

**BIPM comparison BIPM.RI(II)-K1.Ge-68 of**  
**activity measurements of the radionuclide <sup>68</sup>Ge for the LNMRI/IRD, NIST, NIM,**  
**IRA-METAS, LNE-LNHB and the TAEK,**  
**and the linked 2015 CCRI(II)-K2.Ge-68 comparison**

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## **Abstract**

This report summarises the results for the new BIPM.RI(II)-K1.Ge-68 comparison. Six metrology institutes have participated, enabling the first value of the key comparison reference value (KCRV) to be determined. The KCRV was calculated using the power-moderated weighted mean and the results. A link has been made to the CCRI(II)-K2.Ge-68 comparison held in 2015 through the NIST, NIM, IRA-METAS and the LNE-LNHB who participated in both comparisons. One NMI 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 five NMIs in the BIPM.RI(II)-K1.Ge-68 comparison and the 12 other participants in the CCRI(II)-K2.Ge-68 comparison. A graphical presentation is also given.

## **1. Introduction**

The SIR for activity measurements of  $\gamma$ -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 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  $^{226}\text{Ra}$  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 2019, the SIR has measured 1016 ampoules to give 771 independent results for 72 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.Ge-68 key comparison.

In addition, an international comparison was held in 2015 for this radionuclide, CCRI(II)-K2.Ge-68 [3]. Seventeen laboratories took part in this comparison including the NIST, NIM, IRA-METAS and the LNE-LNHB who participated in the SIR at the same time, enabling to link the CCRI(II)-K2 comparison to the BIPM.RI(II)-K1 comparison. One NMI had previously submitted ampoules to the SIR and has updated their result through this CCRI(II) comparison.

## **2. Participants**

Six NMIs submitted ampoules for inclusion in this new BIPM.RI(II)-K1 comparison. The details of the laboratories that have participated in the comparison are given in Table 1a. The date of measurement in the SIR is also included and is used in the KCDB and all references in this report.

The 13 additional NMIs that took part in the CCRI(II) international comparison CCRI(II)-K2.Ge-68 in 2015 and are also eligible for the KCDB are shown in Table 1b. This include the LNMRI/IRD, that used this comparison to update their result, and the TAEK, whose result is superseded by the later participation in the SIR.

**Table 1a. Details of the participants in the BIPM.RI(II)-K1.Ge-68**

| <b>NMI</b>     | <b>Original acronym</b> | <b>Full name</b>  | <b>Country</b> | <b>Regional metrology organization</b> | <b>Date of measurement at the BIPM</b><br>YYYY-MM-DD |
|----------------|-------------------------|---|----------------|--|--|
| LNMRI/<br>IRD* | –                       | Laboratorio Nacional de Metrologia das Radiações Ionizantes/<br>Instituto de Radioproteção e Dosimetria | Brazil         | SIM                                    | 2013-01-17   |
| NIST           | –                       | National Institute of Standards and Technology  | United States  | SIM                                    | 2014-12-15   |
| NIM            | –                       | National Institute of Metrology   | China          | APMP                                   | 2015-03-23   |
| IRA-METAS      | –                       | Institut de radiophysique appliquée -<br>Institut fédéral de métrologie                                 | Switzerland    | EURAMET                                | 2015-04-22   |
| LNE-LNHB       | –                       | Laboratoire national de métrologie et d'essais –<br>Laboratoire national Henri Becquerel                | France         | EURAMET                                | 2015-05-28   |
| TAEK           | –                       | Turkish Atomic Energy Authority   | Turkey         | EURAMET                                | 2018-01-04   |

