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APMP comparison of activity measurements of I-131 (APMP.RI(II)-K2.I-131)

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Abstract

The international comparison of activity measurements of ^{131}I (APMP.RI(II)-K2.I-131) was carried out within the framework of the Asia-Pacific Metrology Programme (APMP). Seven laboratories took part in the comparison, and five of them undertook absolute measurements. One ampoule containing the same radioactive solution as that used in the APMP comparison was sent to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM) in order to link the APMP comparison to the BIPM.RI(II)-K1.I-131 comparison and to evaluate degrees of equivalence with the key comparison reference value (KCRV).

1. Introduction

The plan of the regional key comparison of activity measurements of ^{131}I solution was discussed at the fifth meeting of APMP/TCRI in Korea. It was registered as the official regional metrology organization (RMO) key comparison of APMP.RI(II)-K2.I-131. The coordinating laboratory was the Korea Research Institute of Standards and Science (KRISS). Three laboratories took part in the comparison in 2006. The reported results did not agree well. At the 7th meeting of APMP/TCRI in Australia, the project restarted with more participants. The coordinating laboratory was changed to the National Metrology Institute of Japan (NMIJ). The ampoules of ^{131}I solution were distributed in March 2009 to all the participants. All the participants measured with their own methods and reported to the coordinating laboratory. One ampoule containing the same radioactive solution as that used in the comparison was sent to the SIR at the BIPM [1]. The SIR equivalent activity result of the NMIJ (Pilot laboratory) A_e = 40.43 (10) MBq in the BIPM.RI(II)-K1.I-131 comparison agrees with the 2008 key comparison reference value of 40.40 (4) MBq [2] within one standard uncertainty. The half life of ^{131}I selected for the comparison is 8.0233 day with a standard uncertainty of 0.0019 day [3].

2. Source preparation

The original solution of ^{131}I was provided by PerkinElmer Life and Analytical Sciences, Inc. The chemical form was NaI and the activity concentration was approximately 68 GBq/g on 13 February 2009. After a quick activity measurement and impurity check, the original solution was diluted to be approximately 20 MBq/g by doubly distilled water. 0.05 mg/g NaI, 0.02 mg/g $Na_2S_2O_3$ and 0.02 mg/g LiOH were added as carrier. The diluted solution was then transferred into the NMIJ standard ampoules and the BIPM/NIST - type ampoule. The NMIJ standard ampoules were sent to all the participants. The solution in BIPM/NIST - type ampoule was sent to the SIR at the BIPM [1]. The activities of solution in these ampoules were measured in a well-type pressurized ionization chamber before shipping, and the standard deviation of the specific output current (pA/g) was 0.052 % in relative terms. The ampoule number and mass of solution that each participant received is given in Table 1.

Table 1 Ampoule numbers, mass and the laboratory

Ampoule No.	Laboratory	Mass of solution / g
b0301	BIPM	3.59974
a0301	BARC	5.00045
a0302	KRISS	5.00033
a0303	ANSTO	4.99941
a0306	INER	4.99915
a0307	OAP	5.00985
a0315	NMIJ	5.00262
a0316	PTKMR	5.00800

3. Participants

The details of participating laboratories are listed in Table 2.

4. Results

The standardization methods used by each laboratory, the measured activity, the relative uncertainties associated with the activity measurement, E_n evaluations and the date of measurements are listed in Table 3. The E_n is derived from the following formula [4];

$$E_n = \frac{A_{lab} - A_{ref}}{\sqrt{U_{lab}^2 + U_{ref}^2}},$$

where A_{lab} and U_{lab} are the activity and expanded uncertainty obtained by each participating laboratory, A_{ref} and U_{ref} are the activity and an expanded uncertainty

obtained by the pilot laboratory. The E_n values are greater than 1 for several laboratories. Figure 1 shows the measured activity by each laboratory relative to the mean value, as listed in Table 3.

Table 2. List of the laboratories participating in the APMP.RI(II)-K2. I-131

Name	Acronym	Country	RMO
Australian Nuclear Science and Technology Organisation	ANSTO	Australia	APMP
Bhabha Atomic Research Centre	BARC	India	APMP
Institute of Nuclear Energy Research	INER	Chinese Taipei	APMP
Korea Research Institute of Standards and Science	KRISS	Korea	APMP
Office of Atoms for Peace	OAP	Thailand	APMP
Pusat Teknologi Keselamatan Dan Metrologi Radiasi	PTKMR	Indonesia	APMP
National Metrology Institute of Japan	NMIJ	Japan	APMP

