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Results of an International Comparison of Activity Measurements of ^{68}Ge

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Abstract

An international key comparison, identifier CCRI(II)-K2.Ge-68, has been performed. The National Institute of Standards and Technology (NIST) served as the pilot laboratory, distributing aliquots of a $^{68}\text{Ge}/^{68}\text{Ga}$ solution. Results for the activity concentration, C_A , of ^{68}Ge at a reference date of 12h00 UTC 14 November 2014 were submitted by 17 laboratories, encompassing many variants of coincidence methods and liquid-scintillation counting methods. The first use of 4π (Cherenkov) β - γ coincidence and anticoincidence methods in an international comparison is reported. One participant reported results by secondary methods only. Two results, both utilizing pure liquid-scintillation methods, were identified as outliers. Evaluation using the Power-Moderated Mean method results in a proposed Comparison Reference Value (CRV) of $621.7(11) \text{ kBq}\cdot\text{g}^{-1}$, based on 14 results. The degrees of equivalence and their associated uncertainties are evaluated for each participant. Several participants submitted 3.6 mL ampoules to the BIPM to link the comparison to the International Reference System (SIR) which may lead to the evaluation of a Key Comparison Reference Value and associated degrees of equivalence.

1. Introduction

There has been increasing interest in the use of $^{68}\text{Ge}/^{68}\text{Ga}$ as a surrogate for ^{18}F in quantitative imaging (Zimmerman and Cessna, 2010; Zimmerman et al., 2014; Yamada et al., 2017), as well as increasing interest in the use of ^{68}Ga for radionuclide-based radiotherapy for certain types of cancers (Banerjee and Pomper, 2013). Accurate administrations of drugs using this radionuclide require accurate standards against which instrumentation used in the clinics and radiopharmacies can be calibrated.

A simplified decay scheme adapted from Bé et al. (2013) is shown in **Figure 1**. Germanium-68 decays with a half-life of 270.95(26) days by pure electron capture to the ground state of ^{68}Ga . Gallium-68 decays by both positron emission and by electron capture, mainly to the ground state of ^{68}Zn with a half-life of 67.83(20) minutes. Additionally, there is decay to a 1077 keV excited state with a probability of about 3 % and decay to several higher excited states with a combined probability less than 0.4 %. This decay scheme makes it suitable for analysis using a variety of techniques, including liquid-scintillation counting and coincidence counting.

There exists an ongoing comparison in the International Reference System (SIR), designated BIPM.RI(II)-K1.Ge-68 since 2012. The initial result was submitted by Laboratório Nacional de Metrologia das Radiações Ionizantes, Instituto de Radioproteção e Dosimetria (LNMRI/IRD), a participant in this comparison. Three NMIs had previously reported standardizing this radionuclide (Schönfeld et al., 1994; Grigorescu et al., 2004; Zimmerman et al., 2008), with additional papers having reported standardizing the daughter radionuclide alone (Roteta et al., 2012; Sahagia et al., 2012). In order to provide a means for laboratories to substantiate calibration and measurement capability (CMC) claims for this nuclide, a Key Comparison of ^{68}Ge was proposed, agreed, and carried out, designated CCRI(II)-K2.Ge-68

2. Organization of the comparison

The bulk ^{68}Ge was purchased from International Isotopes, Inc. (Idaho Falls, Idaho) as 130 MBq in 1 mL solution. The production method was not provided and could not be obtained. This master solution was diluted with a carrier for a resulting solution containing 65 μg and 62 μg non-active Ge^{+4} and Ga^{+3} ions, respectively, per gram of 0.5 mol·L⁻¹ HCl. The solution density was measured to be 1.0062(13) g·mL⁻¹ at 20 °C. The carrier solution was chosen based on the recommendations of Mirzadeh and Lambrecht (1996) to avoid the potential loss of germanium from the solution. During production of the carrier solution, concerns were raised over the long-term stability of the proposed composition. Utilizing solutions with approximately the same composition, previously prepared at NIST for a primary standardization, a study of the solution composition was undertaken. Using 2-year old sources, the composition was shown to successfully transfer, ampoule to ampoule, with no change in activity concentration. Additionally, activity determinations were made using the identical method on the same solution after storage for 7 years. There was no discernable change in activity concentration, within uncertainties. Full details can be found in Zimmerman et al. (2016). Despite the long-term stability of the carrier composition, participants were cautioned about the continued possibility of volatility if the solution is taken to dryness.

