# BIPM comparison BIPM.RI(II)-K1.Zn-65 of activity measurements of the radionuclide $^{65}$Zn for the BARC (India) with linked results for the CCRI(II)-K2.Zn-65 comparison

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## Abstract

A new participation in the BIPM.RI(II)-K1.Zn-65 comparison has been added to the previous results. A link has been made to the CCRI(II)-K2.Zn-65 comparison held in 2003 through the measurement of all ampoules of the K2 comparison in the International Reference System (SIR) at the BIPM before despatch to the participants. This has produced a revised value for the key comparison reference value (KCRV), calculated using the power-moderated weighted mean. Six NMIs used the K2 comparison to update their degree of equivalence. The degrees of equivalence between each equivalent activity measured in the International Reference System (SIR) and the KCRV have been calculated and the results are given in the form of a table for the remaining two NMIs in the BIPM.RI(II)-K1.Zn-65 comparison and the 17 other participants in the CCRI(II)-K2.Zn-65 comparison. A graphical presentation is also given.

## 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 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 2013, the SIR has measured 973 ampoules to give 728 independent results for 67 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.Zn-65 key comparison and includes results published previously [3].

In addition, an international comparison was held in 2003 for this radionuclide, CCRI(II)-K2.Zn-65. Seventeen NMIs and one international laboratory took part in this comparison and the measurement of the 18 ampoules of the K2 comparison in the SIR before despatch to the participants enabled the linking of the K2 comparison to the BIPM.RI(II)-K1 comparison. Six NMIs had previously submitted ampoules to the SIR and have updated their results through this CCRI(II) comparison.

## 2. Participants

The BARC has submitted one ampoule for inclusion in this comparison, updating his result from the CCRI(II)-K2.Zn-65 comparison. The laboratory details are given in Table 1a, with the earlier submissions being taken from [3]. In cases where the laboratory has changed its name since the original submission, both the earlier and the current acronyms are given, as it is the latter that are used in the KCDB. The date of measurement in the SIR is also given in Table 1a and is used in the KCDB and all references in this report.

The eleven NMIs and one international laboratory that took part in the CCRI(II) international comparison, CCRI(II)-K2.Zn-65 in 2003 and are also eligible for the KCDB are shown in Table 1b together with the six laboratories that used this comparison to update their results.

**Table 1a. Details of the participants in the BIPM.RI(II)-K1.Zn-65**

| **Original acronym** | **NMI** | **Full name** | **Country** | **Regional metrology organization** | **Date of measurement at the BIPM** YYYY-MM-DD |
|---|---|---|---|---|---|
| AAEC | ANSTO | Australian Nuclear Science and Technology Organisation | Australia | APMP | 1977-10-17 |
| ASMW* | PTB | Physikalisch-Technische Bundesanstalt | Germany | EURAMET | 1977-11-22*<br>1978-03-10<br>1987-12-02 |
| OMH | MKEH | Országos Mérésügyi Hivatal | Hungary | EURAMET | 1978-10-10<br>1985-12-05<br>1995-07-17 |
| – | NPL | National Physical Laboratory | UK | EURAMET | 1979-09-06 |

* another laboratory in the country

continued overleaf.

Table 1a continued. Details of the participants in the BIPM.RI(II)-K1.Zn-65

| **Original acronym** | **NMI** | **Full name** | **Country** | **Regional metrology organization** | **Date of measurement at the BIPM** YYYY-MM-DD |
|---|---|---|---|---|---|
| UVVVR | CMI-IIR | Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation | Czech Republic | EURAMET | 1980-09-03 |
| LMRI<br>BNM-LNHB | LNE-LNHB | Bureau national de métrologie-Laboratoire national Henri Becquerel | France | EURAMET | 1982-12-01<br>1999-11-26 |
| PSPKPR | P3KRBiN ** | Pusat Penelitian & Pengembangan Keselamatan Radiasi & Biomedika Nuklir | Indonesia | APMP | 1993-09-30 |
| ETL | NMIJ | National Metrology Institute of Japan | Japan | APMP | 1994-12-06 |
| – | NIST | National Institute of Standards and Technology | United States | SIM | 1999-05-04<br>2001-11-27 |
| – | BARC | Bhabha Atomic Research Centre | India | APMP | 2006-11-29 |

** not a designated institute for Indonesia.

**Table 1b. Details of the participants in the 2003 CCRI(II)-K2.Zn-65 to be linked to BIPM.RI(II)-K1.Zn-65**

| **Original acronym** | **NMI** | **Full name** | **Country** | **Regional metrology organization** |
|---|---|---|---|---|
| – | BARC | Bhabha Atomic Research Centre | India | APMP |
| – | BEV | Bundesamt für Eich- und Vermessungswesen | Austria | EURAMET |

continued overleaf.

Table 1b continued. Details of the participants in the 2003 CCRI(II)-K2.Zn-65 to be linked to BIPM.RI(II)-K1.Zn-65

| **Original acronym** | **NMI** | **Full name** | **Country** | **Regional metrology organization** |
|---|---|---|---|---|
| – | CMI-IIR | Czech Metrological Institute - Inspectorate for Ionizing Radiation | Czech Republic | EURAMET |
| – | CNEA | Comisión Nacional de Energía Atómica | Argentina | SIM |
| – | ENEA-INMRI | Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile - Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti | Italy | EURAMET |
| – | IFIN-HH | Institutul National de Cercetare - Dezvoltare in Fizica si Inginerie Nucleara- "Horia Hulubei" | Romania | EURAMET |
| – | IRA | Institut de Radiophysique Appliquée | Switzerland | EURAMET |
| – | IRMM | EC-JRC Institute for Reference Materials and Measurements | European Union | EURAMET |
| – | KRISS | Korea Research Institute of Standards and Science | Republic of Korea | APMP |
| BNM-LNHB | LNE-LNHB | Laboratoire national de métrologie et d'essais - Laboratoire national Henri Becquerel | France | EURAMET |
| – | LNMRI /IRD | Laboratorio Nacional de Metrologia das Radiaçoes Ionizantes/ Instituto de Radioproteção e Dosimetria | Brazil | SIM |
| OMH | MKEH | Magyar Kereskedelmi Engedélyezési Hivatal | Hungary | EURAMET |
| – | NMIJ | National Metrology Institute of Japan | Japan | APMP |
| CSIR-NML | NMISA | National Metrology Institute, South Africa | South Africa | SADCMET |

continued overleaf.

Table 1b continued. Details of the participants in the 2003 CCRI(II)-K2.Zn-65 to be linked to BIPM.RI(II)-K1.Zn-65

| **Original acronym** | **NMI** | **Full name** | **Country** | **Regional metrology organization** |
|---|---|---|---|---|
| – | NPL | National Physical Laboratory | United Kingdom | EURAMET |
| – | PTB | Physikalisch-Technische Bundesanstalt | Germany | EURAMET |
| – | SMU | Slovensky Metrologicky Ustav | Slovakia | EURAMET |
| – | VNIIM | D.I. Mendeleyev Institute for Metrology | Russian Federation | COOMET |

## 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 used by the laboratories, the activities submitted, the relative standard uncertainties ($k$ = 1) and the half-life used by the participants for the SIR are given in Table 2. The uncertainty budget for the new submission from BARC is given in Appendix 1, previous uncertainty budgets are given in the earlier K1 report [3]. The uncertainty budgets for all the participants in the CCRI(II)-K2.Zn-65 comparison are given in Appendix 2. The acronyms used for the measurement methods are given in Appendix 3.

