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The NPL contribution to a CCRI international comparison of measurements of neutron source emission rate

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ABSTRACT

This report contains details of the experimental procedures used by NPL to measure the neutron emission rate of the ^{241}Am-Be source used in the CCRI international comparison exercise CCRI(III)-K9.AmBe. Measurements were made using the NPL manganese bath facility in 1999 and again in 2004. A description is also given of the analysis technique and the various corrections applied. The final values for the neutron emission rate into 4π sr of the source corrected to 1 January 2000 are given together with a detailed breakdown of the uncertainties.

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1   INTRODUCTION

This intercomparison exercise was organised by Section III of the Comité Consultatif pour les Rayonnements Ionisants (CCRI). It is the aim of Section III to compare the realisation of standards of all relevant neutron quantities over a ten year cycle. The aim of this particular comparison is to compare primary measurements of the neutron emission rate of an ^{241}Am-Be(α,n) radionuclide source.

The exercise started in 1999 with the first measurement at NPL who provided the source (serial number AMN1000/1096, 37 GBq ^{241}Am-Be in an X3 capsule). Since then it has been to VNIIM (Russian Federation), CMI (Czech Republic), KRISS (Republic of Korea), CIAE (China), IRD (Brazil), and NIST (USA) before returning to NPL in 2004 for a repeat measurement. The purpose of the repeat measurement was to confirm that the source characteristics have not changed during the exercise. It was originally intended for the NPL repeat measurement to be the concluding measurement however, due to scheduling difficulties the repeat measurement was performed in July 2004 and the exercise continued with measurements by LNHB (France) in the Autumn of 2004. Other NMIs may also receive the source for a second time due to problems with the data from their first measurements.

2   THE NPL MANGANESE BATH

2.1   System overview

The NPL manganese bath (shown in Figure 1) is used to measure the emission rates of radionuclide neutron sources for NPL and fee paying customers. The manganese bath is filled with manganese sulphate solution, $MnSO_4$. The system consists of the bath, the pumping system and two counting reservoirs, each with a pair of NaI scintillator detectors. The manganese bath is spherical in shape with a diameter of 98 cm. It is connected, via plastic tubing, to the two reservoirs which are located in an adjacent room and are shielded from photon background and any scattered neutrons by lead and wax blocks. The detectors are located in recesses in either side of the reservoir where they then detect the decay of the active ^{56}Mn. The reservoirs are designed to enable the measurement of sources with different emission rates. The main reservoir used to calibrate the majority of sources that come to NPL, including the one used in the present exercise, has the larger volume of the two, effectively making it more efficient at detecting the decay of ^{56}Mn. The second reservoir has a smaller volume and is used only when the emission rate of a source is sufficiently high that using the larger reservoir would incur problems due to excessive dead-time. Because of the smaller volume, and hence lower detection efficiency, the aforementioned dead-time problems are avoided.

During the entire process of a source measurement, the manganese sulphate solution is continuously pumped around the system whilst an individual stirring pump separately stirs the solution within the bath. This enables the growth of the activity to be measured in addition to the decay. When measuring a source, it is first placed in one of two stainless steel cavities depending on the physical size of the source itself. The cavity is evacuated using a vacuum pump that pumps for the whole time the cavity is immersed in the bath. The cavities serve two purposes, the first being to protect the source from the $MnSO_4$ solution (and vice-versa). The second is to avoid a high thermal neutron flux around the source, which would increase the chances of neutrons being captured by the source. The presence of the vacuum helps in this regard. A long stainless steel rod connects to the cavity (see Figure 1) and

enables the automatic source transfer system to take the source from the handling cell and place it at the exact centre of the bath. Neutron leakage from the bath is monitored by a De Pangher long counter at a fixed geometry on the surface of the bath. However, since the introduction of MCNP[1] based correction factors the long counter is no longer used to derive the leakage correction factor.

Figure 1: The NPL manganese bath facility

2.2   Absolute calibration of the efficiency

To provide traceability of the manganese bath to an absolute standard, the counting efficiency is determined using a standardised solution of active $MnSO_4$. A sample of solid ^{55}Mn is irradiated at the CONSORT research reactor at Ascot[†] in order to produce active ^{56}Mn. It is then made up into $MnSO_4$ solution at NPL with samples being taken for ion chamber counting or, once a year, for $4\pi\beta$-γ standardisation. The rest of the solution is added to the manganese bath and stirred for at least 5 minutes to achieve homogeneity before being pumped through the counting reservoirs. Further time is allowed once pumping begins for the active solution to become homogenously distributed around the system. The decay of the solution is measured using all four NaI detectors, with correction factors applied during the analysis to allow for the different system volumes visible to each pair of detectors. The efficiency of the detectors can therefore be obtained in terms of counts per decay of ^{56}Mn anywhere in the system.