The comparison solution was dispensed into 5-mL ampoules using an automated dispenser, with individually determined masses. Homogeneity was checked by measurement in two overlapping

batches in the NIST 4 π automated ionization chamber against a ^{226}Ra reference standard. The standard deviation on the calculated activity concentration over all ampoules was 0.02 %.

Table 1 lists the participant laboratories and their respective Regional Metrology Organizations. The majority of sources were received by the participants over a period from 10 October 2014 to 14 November 2014. Due to bureaucratic delay and a temporary shipping embargo two sources were delayed until 1 January 2015 and 22 April 2015. The reporting period opened on 28 May 2015. Final results were received until 23 November 2015, although clarifications were allowed, for one result, on 11 November 2016. All activities were to be reported as ^{68}Ge massic activity at a comparison reference time of 12h00 UTC 14 November 2014. Decay correction were to be made using the half-life of 270.95(25) d (Bé et al., 2013). Nuclear data from the 2013 evaluation by Bé et al. (2013) were to be used for calculations. All participants were encouraged to send an ampoule containing 3.6 g of solution to the BIPM for measurement in the SIR to create a link to BIPM.RI(II)-K1.Ge-68.

3. Results and Discussion

Participants were encouraged to perform preliminary measurement to check the comparison ampoule for adsorption and to identify any radionuclidic impurities. Results of these measurements by those participants performing them are summarized in **Table 2**. Reported measurements of adsorption ranged from 0.017 % to 0.11 % of the total ampoule activity, with the largest being a measurement of initial rinse solution. The next largest measured adsorption corresponds to 0.03 %. The comparison solution can be considered stable. See footnotes to the table for details of the adsorption measurements. No participants reported any photon-emitting impurities, with the majority of those measurements being by high purity germanium spectroscopy. Additional details for some measurements, where reported, can be found in the footnotes to the table. The comparison solution can be considered free from measurable photon-emitting impurities.

All reported results comprising those reported for inclusion in the Key Comparison Database (KCDB) and reported confirmatory measurements, by primary methods, are listed in **Table 3**. For an explanation of the acronyms used to denote the measurement method see online supplement **Appendix A**. As part of the comparison protocol, participants are instructed to also report a single value that will be taken as their comparison result. The single value results from each lab are shown graphically in **Figure 2**. All individually reported results, by method and including confirmatory measurements, are shown in **Figure 3**. See figure captions for description of symbols. Full uncertainty budgets for all reported values are presented in online supplement **Appendix B**.

All participant single value results based on primary methods were plotted and visually examined for outlying data. The SMU result was initially identified as an outlier on this basis. Next, results were evaluated by the Power Moderated weighted Mean (PMM) methodology (Pommé, 2012) using the spreadsheet tool provided in the online supplement to Pommé (2015). Auto-rejection, using a criterion of 2.5 (CCRI(II), 2013), and auto-selection of alpha-parameter (power of the uncertainties in weighting factors, see Pommé, 2015) setting were used. This evaluation identified the results of two labs, SMU and NIM as outliers. Starting with the most extreme point, outliers were removed from the data set and the evaluation was redone until no extreme data remained. No additional outliers were identified. Finally, the data were subjected to a generalized extreme studentized deviate test, used to identify multiple outliers, in Dataplot (Filliben, 1984). Again, two outliers were identified with a test statistic on the second outlier of 2.5929 versus a critical value of 2.548 for 95 %

confidence. Both laboratories with outlying results were notified, without revealing the magnitude or direction of the difference from the CRV, and given the opportunity to examine their results for numerical errors. NIM responded that no errors were found. SMU submitted a new value with the inclusion of a missing correction factor. This report contains the new value; however, it remains an outlier. It is noted that all individual values included in the outlying data are from liquid-scintillation counting based methods, both CNET and TDCR.

A proposed CRV was calculated of 621.7(11) kBq·g⁻¹ at the reference time, based on 14 values. The reported uncertainty is a combined standard uncertainty. The results from TAEK are not included in the calculation because they are reported from secondary measurement methods. This proposed CRV, calculated by the PMM method, is in agreement with both the arithmetic mean 622.4(12) and the weighted mean 621.6(11).