Table 3. Standardization methods of the participants and their results

Laboratory	Method used (see Appendix 1)	Activity / (kBq/g)*	Relative uncertainty ($k = 2$) / %	E_n value	Date of measurement
ANSTO	4P-PC-BP-NA-GR-CO	891.7	1.1	0.04	03 April 2009
BARC	4P-PC-BP-NA-GR-CO 4P-LS-BP-00-00- CN**	883.18	0.92	-0.89	30 March 2009
INER	4P-PC-BP-NA-GR-CO	907.9	0.80	1.97	13 March 2009
KRISS	4P-PP-BP-NA-GR-CO	910.3	0.46	3.18	01 April 2009
OAP	4P-IC-GR-00-00-00 #	901.5	3.1	0.36	09 March 2009
PTKMR	UA-GH-GR-00-00-00	879.47	4.5	-0.30	05 April 2009
NMIJ	4P-PC-BP-NA-GR-CO	891.3	0.48	-------	20 March 2009
Arithmetic mean value		895.1			

* reference date = 0:00 h UTC of 20 March 2009,

** not used for the laboratory final result

calibrated by ^{51}Cr, ^{57}Co, ^{60}Co, ^{85}Sr, ^{134}Cs and ^{137}Cs solutions traceable to the NMIJ

The uncertainty budgets of all the participants are listed in Table 4. The ANSTO, BARC, INER and NMIJ measured the activity using a proportional counter for the β-counter and made coincidence measurements with a NaI(Tl) scintillation γ-counter. In addition, the BARC measured the activity using a liquid scintillation counter with the CIEMAT/NIST method (CN2003) with ^{3}H used as a tracer. However, the BARC used only the coincidence method for their final result. The KRISS measured the activity using a pressurized proportional counter for the β-counter and made coincidence measurements with a NaI(Tl) scintillation γ-counter. The OAP measured the activity using a well-type pressurized ionization chamber, whose response was determined by using ^{51}Cr, ^{57}Co, ^{60}Co, ^{85}Sr, ^{134}Cs and ^{137}Cs solutions calibrated by NMIJ. The PTKMR measured the activity using a high-purity Germanium detector whose efficiency was determined by a Eu-152 point source.

The ANSTO reported that the activity ratio of gamma-ray emitting impurity relative to ^{131}I was less than 0.01 %. The other laboratories reported that no impurity was detected. All participating laboratories used a high-purity Germanium detector to identify the impurity.

The 2008 KCRV (X_R) of the BIPM.RI(II)-K1.I-131 comparison is 40.40 (4) MBq [2], to which the results of the APMP comparison will be linked by using the SIR result of the NMIJ (Pilot laboratory) A_e = 40.43 (10) MBq. Degrees of equivalence between the APMP comparison results and the KCRV will be published in the BIPM.RI(II)-K1.I-131 final report [5].

5. Conclusion

In conclusion, the APMP key comparison APMP.RI(II)-K2.I-131 of radionuclide activity measurements was completed successfully. In total, seven laboratories from APMP participated in the comparison. The measurement results are linked to the KCRV through the result of the pilot laboratory in the BIPM.RI(II)-K1.I-131 comparison.

