

## **APMP comparison of the activity measurements of Cs-134**

Y. HINO<sup>1)</sup>, M. Moune<sup>2)</sup>, I. A. Lanièce<sup>2)</sup>, M.C. Yuen<sup>3)</sup>, I. A. Kharitonov<sup>4)</sup>

<sup>1)</sup>NMIJ/AIST, Japan <sup>2)</sup>LNE-LNHB, France <sup>3)</sup>INER, Chinese Taipei <sup>4)</sup>VNIIM, Russian Federation

### **Abstract**

An international comparison on radioactivity measurements of <sup>134</sup>Cs was carried out in 2005 between the NMIJ/AIST, LNE-LNHB, INER and the VNIIM, identified as the APMP.RI(II)-K2.Cs-134 comparison. The original solution was prepared by the NMIJ and sent to the BIPM/SIR and eleven other institutes. The results were in agreement, using the half life value of 753.1 days (2.062 y) recommended for this study. These results will be linked to the BIPM.RI(II)-K1.Cs-134 comparison in due course to provide degrees of equivalence for the APMP participants.

### **1. Introduction**

The NMIJ provides a periodic calibration service for the Japan Radio Isotope Association (JRIA), and sufficient ampoules were prepared at the same time to enable other calibrations and also a comparison to be held. These ampoule sources were distributed to the APMP colleagues to provide calibration factors of their well-type ionization chambers. The NMIJ sent one ampoule to the BIPM/SIR in 2004, for the reference, and after this, a primary measurement comparison was carried out using the stock ampoules. As almost a year passed since the first submission to the SIR and the comparison, the NMIJ submitted an independent absolute measurement for this stock solution, and the LNE-LNHB also sent an ampoule of the comparison stock solution to the International Reference System (SIR) at the Bureau International des Poids et Mesures (BIPM). All the results were in agreement, adopting the half life of 753.1 (1.8) days (2.062 y) [1].

### **2. Source preparation**

The original solution of <sup>134</sup>Cs was provided by Isotope Products Laboratories, U.S.A. with the chemical form of CsCl. After impurity checks, this original solution was diluted by 0.1N HCl with 100µg/g of CsCl carrier to give a specific activity of about 0.5 MBq/g. The diluted solution was transferred to several different types of ampoules as listed in the Table 1. All these ampoules were measured using a well-type ionization chamber, and the standard deviation of the specific output current (pA/g) was within 0.1 %.

At that time, the APMP carried out the project of “portability of the calibration factors of ionization chambers” to have relative response functions to the reference <sup>166</sup>Ho<sup>m</sup> sealed source. The NMIJ sent several ampoule sources to 8 countries of APMP colleagues and one to the BIPM/SIR in the year of 2004. At that time <sup>51</sup>Cr, <sup>57</sup>Co, <sup>60</sup>Co, <sup>88</sup>Y, <sup>109</sup>Cd, <sup>137</sup>Cs and <sup>139</sup>Ce were selected as simple decay mono-energetic sources, and <sup>134</sup>Cs was sent as the multi-gamma source. These sources were measured by each IC, and the response function of each type of chamber has been compared [2].

After this, the NMIJ and the VNIIM discussed the possibility of making absolute measurements of  $^{134}\text{Cs}$  because the VNIIM had installed two NaI(Tl) crystals of 20cm diameter by 10cm in height to construct a complete  $4\pi$  counter, and they wanted to make a comparison for this  $4\pi\gamma$  measurement system. The LNE-LNHB also kindly joined in this comparison, and the NMIJ sent stock ampoules, #0306 to the VNIIM and #0309 to the LNE-LNHB, in 2005. The NMIJ made a completely new absolute measurement using a stock ampoule, #0312. The INER had kept their ampoule #0303 in the mean time, and they also joined this absolute measurement comparison. The four comparison participants are indicated in bold type in Table 1, the others, whose details are given in Appendix 1, having used the ampoules for calibration purposes.