\*Participation superseded by the 2015 CCRI(II)-K2.Ge-68 comparison below

**Table 1b. Details of the participants in the 2015 CCRI(II)-K2.Ge-68 to be linked to BIPM.RI(II)-K1.Ge-68 comparison**

| <b>NMI</b> | <b>Full name</b>   | <b>Country</b> | <b>Regional metrology organization</b> |
|------------|--|----------------|--|
| ANSTO      | Australian Nuclear Science and Technology Organisation   | Australia      | APMP                                   |
| BARC       | Bhabha Atomic Research Centre  | India          | APMP                                   |
| CIEMAT     | Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas                               | Spain          | EURAMET                                |
| IFIN-HH    | Institutul National de Cercetare - Dezvoltare in Fizica si Inginerie Nucleara - "Horia Hulubei"      | Romania        | EURAMET, COOMET                        |
| INER       | Institute of Nuclear Energy Research   | Taiwan         | APMP                                   |
| KRISS      | Korea Research Institute of Standards and Science  | Korea          | APMP                                   |
| LNMRI/IRD  | Laboratório Nacional de Metrologia das Radiações Ionizantes, Instituto de Radioproteção e Dosimetria | Brazil         | SIM                                    |
| NMIJ       | National Metrology Institute of Japan  | Japan          | APMP                                   |
| NPL        | National Physical Laboratory   | United Kingdom | EURAMET                                |
| POLATOM    | National Centre for Nuclear Research, Radioisotope Centre POLATOM                                    | Poland         | EURAMET                                |
| PTB        | Physikalisch-Technische Bundesanstalt  | Germany        | EURAMET, COOMET                        |
| SMU        | Slovenský Metrologický Ústav   | Slovakia       | EURAMET, COOMET                        |
| TAEK*      | Turkish Atomic Energy Authority  | Turkey         | EURAMET, COOMET                        |

\*Participation superseded by the submission of an ampoule to the SIR in 2018

### 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, the activities submitted, the relative combined standard uncertainties and the half-life used by the participants in the SIR are given in Table 2. Most participants performed primary standardizations of  $^{68}\text{Ga}$  from which the activity of  $^{68}\text{Ge}$  was inferred. The uncertainty budgets for the SIR submissions from LNMRI/IRD and TAEK are given in Appendix 1. The uncertainty budgets for the other SIR participants and all the participants in the CCRI(II)-K2.Ge-68 comparison were published as Appendix B of the final report [3]. The acronyms used for the measurement methods are given in Appendix 2.

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.

**Table 2. Standardization methods of the SIR participants for  $^{68}\text{Ge}$**

| NMI       | Method used and acronym (see Appendix 2)   | Half-life / d          | Activity $A_i$ / kBq | Reference date<br>YYYY-MM-DD | Relative standard uncertainty / $10^{-2}$ by method of evaluation |      |
|-----------|--|------------------------|----------------------|------------------------------|---|------|
|           |  |                        |                      |                              | A   | B    |
| LNMRI/IRD | $4\pi(\text{LS})\beta^+-\gamma$ anti-coinc. counting<br>4P-LS-PO-NA-GR-AC            | 270.95(16)             | 867.0                | 2012-04-03<br>12h UTC        | 0.05  | 0.17 |
| NIST      | 4P-LS-PO-NA-GR-AC  | 270.95                 | 2244                 | 2014-11-14<br>12h UTC        | 0.19  | 0.59 |
| NIM       | Liquid scint. TDCR and CIEMAT/NIST<br>4P-LS-PO-00-00-TD<br>4P-LS-PO-00-00-CN         | 270.95(26)<br>[4]      | 1448 <sup>#</sup>    | 2014-11-14<br>12h UTC        | 0.57  |      |
| IRA-METAS | $4\pi$ plastic scintill. - $4\pi$ well-type NaI coinc. counting<br>4P-SP-PO-NA-GR-CO | 270.95(26)<br>[4]<br>* | 697.7                | 2014-11-14<br>12h UTC        | 0.15  | 0.51 |
| LNE-LNHB  | 4P-LS-PO-NA-GR-AC  | 270.95(26)<br>[4]      | 2295                 | 2014-11-14<br>12 h UT        | 0.18  | 0.47 |
| TAEK      | Pressurized $4\pi\gamma$ **<br>4P-IC-GR-00-00-00                                     | 270.95(26)<br>[4]      | 481.3                | 2016-04-01<br>12 h UT        | 0.33  | 1.45 |

