Final Report, 6 January 2022

SUBSEQUENT COMPARISON OF MEASUREMENTS OF NEUTRON SOURCE EMISSION RATE (2012) – CCRI(III)-K9.AmBe.1

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ABSTRACT

Section III (neutron measurements) of the Comité Consultatif des Rayonnements Ionisants, CCRI, conducted a subsequent comparison of primary measurements of the neutron emission rate of an ^{241}Am-Be(α,n) radionuclide source. The aim of this comparison was to compare primary measurements of the neutron emission rate of an ^{241}Am-Be(α,n) radionuclide source for laboratories who were outliers in the previous comparison (K9.AmBe) or who were unable to participate. For various reasons it was only possible for two laboratories to participate – NIM (China) and NPL (UK) – with NPL as the pilot institute and the link to the K9.AmBe comparison. Measurements were made in 2012. Each laboratory reported the neutron emission rate into 4π sr together with a detailed uncertainty budget. Both participants used the manganese bath technique.

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1 INTRODUCTION

It is the aim of Section III to compare the realisation of standards of all relevant neutron quantities over a ten year cycle. Each comparison exercise should take less than four years from initial planning to publication of report in open literature. The previous comparison of measurements of source emission rate started in 1999 and took six years to complete[1]. Difficulties in transporting radionuclide sources between countries and some participants requiring the source for a second measurement were the main factors in the time taken for the previous exercise.

The aim of this comparison is to compare primary measurements of the neutron emission rate of an ^{241}Am-Be(α,n) radionuclide source for laboratories who were outliers in the previous comparison (K9.AmBe) or who were unable to participate. The NPL were the pilot institute and, as participant in the previous key comparison K9.AmBe, provided a link to that exercise.

Five laboratories were scheduled to participate:

China Institute of Atomic Energy (CIAE), Beijing, PR of China

Ente per le Nuove Tecnologie, l'Energia e l'Ambiente -Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti (ENEA-INMRI), Rome, Italy

Laboratoire National Henri Becquerel (LNE-LNHB), Paris, France

National Institute of Metrology (NIM), Beijing, PR of China

National Physical Laboratory (NPL), Teddington, UK

Ultimately only two laboratories participated, the NIM and the NPL. Issues with the special form certification for the source meant it was not possible to send the source to the LNE-LNHB. The time spent attempting to resolve both this and issues relating to new regulations in the UK governing the checking of radioactive air-freight meant that it was not possible to the ENEA-INMRI to participate during the temporary import period while the source was in Europe.

2 THE ^{241}Am-Be(α,n) RADIONUCLIDE SOURCE

The neutron source used was a sealed ^{241}Am-Be(α,n) source (serial number AB3001) owned by the NIM, which had a nominal activity of 142.9 GBq. It is in a cylindrical capsule (outer length 24 mm, outer diameter 24 mm) and was manufactured by HTA Co. Ltd., China.

The source was chosen as it is a similar type as that used in the K9.AmBe comparison. It has a long half-life and stable decay process, and also because it is representative of the type and size of neutron sources commonly used at the present time in calibration laboratories. After the NIM had made the first measurement the source was sent to the NPL in 2012. In 2014 the source was returned from the NPL to the NIM.

3 NEUTRON EMISSION RATE MEASUREMENT TECHNIQUES OF THE LABORATORIES

3.1 NIM

The NIM manganese bath is a sphere 110 cm in diameter and contained solution with a hydrogen to manganese number density ratio (N_H/N_{Mn}) of 71.203 for the comparison measurements. N_H/N_{Mn} was determined gravimetrically (i.e. comparing the mass of a sample of the solution with the mass of the residue after evaporation) and by measuring the density of the solution.

The neutron source was placed in the centre of the bath in a tube with an inner diameter of 32 mm made of 1 mm thick stainless steel. The solution was circulated continuously between the bath and a shielded sample vessel with two ϕ40 mm $\times$ 40 mm NaI(Tl) scintillation detectors mounted one at either side. Six measurements in total were made of the solution activity with each NaI detector.