Table 1  $^{134}\text{Cs}$  ampoule sources and their shipping list

Ampoule No.	Mass of solution /g	Recipient Institute	Country
#0301	5.07169	(KRISS)	Republic of Korea
#0302	5.08145	(NIM)	China
#0303	5.07395	<b>INER</b>	<b>Chinese Taipei</b>
#0304	5.05109	(PTKMR <sup>1</sup> )	Indonesia
#0305	5.09302	(OAP)	Thailand
#0306	5.00137	<b>VNIIM</b>	<b>Russian Federation</b>
#0307	4.95401	(stock)	
#0308	5.01742	(NMISA <sup>2</sup> )	South Africa
#0309	5.01680	<b>LNE-LNHB</b>	<b>France</b>
#0310	4.97961	(ANSTO)	Australia
#0311	4.98314	(BARC)	India
#0312	5.10718	<b>NMIJ*</b>	<b>Japan</b>
B0301	3.58936	<b>BIPM**</b>	—
B0302	3.55324	(stock)	
J0301	4.98450	(JRIA)	Japan
J0302	4.97197	(stock)	

<sup>1</sup> Previously known as P3KRBiN

<sup>2</sup> Previously known as the CSIR

\* This ampoule was opened for the second measurement of NMIJ.

\*\*BIPM/NIST type of B0301 was sent to the BIPM/SIR in Feb. 2004.

### 3. Participants and standardization methods

Table 2 shows the list of participating laboratories for the absolute measurement of  $^{134}\text{Cs}$ . The NMIJ, INER and the VNIIM used the  $4\pi(\text{pc})\beta\text{-}\gamma$  coincidence method, and the LNE-LNHB and also the VNIIM made a  $4\pi\gamma$  measurement using large well NaI(Tl) detectors, as indicated in Table 3.

Table 2. List of the participants in the APMP-RI(II)-K2.Cs134

NMI	Full name	Country	Regional metrology organization
NMIJ/AIST	National Metrology Institute of Japan / National Institute of Advanced Industrial Science and Technology	Japan	APMP
LNE-LNHB	Laboratoire National de Métrologie et d'Essais - Laboratoire National Henri Becquerel	France	EUROMET
INER	Institute of Nuclear Energy Research	Chinese Taipei	APMP
VNIIM	D. I. Mendeleev Institute for Metrology	Russia	COOMET

#### 4. Result and Conclusion

Table 3 shows the standardization methods and the results with their uncertainties. The result of NMIJ (1<sup>st</sup>) was reported to the BIPM/SIR as an ionization chamber measurement when shipping the source in the year of 2004. The other results (coloured blue) were obtained by absolute measurement using  $4\pi\beta\text{-}\gamma$  coincidence or  $4\pi\gamma$  methods. Uncertainty components and estimated values are listed in the Table 4.

In parallel with their measurement, the LNE-LNHB transferred 3.6 ml of this solution to a BIPM/NIST type ampoule and sent this to the SIR. In order to make a link to the KCRV, the INER and VNIIM results are normalized with the LNE-LNHB value as plotted in the Figure 1. (Note that this does not represent the final link made for the APMP comparison.)

As shown in the Figure 1, the present APMP.RI(II)-K2.Cs-134 comparison was successful, and using the half life of 753.1 day (2.062 year) resulted in good agreement between the two measurements of the NMIJ (1<sup>st</sup>) and (2<sup>nd</sup>) in Table 3,

The CCRI(II) agreed in May 2007 that the results of this comparison will be linked to the BIPM.RI(II)-K1.Cs-134 comparison through the SIR results of the NMIJ and the LNE-LNHB, and published in the CIPM MRA key comparison database.

#### Acknowledgements

The authors would like to thank all the participants in this comparison and also the BIPM for their efforts in maintaining the SIR, and keeping radioactive ampoule sources, for such a long period.