Efficiency measurements are always performed within 6 weeks either side of a neutron source emission rate measurement. These are plotted against time and linear fits are made to enable the efficiency to be extrapolated or interpolated to the time of the measurement.

2.3   Pre-measurement checks

Before each measurement commences checks are performed to test the condition of the electronics and detectors, which can alter over a long period of time. Two ^{137}Cs check sources are inserted into fixed positions near the NaI crystals, and an ^{241}Am-Be check source is placed in the De Pangher long counter. The NaI operating voltages are then altered by +100V, -100V and then reset to the original voltage, with three counting cycles being recorded for each voltage setting. Hence the operating voltage plateau is checked to see if any

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[†] Reactor Centre, Imperial College at Silwood Park, Buckhurst Road, Ascot, Berkshire, SL5 7TE, UK

change in the electronics has occurred. A Fortran program, CSCHECK, is used to compute the slope of the plateau and also to compare the count rate of the sources corrected back to a reference time with the original count rates. Plots of the decay corrected count rates for each NaI detector are made to check their stability.

2.4   Dead-time

Prior to 2002, the dead-time in each NaI channel was determined before each measurement using the two-source method and these values were averaged to give the values used in the program MNBATH6 which calculates saturation count rates for each cycle from the uncorrected NaI counts . The dead-time values used in the 1999 measurement therefore come from this method.

Since then the decaying source method described by Knoll[2] has become the preferred method at NPL. The count data from the calibration of the bath with active $MnSO_4$ solution can be used as the dead-time correction can be quite high (~5%). Values of the dead-time from measurements made during the preceding year are averaged to give the value used by MNBATH6. The 2004 measurement uses dead-times from this method. The decaying source method gave dead-times which were approximately 6% lower than those from the two-source method. The effect on the final result is therefore small as the dead-time correction at saturation is around only 0.2%.

2.5   Solution concentration

Concentration measurements of the $MnSO_4$ aqueous solution are carried out routinely throughout the year. A gravimetric method is used where two platinum crucibles are filled with approximately 25 ml of solution and evaporated to complete dryness in a furnace. The furnace temperature is increased over a period of weeks from $90^{\circ}C$ to $300^{\circ}C$. Once at $300^{\circ}C$, the containers are weighed every few days until there is no decrease in the weights of the residues. The weights of the solution and of the residue are used to calculate the hydrogen to manganese number density ratio (N_H/N_{Mn}). The ratios are plotted against time and a linear fit is made to enable the solution concentration to be extrapolated forward to the time of a measurement. A correction is made to allow for the contribution of impurities to the weight of the $MnSO_4$ residue. The extrapolated value of N_H/N_{Mn} is divided by 0.999769[3] to obtain the impurity-corrected value.

2.6   Solution impurities

Chemical analysis of the manganese sulphate solution is performed periodically to monitor the levels of neutron absorbing impurities such as boron, cadmium, and lithium. The most recent analysis was performed by the Laboratory of the Government Chemist (LGC), Teddington, UK in May 2003[4]. A semi-quantitative ICP-MS (Inductively Coupled Plasma Mass Spectrometry) scan showed that boron, cadmium, and lithium were below the reporting limit of 0.15 µg/g. A further quantitative scan by ICP-OES (Inductively Coupled Plasma Optical Emission Spectrometry) verified that levels of boron were below the reporting limit of 1.2 µg/g. Other neutron absorbing impurities were not present in significant quantities in either scan, and no identifiable change in the impurity levels with time was found. For the purposes of calculating the fraction of neutrons absorbed by the impurities, the analysis results given in reference 3 have been used.

2.7 Flow rate

The flow rate of the manganese bath system is continually monitored using a float gauge. However, the float gauge cannot be used to determine the exact flow rate as it is calibrated for water, and therefore doesn't take account of the different density and viscosity of the MnSO$_4$ solution. To measure the flow rate accurately, a method has been devised which looks at the arrival of the active solution at the NaI detectors at the start of a measurement of a high output neutron source. Different values of the flow rate are used in the analysis and the one that produces the lowest standard error in the mean saturated count rate is taken to be the correct value.

2.8 Stability of the system

The long-term stability of the manganese bath measurement system is checked by annual calibrations of the national standard ^{226}Ra-Be photo-neutron source (approx. 22.2 GBq, identifier NASTRABE). A ^{226}Ra-Be source was chosen because of the long half-life of ^{226}Ra (1600 years). The calibrations of the ^{226}Ra-Be source are carried out and analysed in the same way as any other source measurement. The emission rate results are plotted in Figure 2 corrected for decay back to 1st January 1990 for easier comparison. It can be seen that the variations are well within the measurement uncertainties.