The efficiency of the NIM manganese bath system was determined using ^{56}Mn produced by activation in a reactor. It was then dissolved into a solution and added to the bath. The specific activity of the ^{56}Mn solution was determined by $4\pi\beta$-γ coincidence counting.

Corrections were made for neutron leakage, fast neutron losses due to interactions in the oxygen and sulphur, and thermal neutrons absorbed by the neutron source and the container using MCNP with ENDF/B-VII cross-sections. Thermal neutron capture by hydrogen and sulphur was calculated using thermal cross sections with appropriate Westcott parameters to allow for epithermal resonance capture.

3.2 NPL

The NPL manganese bath is a stainless steel sphere 98 cm in diameter. A solution with a hydrogen to manganese number density ratio (N_H/N_{Mn}) of 34.28 was used, the concentration of which was determined gravimetrically.

The solution was continuously circulated through a shielded reservoir where two ϕ50 mm $\times$ 50 mm NaI scintillators were used to measure the activity of the solution, before being pumped back into the bath. The saturated count rate was obtained from the counting cycles when the source was in the bath as well as from those after the source had been removed. Both NPL measurements consisted of two separate bath irradiations performed within a week of each other.

The NaI detectors were calibrated by adding an active solution of ^{56}Mn to the bath, the activity concentration of which had been determined by ionisation chamber measurements at the NPL. The active solution was produced by irradiating a bottle of $MnSO_4$ solution extracted from the bath in the NPL thermal pile. This is repeated approximately every 3 months with a periodic confirmation of the ionisation chamber calibration by absolute $4\pi\beta$-γ coincidence counting. A linear fit is made to the efficiency measurements to interpolate or extrapolate to the day of a neutron source measurement.

MCNP5 was used to calculate the leakage fraction, the capture in the source and source mounting assembly, and the capture by (n,p) and (n,α) reactions in sulphur and oxygen using ENDF/B-VI cross-sections where available. Thermal neutron capture by hydrogen, sulphur, and solution impurities was calculated using thermal cross sections with appropriate Westcott parameters to allow for epithermal resonance capture. A hydrogen to manganese cross-section ratio derived from measurements in the NPL manganese bath was used. The impurity levels were taken from a chemical analysis of the solution.

Table 1: Summary of manganese bath parameters

Laboratory	Bath size	N_H/N_{Mn} at time of measurement	Correction factor method	Oxygen cross-section	Impurities considered?	Activity counting system
NIM	110 cm diameter	71.203	MCNP+thermal calculation	ENDF/B-VII	No	Circulation of solution
NPL	98 cm diameter	34.28	MCNP+thermal calculation	ENDF/B-VI	Yes	Circulation of solution

4 MEASUREMENTS

4.1 Emission rates as reported by participants

The emission rates submitted by each participant with uncertainties at $k = 2$ are given in Table 2. All have been corrected to the reference date of 1st January 2012.

Table 2: Emission rates corrected to 1st January 2012 with uncertainties at $k = 2$

Laboratory	Measurement date	Emission rate ($\times 10^6$ s^{-1})	Uncertainty [$k = 2$] ($\times 10^6$ s^{-1})
NIM	12 April 2012	8.167	0.127
NPL	11-15 October 2012	8.145	0.102

The results are plotted in Figure 1 with uncertainties at the $k = 2$ level.

