. The CCRI(II) agreed in 1999 [4] that this method should be followed according to the protocol described in [5] when an NMI makes such a request or when there appear to be discrepancies. No impurity was identified in this case.

Table 3. Details of the solution of ^{64}Cu submitted

NMI	Chemical composition	Solvent conc. (mol dm^{-3})	Carrier: conc. ($\mu\text{g g}^{-1}$)	Density $/(g\text{ cm}^{-3})$	Relative activity of impurity ¹
PTB	CuCl_2 in HCl	0.1	CuCl_2 : 30	1.000	none detected

¹ the ratio of the activity of the impurity to the activity of ^{64}Cu at the reference date

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "master-file". The activity measurement for ^{64}Cu arises from one ampoule and the SIR equivalent activity for each ampoule, A_{ei} , is given in Table 4 for the NMI, i . The relative standard uncertainties arising from the measurements in the SIR are 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 half-life of 0.529 21 (8) d [6] used by the BIPM is in agreement with the PTB evaluation of 0.529 17 (9) days. The small difference has no significant effect on the SIR result. The date of measurement in the SIR is given in Table 1.

The ampoule was measured fourteen times in the SIR, up to 4 days after the first measurement, each measurement giving the same result. These measurements confirm the validity of the half-life value used and the lack of impurity content.

It is interesting to note that the PTB result is in agreement within the standard uncertainties with the value of 81 910 (650) kBq deduced from the efficiency curve of the SIR [7, 8] and the nuclear data from [6].

As this submission has been identified for the purpose of equivalence, the result of the PTB is eligible for inclusion in the KCDB of the CIPM MRA. However, as the PTB is the only NMI that has submitted an activity for this comparison, a key comparison reference value cannot be computed and consequently no degrees of equivalence can be derived. Nevertheless, details of how this will be done once another NMI submits a sample are given in the next section of this report for completeness.

Table 4. Results of SIR measurements of ^{64}Cu

NMI	Mass of solution /g	Activity submitted/ kBq	N° of Ra source used	SIR A_{ei} /kBq	Relative uncertainty from the SIR	Total standard uncertainty u_i / kBq
PTB	3.635 78	9311	4	81 400	6×10^{-4}	390

4.1 The key comparison reference value

The key comparison reference value is derived from the unweighted mean of all the results submitted to the SIR with the following provisions:

- only primary standardized solutions 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 included¹;
- each NMI or other laboratory has only one result (normally the most recent result or the mean if more than one ampoule is submitted);
- any outliers are identified using a reduced chi-squared test and, if necessary, excluded from the KCRV using the normalized error test with a test value of four;
- exclusions must be approved by the CCRI(II).

The reduced data set used for the evaluation of the KCRVs is known as the KCRV file and is the reduced data set from the SIR master-file. 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 only made by the CCRI(II), normally during one of its biennial meetings.

Although the PTB value is eligible to be included in a KCRV for ^{64}Cu , this cannot be computed until another NMI fulfils the criteria described.

4.2 Degrees of equivalence

Every NMI that has submitted ampoules to the SIR 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. Normally, the most recent result is the one included. Any NMI may withdraw its result only if all the participants agree.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the key comparison reference value [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.

¹ Rule modified at the CCRI(II) meeting in 2005.

However, as no KCRV can be determined at present for this radionuclide, no degrees of equivalence can be expressed.

Conclusion

The BIPM ongoing key comparison for ^{64}Cu , BIPM.RI(II)-K1.Cu-64 currently comprises one result from the PTB. Although with only one result, no KCRV can be determined for this radionuclide, nor can degrees of equivalence be calculated for publication in the BIPM key comparison database, the result of the PTB agrees with the KCRV computed using the SIRIC efficiency calculation. Once another NMI contributes ^{64}Cu activity measurements to this comparison, the KCRV and degrees of equivalence will be determined and the approval of the CCRI(II) sought prior to publication.

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Appendix 1. Uncertainty budget for the activity of ^{64}Cu for the PTB**Liquid scintillation counting method 4P-LS-MX-00-00-CN**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	4	-
weighing	-	6
dead time	-	10
background	-	5
adsorption	-	5
impurities	-	5
tracer	-	32
input parameters and model	-	30
quenching	-	10
half life	-	4
other effects	-	7
Quadratic summation	4	48
Relative combined standard uncertainty, u_c	49	

Coincidence counting method 4P-PC-MX-NA-GR-CO

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	40	-
weighing	-	18
background	4	-
half life	-	13
extrapolation of efficiency curve	-	62
other effects (if relevant)		
efficiency variation	-	17
coincidence resolving time	-	2
Gandy-effect	-	1
Quadratic summation	40	68
Relative combined standard uncertainty, u_c	79	

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
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	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
bremsstrahlung	BS	coincidence	CO
gamma ray	GR	anti-coincidence	AC
X - rays	XR	coincidence counting with efficiency tracing	CT
alpha - particle	AP	anti-coincidence counting with efficiency tracing	AT
mixture of various radiation e.g. X and gamma	MX	triple-to-double coincidence ratio counting	TD
		selective sampling	SS

Examples	method	acronym
4 π (PC) β - γ -coincidence counting		4P-PC-BP-NA-GR-CO
4 π (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
4 π (PPC)AX- γ (GeHP)-anticoincidence counting		4P-PP-MX-GH-GR-AC
4 π CsI- β ,AX, γ counting		4P-CS-MX-00-00-00
calibrated IC		4P-IC-GR-00-00-00
internal gas counting		4P-PC-BP-00-00-IG