Table 3. Standardization methods of the participants and their results

NMI	Method used	Activity in the report (kBq/g)	Activity at the reference date* (kBq/g)	Relative uncertainty ( $k=2$ )
NMIJ 1st	4P-IC-GR-00-00-00 (calibrated in March 2000)	582.9 (2004/02/01)	410.9**	0.70%
NMIJ 2nd	4P-PC-BP-NA-GR-CO	409.5 (2005/02/15)	409.5	0.40%
INER	4P-PC-BP-NA-GR-CO	396.33 (2005/03/30)	412.3**	0.32%
LNHB	4P-NA-GR-00-00-00	409.9 (2005/02/15)	409.9	0.38%
VNIIM	4P-PC-BP-NA-GR-CO	410.06 (2005/02/15)	410.1	0.50%
	4P-NA-GR-00-00-00	407.6 (2005/02/15)	407.6	0.50%

\* reference date = 0:00 h UTC of 15 February 2005

\*\*adopted half life = 753.1 days

Table 4. Reported uncertainty components and estimated values of all measurements.

Item No.	Function	NMIJ/ 1st	NMIJ/ 2nd	LNHB	INER	VNIIM $4\pi\beta-\gamma$	VNIIM $4\pi\gamma$
1	counting statistics	0.05 (current)	0.01	0.01	0.1	0.07	0.022
2	weighing	Negligible	0.05	0.05	0.01	0.07	0.02
3	dead time	--	0.01	Negligible	0.01	0.009	0.14
4	background	0.03	0.03	0.02	0.01	0.19	0.037
5	resolving time	--	0.01	--	0.01	0.0021	--
6	Gandy effect	--	0.01	--	--	0.07	--
7	counting time	--	0.02	<0.01	0.01	--	--
8	adsorption	<0.01	--	--	--	--	--
9	impurities	<0.01	--	--	--	0.002	0.002
10	decay-scheme	--	--	--	--	--	0.08
11	half life $T_{1/2}$ =753.1 d	0.1	0.01	Negligible	0.01	0.0035	0.001
12	self absorption	--	--	--	--	--	--
13	extrapolation	--	0.18	0.16	0.12	0.11	0.18
14	other effects	0.33 (calibration factor)	0.05*	0.07** 0.03***	--	--	--
combined uncertainty ( $1\sigma$ )		0.35	0.20	0.18	0.16	0.25	0.25