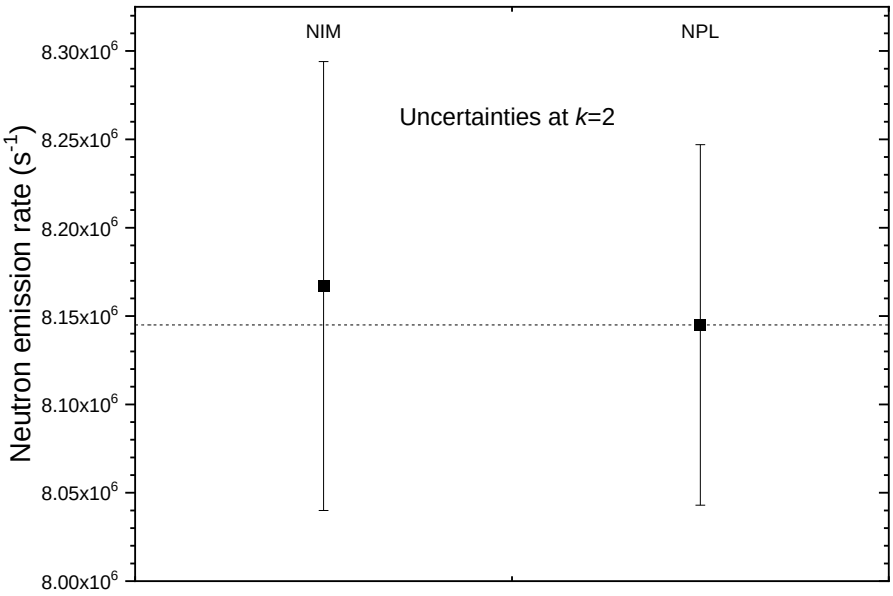

Figure 1: Graph of emission rates with uncertainties at $k = 2$

4.2 Degree of equivalence between participants

According to the CIPM MRA rules for measurement comparisons[23], no key comparison reference value is computed for these new results and the previous key comparison value is kept. The previous NPL results in the original comparison remain valid and the new results are used to link

the NIM results to the original key comparison and to compute their degrees of equivalence with respect to the previous key comparison reference value.

The degree of equivalence between the two participating laboratories was calculated as the difference between the values of emission rate (Q) reported by the participants

$$D_{NPL,NIM} = Q_{NPL} - Q_{NIM} \quad (1)$$

with the expanded uncertainty as follows

$$U(D_{NPL,NIM}) = 2\sqrt{u^2(Q_{NPL}) + u^2(Q_{NIM})} \quad (2)$$

For the above formula, the correlation between results reported by NPL and NIM are considered not significant. The normalised error (En) in the degree of equivalence was then calculated as follows

$$En = \frac{|D_{NPL,NIM}|}{U(D_{NPL,NIM})} \quad (3)$$

A value of En in the range 0 to 1 indicates that the difference between the measured values of the participating laboratories is less than or equal to the combined expanded uncertainties of the two laboratories.

The degree of equivalence with the associated uncertainty is given in Table 3.

Table 3: Degree of equivalence between NPL and NIM

$D_{NPL,NIM}$	$U(D_{NPL,NIM})$	En
2.205×10^4 s^{-1}	1.633×10^5 s^{-1}	0.135

The results presented in Figure 1 and Table 3 indicate that the measurements by the NIM and the NPL are in excellent agreement.

4.3 Using same oxygen (n,α) cross-section evaluation

It is well known that the choice of O(n,α) cross-section has a significant influence on the neutron emission rate obtained from the manganese bath technique, the difference being around 0.8 % between ENDF/B-VI.0 and ENDF/B-VII.0[3,4]. NIM used the ENDF/B-VII.0 evaluation, whereas NPL used the ENDF/B-VI.0 evaluation, so to quantify the influence of the O(n,α) cross-section on the level of agreement the NPL results were recalculated using correction factors based on ENDF/B-VII.0 evaluations. The NPL values obtained using both cross-section evaluations are plotted alongside the NIM value in Figure 2.