The half-life used by the BIPM is 244.1(2) days [4], which is in agreement with 244.01(9) d, the value published in the BIPM Monographie 5 [5]. The SIR data could be revised using the new half-life. However, the updated degrees of equivalence would not differ significantly as the SIR measurements were performed within a few weeks following the reference date. In the extreme case of 250 days for the CCRI(II)-K2 ampoules measured in the SIR, the relative change in $A_e$ would be about $2.6 \times 10^{-4}$. The half-life used in the CCRI(II)-K2.Zn-65 comparison is 244.26(26) d [6].

**Table 2. Standardization methods of the SIR participants for $^{65}$Zn**

| NMI | Method used and acronym (see Appendix 3) | Half-life /d | Activity $A_i$ / kBq | Reference date YYYY-MM-DD | Relative standard uncertainty / $10^{-2}$ by method of evaluation | |
|---|---|---|---|---|---|---|
| | | | | | A | B |
| ANSTO | 4πβ-γ coincidence<br>4P-PC-MX-NA-GR-CO | 243.8 | 454.9 | 1977-10-03<br>0 h UT | 0.20 | 0.26 |
| PTB | 4πEC-γ coinc.<br>4P-PC-MX-NA-GR-CO | – | 2 793†<br>2 786 | 1977-10-01<br>12 h UT | 0.3 | 0.3 |
| | Pressurized IC *<br>4P-IC-GR-00-00-00 | – | 11 326 | 1978-01-01<br>0 h UT | 0.02 | 0.14 |
| | Pressurized IC<br>4P-IC-GR-00-00-00<br>calibrated in 1980 by 4πPC-γ and 4πPPC-γ coinc.<br>4P-PC-MX-NA-GR-CO<br>4P-PP-MX-NA-GR-CO | – | 4 246 | 1987-11-01<br>0 h UT | 0.07 | 0.68 |
| MKEH | 4π(x,$e_A$,$\beta^+$)-γ coincidence<br>4P-PP-MX-NA-GR-CO | 244.1(2) [4] | 2 015†<br>2 016 | 1978-10-01<br>12 h UT | 0.05 | 0.20 |
| | | 243.9(2) [7] | 3 621 | 1985-12-15<br>12 h UT | 0.05 | 0.21 |
| | | 244.26(26) [6] | 3 045 | 1995-07-01<br>0 h UT | 0.05 | 0.20 |
| NPL | Pressurized IC *<br>4P-IC-GR-00-00-00 | – | 5 587†<br>5 721 | 1979-09-01<br>0 h UT | 0.12 | 0.69 |
| CMI-IIR | 4πx-γ<br>4P-PC-MX-NA-GR-CO | 243.9 | 3 521 | 1980-07-31<br>10 h UT | 0.07 | 0.67 |
| LNE-LNHB | 4πPPC-γ<br>4P-PP-MX-NA-GR-CO | – | 2 290†<br>2 289 | 1982-11-08<br>12 h UT | 0.06 | 0.21 |
| | 4πx-γ coincidence<br>4P-PP-XR-NA-GR-CO | 244.06(10) [8] | 1 877 | 1999-07-12<br>12 h UT | 0.10 | 0.20 |
| P3KRBiN | 4πPC-γ<br>4P-PC-MX-NA-GR-CO | – | 445.1 | 1993-09-01<br>12 h UT | 0.20 | 0.21 |
| NMIJ | 4π(x,$e_A$,$\beta^+$)-γ coincidence<br>4P-PC-MX-NA-GR-CO | – | 1 694 | 1994-12-01<br>12 h UT | 0.11 | 0.32 |
| NIST | Pressurized IC<br>4P-IC-GR-00-00-00<br>calibrated in 1970 using 4π($e^+$,x)-γ coincidence<br>4P-PC-MX-NA-GR-CO | 244.06(10) [8] | 1 712 | 1999-04-22<br>19 h UT | 0.06 | 0.63 |
| | | | 14 859 | 2001-11-15<br>12 h UT | 0.08 | 0.63 |
| BARC | 4πβ-γ coincidence<br>4P-PC-BP-NA-GR-CO | 243.9 | 1 582 | 2006-05-05<br>6 h 30 UT | 0.99 | 0.36 |

† two ampoules submitted.

* calibrated by primary measurements of $^{65}$Zn in 1977 by the method 4P-PP-MX-NA-GR-CO for the PTB and in 1979 by the method 4P-PC-MX-NA-GR-CO for the NPL.

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. The BIPM has developed a standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [9]. The CCRI(II) agreed in 1999 [10] that this method should be followed according to the protocol described in [11] when an NMI makes such a request or when there appear to be discrepancies. No impurity measurements were carried out at the BIPM in the present case.

**Table 3. Details of the solution of $^{65}$Zn submitted**

| **NMI / SIR year** | **Chemical composition** | **Solvent conc. / (mol dm$^{-3}$)** | **Carrier: conc. /(μg g$^{-1}$)** | **Density /(g cm$^{-3}$)** | **Relative activity of impurity†** |
|---|---|---|---|---|---|
| ANSTO 1977 | $ZnCl_2$ in HCl | 0.1 | $ZnCl_2$ < 50 | 1.00 | < 0.1 % |
| PTB 1977 | $ZnCl_2$ in HCl | 0.1 | $ZnCl_2$: 20 | - | < 0.1 % |
| 1978 | | | $ZnCl_2$: 30 | 1.00 | < 0.01 % |
| 1987 | | | $ZnCl_2$: ~30 | 1.00 | – |
| MKEH 1978 | $ZnCl_2$ in HCl | 0.1 | Zn: 108 | - | $^{60}$Co: 0.005(1) % |
| 1985 | | | Zn: 40 | - | $^{60}$Co: 0.0050(15) % |
| 1995 | | | $ZnCl_2$: 25 | - | – |
| NPL 1979 | $ZnCl_2$ in HCl | 0.1 | $ZnCl_2$: 60 | 1.001 | – |
| CMI-IIR 1980 | $ZnCl_2$ in HCl | 0.08 | $ZnCl_2$: 50 | - | < 0.1 % |
| LNE-LNHB 1982 | $ZnCl_2$ in HCl | 0.11 | $ZnCl_2$: 64 | 1.000 | $^{75}$Se: 0.003(2) %<br>$^{60}$Co: 0.007(3) % |
| 1999 | | 0.1 | $Zn^{++}$: 10 | 1.001 | $^{60}$Co: 0.0010(1) % |
| P3KRBiN 1993 | $ZnCl_2$ in HCl | 1 | $ZnCl_2$: 10 | 1.0 | – |
| NMIJ 1994 | $ZnCl_2$ in HCl | 0.1 | $ZnCl_2$: 50 | 1.00 | – |
| NIST 1999 | $ZnCl_2$ in HCl | 1 | $ZnCl_2$: 300 | 1.014(1) | – |
| 2001 | | | $ZnCl_2$: 400 | 1.014(1) | – |
| BARC 2006 | $ZnCl_2$ in HCl | 0.1 | $ZnCl_2$: 20 | 1 | – |
| CCRI(II)-K2 2003 | $ZnCl_2$ in HCl | 0.5 | $Zn^{++}$: 20 | 1.007 | – |