The degrees of equivalence, D_i , with the proposed CRV and associated expanded ($k = 2$) uncertainties, U_i , for each participant i are shown in **Figure 4** and listed in **Table 4**. **Figure 4** includes approximate relative degrees of equivalence. It can be seen that most results fall within 1 % of the proposed CRV.

PTB and LNE-LNHB report coincidence and anticoincidence results, respectively, with Cherenkov detection in the beta channel. Although one result is reported as a confirmatory measurement, it can be seen in **Figure 3** that both results are in excellent agreement with the proposed CRV. Full details of these measurements are reported in Marganec-Gałązka et al. (2017) and Bobin et al. (2017).

BARC reported using DDEP data from 2003 (Kulkarni et al., 2017). It could be expected that coincidence counting results would increase by approximately 0.3 % due to the change in positron branching ratios from the old DDEP data to that specified for the comparison. Re-evaluating the PMM for this approximate correction would change the proposed CRV from 621.7(11) to 621.8(10), with no additional outliers identified.

Two results were submitted that included the production of solid sources – see triangle symbols in **Figure 3**. Solid sources require care in their production due to the volatility of germanium. IFIN-HH has previously described their approach of precipitation of germanium as a sulphide and monitoring loss by comparison of gamma-ray measurements, using the 1077 keV photons, to a similarly prepared, sealed, liquid sample (Grigorescu et al., 2004). A correction for loss is made based on this comparison. The method was modified for sources produced as part of this comparison by mixing CuSO₄, used as a stabilizer, directly with the comparison solution rather than sequential deposition before addition of saturated H₂S, bubbled in H₂O. For full details see Sahagia et al. (2017). IRA initially observed losses of 1 % to 7 % in the drying of their solid sources, prior to sealing together the two halves of their plastic scintillator – their normal production method. IRA modified their source production method to immediately seal together the top and bottom halves of the plastic scintillator and then heat the sealed source in an oven at 45 °C for a few days to dry the source. In some sources the drop was sandwiched between two aluminum foils. The comparison results indicate success for both the IFIN-HH and IRA source production methods.

Agreement within uncertainties ($k = 2$) by the TDCR method was reported in a separate comparison between LNMRI/IRD, using a modified version of TDCR07c, and LNE-LNHB in da Cruz et al. (2016).

However, LNMRI/IRD does not report a TDCR value in the present comparison. LNE-LNHB reports a TDCR-based value, using the Fortran code gega68. Details of the LNMRI/IRD reported measurements for this comparison are presented in da Silva et al. (2017), additionally TDCR measurements on the comparison solution are reported as a comparison to their anticoincidence counting measurements. By linking the two comparisons through the LNMRI/IRD anticoincidence measurements, it can be seen that LNMRI would expect a reported TDCR result of $627.1(51) \text{ kBq}\cdot\text{g}^{-1}$. This value would again be in agreement with the LNE-LNHB value of $617.7(31) \text{ kBq}\cdot\text{g}^{-1}$, if evaluated with expanded ($k = 2$) uncertainties.

Additional participants have published details of their measurements separately (van Wyngaardt et al. 2017). Finally, some participants also reported values based on secondary methods, either as the comparison value or as confirmatory measurements. Only the IRA ionization chamber value is directly correlated to the participant's standard for ^{68}Ge . The remaining results are based on established calibration curves or Monte Carlo modeling. The agreement with the proposed CRV as seen in **Figure 3** gives evidence that $^{68}\text{Ge} + ^{68}\text{Ga}$ can be successfully measured by these methods.

4. Conclusion

A key comparison of the ^{68}Ge activity concentration of a $^{68}\text{Ge}/^{68}\text{Ga}$ solution has been successfully carried out. Samples were distributed to 18 participants. Results were received from 17 participants. A Comparison Reference Value (CRV) of $621.7(11) \text{ kBq}\cdot\text{g}^{-1}$ is proposed, based on 14 values. Two results are identified as outliers. One result is not from primary methods. Both outlying results are from liquid-scintillation based methods. Results based on solid sources show these sources can be successfully produced, if necessary precautions are taken. Coincidence and anticoincidence results utilizing Cherenkov detection in the beta channels have been utilized in an international comparison for the first time, with success. Degrees of equivalence with the proposed CRV are reported.