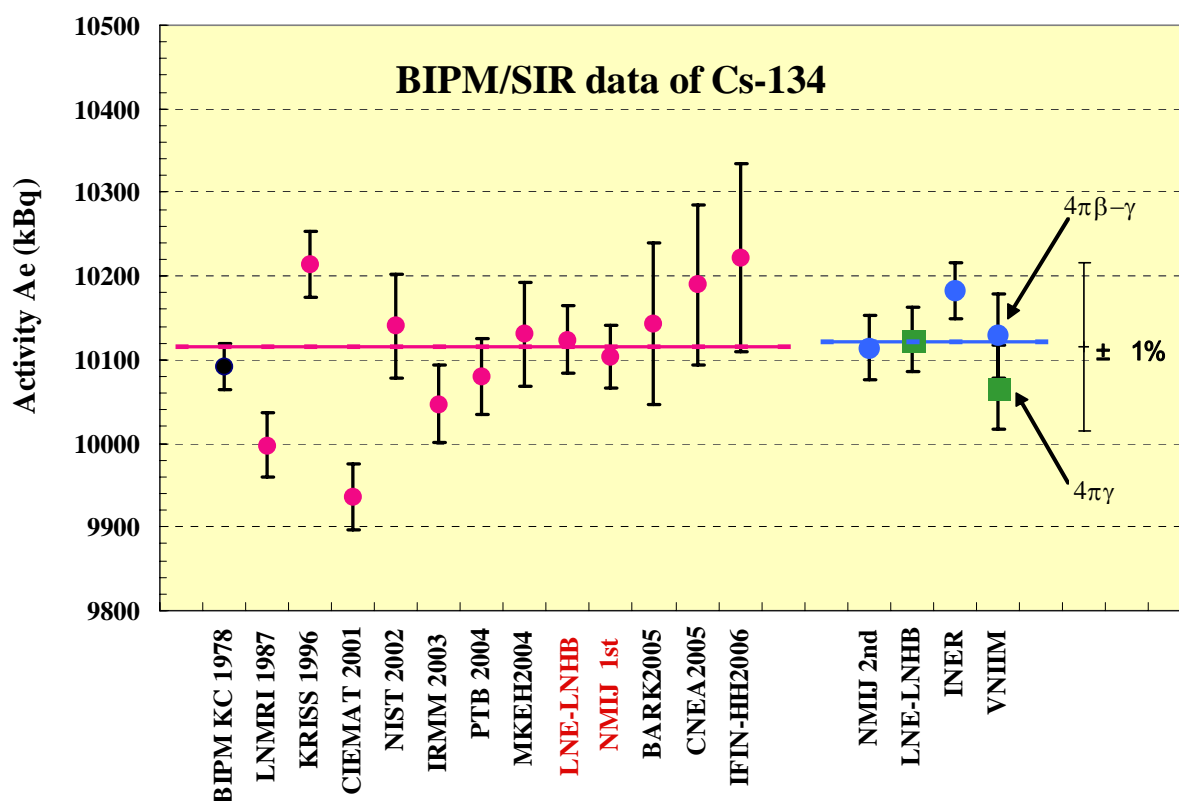


Figure 1. The BIPM/SIR key comparison results of  $^{134}\text{Cs}$  and the APMP.RI(II)-K2.Cs-134 comparison. The mean value of the SIR (red colour) shows the KCRV of  $A_e=10\ 116$  [3, 4], and the blue line shows the mean of the five present results plotted in this figure. The NMIJ 1st value of  $A_e = 10\ 104$  is the result of ampoule B0301 submitted to the BIPM/SIR. The NMIJ 2nd, INER and VNIIM results are normalized with the LNE-LNHB value of  $A_e = 10\ 124$  and  $409.9\ \text{kBq/g}$  at 15 February 2005.

## References

- [1] Dietz L.A. and Pachucki C.F.,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  half-lives determined by mass spectrometry, 1973, *J. Inorg. Nucl. Chem.* **35**, 1769.
- [2] Y. Sato, A. Yunoki, Y. Hino, T. Yamada, "Response calculation for standard ionization chambers in APMP using EGS4 Monte Carlo code", 2006, *Applied Radiation and Isotopes*, **64**, 1211-1214.
- [3] BIPM comparison BIPM.RI(II)-K1.Cs-134 of activity measurements of the radionuclide  $^{134}\text{Cs}$  and links for the 1978 international comparison CCRI(II)-K2.Cs-134, 2003 [Metrologia 40 Technical Supplement 06024](#)
- [4] Ratel G, Michotte C, Wätjen U, Janßen H, Szücs L, Coursol N, Hino Y, Activity measurements of the radionuclide  $^{134}\text{Cs}$  for the IRMM, Geel, PTB, Germany, OMH, Hungary, LNE-LNHB, France and the NMIJ, Japan in the ongoing comparison BIPM.RI(II)-K1.Cs-134, 2005 [Metrologia 42 Technical Supplement 06013](#)

Appendix 1 Details of other institutes identified in this report

<b>Institute acronym</b>	<b>Institute</b>	<b>Country</b>
KRISS	Korea Research Institute of Standards and Science	Republic of Korea
NIM	National Insitute of Metrology	China
PTKMR	Center for Technology of Radiation Safety and Metrology	Indonesia
OAP	Office of Atoms for Peace	Thailand
NMISA	National Metrology Institute of South Africa	South Africa
ANSTO	Australian Nuclear Science and Technology Organisation	Australia
BARC	Bhabha Atomic Research Centre	India
JRIA	Japan Radio Isotope Association	Japan