\* Ga-68 half-life is 67.83(20) min

\*\* calibrated at the Physikalisch-Technische Bundesanstalt, Germany in 2014

# arithmetic mean result from two measurements methods, and combined uncertainty

**Table 3. Details of the solution of  $^{68}\text{Ge}$  submitted**

| NMI/<br>SIR year                   | Chemical<br>composition                           | Solvent<br>conc. /<br>(mol dm <sup>-3</sup> ) | Carrier:<br>conc.<br>/( $\mu\text{g g}^{-1}$ )   | Density<br>/(g cm <sup>-3</sup> ) | Relative activity<br>of impurity <sup>a</sup> |
|------------------------------------|---|---|--|-----------------------------------|---|
| LNMRI/<br>IRD<br>2013              | HCl   | 0.1   | –  | 0.99                              | –   |
| NIST<br>2014 <sup>b</sup>          | Ge and Ga in<br>HCl                               | 0.5   | Ge <sup>4+</sup> : 65<br>Ga <sup>3+</sup> : 62   | 1.006                             | –   |
| NIM<br>2015 <sup>c</sup>           | Ge <sup>+4</sup> and<br>Ge <sup>+3</sup> in HCl   | 0.5   | Ge <sup>4+</sup> : 65<br>Ga <sup>3+</sup> : 65   | 1.0                               | –   |
| IRA-<br>METAS<br>2015 <sup>d</sup> | Ge <sup>+4</sup> and<br>Ge <sup>+3</sup> in HCl   | 0.5   | Ge <sup>4+</sup> : 65<br>Ga <sup>3+</sup> : 65   | 1.007                             | –   |
| LNE-<br>LNHB<br>2015 <sup>b</sup>  | Ge and Ga in<br>HCl                               | 0.5   | Ge <sup>4+</sup> : 65<br>Ga <sup>3+</sup> : 62   | 1.007                             | –   |
| TAEK<br>2018                       | GeCl <sub>4</sub> and<br>GaCl <sub>3</sub> in HCl | 0.5   | GeCl <sub>4</sub> : 65<br>GaCl <sub>3</sub> : 62 | 1.006                             | –   |

<sup>a</sup> The ratio of the activity of the impurity to the activity of  $^{68}\text{Ge}$  at the reference date.

<sup>b</sup> Same solution as for the CCRI(II)-K2.Ge-68 comparison.

<sup>c</sup> solution from the CCRI(II)-K2.Ge-68 comparison diluted by a factor of 1.504 00(8)

<sup>d</sup> solution from the CCRI(II)-K2.Ge-68 comparison diluted by a factor of 3.185 50(13)

#### 4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "master-file". The recent submissions have provided six ampoules for the activity measurements for  $^{68}\text{Ge}$ . 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  $^{226}\text{Ra}$ , all the SIR results are normalized to the radium source number 5 [1].

The half-life used by the BIPM is 270.95(26) d from the BIPM Monographie 5 [4] and is the same half-life used in the CCRI(II)-K2.Ge-68 comparison.

**Table 4a. Results of SIR measurements of <sup>68</sup>Ge**

| NMI/<br>SIR<br>year   | Mass of<br>solution <i>m</i><br>/g | Activity<br>submitted<br><i>A</i> / kBq | No. of<br>Ra<br>source<br>used | SIR<br><i>A<sub>e</sub></i> / kBq | Relative<br>standard<br>uncertainty<br>from SIR<br>/10 <sup>-4</sup> | Combined<br>uncertainty<br><i>u(A<sub>e</sub>)</i> / kBq |
|-----------------------|------------------------------------|---|--------------------------------|-----------------------------------|--|--|
| LNMRI/<br>IRD<br>2013 | 3.486 97                           | 867.0                                   | 2                              | 15 772                            | 13   | 36   |
| NIST<br>2014          | 3.595 5                            | 2244                                    | 3                              | 15 829 <sup>b</sup>               | 5  | 98   |
| NIM<br>2015           | 3.596 88                           | 1448                                    | 3                              | 15 338 <sup>b</sup>               | 8  | 88   |
| IRA-<br>METAS<br>2015 | 3.566 10(21)                       | 697.7                                   | 2                              | 15 797 <sup>b</sup>               | 10   | 86   |
| LNE-<br>LNHB<br>2015  | 3.6702                             | 2295                                    | 3                              | 15 855 <sup>b</sup>               | 8  | 81   |
| TAEK<br>2018          | 3.6164                             | 481.3                                   | 1                              | 15 960                            | 22 <sup>a</sup>  | 240  |

<sup>a</sup> Uncertainty dominated by the contribution of the decay correction for <sup>68</sup>Ge

<sup>b</sup> Results used to link the CCRI(II)-K2 comparison

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.

An international comparison for this radionuclide, CCRI(II)-K2.Ge-68 was held in 2015 [3] and the twelve laboratories from this comparison to be added to the matrix of degrees of equivalence are given in Table 1b. The LNMRI/IRD used the CCRI(II) comparison to update their degree of equivalence from earlier SIR participation.