Disclaimer

Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

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doi:10.1016/j.apradiso.2015.11.078

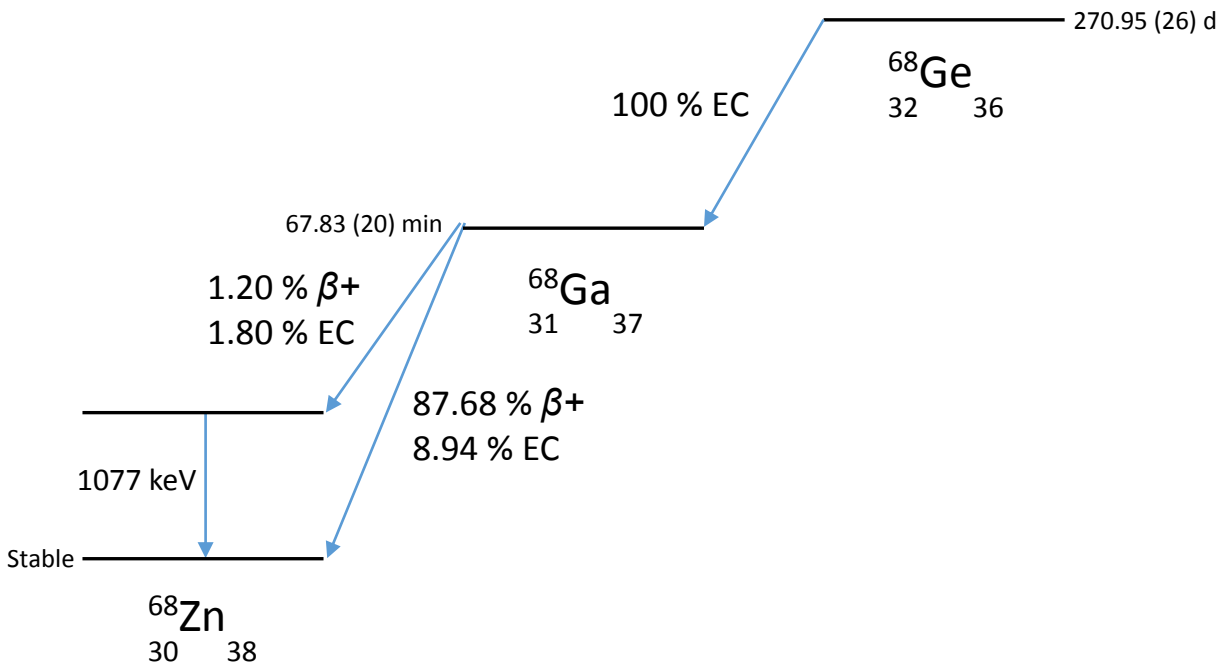


Figure 1 – Simplified decay scheme of $^{68}\text{Ge}/^{68}\text{Ga}$, data taken from Bé et al. (2013).