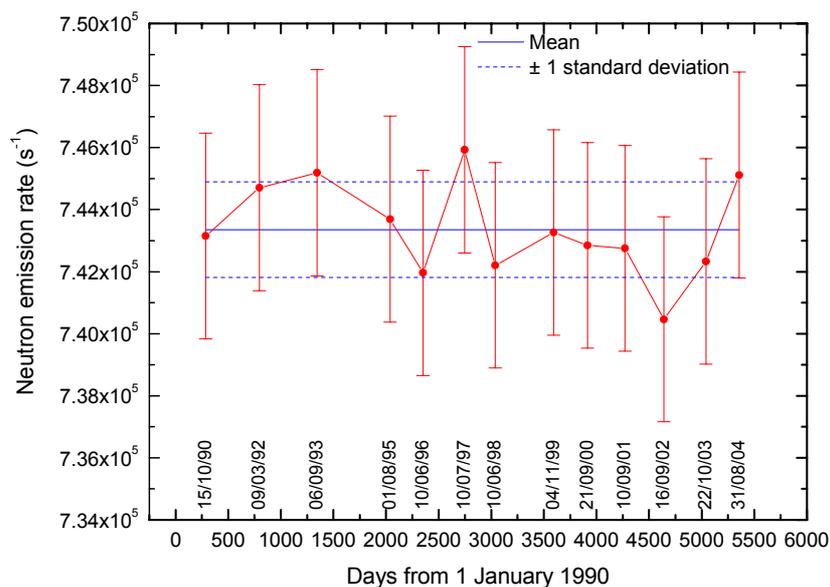**Figure 2: Annual measurements of the national standard ^{226}Ra-Be source, decay corrected to 1 January 1990 (error bars correspond to only the uncorrelated uncertainty components at 1σ)**

2.9 Backgrounds

Backgrounds are continually recorded whenever a measurement is not being performed. The mean of the preceding 10 count cycles (usually of 10000s duration) is taken before a measurement.

3 NEUTRON EMISSION RATE MEASUREMENTS

3.1 Data acquisition

The basic method for the 1999 and 2004 measurements was the same. The only differences are in the number of growth and decay cycles logged for each run.

The source was mounted in a stainless steel cup designed for X3 source capsules at the centre of the thin walled stainless steel cavity of diameter 88 mm. After evacuating the sphere, the source was placed at the centre of the bath using the automatic mechanical transfer system. The entry of the source into the bath was viewed using a CCTV system and a stopwatch was started as the sphere enters the solution. The stopwatch was stopped at a fixed time by a radio controlled clock previously checked against the UK telephone based time service. This enabled the exact time that the source entered the bath to be calculated.

The source was then left in the bath for about 24 hours to allow the activity of the manganese solution to reach saturation (see Table 1 for details of the number of counting cycles). The counts in the NaI detectors were recorded in 1000s count periods, separated by a 20s wait period. The scalers used at NPL require a wait period between count cycles, and 20 seconds is chosen because with a 1000s count period it gives a combined cycle time of exactly 17 minutes. When removing the source from the manganese bath, a stopwatch was again started as the sphere left the solution and stopped at a fixed time by the radio clock to calculate the true 'source out' time. Counting continued while the activity decayed until the count per cycle became too low. The solution was then allowed to decay back down to background levels for a further period of at least 24 hours. A second measurement run was then begun with the pre-measurement checks.

Table 1: Number of growth and decay cycles for both runs and both measurements

Measurement date	Run 1		Run 2	
	Growth	Decay	Growth	Decay
1999	60	19	83	19
2004	84	58	95	67

3.2 Data analysis

The NaI readings were processed using the Fortran program MNBATH6. The program corrects for background, source decay, solution circulation, and dead-time effects, and calculates a value for the saturated count rate at the reference time from each growth and decay cycle count. The equations used to calculate the saturated counting rates for growth and decay were derived by Axton *et al*[5]. The final outputs of the program are eight values of the saturated count rate (2 measurement runs × 2 NaI detector channels × growth & decay) together with uncertainties.