† the ratio of the activity of the impurity to the activity of $^{65}$Zn 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 present submission has added one ampoule for the activity measurements for $^{65}$Zn giving rise to twenty ampoules in total. The SIR equivalent activity, $A_{ei}$, for each ampoule for the previous and new results is given in Table 4a for each NMI, *i*. The relative standard uncertainty arising from the measurements in the SIR is also shown. This uncertainty is additional to that declared by the NMI 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 4a. Results of SIR measurements of $^{65}$Zn**

| NMI / SIR year | Mass of solution /g | Activity submitted $A_i$ / kBq | N° of Ra source used | SIR $A_{ei}$ / kBq | Relative standard uncertainty from SIR | Combined standard uncertainty $u(A_{ei})$ / kBq |
|---|---|---|---|---|---|---|
| ANSTO 1977 | 3.635 1 | 454.9 | 2 | 29 610 | $9 \times 10^{-4}$ | 100 |
| PTB 1977 | 3.510 45 | 2 793 | 3 | 29 494 | $6 \times 10^{-4}$ | 130 |
| | 3.501 40 | 2 786 | | 29 473 | | 130 |
| 1978 | 3.659 5(1) | 11 326 | 4 | 29 680 | $4 \times 10^{-4}$ | 44 |
| 1987 | 3.627 7 | 4 246 | 3 | 29 670 | $5 \times 10^{-4}$ | 200 |
| MKEH 1978 | 3.602 0 | 2 015 | 3 | 29 699 | $7 \times 10^{-4}$ | 65 |
| | 3.602 7 | 2 016 | | 29 691 | | 64 |
| 1985 | 3.603 7 | 3 621 | 3 | 29 763 | $5 \times 10^{-4}$ | 66 |
| 1995 | 3.610 0 | 3 045 | 3 | 29 690 | $5 \times 10^{-4}$ | 63 |
| NPL 1979 | 3.482 9 | 5 587 | 3 | 29 799 | $6 \times 10^{-4}$ | 210 |
| | 3.566 8 | 5 721 | | 29 783 | $5 \times 10^{-4}$ | 210 |
| CMI-IIR 1980 | 3.557 59 | 3 521 | 3 | 29 780 | $5 \times 10^{-4}$ | 200 |
| LNE-LNHB | 3.621 38 | 2 290 | 3 | 29 695 | $8 \times 10^{-4}$ | 69 |
| 1982 | 3.620 35 | 2 289 | | 29 682 | $7 \times 10^{-4}$ | 67 |
| 1999 | 3.552 07 | 1 877 | 2 | 29 800 | $9 \times 10^{-4}$ | 69 |
| P3KRBiN 1993 | 3.537 5 | 445.1 | 1 | 28 540 | $11 \times 10^{-4}$ | 89 |
| NMIJ 1994 | 3.595 8 | 1 694 | 3 | 29 750 | $6 \times 10^{-4}$ | 100 |
| NIST 1999 | 3.706 8(2) | 1 712 | 3 | 29 840 | $6 \times 10^{-4}$ | 190 |
| 2001 | 3.783 9(2) | 14 859 | 4 | 29 680 | $4 \times 10^{-4}$ | 190 |
| BARC 2006 | 3.5579 2 | 1 582 | 2 | 29 126 | $10 \times 10^{-4}$ | 310 |

The BARC SIR measurement was repeated after three months producing the same comparison results.

No recent submission has been identified as a pilot study so the result of each NMI is normally eligible for the key comparison database (KCDB) of the CIPM MRA. However, the P3KRBiN is not a designated laboratory of the Puslit KIM-LIPI, Indonesia, therefore this result is not included in the KCDB.

An international comparison for this radionuclide, CCRI(II)-K2.Zn-65 was held in 2003 and the 18 laboratories from this comparison to be added to the matrix of degrees of equivalence are given in Table 1b.

The results of the CCRI comparison have been linked to the BIPM.RI(II)-K1.Zn-65 comparison through the measurement in the SIR of the 18 ampoules of the K2 comparison, before despatch to the participants. The uncertainties for the CCRI(II) comparison results linked to the SIR are comprised of the original NMI uncertainties combined with the uncertainty of the SIR measurement using the source of $^{226}$Ra number 1 as given in Table 4b. The main contribution to the SIR uncertainty is the statistical uncertainty of the measurement of the ionization current.

Except for the BARC, the results in the CCRI(II)-K2.Zn-65 comparison agree within the combined uncertainty with the corresponding earlier SIR result when available. The BARC result in the SIR improved when comparing with their earlier CCRI(II)-K2.Zn-65 result and agrees with the key comparison reference value within expanded uncertainty ($k = 2$) as shown below.

**Table 4b. Results of the 2003 CCRI(II) comparison of $^{65}$Zn and links to the SIR**

| NMI | Measurement method and acronym (see Appendix 3) | Activity** concentration / (kBq g$^{-1}$) | Activity concentration selected for the KCDB / (kBq g$^{-1}$) | Mass of solution in the SIR ampoule / g | Relative standard uncertainty from SIR × 10$^{-4}$ | SIR equivalent activity $A_{ei}$ / kBq | Combined standard uncertainty $u(A_{ei})$ / kBq |
|---|---|---|---|---|---|---|---|
| BARC[#] | 4πICγ (efficiency curve obtained in 1982 using BARC standards)<br>4P-IC-GR-00-00-00 | 52.77(53) | | 3.561 89 | 19 | 28 480 | 290 |
| BEV | 4πICγ traceable to NPL primary activity standards<br>4P-IC-GR-00-00-00 | 55.07(62) | | 3.630 73 | 15 | 29 670 | 330 |
| CMI-IIR | 4π(PC)β$^+$,e$_A$,x$_e$-γ coincidence<br>4P-PC-MX-NA-GR-CO | 55.28(31) | | 3.605 12 | 14 | 29 850 | 170 |
| CNEA | 4π(PPC)β$^+$,e$_A$,x$_e$-γ coincidence<br>4P-PP-MX-NA-GR-CO | 55.71(23) | | 3.682 10 | 14 | 30 030 | 130 |
| ENEA | 4πICγ<br>4P-IC-GR-00-00-00 | 54.92(29) [a]<br>55.00(29) [b] | 54.96(20) [##] | 3.659 66 | 17 | 29 660 | 120 |
| IFIN-HH | 4π(PC)β$^+$,e$_A$,x$_e$-γ coincidence [h] [18, 19]<br>4P-PC-MX-NA-GR-CO | 54.78(27) | | 3.593 39 | 13 | 29 550 | 150 |
| IRA | 4π(PC)β$^+$,e$_A$,x$_e$-γ coincidence<br>4P-PC-MX-NA-GR-CO | 55.20(24) [g] | | 3.607 66 | 16 | 29 720 | 140 |
| IRMM | 4π(PPC) β$^+$,e$_A$,x$_e$-γ coincidence [i]<br>4P-PP-MX-NA-GR-CO<br>4π(LS) CIEMAT/NIST<br>4P-LS-MX-00-00-CN | 55.06(10)<br><br><br>55.1(5) | 55.06(10) [##] | 3.643 16 | 14 | 29 661 | 68 |