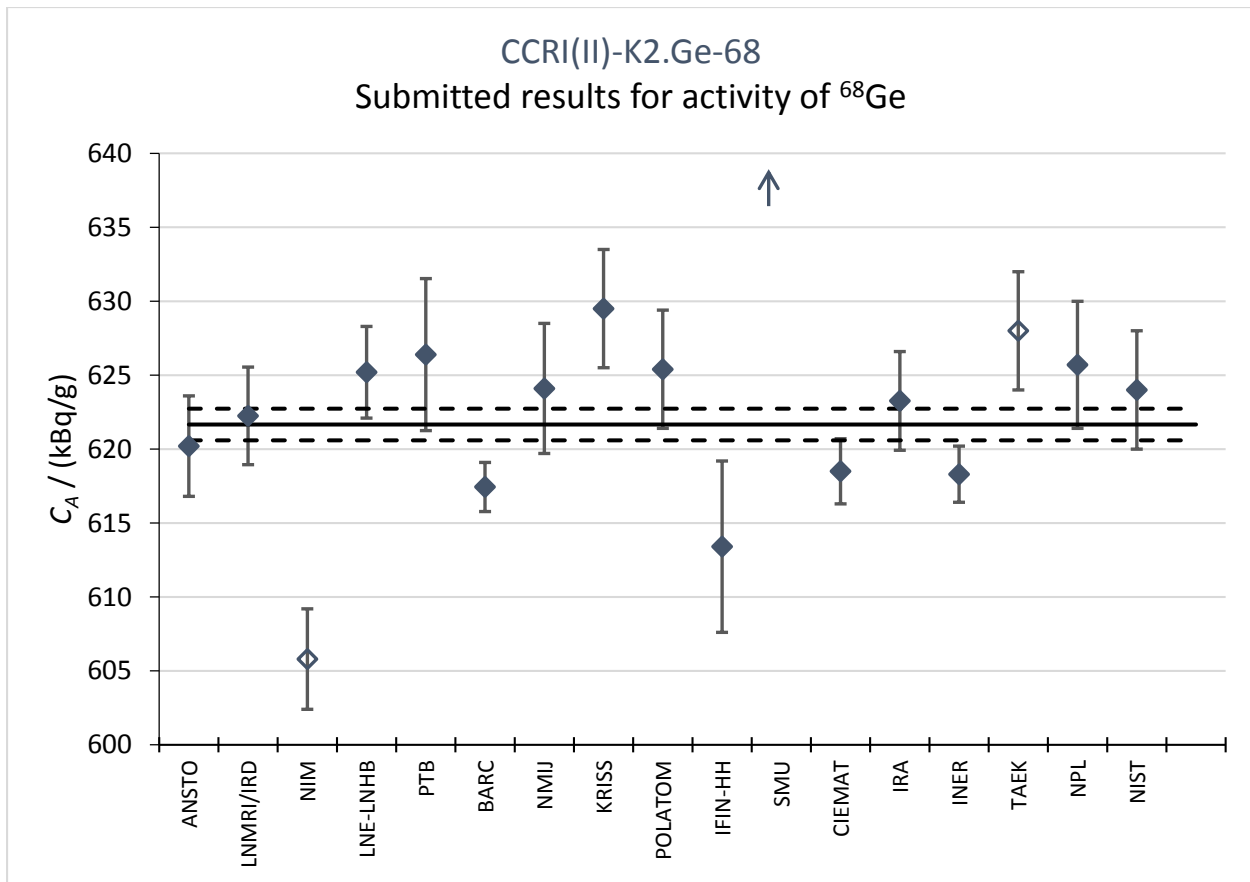


Figure 2. Final results of the international comparison of ^{68}Ge activity concentration of a $^{68}\text{Ge} + ^{68}\text{Ga}$ solution; one value per each designated institute. The uncertainty bars correspond to the combined standard uncertainty on each respondent's value. The solid line represents the proposed Comparison Reference Value (CRV) of $621.7 \text{ kBq}\cdot\text{g}^{-1}$ and the dashed lines represent the combined standard uncertainty of $1.1 \text{ kBq}\cdot\text{g}^{-1}$ on the CRV. Open symbols are not included in the proposed Comparison Reference Value. NIM and SMU (above graph) results are identified as outliers. TAEK results are not from primary methods.