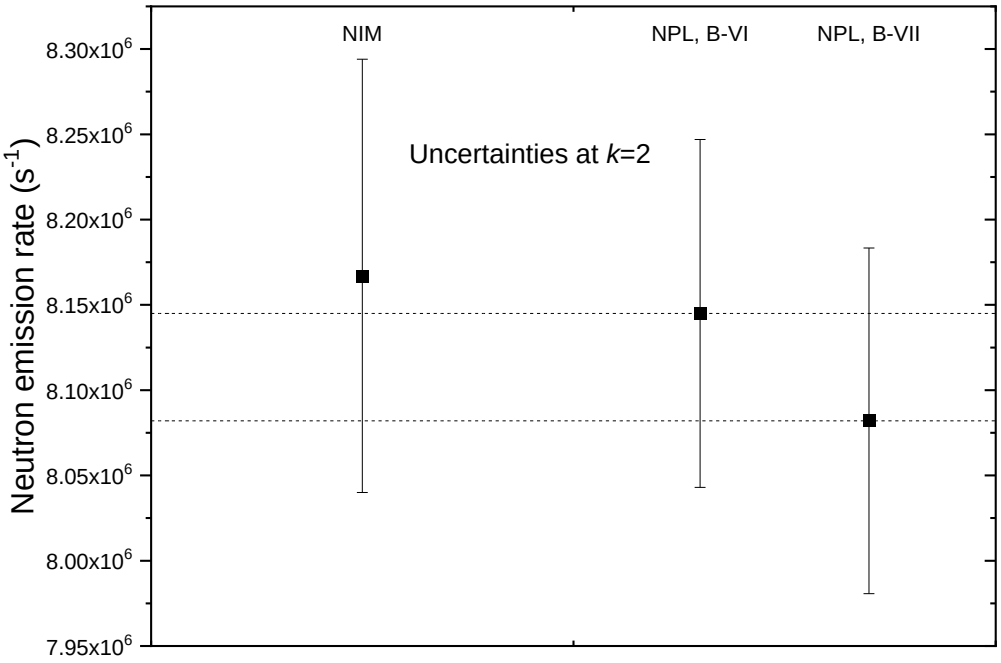

Figure 2: Graph of emission rates, including the NPL rate using correction factors based on ENDF/B-VII cross-sections, with uncertainties at k = 2

The degree of equivalence when both laboratories use the same oxygen cross-section data is given in Table 4.

Table 4: Degree of equivalence between NPL and NIM when both use the same oxygen cross section data

$D_{NPL,NIM}$	$U(D_{NPL,NIM})$	En
8.477×10^4 s^{-1}	1.633×10^5 s^{-1}	0.519

Although the degree of equivalence has increased by a factor of 3.8 the normalised error shows that the difference between the measured values of the participating laboratories is still much less than the combined expanded uncertainties of the two laboratories. The increase in the degree of equivalence of 6.272×10^4 s^{-1} has the same magnitude as the reduction in the NPL measured rate when the ENDF/B-VII evaluation is used, which corresponds to a 0.77 % reduction in the source emission rate. In keeping with the approach taken for the original CCRI(III).K9.AmBe evaluation, the degrees of equivalence for this exercise are based on the reported values of the participants regardless of which cross-sections were used in their analysis.

4.4 Long term stability of the NPL Mn bath

NPL routinely measures the emission rates of its neutron sources both to confirm the decay is as expected and to confirm the long-term stability of the manganese bath system. The decay-corrected emission rates of 5 Am-based sources are plotted in Figure 3 for the period from 1997 to 2017, covering the K9.AmBe and K9.AmBe.2 exercises. Source number 1096 was the Am-Be source used in the K9.AmBe comparison.