The results  $(A/m)_i$  of the CCRI(II) comparison have been linked to the BIPM.RI(II)-K1.Ge-68 comparison through the measurement in the SIR of four ampoules of the CCRI(II) solution (or a dilution of this solution), standardized by the NIST, NIM, IRA-METAS and LNE-LNHB. The linking factor  $L_j$  for each linking laboratory  $j$  is shown in Table 4b and is defined to be  $L_j = A_{e,j} / d_j(A/m)_j$  where the activity, mass and equivalent activity are taken from Table 4a and the dilution factor  $d_j$  for the NIM and IRA-METAS from Table 3. The uncertainty of  $L_j$  shown in Table 4b is obtained by combining in quadrature the uncertainty of the dilution factor and the uncertainty from the SIR measurement. The linking factor for the CCRI(II) comparison is finally obtained as the weighted mean of the factors  $L_j$ , where  $w_j = 1/u(L_j)^2$ :

$$A_{e,i} = (A/m)_i \times \sum_{j=1}^4 w_j \left[ A_{e,j} / d_j (A/m)_j \right] = (A/m)_i \times 25.355(9),$$

**Table 4b. Details of the link calculation**

| <b>NMI/<br/>SIR<br/>year</b> | <b>Activity conc.<br/>corrected for<br/>dilution<br/>(A/m)<sub>j</sub> × d<sub>j</sub><br/>/ kBq g<sup>-1</sup></b> | <b>Relative<br/>uncertainty<br/>of dilution<br/>factor d<sub>j</sub><br/>/10<sup>-4</sup></b> | <b>SIR<br/>A<sub>e,j</sub><br/>/ kBq</b> | <b>Relative<br/>standard<br/>uncertainty<br/>from SIR<br/>/10<sup>-4</sup></b> | <b>Linking<br/>factor<br/>L<sub>j</sub></b> |
|------------------------------|---|---|--|--|---|
| NIST<br>2014                 | 624   | –   | 15 829                                   | 5  | 25.37(1)                                    |
| NIM<br>2015                  | 605.7   | 0.5   | 15 338                                   | 8  | 25.32(2)                                    |
| IRA-<br>METAS<br>2015        | 623.3   | 0.4   | 15 797                                   | 10   | 25.35(3)                                    |
| LNE-<br>LNHB<br>2015         | 625.2   | –   | 15 855                                   | 8  | 25.36(2)                                    |

The linked results are detailed in Table 4c. The uncertainties for the CCRI(II) comparison results linked to the SIR are comprised of the original uncertainties together with the relative uncertainty in the link,  $4 \times 10^{-4}$ , given by the internal standard deviation of the linking values in Table 4b. Further details of the results of the CCRI(II)-K2.Ge-68 comparison are available in [3].



**Table 4c. Results of the 2015 CCRI(II) comparison of  $^{68}\text{Ge}$  and links to the SIR**

| NMI       | Measurement method acronym (see Appendix 2)  | Activity* concentration measured $(A/m)_i$ / (kBq g $^{-1}$ ) | Standard uncertainty $u_i$ / (kBq·g $^{-1}$ ) | Equivalent SIR activity $A_{ei}$ / MBq | Combined standard uncertainty $u_i$ / MBq |
|-----------|--|---|---|--|---|
| ANSTO     | 4P-LS-PO-NA-GR-CO  | 620.2   | 3.4   | 15 725                                 | 86  |
| BARC      | Mean result of 4P-LS-PO-NA-GR-CO (3 setups) and 4P-LS-MX-00-00-CN (see [3])              | 617.4   | 1.7   | 15 655                                 | 42  |
| CIEMAT    | Mean result of 4P-LS-MX-00-00-TD 4P-LS-MX-NA-GR-CO                                       | 618.5   | 2.2   | 15 682                                 | 56  |
| IFIN-HH   | 4P-PC-PO-NA-GR-CO  | 613.4   | 5.8   | 15 550                                 | 150                                       |
| INER      | 4P-LS-MX-00-00-CN  | 618.3   | 1.9   | 15 677                                 | 49  |
| KRISS     | 4P-LS-PO-NA-GR-CO  | 629.5   | 4.0   | 15 960                                 | 100                                       |
| LNMRI/IRD | 4P-LS-PO-NA-GR-AC  | 622.2   | 3.3   | 15 777                                 | 84  |
| NMIJ      | 4P-LS-MX-00-00-CN  | 624.1   | 4.4   | 15 820                                 | 110                                       |
| NPL       | 4P-LS-MX-GH-GR-CO  | 625.7   | 4.3   | 15 860                                 | 110                                       |
| POLATOM   | 4P-LS-PO-NA-GR-CO  | 625.4   | 4.0   | 15 860                                 | 100                                       |
| PTB       | Mean result of 4P-CD-PO-NA-GR-CO using double or triple coincidences in the beta channel | 626.4   | 5.1   | 15 880                                 | 130                                       |
| SMU       | 4P-LS-PO-00-00-TD  | 689.7   | 2.8   | 17 487                                 | 71  |
| TAEK**    | Weighted mean of 4P-NA-GR-00-00-HE 4P-IC-GR-00-00-00                                     | 628.0   | 4.0   | 15 920                                 | 100                                       |