The main equation used to determine the emission rate of the source (Q) is given in a paper by Axton and Bardell[6] and is as follows:

$$Q = \frac{A}{\varepsilon f(1 - L - S - O)} \qquad (1)$$

where A is the saturation count rate of the ^{56}Mn produced in the bath,

ε is the counting efficiency, defined as the counting rate per unit ^{56}Mn disintegration rate throughout the system,

L is the fraction of neutrons which escape from the boundaries of the bath,

S is the fraction of neutrons which are captured by the source and its mounting assembly,

O is the fraction of neutrons lost due to capture in fast neutron reactions in oxygen and sulphur,

f is the fraction of the remaining neutrons which are captured by the manganese.

A second Fortran program, SOURCEIMP, was used to calculate the neutron emission rate of the source by applying equation 1. The program calculates the value of f using the convention of Westcott et al[7] in the following equation taken from reference 6:

$$\frac{1}{f} = 1 + \frac{N_{Im}}{N_{Mn}} \frac{\sigma_{Im}}{\sigma_{Mn}} \frac{(1+\bar{r}s)_{Im}}{(1+G\bar{r}s)_{Mn}} + \left[1 + \frac{N_{Im}}{N_{Mn}}\right]\left[\frac{\sigma_S}{\sigma_{Mn}} \frac{(1+\bar{r}s)_S}{(1+G\bar{r}s)_{Mn}}\right] + \frac{N_H}{N_{Mn}} \frac{\sigma_H}{\sigma_{Mn}} \frac{1}{(1+G\bar{r}s)_{Mn}} \quad (2)$$

where s is the Westcott parameter representing the resonance activation integral normalized to the thermal neutron capture cross-section,

$\bar{r}$ is the Westcott epithermal-flux parameter averaged over the system,

G is a resonance self-shielding factor,

N_H and N_{Mn} are the number of hydrogen and manganese nuclei per unit volume of solution,

N_{Im} is the effective number of impurity nuclei per unit volume of solution,

σ_{Mn}, σ_{Im}, σ_S and σ_H are the capture cross-sections for manganese, impurities, sulphur and hydrogen respectively.

The values of the constants used in equation 2 are given in Table 2.

Table 2: Cross-sections, Westcott parameters, and impurity ratio used in equation 2

	Value	Reference
σ_{Im}/σ_{Mn}	3.297 ± 0.406	8
σ_{Mn}	(13.408 ± 0.037) barn	9
σ_S	(0.5350 ± 0.0055) barn	9
σ_H/σ_{Mn}	(0.024799 ± 0.00005)	9
$(1+G\bar{r}s)_{Mn}$	$1.0159 \pm 0.0007^*$	5
$(1+\bar{r}s)_S{}^\dagger$	$1.000852 \pm 0.000176^\ddagger$	8
$(1+\bar{r}s)_{Im}$	$1.007063 \pm 0.00039^\ddagger$	3
N_{Im}/N_{Mn}	$(3.1 \pm 0.03^\ddagger) \times 10^{-4}$	3

[*] The uncertainty in $(1+G\bar{r}s)_{Mn}$ has been estimated from difference in values of $(G\bar{r}s)_{Mn}$ between references 5 and 10.

[†] A value for s_S of 0.025 has been used which is given in reference 8. According to the reference it has been calculated from the resonance parameters for sulphur given by Mughabghab et al[11]. The individual resonance parameters for ^{32}S were summed and then

multiplied by $\sqrt{\dfrac{4}{\pi}\dfrac{T}{T_0}}$ (where T is the neutron temperature and T_0 is 293.6 K) to give the reduced resonance integral. Dividing by the thermal cross section for ^{32}S then gives s_S. Note that the value for the reduced resonance integral derived in this way is inconsistent with the value for the total resonance integral given by Mughabghab in reference 11. However, a more recent evaluation of the resonance integral has been made[12], and using this value (246 ± 10 mb) gives a value for s_S broadly in line with the figure of 0.025 given in reference 8. The uncertainties in s_S are essentially negligible as a component in the final uncertainty.

‡ The uncertainties in $\bar{r}$, s_S, $s_{Im,}$ and N_{Im}/N_{Mn} have come from reference 8.

The three correction factors $(1-O)$, $(1-S)$, and $(1-L)$ are input to the program and these were calculated using the new MCNP based system[13] with the ISO ^{241}Am-Be energy spectrum[14].