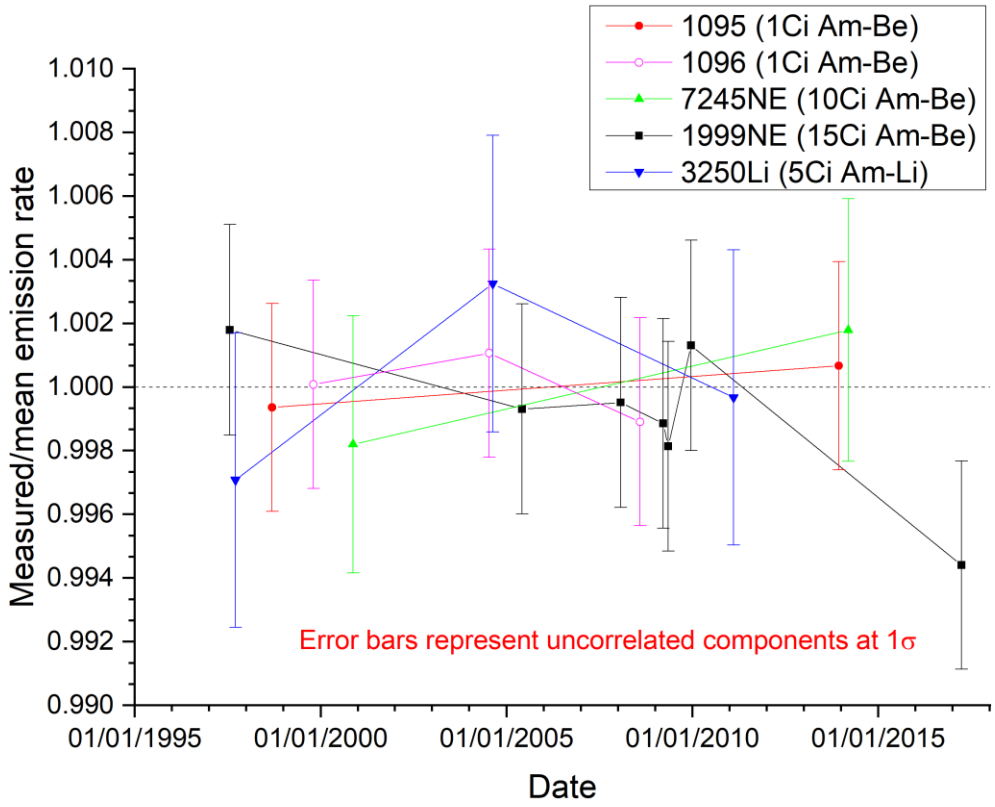

Figure 3: Normalised decay-corrected measurements of Am-based neutron source emission rates at NPL over the period between the K9.AmBe and K9.AmBe.1 comparison exercises

5 LINK WITH CCRI(III)-K9.AMBE

The new result obtained by NIM can be linked to the CCRI(III)-K9.AmBe reference value via the NPL results. The ratio of the participant's value in this comparison to that of NPL in this comparison can be multiplied by the NPL value from the original CCRI(III)-K9.AmBe comparison to yield the linked value for the participant ($Q_{participant,K9.AmBe}$) as shown in equation 4.

$$Q_{participant,K9.AmBe} = Q_{NPL,K9.AmBe} \frac{Q_{participant,K9.AmBe.2}}{Q_{NPL,K9.AmBe.2}} \qquad (4)$$

The linked value can then be used to derive the unilateral degrees of equivalence and associated uncertainties linked to the CCRI(III)-K9.AmBe KCRV. This is given in Table 5 and plotted alongside the DoEs for the other participants in the CCRI(III)-K9.AmBe exercise in Figure 4.

Table 5: Unilateral degrees of equivalence linked to the CCRI(III)-K9.AmBe KCRV

Participant	$D_{participant}$	$U(D_{participant})$
NIM	-0.0061×10^6 s^{-1}	0.0587×10^6 s^{-1}