\* referenced to 12:00 UTC 14 November 2014

\*\* result superseded by the SIR participation of this laboratory in 2018

#### 4.1 The Key Comparison Reference Value

In May 2013 the CCRI(II) decided to calculate the key comparison reference value (KCRV) using the power-moderated weighted mean [5] rather than an unweighted mean, as had been the policy. 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) results more than 20 years old are included in the calculation of the KCRV (but are not included in data shown in the KCDB or in the plots in this report as they have expired);
- d) 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;
- e) results can also be excluded for technical reasons; and
- f) 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  $^{68}\text{Ge}$  has been calculated for the first time and is equal to 15 800(31) kBq with the power  $\alpha = 1.25$  on the basis of the SIR results from the NIST, IRA-METAS, LNE-LNHB and LNMRI/IRD. The NIM result is not included as it has been identified as an outlier using the normalized error test as detailed in bullet d) above.

#### 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

$$D_i = A_{e_i} - \text{KCRV} \quad (1)$$

and the expanded uncertainty ( $k = 2$ ) of this difference,  $U_i$ , known as the equivalence uncertainty; hence

$$U_i = 2u(D_i). \quad (2)$$

When the result of the NMI  $i$  is included in the KCRV with a weight  $w_i$ , then

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

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

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

#### 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

$$D_{ij} = D_i - D_j = A_{e_i} - A_{e_j} \quad (5)$$

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

$$u_{D_{ij}}^2 = u_i^2 + u_j^2 - 2u(A_{e_i}, A_{e_j}) \quad (6)$$

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  $u(A_{e_i}, A_{e_j})$  (see [6] 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  $A_{e_i}$  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  $x_R$  in the KCDB), is shown in Figure 1 in relative terms. 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  $^{68}\text{Ge}$ , BIPM.RI(II)-K1.Ge-68 currently comprises five valid results. The key comparison reference value has been evaluated using the power-moderated weighted mean to include the NIST, IRA-METAS, LNE-LNHB and the LNMRI/IRD results. The SIR results have been analysed with respect to the first KCRV determined for this radionuclide, providing degrees of equivalence for five participants.

The results of twelve other NMIs that took part in the CCRI(II)-K2.Ge-68 comparison in 2015 have been linked to the BIPM ongoing key comparison through four ampoules of the comparison measured in the SIR. These results superseded one earlier result from the BIPM.RI(II)-K1.Ge-68 comparison. The linked results are included in the matrix of degrees of equivalence and shown on the graph. This has enabled the table of degrees of equivalence to include 17 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  $^{68}\text{Ge}$  activity measurements to the ongoing K1 comparison or take part in other linked comparisons.

## References

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**Table 5. Table of degrees of equivalence and introductory text for <sup>68</sup>Ge**

**Key comparison BIPM.RI(II)-K1.Ge-68**

**MEASURAND :**                      **Equivalent activity of <sup>68</sup>Ge**

**Key comparison reference value: the SIR reference value for this radionuclide is  $x_R = 15\,800$  kBq with a standard uncertainty,  $u_R = 31$  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**

**$U_i = 2((1 - 2w_i)u_i^2 + u_R^2)^{1/2}$ , where  $w_i$  is the weight of laboratory  $i$  contributing to the calculation of  $x_R$ .**

**Linking CCRI(II)-K2.Ge-68 (2015) to BIPM.RI(II)-K1.Ge-68**

**The value  $x_i$  is the equivalent activity for laboratory  $i$  participant in CCRI(II)-K2.Ge-68 having been normalized using the NIST, NIM, IRA-METAS and LNE-LNHB as linking laboratory (see Section 4 of the Final report).**

