BIPM comparison BIPM.RI(II)-K1.Rn-222 of the activity measurements of the radionuclide ^{222}Rn

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Abstract

Since 2001, a national metrology institute, the Institut de Radiophysique Appliquée (IRA), Switzerland, has submitted two samples of known activity of ^{222}Rn to the international reference system (SIR) for activity comparison at the BIPM. The activities ranged from about 13 kBq to 370 kBq. The result of this comparison has been approved for publication by Section II of the Consultative Committee for Ionizing Radiation (CCRI(II)), with comparison identifier BIPM.RI(II)-K1.Rn-222.

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 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 are all given in [1].

Since its inception until 31 December 2003, the SIR has measured 849 ampoules to give 615 independent results for 62 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 Mutual Recognition Arrangement (MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Rn-222 key comparison.

2. Participants

The IRA is the only participant to date for the comparison of ^{222}Rn activity measurements and one ampoule was submitted in 2001, as a pilot study and another in 2003. The laboratory details are given in Table 1.

Table 1. Details of the participants in the BIPM.RI(II)-K1.Rn-222

NMI	**Full name**	**Country**	**Regional metrology organization**	**Date of measurement at the BIPM**
IRA	Institut de Radiophysique Appliquée	Switzerland	EUROMET	2001-03-09 12 h 38 UT 2003-01-27 14 h 44 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 method, the activities submitted and the relative standard uncertainties (k = 1) are given in Table 2. A full uncertainty budget is given in Appendix 1.

Table 2. Standardization method of the participant for ^{222}Rn

NMI	Method used and acronym (Appendix 2)	Half-life / d	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty × 100 by method of evaluation	
					A	B
IRA	4π NaI(Tl) γ counting 4P-NA-GR-00-00-00	3.8235 (4)	12.62	01-03-06 08 h 36 UT	0.4	
	Calibrated ionization chamber 4P-IC-GR-00-00-00	3.8230 (4)	367.07* 368.16	03-01-22 12 h 00 UT	0.04* 0.04	0.30* 0.48

* values obtained using the defined solid angle SA-PS-AP-00-00-00 counting method and the 4π NaI(Tl) γ 4P-NA-GR-00-00-00 respectively to calibrate the ionization chamber.

In 2003, the IRA submitted two activity values obtained using an ionization chamber calibrated by two different methods. However, the value taken as the Swiss reference and to be used for equivalence purposes is the first value based on the defined solid angle counting ("Picolo") method [3].

Details regarding the gas sample submitted in 2003 are shown in Table 3, including any impurities, when present, as identified by the laboratory. When given, the standard uncertainties on the evaluations are shown. Recently the BIPM has developed a standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [4]. The CCRI(II) agreed in 1999 [5] that this method should be followed according to the protocol described in [6] when an NMI makes such a request or when there appear to be discrepancies.

Table 3. Details of the sample of ^{222}Rn gas submitted in 2003

NMI	Chemical composition	Amount / mol	Volume of gas / cm^3	Gas pressure / Pa in the ampoule at 20.5 °C	Relative activity of impurity†
IRA	^{222}Rn gas	2.96×10^{-13}	5.16	1.40×10^{-4}	–

† the ratio of the activity of the impurity to the activity of ^{222}Rn 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 "mother-file". The activity measurement for ^{222}Rn arises from two ampoules and the SIR equivalent activity for each ampoule, A_{ei}, is given in Table 4 for the NMI, i. The assumption is made that the ^{222}Rn decay chain (^{218}Po / ^{214}Pb / ^{214}Bi / ^{214}Po / ^{210}Pb) is in equilibrium with the parent at the SIR measurement date. 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 used by the BIPM is 3.8235 (3) days [7]. The date of measurement in the SIR is given in Table 1.

The ampoules submitted in 2001 and 2003 were re-measured in the SIR, up to 5 days and 2 days respectively after the first measurement and the results agreed within two combined uncertainties in each case.

The results of 2001 and 2003 agree within the combined standard uncertainties.

Although the IRA submission in 2001 was submitted as a pilot study, the 2003 result is eligible for Appendix B of the MRA. However, as only one NMI 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 ^{222}Rn

NMI	Activity submitted/ kBq	N° of Ra source used	SIR A_{ei} /kBq	Relative uncertainty from the SIR	Total standard uncertainty u_i / kBq
IRA	12.62	1	9 996	37×10^{-4}	55
	367.07	1	10 013*	17×10^{-4}	35*
	368.16		10 043		52

* value to be used for the equivalence (see section 3 and [3])

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:

a) only primary standardized solutions are accepted, or ionization chamber measurements that are directly traceable to a primary measurement in the laboratory;
b) each NMI or other laboratory has only one result (normally the most recent result or the mean if more than one ampoule is submitted);
c) 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;
d) 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 mother-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 IRA value is eligible to be included in a KCRV for ^{222}Rn, 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 Appendix B of the KCDB as long as the NMI is a signatory or designated institute listed in the 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.

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 ^{222}Rn, BIPM.RI(II)-K1.Rn-222 currently comprises one result from the IRA. Consequently, no KCRV can be determined for this radionuclide, nor can degrees of equivalence be calculated for publication in the BIPM key comparison database. Once another NMI contributes ^{222}Rn 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 ^{222}Rn submitted to the SIR

The uncertainty budget submitted by the IRA in 2003 is as follows:

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
current of reference source	1.8	–
current of ^{222}Rn	3.7	–
reference source substitution factor	–	4.5
reference source decay correction	–	2.4
^{222}Rn decay correction	–	0.02
calibration factor of ionization chamber using		
4π NaI γ counting	–	48
or using defined solid angle alpha counting	–	or 30
Quadratic summation	**4.1**	**48.3 or 30.4**
Relative combined standard uncertainty, u_c	**48 or 31**	

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	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**
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(GeHP)-anticoincidence counting	4P-PP-MX-GH-GR-AC
4π 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