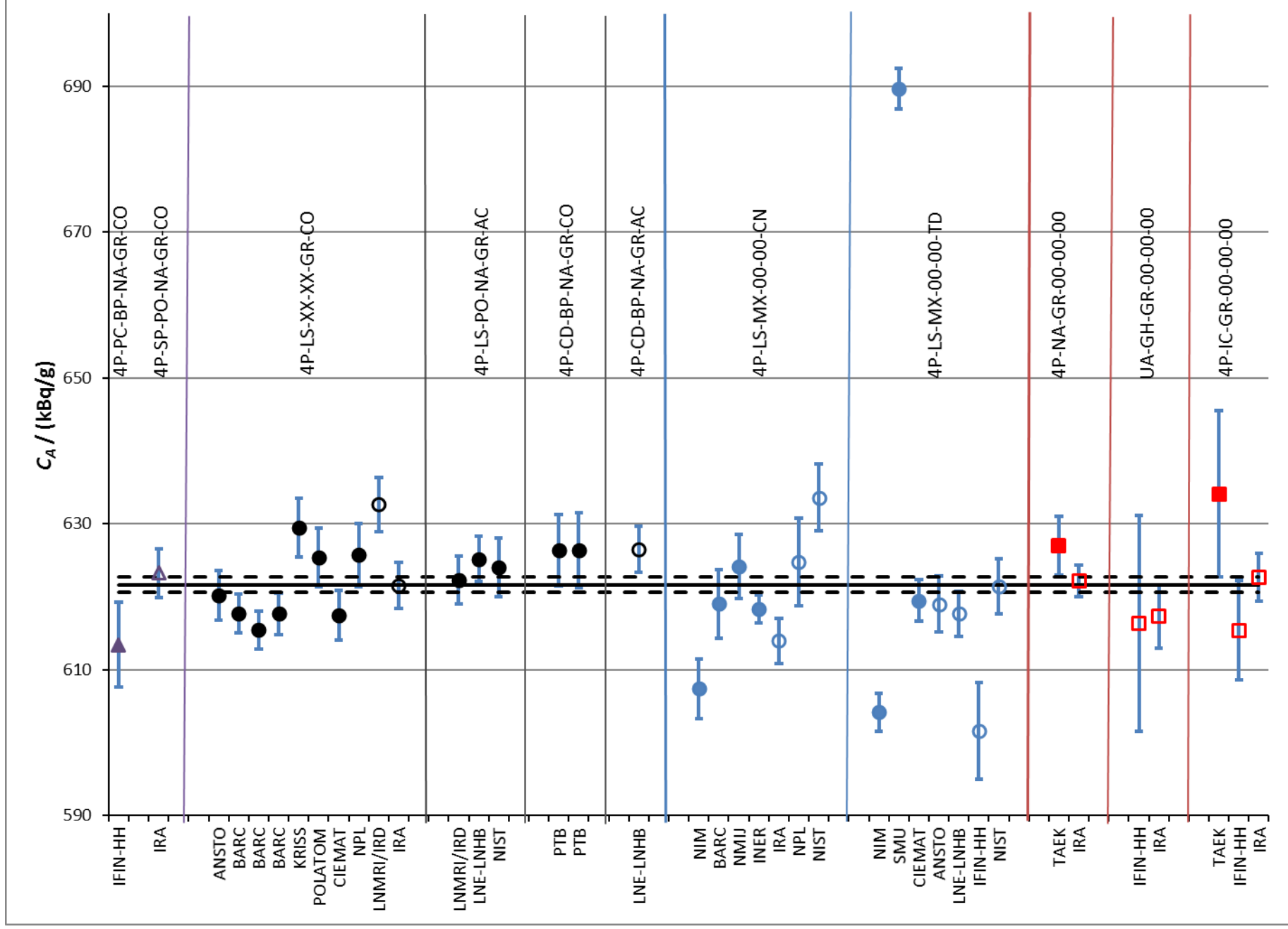


Figure 3. All reported individual results by measurement method (see acronyms in online supplement Appendix 1, XX is used here to combine multiple coincidence counting methods). Error bars are combined standard uncertainties, as reported by participants. Closed symbols represent results that were reported as, or combined in, laboratory single results. Open symbols represent results reported as confirmatory. Triangles are results determined using solid sources. Squares indicate results from secondary measurement methods.