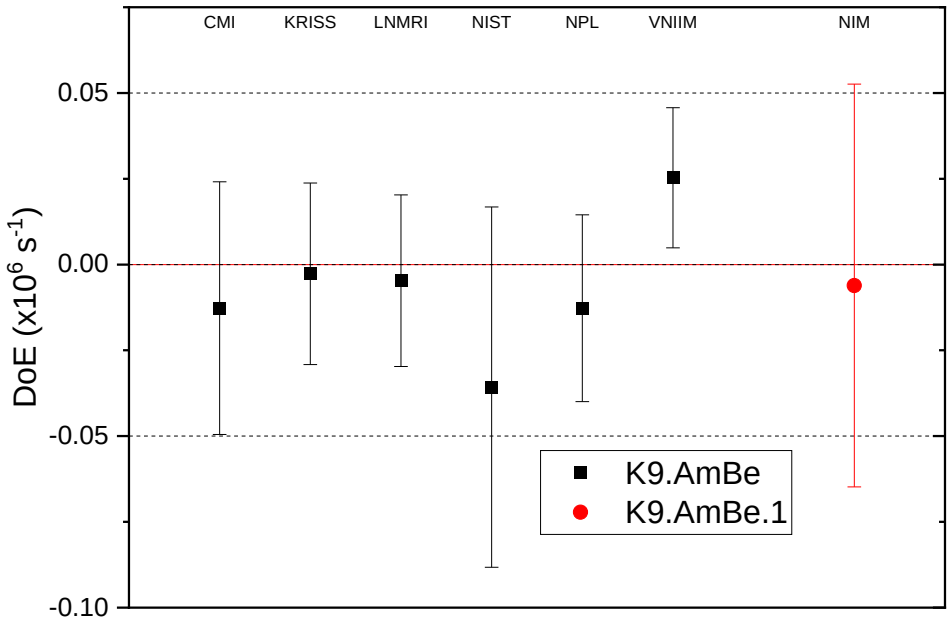

Figure 4: Degrees of Equivalence for the original K9.AmBe comparison and those linked to it through the subsequent K9.AmBe.2 comparison, with expanded (k = 2) uncertainties. All participants in the K9.AmBe comparison used the Mn bath technique, although the VNIIM value is a mean of the Mn bath and associated particle techniques. The correction factors for all K9.AmBe participants were based on ENDF/B-VI cross-sections, except for NIST and LNMRI whose were based on ENDF/B-V.

6 SUMMARY AND CONCLUSION

The subsequent comparison of neutron emission rate measurements, CCRI(III)-K9.AmBe.1, has concluded with only NIM (China) and NPL (UK) able to participate. Both participants used the manganese bath technique to measure the neutron emission into 4π sr of an Am-Be source. The reported values of the participants are consistent within the uncertainties, with the normalised error on the degree of equivalence equal to 0.135.

When linked to the KCRV of the original CCRI(III)-K9.AmBe comparison, the unilateral degree of equivalence and expanded uncertainty for NIM is $(-0.0061 \pm 0.0587) \times 10^6$ s^{-1}.

7 ACKNOWLEDGEMENTS

The authors would like to thank Dr José Maria Los Arcos for collating and checking the participants' reports before the evaluation.

The evaluation of the intercomparison was funded by the UK National Measurement System Policy Unit and was Deliverable (v) of Project N1 in its 2012 to 2015 Programme for Ionising Radiation Monitoring Metrology.

8 APPENDIX

8.1 Uncertainty Budgets

Table 6: Full uncertainty budget for NIM measurement

Source	Uncertainty / %	c_i	Probability Distribution	Component of Uncertainty / %	Type of uncertainties
Detection efficiency	0.64	1	Normal	0.64	B
Counting statistical	0.11	1	Normal	0.11	A
Background correction	0.31	0.0164	Normal	0.01	A
Leakage	7.75	0.0087	Rectangular	0.04	B
Oxygen and sulphur capture	20.12	0.0208	Rectangular	0.24	B
Source and cavity capture	5.09	0.0208	Rectangular	0.06	B
Fraction of thermal neutrons captured by ^{55}Mn	0.35	1	Normal	0.35	B
Combined Standard Uncertainty / %				0.78	
Expanded Uncertainty (k = 2) / %				1.56	

Table 7: Full uncertainty budget for NPL measurement

Source	Uncertainty (%)	Distribution	Sensitivity	Uncertainty component (%)	Degrees of freedom
Counting	0.01	Normal	1	0.01	∞
Cross-section ratio	0.2	Normal	1	0.2	∞
Efficiency	0.4	Normal	1	0.4	∞
Oxygen & sulphur losses	20	Rectangular	0.03530	0.4	∞
Cavity and source capture	5	Rectangular	0.01523	0.04	∞
Leakage	4	Rectangular	0.01420	0.03	∞
Timing	0.05	Rectangular	1	0.03	∞
Mixing	0.2	Rectangular	1	0.12	∞
Solution concentration	0.1	Normal	1	0.1	9
Background	10	Normal	0.001	0.01	9
Dead time	5	Normal	0.01	0.05	9
Half life of source	0.15	Rectangular	0.0012	0.0001	∞
Combined standard uncertainty				0.63	13235
Expanded uncertainty				1.26	

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