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Table 4b continued. Results of the 2003 CCRI(II) comparison of $^{65}$Zn and links to the SIR

| **NMI** | **Measurement method and acronym (see Appendix 3)** | **Activity** concentration / (kBq g$^{-1}$)** | **Activity concentration selected for the KCDB / (kBq g$^{-1}$)** | **Mass of solution in the SIR ampoule / g** | **Relative standard uncertainty from SIR × 10$^{-4}$** | **SIR equivalent activity $A_{ei}$ / kBq** | **Combined standard uncertainty $u(A_{ei})$ / kBq** |
|---|---|---|---|---|---|---|---|
| KRISS | $4\pi(PPC)\beta^+,e_A,x_e$-$\gamma$ coincidence<br>4P-PP-MX-NA-GR-CO | 55.20(22) | | 3.654 34 | 17 | 29 780 | 130 |
| LNE-LNHB [d] | $4\pi$(LS) TDCR [20]<br>4P-LS-MX-00-00-TD | 54.92(29) | 55.28(22) [c] | 3.673 49 | 15 | 29 810 | 130 |
| | $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ anticoincidence [21]<br>4P-PC-PO-GH-GR-AC | 55.42(22) | | | | | |
| | $4\pi(LS)\beta^+,e_A,x_e$-$\gamma$ coincidence [j]<br>4P-LS-MX-NA-GR-CO | 55.43(31) | | | | | |
| LNMRI /IRD | $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence<br>4P-PC-MX-NA-GR-CO | 55.68(28) [e] | 55.68(28) | 3.624 85 | 19 | 30 040 | 160 |
| | $4\pi$(LS) CIEMAT/NIST<br>4P-LS-MX-00-00-CN | 57.90(32) | | | | | |
| CNEN-IPEN* | $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence<br>4P-PC-MX-NA-GR-CO | 56.00(29) [f] | | | | | |
| MKEH | $4\pi(PPC)\beta^+,e_A,x_e$-$\gamma$ coincidence and anticoincidence<br>4P-PP-MX-NA-GR-CO<br>4P-PP-MX-NA-GR-AC | 54.91(20) | | 3.650 86 | 14 | 29 590 | 120 |

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Table 4b continued. Results of the 2003 CCRI(II) comparison of $^{65}$Zn and links to the SIR

| **NMI** | **Measurement method and acronym (see Appendix 3)** | **Activity** concentration / (kBq g$^{-1}$)** | **Activity concentration selected for the KCDB / (kBq g$^{-1}$)** | **Mass of solution in the SIR ampoule / g** | **Relative standard uncertainty from SIR × 10$^{-4}$** | **SIR equivalent activity $A_{ei}$ / kBq** | **Combined uncertainty $u(A_{ei})$ / kBq** |
|---|---|---|---|---|---|---|---|
| NMIJ | $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence<br>4P-PC-MX-NA-GR-CO | 55.12(28) | | 3.525 77 | 13 | 29 700 | 150 |
| NMISA | $4\pi(LS)\beta^+,e_A,x_e$-$\gamma$ coincidence [12]<br>4P-LS-MX-NA-GR-CO | 55.48(17) | | 3.668 22 | 16 | 29 870 | 110 |
| NPL | $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence<br>4P-PC-MX-NA-GR-CO<br>$4\pi(LS)\beta^+,e_A,x_e$-$\gamma$ coincidence with DCC [k]<br>4P-LS-MX-NA-GR-CO | 55.58(33)<br><br><br>55.58(22) | 55.58(18) [##] | 3.630 22 | 14 | 29 990 | 110 |
| PTB | $4\pi(PPC)\beta^+,e_A,x_e$-$\gamma$ coincidence<br>4P-PP-MX-NA-GR-CO<br>$4\pi$(LS) CIEMAT/NIST<br>4P-LS-MX-00-00-CN | 55.14(23)<br><br><br>54.7(6) | 55.14(23) | 3.625 07 | 16 | 29 710 | 130 |
| SMU | $4\pi$ICγ calibrated at the PTB in Nov. 2002<br>4P-IC-GR-00-00-00 | 54.0(1.2) | | 3.616 07 | 15 | 29 200 | 670 |
| VNIIM | $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence [18]<br>4P-PC-MX-NA-GR-CO<br>Kx-γ coincidence [m] [22, 23]<br>UA-NA-XR-NA-GR-CO | 55.05(69)<br><br>55.17(14) | 55.17(14) [##] | 3.656 33 | 15 | 29 727 | 87 |

[#] superseded in the KCDB by the SIR submission in 2006
[##] weighted mean result
[*] another laboratory in the country

** referenced to 00:00 UTC 1 January 2003

[a] IC calibrated in 2002 with another $^{65}$Zn solution by the 4π(LS) CIEMAT/NIST method.

[b] IC calibrated in 2002 with another $^{65}$Zn solution by the 4π(PC)$\beta^+$,$e_A$,$x_e$-γ coincidence method.

[c] weighted mean value with the uncertainty taken as the uncertainty of the anticoincidence measurement

[d] a value of 54.40 kBq g$^{-1}$ , $u$ = 0.32 kBq g$^{-1}$ was obtained by γ spectrometry. This motivated the EUROMET action 721 [13] which enabled to update the DDEP data sheet for $^{65}$Zn in 2006 [5].

[e] obtained with a γ window set between 300 keV and 1250 keV

[f] obtained with a γ window set between 970 keV and 1250 keV

[g] other determinations were carried out in order to characterise the dependence of the results on the γ emission intensity values used

[h] a lead absorber of 9 mm thickness was placed between the proportional counter and the scintillation detector to ensure the fulfilment of the linearity conditions

[i] using a 6”x 6”well-type NaI(Tl) scintillation detector in the gamma channel

[j] using a TDCR system in the beta channel

[k] with a two-dimensional extrapolation from two γ channels

[m] using two NaI(Tl) detectors of different thicknesses to detect the x-rays and the γ-rays respectively under small solid angles

## 4.1 The key comparison reference value

In May 2013 the CCRI(II) decided to no longer calculate the key comparison reference value (KCRV) by using an unweighted mean but rather by using the power-moderated weighted mean [14]. 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 smaller than two in the weighting factor. Therefore, all SIR key comparison results can be selected for the KCRV with the following provisions:

a) only results for solutions standardized by 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 has only one result (normally the most recent result or the mean if more than one ampoule is submitted);
c) 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;
d) results can also be excluded for technical reasons.
e) The CCRI(II) is always the final arbiter regarding excluding any data from the calculation of the KCRV.