**The degree of equivalence of laboratory  $i$  participant in CCRI(II)-K2.Ge-68 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.**

**The approximation  $U_i = 2(u_i^2 + u_R^2)^{1/2}$  is used in the following table.**

**These statements make it possible to extend the BIPM.RI(II)-K1.Ge-68 matrices of equivalence to the other participants in CCRI(II)-K2.Ge-68.**

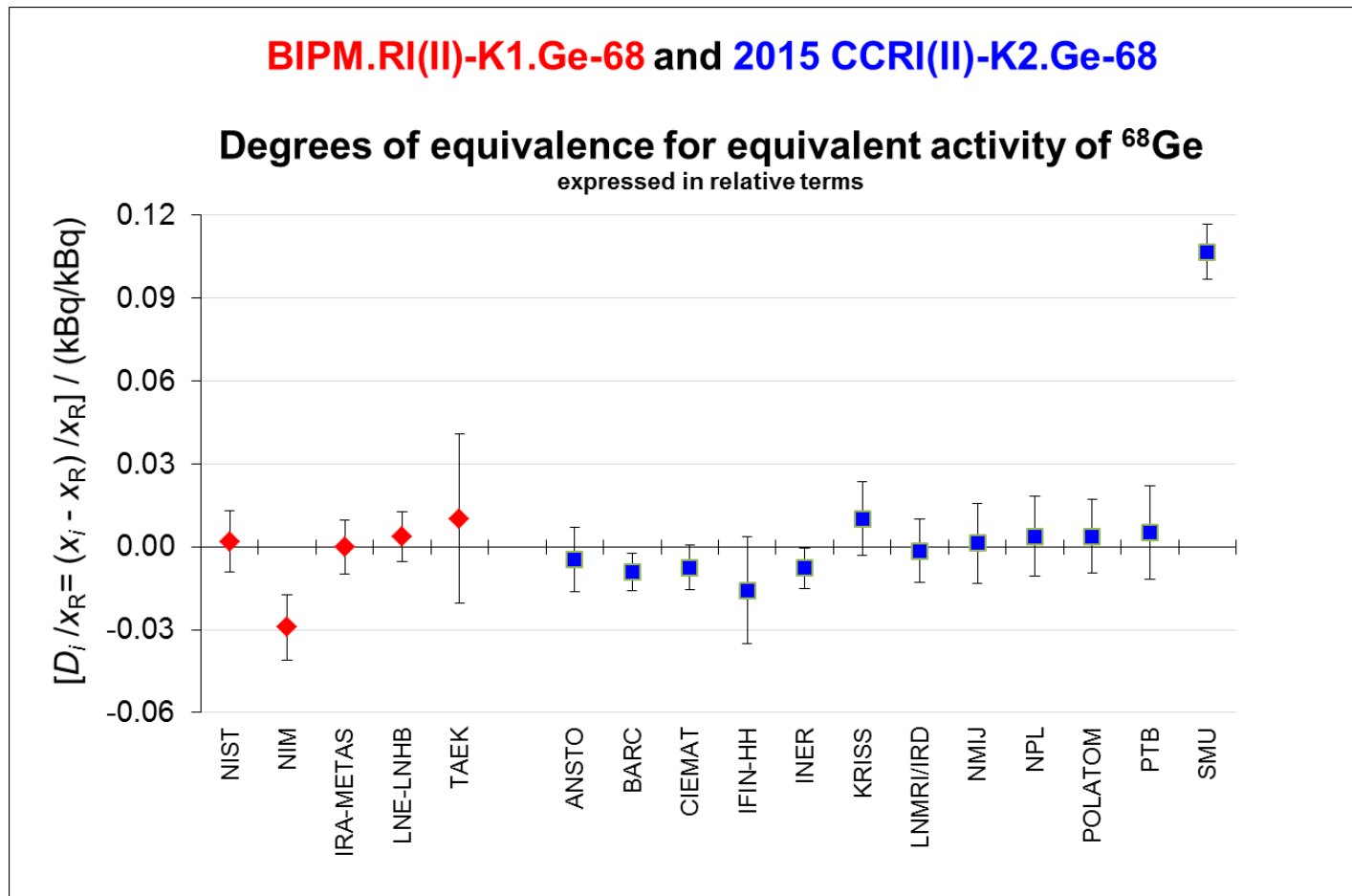
**Table 5 continued**

Lab *i*

|           | <i>D<sub>i</sub></i> | <i>U<sub>i</sub></i> |
|-----------|----------------------|----------------------|
|           | / MBq                |                      |
| NIST      | 0.03                 | 0.18                 |
| NIM       | -0.46                | 0.19                 |
| IRA-METAS | 0.00                 | 0.15                 |
| LNE-LNHB  | 0.06                 | 0.14                 |
| TAEK      | 0.16                 | 0.48                 |