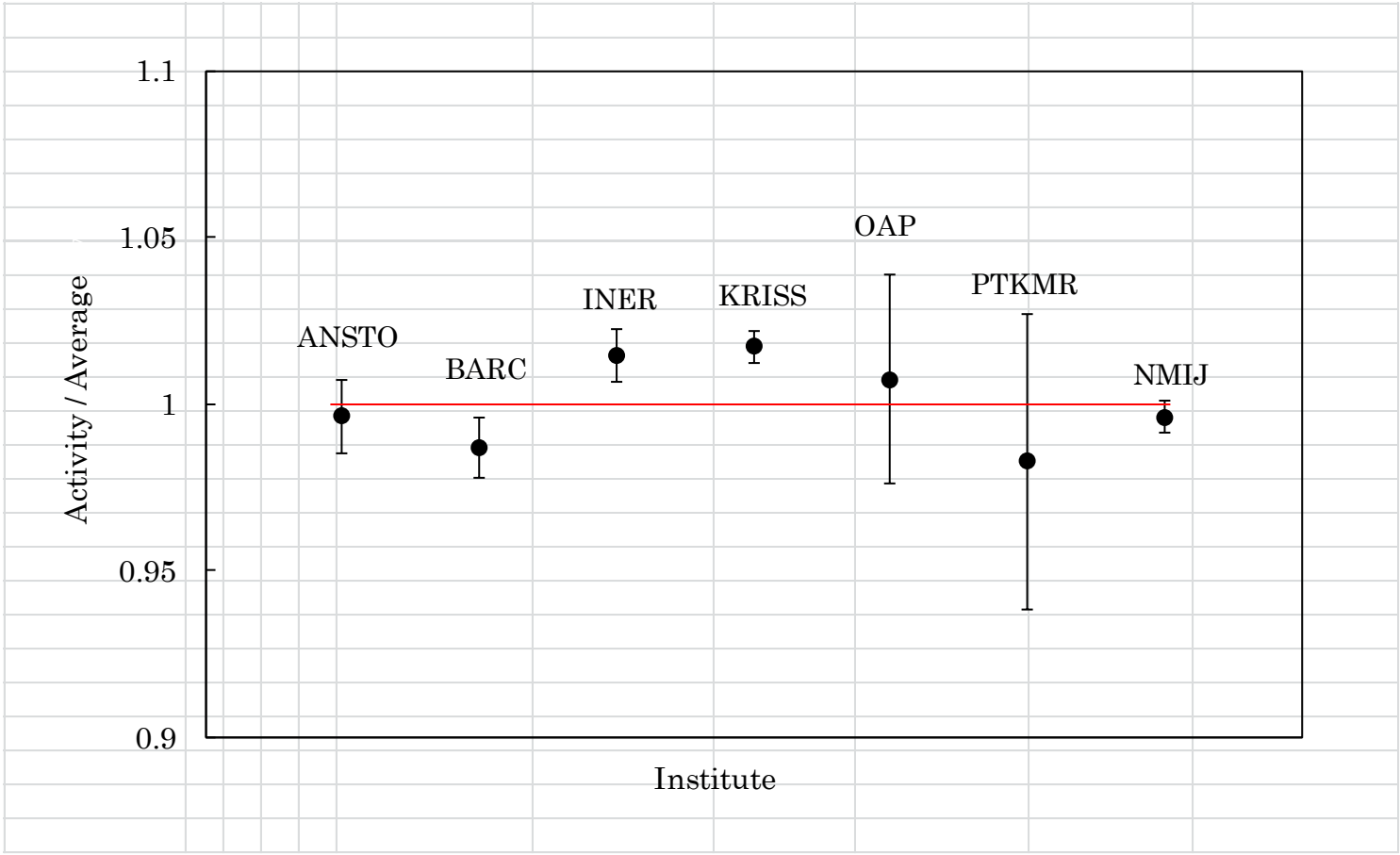

Figure 1. Results of APMP.RI(II)-K2.I-131. Relative expanded uncertainties at k=2 level are shown. The red line indicates the arithmetic mean of all the results. The 2008 KCRV of the linked BIPM.RI(II)-K1.I-131 comparison is 40.40 (4) MBq and the SIR result of NMIJ in this latter comparison is A_e = 40.43 (10) MBq.

Table 4. Uncertainty components and estimated relative values / 10^{-2}.

Item No.	Effect	ANSTO	BARC[*1]	BARC[*2]	INER	KRISS	OAP
1	counting statistics	0.22	0.43	0.34	0.27	0.12	0.1
2	weighing	0.01	0.03	0.03	0.02	0.13	0.02
3	dead time	< 0.01	0.03	--	0.01	0.01	--
4	background	--	0.09	--	0.01	0.10	0.01
5	pile up	--	--	--	--	--	--
6	resolving time	< 0.01	0.12	--	0.01	0.02	--
7	Gandy effect	--	--	--	0.01	--	--
8	counting time	--	--	--	0.01	0.01	--
9	adsorption	0.03	--	--	--	--	--
10	impurities	0.48	--	--	--	0.00	--
11	tracer	--	--	0.66	--	--	--
12	input parameters and statistical model	--	--	--	--	--	--
13	quenching	--	--	--	--	--	--
14	interpolation from calibration curve	--	--	--	--	--	--
15	decay-scheme parameters	--	--	--	--	--	--
16	half life $T_{1/2}$ = 8.0233 d	< 0.01	0.02	0.05	0.02	0.06	0.02
17	self absorption	--	--	--	--	--	--
18	extrapolation of efficiency curve	0.08	--	--	0.28	0.08	--
19	other effects - ref source stability - drift of reference source response - calibration factor	--	--	--	--	--	0.02 0.2 1.5
combined uncertainty (1σ)		0.53	0.46	0.74	0.40	0.23	1.52

[*1] Uncertainty of measurement using the 4πβ-γ coincidence counter

[*2] Uncertainty of measurement using the liquid scintillation counter with CIEMAT/NIST method

Table 4 (continued)

Item No.	Effect	PTKMR	NMIJ
1	counting statistics	0.5	0.1(1)
2	weighing	0.5	0.05(1)
3	dead time	0.05	0.01
4	background	0.02	0.05
5	pile up	--	0.02(1)
6	resolving time	0.02	0.01
7	Gandy effect	--	0.02
8	counting time	0.01	0.01
9	adsorption	--	--
10	impurities	--	--
11	tracer	--	--
12	input parameters and statistical model	--	--
13	quenching	--	--
14	interpolation from calibration curve	0.5	--
15	decay-scheme parameters	1.4	--
16	half life $T_{1/2}$ = 8.0233 d	0.0237	0.02
17	self absorption	--	--
18	extrapolation of efficiency curve	--	0.23
19	other effects - ref source stability - drift of reference source response - calibration factor	1.5	--
combined uncertainty (1σ)		2.23	0.24

Note (1): included in extrapolation of efficiency curve

Acknowledgements

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Appendix 1 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. tracing	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