The data set used for the evaluation of the KCRVs is known as the "KCRV file" and is a 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 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.

Consequently, the KCRV for $^{65}$Zn has been calculated as 29 740(43) kBq on the basis of the SIR results from the ASMW (1977), ANSTO, NIST (1999) and the BARC and of the SIR results for the following CCRI(II)-K2 participants: the CMI-IIR, CNEA, IFIN-HH, IRA, IRMM, KRISS, LNE-LNHB, LNMRI/IRD, MKEH, NMIJ, NMISA, NPL, PTB and the VNIIM. The P3KRBiN SIR result has been identified as an outlier. This can be compared with the previous KCRV value of 29 710(40) kBq published in 2004 [3] and the value of 29 694(65) kBq obtained using the SIRIC efficiency curve of the SIR [15, 16] and nuclear data from [5].

## 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). Normally, the most recent result is the one included. 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

$$D_i = A_{\mathrm{e}\,i} - \mathrm{KCRV} \tag{1}$$

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

$$u^2(D_i) = (1\text{-}2w_i)\,u_i^2 + u^2(\mathrm{KCRV})\,. \tag{3}$$

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

$$u^2(D_i) = u_i^2 + u^2(\mathrm{KCRV}). \tag{4}$$

### 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

$$D_{ij} = D_i - D_j = A_{\mathrm{e}\,i} - A_{\mathrm{e}\,j} \tag{5}$$

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

$$u_{D\,ij}{}^2 = u_i{}^2 + u_j{}^2 - 2u(A_{\mathrm{e}i}, A_{\mathrm{e}j}) \tag{6}$$

where any obvious correlations between the NMIs (such as a traceable calibration, or correlations normally coming from the SIR or from the linking factor in the case of linked comparison) are subtracted using the covariance $u(A_{\mathrm{e}i}, A_{\mathrm{e}j})$ (see [17] 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.

Table 5 shows 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 $A_{\mathrm{e}i}$ 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 $x_{\mathrm{R}}$ in the KCDB), is shown in Figure 1. 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.

## Conclusion

The BIPM ongoing key comparison for $^{65}$Zn, BIPM.RI(II)-K1.Zn-65 currently comprises two results. These have been analysed with respect to the updated KCRV determined for this radionuclide. The results of the CCRI(II)-K2.Zn-65 comparison held in 2003 have been linked to the BIPM comparison through the measurement in the SIR of the ampoules of the K2 comparison, before despatch to the participants. This has enabled the table of degrees of equivalence to include nineteen results in total.

The degrees of equivalence have been approved by the CCRI(II) and are published in the BIPM key comparison database. Further results may be added when other NMIs contribute $^{65}$Zn activity measurements to the ongoing K1 comparison or take part in other linked comparisons.

## Acknowledgements

The authors would like to thank Anuradha and D.B. Kulkarni from the BARC for their participation in this comparison, all the participants in the CCRI(II)-K2.Zn-65 comparison and Dr J.M. Los Arcos of the BIPM for editorial assistance.

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## Table 5. Table of degrees of equivalence and introductory text for $^{65}$Zn

**Key comparison BIPM.RI(II)-K1.Zn-65**

**MEASURAND : Equivalent activity of $^{65}$Zn**

**Key comparison reference value: the SIR reference value for this radionuclide is $x_R$ = 29 740 kBq with a standard uncertainty, $u_R$ = 43 kBq (see Section 4.1 of the Final Report). The value $x_i$ is the equivalent activity for laboratory *i*.**

**The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms: $D_i = (x_i - x_R)$ and $U_i$, its expanded uncertainty ($k$ = 2), both expressed in MBq, and $U_i = 2((1 - 2w_i)u_i^2 + u_R^2)^{1/2}$ when each laboratory has contributed to the calculation of $x_R$.**

**When required, the degree of equivalence between two laboratories is given by a pair of terms: $D_{ij} = D_i - D_j = (x_i - x_j)$ and $U_{ij}$, its expanded uncertainty ($k$ = 2), both expressed in MBq. The approximation $U_{ij} \sim 2(u_i^2 + u_j^2)^{1/2}$ may be used in the following table.**

**Linking CCRI(II)-K2.Zn-65 (2003) to BIPM.RI(II)-K1.Zn-65**

**The value $x_i$ is the equivalent activity for laboratory *i* participant in CCRI(II)-K2.Zn-65 whose ampoule was also measured in the SIR (see Final report).**

**The degree of equivalence of laboratory *i* participant in CCRI(II)-K2.Zn-65 with respect to the key comparison reference value is given by a pair of terms: $D_i = (x_i - x_R)$ and $U_i$, its expanded uncertainty ($k$ = 2), both expressed in MBq and $U_i = 2((1 - 2w_i)u_i^2 + u_R^2)^{1/2}$ when each laboratory has contributed to the calculation of $x_R$.**

**When required, the degree of equivalence between two laboratories *i* and *j*, one participant in BIPM.RI(II)-K1.Zn-65 and one in CCRI(II)-K2.Zn-65, or both participant in CCRI(II)-K2.Zn-65, is given by a pair of terms: $D_{ij} = D_i - D_j$ and $U_{ij}$, its expanded uncertainty ($k$ = 2),**