|           |       |      |
|-----------|-------|------|
| ANSTO     | -0.07 | 0.18 |
| BARC      | -0.14 | 0.10 |
| CIEMAT    | -0.12 | 0.13 |
| IFIN-HH   | -0.25 | 0.31 |
| INER      | -0.12 | 0.11 |
| KRISS     | 0.16  | 0.21 |
| LNMRI/IRD | -0.02 | 0.18 |
| NMIJ      | 0.02  | 0.23 |
| NPL       | 0.06  | 0.23 |
| POLATOM   | 0.06  | 0.21 |
| PTB       | 0.08  | 0.27 |
| SMU       | 1.69  | 0.16 |

**Figure 1. Graph of degrees of equivalence with the KCRV for  $^{68}\text{Ge}$ , expressed in relative terms**  
 (as it appears in Appendix B of the MRA)



## Appendix 1. Uncertainty budgets for the activity of $^{68}\text{Ge}$ submitted to the SIR

The LNMRI/IRD submitted a detailed uncertainty budget as follows:

| Relative standard uncertainties                                 | $u_{rel,i} \times 10^4$<br>evaluated by method |           |
|---|--|-----------|
|   | A  | B         |
| <b>Contributions due to</b>                                     |  |           |
| counting statistics   | 2  | –         |
| weighing, dilution  | –  | 5         |
| dead time   | –  | 1         |
| decay scheme parameters   | –  | 13        |
| decay correction  | –  | 3         |
| extrapolation   | 5  | –         |
| interference of 1077 keV gamma-ray in the $\gamma$ -counting    | –  | 10        |
| <b>Quadratic summation</b>                                      | <b>5</b>                                       | <b>17</b> |
| <b>Relative combined standard uncertainty, <math>u_c</math></b> | <b>18</b>                                      |           |

The TAEK submitted a detailed uncertainty budget as follows:

| Relative standard uncertainties                                 | Comment  | $u_{rel,i} \times 10^4$<br>evaluated by method |            |
|---|--|--|------------|
|   |  | A  | B          |
| <b>Contributions due to</b>                                     |  |  |            |
| counting statistics   | Current measurement                                      | 22   | –          |
| weighing, dilution  | 2 $\mu\text{g}$ precision microbalance                   | –  | 0.3        |
| background  | Included in counting statistics                          | –  | –          |
| Cs-137 ref. source current measurement                          |  | 25   |            |
| ionization chamber calibration factor                           |  |  | 145        |
| adsorption  | Negligible (rinsed ampoules counting with HPGe detector) | –  | –          |
| impurities  | Negligible (not detectable at 20 cm with HPGe detector)  | –  | –          |
| Decay correction  |  | –  | 12         |
| <b>Quadratic summation</b>                                      |  | <b>33</b>                                      | <b>145</b> |
| <b>Relative combined standard uncertainty, <math>u_c</math></b> |  | <b>150</b>                                     |            |



## 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 |
|------------------------------|---------|---|---------|
| $4\pi$                       | 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      |
|                              |         | Cerenkov detector                                 | CD      |
|                              |         | calorimeter                                       | CA      |
|                              |         | solid plastic scintillator                        | SP      |
|                              |         | 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 radiation | MX      | high efficiency                                   | HE      |

| Examples  | method | acronym           |
|---|--------|-------------------|
| $4\pi(\text{PC})\beta\text{-}\gamma\text{-coincidence counting}$                  |        | 4P-PC-BP-NA-GR-CO |
| $4\pi(\text{PPC})\beta\text{-}\gamma\text{-coincidence counting eff. trac.}$      |        | 4P-PP-MX-NA-GR-CT |
| defined solid angle $\alpha$ -particle counting with a PIPS detector              |        | SA-PS-AP-00-00-00 |
| $4\pi(\text{PPC})\text{AX-}\gamma(\text{Ge(HP)})\text{-anticoincidence counting}$ |        | 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 |