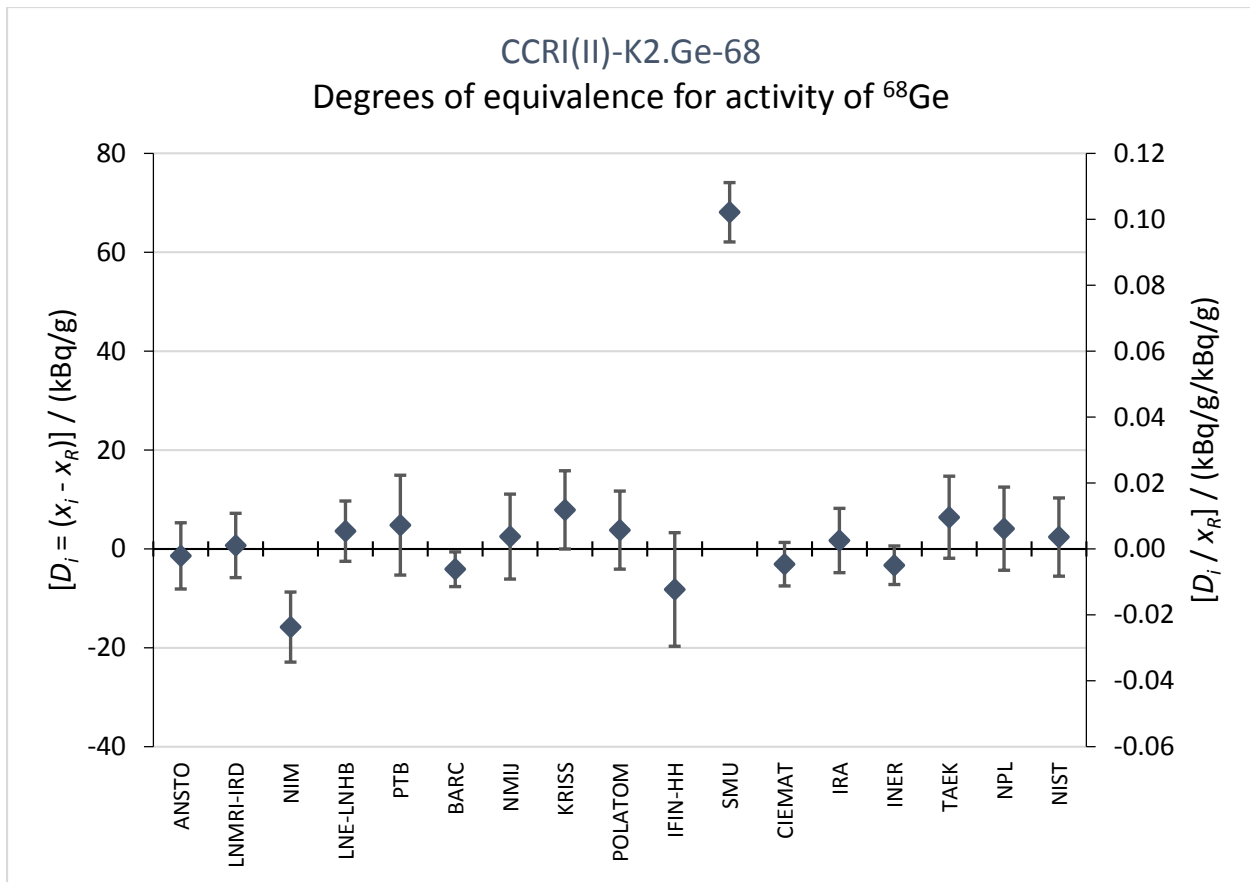


Figure 4. Degrees of Equivalence, D_i , for participants in the CCRI Key Comparison CCRI(II)-K2.Ge-68. The value of D_i is computed as $x_i - x_R$, where x_i is the laboratory reported result and x_R is the proposed Comparison Reference Value of $621.7 \text{ kBq}\cdot\text{g}^{-1}$. The uncertainty bars correspond to the expanded uncertainty ($k = 2$), U_i , on D_i . N.B.: Right-hand axis shows approximate relative values.

Table 1. Laboratories participating in CCRI(II) Key Comparison CCRI(II)-K2.Ge-68 for ^{68}Ge , listed alphabetically by country.

Laboratory Name	Acronym	Country	Regional Metrology Organization
Australian Nuclear Science and Technology Organisation	ANSTO	Australia	Asia-Pacific Metrology Programme (APMP)
Laboratório Nacional de Metrologia das Radiações Ionizantes, Instituto de Radioproteção e Dosimetria	LNMRI-IRD	Brazil	Inter-American Metrology System (SIM)
National Institute of Metrology	NIM	China	APMP, Euro-Asian Cooperation of National Metrological Institutions (COOMET)
Laboratoire national de métrologie et d'essais-Laboratoire national Henri Becquerel	LNE-LNHB	France	European Collaboration in Measurement Standards (EURAMET)
Physikalisch-Technische Bundesanstalt	PTB	Germany	EURAMET, COOMET
Bhabha Atomic Research Centre	BARC	India	APMP
National Metrology Institute of Japan	NMIJ	Japan	APMP
Korea Research Institute of Standards and Science	KRISS	Korea	APMP
National Centre for Nuclear Research Radioisotope Centre POLATOM, Laboratory of Radioactivity Standards	POLATOM	Poland	EURAMET
National Institute of Research and Development for Physics and Engineering "Horia Hulubei"	IFIN-HH	Romania	EURAMET, COOMET
Slovenský Metrologický Ústav	SMU	Slovakia	EURAMET, COOMET
Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	CIEMAT	Spain	EURAMET
Institut Universitaire de Radiophysique Appliquée	IRA	Switzerland	EURAMET

Institute of Nuclear Energy Research	INER	Taiwan	APMP
Turkish Atomic Energy Authority	TAEK	Turkey	EURAMET, COOMET
National Physical Laboratory	NPL	United Kingdom	EURAMET
National Institute of Standards and Technology	NIST	United States of America	SIM