**both expressed in MBq, where the approximation $U_{ij} \sim 2(u_i^2 + u_j^2)^{1/2}$ may be used.**

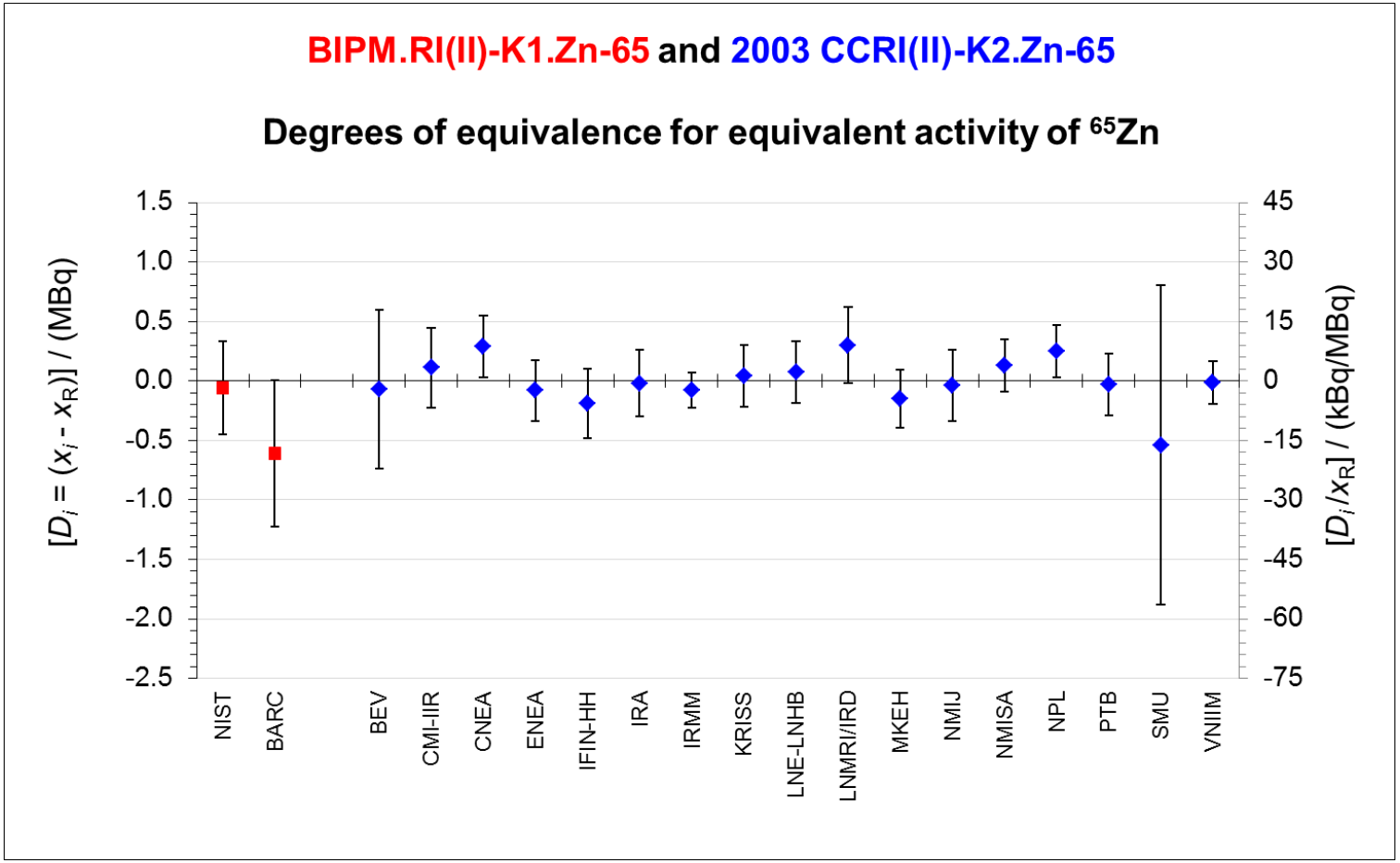
**These statements make it possible to extend the BIPM.RI(II)-K1.Zn-65 matrices of equivalence to the other participants in CCRI(II)- K2.Zn-65.**

**Lab *i*** ⇩

| | $D_i$ / MBq | $U_i$ / MBq |
|---|---|---|
| **NIST** | **-0.06** | 0.39 |
| **BARC** | **-0.61** | 0.61 |

| | | |
|---|---|---|
| **BEV** | **-0.07** | 0.67 |
| **CMI-IIR** | **0.11** | 0.34 |
| **CNEA** | **0.29** | 0.26 |
| **ENEA** | **-0.08** | 0.25 |
| **IFIN-HH** | **-0.19** | 0.29 |
| **IRA** | **-0.02** | 0.28 |
| **IRMM** | **-0.08** | 0.15 |
| **KRISS** | **0.04** | 0.26 |
| **LNE-LNHB** | **0.07** | 0.26 |
| **LNMRI/IRD** | **0.30** | 0.32 |
| **MKEH** | **-0.15** | 0.24 |
| **NMIJ** | **-0.04** | 0.30 |
| **NMISA** | **0.13** | 0.22 |
| **NPL** | **0.25** | 0.22 |
| **PTB** | **-0.03** | 0.26 |
| **SMU** | **-0.54** | 1.34 |
| **VNIIM** | **-0.01** | 0.18 |

**Figure 1. Graph of degrees of equivalence with the KCRV for $^{65}$Zn**
(as it appears in Appendix B of the MRA)




N.B. The right hand axis shows approximate values only

## Appendix 1. Uncertainty budgets for the activity of $^{65}$Zn submitted to the SIR

### BARC, 2006

| **Relative standard uncertainties** | $u_i \times 10^4$ evaluated by method | |
|---|---|---|
| **Contributions due to** | **A** | **B** |
| counting statistics | 50 | – |
| weighing | – | 5 |
| dead time | – | 3 |
| resolving time of coincidence analyser | – | 34 |
| background | – | 6 |
| half-life | – | 8 |
| extrapolation | 85 | – |
| **Quadratic summation** | **99** | **36** |
| **Relative combined standard uncertainty, $u_c$** | **105** | |

## Appendix 2. Uncertainty budgets for the activity measurements of $^{65}$Zn in the CCRI(II)-K2.Zn-65 comparison

| **Uncertainty components** | Laboratory Method | | | |
|---|---|---|---|---|
| | **BARC 4πICγ** | | **BEV 4πICγ** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.45 | A | 0.37 | |
| weighing | | | 0.05 | |
| background | | | 0.81 | |
| half-life | | | 0.11 | |
| calibration factor | 0.8 | B | 0.67 | |
| source positioning | 0.2 | B | | |
| source volume | 0.2 | B | | |
| collection efficiency | 0.1 | B | | |
| electrometer non-linearity | 0.1 | B | | |
| wall thickness of source container | 0.15 | B | | |
| current from radium source | 0.1 | B | | |
| current measurement | | | 0.10 | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **1.0** | | **1.12** | |

| **Uncertainty components** | Laboratory Method | | | |
|---|---|---|---|---|
| | **CMI-IIR $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | | **CNEA $4\pi(PPC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.3 | | 0.2 | |
| weighing | 0.05 | | 0.15 | |
| dead time | 0.01 | | < 0.01 | |
| background | 0.1 | | 0.2 | |
| pile-up | 0.01 | | | |
| resolving time | 0.05 | | 0.05 | |
| Gandy effect | 0.02 | | | |
| counting time | 0.02 | | 0.01 | |
| adsorption | 0.05 | | | |
| impurities | 0.01 | | | |
| half-life | 0.04 | | 0.2 | |
| extrapolation of efficiency curve | 0.45 | | 0.16 | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.56** | | **0.41** | |

| **Uncertainty components** | Laboratory Method | | | | | |
|---|---|---|---|---|---|---|
| | **ENEA 4πICγ[1)]** | | **ENEA 4πICγ[2)]** | | **IFIN-HH 4π(PC)β$^+$,e$_A$,x$_e$-γ coincidence** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.3 | A | 0.42 | A | 0.37 | |
| weighing | 0.05 | A | 0.03 | B | 0.05 | |
| dead time | 0.1 | B | 0.01 | B | 0.015 | |
| background | 0.006 | A | 0.02 | B | 0.23[3)] | |
| resolving time | | | 0.01 | B | 0.003 | |
| Gandy effect | | | 0.01 | B | | |
| counting time | 0.01 | B | | | | |
| adsorption | 0.02 | B | | | | |
| tracer ($^3$H standard) | 0.01 | B | | | | |
| input parameters and statistical model | 0.1 | B | | | | |
| quenching | 0.1 | A | | | | |
| decay-scheme parameters | 0.01 | B | | | | |
| half-life | < 0.01 | B | | | 0.013 | |
| photomultipliers asymmetry | 0.1 | B | | | | |
| ionization quench | 0.2 | B | | | | |
| scintillator stability | 0.1 | A | | | | |
| TSIE determination | 0.04 | A | | | | |
| mass determination | 0.1 | B | | | | |
| extrapolation of efficiency curve | | | | | 0.22 | |
| ionization chamber measurement | 0.3 | B | 0.3 | B | | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.53** | | **0.52[4)]** | | **0.49** | |

[1)] ionization chamber calibrated with another solution standardized by the 4π(LS) CIEMAT/NIST method in 2002.

[2)] ionization chamber calibrated with another solution standardized by the 4π(PC)β$^+$,e$_A$,x$_e$-γ coincidence method in 2002 [14].

[3)] in the γ channel. 0.042 % in the 4π (PC) channel.

[4)] the $u_A$ value is prevalent because of the low coincidence counting rates originating in low counting efficiencies in the β and γ channels.

| **Uncertainty components** | Laboratory Method | | | | | |
|---|---|---|---|---|---|---|
| | **IRA $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | | **IRMM $4\pi(PPC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | | **IRMM $4\pi$(LS) CIEMAT/NIST** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | | | 0.05 | | 0.1[5)] | |
| weighing | 0.03[6)] | B | 0.05 | | 0.1 | |
| dead time | 0.001[7)] | B | < 0.01 | | 0.1 | |
| background | 0.31[8)] | B | < 0.02[9)] | | 0.005 | |
| pile-up | [10)] | | | | | |
| resolving time | | | < 0.01 | | | |
| timing | 0.002[11)] | B | | | | |
| Gandy effect | | | 0.01 | | | |
| counting time | | | < 0.001 | | 0.05 | |
| adsorption | | | 0.06 | | 0.06 | |
| interpolation from calibration curve | | | | | 0.9 | |
| decay scheme parameters | | | | | 0.23 | |
| half-life | 0.02[12)] | B | 0.02 | | 0.02 | |
| extrapolation of efficiency curve | 0.30[13)] | A | 0.17[14)] | | | |
| wall effect | | | | | 0.05 | |
| *kB* | | | | | 0.3 | |
| sample stability | | | | | 0.1 | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.43** | | **0.18** | | **1.0** | |

[5)] for individual measurements.
[6)] $\Delta m/m_{\min}$.
[7)] $\Delta\tau N_{\beta\max} / (1 - N_{\beta\max}\tau)$.
[8)] $\Delta N_{z\gamma}/N_{\gamma\min}$.
[9)] included in counting statistics.
[10)] included in type A evaluation.
[11)] worst case time base error.
[12)] $\Delta$*decay factor*/*decay factor*.
[13)] standard deviation of the weighted mean of intercepts for three $\gamma$ gate settings i.e.: 467 keV – $\infty$; 561 keV – $\infty$ and 1015 keV – $\infty$.
[14)] including statistical uncertainty, mass uncertainty, confidence limits.

| **Uncertainty components** | Laboratory Method | | | |
|---|---|---|---|---|
| | **KRISS $4\pi(PPC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | | **LNE-LNHB $4\pi$(LS) TDCR** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.08[15)] | | 0.1 | |
| weighing | 0.07[16)] | | 0.05 | |
| dead time | 0.04[17)] | | < 0.01 | |
| background | 0.13[18)] | | < 0.02 | |
| pile-up | | | 0.03 | |
| resolving time | 0.05[19)] | | < 0.000001 | |
| counting time | 0.01[20)] | | < 0.01 | |
| adsorption | 0.08 | | | |
| scheme correction | | | | |
| impurities | | | | |
| quenching (*kB*) | | | 0.2 | |
| decay-scheme parameters | | | 0.48[21)] | |
| half-life | 0.01 | | 0.0059 | |
| extrapolation of efficiency curve | 0.34[22)] | | | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.39** | | **0.53** | |

[15)] standard uncertainty of 7 results.
[16)] calculated from $\Delta m/m$.
[17)] estimated from measured uncertainty.
[18)] calculated from measured uncertainty.
[19)] estimated from measured uncertainty.
[20)] estimated from time distribution.
[21)] $P_K$, 0.1 % ; $I_{\beta+}$, 0.2 %, $P_{absorption}$ $K_\alpha$, 0.3 % ; $P_{absorption}$ $K_\beta$, < 0.001 %; $P_{absorption}\gamma$, 0.2 %.
[22)] from residuals for linear extrapolations.

| **Uncertainty components** | Laboratory Method | | | |
|---|---|---|---|---|
| | **LNE-LNHB $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ anticoincidence** | | **LNE-LNHB $4\pi(LS)\beta^+,e_A,x_e$-$\gamma$ coincidence** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.23 | | 0.22 | |
| weighing | 0.05 | | 0.05 | |
| dead time | [23)] | | [24)] | |
| background | 0.15 | | [25)] | |
| pile-up | | | | |
| resolving time | | | | |
| counting time | 0.01 | | 0.01 | |
| scheme correction | 0.13[26)] | | 0.13[27)] | |
| impurities | | | | |
| quenching (*kB*) | | | | |
| decay-scheme parameters | | | | |
| half-life | 0.015 | | 0.015 | |
| extrapolation of efficiency curve | 0.24 | | 0.5 | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.39** | | **0.56** | |

[23)] neglected because acquisition of data in live time mode.
[24)] neglected because acquisition of data in live time mode
[25)] included in statistics.
[26)] due to uncertainty of $P_K$.
[27)] due to uncertainty of $P_K$.

| **Uncertainty components** | Laboratory Method | | | | | |
|---|---|---|---|---|---|---|
| | **LMNRI/IRD $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | | **LMNRI/IRD $4\pi$(LS) CIEMAT/NIST** | | **CNEN-IPEN $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.46[28)] | | 0.24 | | [29)] | |
| weighing | 0.10 | | < 0.01 | | 0.1 | |
| dead time | 0.17 | | | | 0.1 | |
| background | 0.02 | | | | 0.3 | |
| resolving time | 0.10 | | | | | |
| Gandy effect | 0.17 | | | | | |
| quench determination of $^3$H standards | | | 0.23 | | | |
| $^3$H counting | | | 0.04 | | | |
| photomultiplier efficiency loss | | | 0.07 | | | |
| photomultiplier asymmetry | | | 0.10 | | | |
| weighing of $^3$H samples | | | < 0.01 | | | |
| quench determination of $^{65}$Zn samples | | | 0.21 | | | |
| ionization quench | | | 0.10 | | | |
| $^3$H activity standardization | | | 0.19 | | | |
| decay-scheme parameters | | | 0.30 | | 0.14 | |
| half-life | 0.02 | | | | 0.11 | |
| extrapolation of efficiency curve | 0.43 | | | | 0.35 | |
| satellite pulses | | | | | | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.51** | | **0.55** | | **0.51** | |

[28)] included in the fitting of efficiency extrapolation curve.
[29)] included in the extrapolation procedure.

| **Uncertainty components** | Laboratory Method | | | | | |
|---|---|---|---|---|---|---|
| | **MKEH $4\pi(PPC)\beta^+,e_A,x_e$-$\gamma$ coincidence and anticoincidence** | | **NMIJ $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | | **NMISA $4\pi(LS)\beta^+,e_A,x_e$-$\gamma$ coincidence** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.26 | | 0.1 | | 0.11 | |
| weighing | 0.005 | | 0.1 | | 0.02 | |
| dead time | 0.005 | | 0.05 | | 0.006 | |
| background | 0.05 | | 0.05 | | 0.10 | |
| pile-up | | | 0.05 | | | |
| resolving time | 0.01 | | 0.05 | | < 0.001 | |
| Gandy effect | 0.05 | | 0.1 | | | |
| counting time | 0.005 | | 0.05 | | | |
| adsorption | 0.02 | | 0.05 | | | |
| impurities | 0.04 | | 0.1[30)] | | < 0.012 | |
| interpolation from calibration curve | | | 0.1 | | | |
| decay-scheme parameters | | | 0.1 | | | |
| half-life | < 0.02 | | 0.1 | | 0.25 | |
| self-absorption | | | 0.1 | | | |
| extrapolation of efficiency curve | 0.25 | | 0.4 | | 0.26 | |
| satellite pulses | | | | | 0.06 | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.37** | | **0.5** | | **0.31** | |

[30)] no impurity found by HP Ge spectrometry.

| **Uncertainty components** | Laboratory Method | | | |
|---|---|---|---|---|
| | **NPL $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | | **NPL $4\pi(LS)\beta^+$-$\gamma$ coincidence with DCC** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.27 | | 0.23 | |
| weighing | 0.05 | | 0.05 | |
| dead time | 0.01 | | 0.01 | |
| background | 0.05 | | 0.05 | |
| pile-up | < 0.01 | | < 0.01 | |
| resolving time | 0.004 | | 0.004 | |
| Gandy effect | < 0.001 | | < 0.001 | |
| counting time | < 0.01 | | < 0.01 | |
| adsorption | 0.01 | | 0.01 | |
| impurities | < 0.001 | | < 0.001 | |
| half-life | 0.018 | | 0.01 | |
| extrapolation of efficiency curve | 0.52 | | 0.30 | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.59** | | **0.39** | |

| **Uncertainty components** | Laboratory Method | | | |
|---|---|---|---|---|
| | **PTB $4\pi(PC)\beta^+,e_A,x_e$-$\gamma$ coincidence** | | **PTB $4\pi$(LS) CIEMAT/NIST** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 0.15 | | 0.061 | |
| weighing | 0.03 | | 0.062 | |
| dead time | < 0.02[31)] | | 0.1 | |
| background | 0.40 | | 0.01 | |
| resolving time | < 0.02[32)] | | | |
| Gandy effect | 0.02 | | | |
| counting time | < 0.001[33)] | | < 0.01 | |
| impurities | [34)] | | | |
| tracer | | | 0.923 | |
| input parameters and statistical model | | | 0.1 | |
| quenching | | | 0.05 | |
| interpolation from calibration curve | | | [35)] | |
| decay-scheme parameters | | | 0.01 | |
| half-life | 0.03 | | 0.037[36)] | |
| source stability | | | 0.2 | |
| extrapolation of efficiency curve | 0.01 | | | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **0.41[37)]** | | **0.96** | |

[31)] negligible.
[32)] negligible.
[33)] negligible.
[34)] no impurities detected.
[35)] part of tracer uncertainty.
[36)] $\Delta t$ = 122 d.
[37)] coefficients of correlation taken into account in the summation reducing the combined uncertainty by about 0.02 %.

| **Uncertainty components** | Laboratory Method | | | | | |
|---|---|---|---|---|---|---|
| | **SMU 4πICγ** | | **VNIIM 4π(PC)β$^+$,e$_A$,x$_e$-γ coincidence** | | **VNIIM Kx-γ coincidence** | |
| | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method | Relative value / (%) | Evaluation method |
| counting statistics | 1.8[38)] | | 0.13 | | 0.235 | |
| weighing | 0.0003 | | 0.1 | | 0.05 | |
| dead time | | | < 0.001 | | < 0.001 | |
| background | 1.0[39)] | | 0.1 | | 0.02 | |
| resolving time | | | 0.003 | | 0.006 | |
| interpolation from calibration curve | 1.0[40)] | | | | | |
| decay-scheme parameters | 0.4[41)] | | | | 0.02[42)] | |
| half-life | 0.012[43)] | | 0.004 | | 0.004 | |
| self-absorption | 0.2[44)] | | | | | |
| extrapolation of efficiency curve | | | 0.16 | | | |
| 511 keV to 1115 keV γ-ray efficiency ratio setting | | | 1.2 | | | |
| γ-ray efficiency of the Kx-ray detector | | | | | 0.04 | |
| **Combined uncertainty** (as quadratic sum of all uncertainty components) | **2.3** | | **1.25** | | **0.25** | |

[38)] standard deviation of sample measurements.
[39)] standard deviation of background measurements relative to sample.
[40)] estimated overall calibration curve uncertainty.
[41)] according to PTB tables used.
[42)] $\beta^+$ emission probability: $P_{\beta+} = 1.42$ %; $u = 0.02$ %.
[43)] including uncertainty in decay time.
[44)] differences in ampoule dimensions and filling.

## Appendix 3. 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** |
|---|---|---|---|
| $4\pi$ | 4P | proportional counter | PC |
| defined solid angle | SA | press. prop. counter | PP |
| $2\pi$ | 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 |
| | | 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 |
| mixed electrons | ME | coincidence | CO |
| bremsstrahlung | BS | anti-coincidence | AC |
| gamma rays | GR | coincidence counting with efficiency tracing | CT |
| X - rays | XR | anti-coincidence counting with efficiency tracing | AT |
| photons (x + $\gamma$) | PH | triple-to-double coincidence ratio counting | TD |
| alpha - particle | AP | selective sampling | SS |
| mixture of various radiations | MX | high efficiency | HE |
| | | | |

| **Examples** **method** | **acronym** |
|---|---|
| $4\pi$(PC)$\beta$–$\gamma$-coincidence counting | 4P-PC-BP-NA-GR-CO |
| $4\pi$(PPC)$\beta$–$\gamma$-coincidence counting eff. trac. | 4P-PP-MX-NA-GR-CT |
| defined solid angle $\alpha$-particle counting with a PIPS detector | SA-PS-AP-00-00-00 |
| $4\pi$(PPC)AX-$\gamma$(Ge(HP))-anticoincidence counting | 4P-PP-MX-GH-GR-AC |
| $4\pi$ CsI-$\beta$,AX,$\gamma$ counting | 4P-CS-MX-00-00-HE |
| calibrated IC | 4P-IC-GR-00-00-00 |
| internal gas counting | 4P-PC-BP-00-00-IG |