4   ANALYSIS AND RESULTS

4.1   1999 measurement

Table 3: Saturated count rates from MNBATH6 (referenced to 12 noon GMT on 21.10.99):

Measurement	Channel 1		Channel 2	
date	Growth	Decay	Growth	Decay
21.10.99	562.43 ± 0.11	561.56 ± 0.24	561.59 ± 0.11	561.44 ± 0.24
25.10.99	561.96 ± 0.09	560.14 ± 0.22	561.32 ± 0.09	560.20 ± 0.22

Table 4: Efficiencies and concentrations extrapolated from linear fits to recent measurements

Measurement	Efficiencies		Solution concentration
date	Channel 1	Channel 2	(N_H/N_{Mn})
21.10.99	4.581×10^{-4}	4.575×10^{-4}	33.50
25.10.99	4.581×10^{-4}	4.575×10^{-4}	33.50

(Efficiencies are in terms of counts per decay of ^{56}Mn anywhere in the solution.)

Fraction of thermal neutrons captured in manganese, $f = 0.53820$ (from equation 2)

Table 5: Correction factors for 1999 measurement calculated using MCNP

	Percentage of emitted neutrons
Oxygen and sulphur fast capture	3.33%
Cavity and source capture	1.57%
Bath leakage	1.41%

Combined correction factor $\dfrac{1}{(1-O-S-L)} = 1.0674$

Mean source emission rate from Channel 1 $= 2.4310 \times 10^6$ s^{-1}
Mean source emission rate from Channel 2 $= 2.4327 \times 10^6$ s^{-1}

Overall mean emission rate = 2.432×10^6 s^{-1} (at 12 noon GMT on the 21st October 1999)

4.2   2004 measurement

Table 6: Saturated count rates for 2004 measurement from MNBATH6

(referenced to 12 noon GMT on 14.7.04)

Measurement	Channel 1		Channel 2	
date	Growth	Decay	Growth	Decay
14.7.04	555.35± 0.09	553.76 ± 0.21	554.21 ± 0.09	552.73 ± 0.21
19.7.04	555.42 ± 0.08	553.91± 0.21	553.92 ± 0.08	553.15 ± 0.21

Table 7: Efficiencies and concentrations for 2004 measurement extrapolated from linear fits to recent measurements

Measurement	Efficiencies		Solution concentration
date	Channel 1	Channel 2	(N_H/N_{Mn})
14.7.04	4.572×10^{-4}	4.565×10^{-4}	33.81
19.7.04	4.572×10^{-4}	4.565×10^{-4}	33.81

(Efficiencies are in terms of counts per decay of ^{56}Mn anywhere in the solution.)

Fraction of thermal neutrons captured in manganese, $f = 0.53602$ (from equation 2)

Table 8: Correction factors for 2004 measurement calculated using MCNP

	Percentage of emitted neutrons
Oxygen and sulphur fast capture (O)	3.33%
Cavity and source capture (S)	1.57%
Bath leakage (L)	1.41%

Combined correction factor $\dfrac{1}{(1-O-S-L)} = 1.0674$

Mean source emission rate from Channel 1 = 2.4155×10^6 s^{-1}
Mean source emission rate from Channel 2 = 2.4146×10^6 s^{-1}

Overall mean emission rate = 2.415×10^6 s^{-1} at 12 noon GMT on the 14th July 2004

5   UNCERTAINTIES

The evaluation of uncertainties was carried out in accordance with UKAS requirements[15]. The UKAS publication is consistent with the ISO *Guide to the Expression of Uncertainty in Measurement*[16].

5.1   Explanation of components

5.1.1   Counting

The emission of neutrons by the source and the decay of ^{56}Mn nuclei are both random processes. The uncertainties associated with these effects are calculated from the count data by the program SOURCEIMP. The standard error of the source emission rates calculated

from each channel, each run, and for growth and decay (i.e. 8 values for a 2 run measurement) is obtained.

The calculated counting uncertainty is typically less than ± 0.1% at the 67% confidence level. The uncertainty effectively has an infinite number of degrees of freedom.

5.1.2   Cross section ratios

Cross section ratios are used to determine the fraction of neutrons captured by ^{55}Mn in the solution. This uncertainty is dominated by the uncertainty in the H:Mn capture cross section ratio[9], but also includes the other uncertainties given in Table 2. It is taken as ± 0.2% at the 67% confidence level with a normal distribution.

5.1.3   Counter efficiencies

This uncertainty is dominated by the uncertainty in the ionisation chamber and $4\pi\beta$-γ coincidence measurements of the ^{56}Mn activity added to the solution. A linear fit to previous efficiency measurements is made as a function of time to enable interpolation or extrapolation to the time of the neutron source calibration. A typical value is ± 0.4% at the 67% confidence level with a normal distribution.

5.1.4   Fast neutron losses to oxygen and sulphur reactions [O]

This correction is neutron energy spectrum dependent and the sensitivity coefficient c_O has to be considered for each calibration. Partial differentiation of equation 1 yields:

$$c_O = \frac{O}{1-O} \qquad (3)$$

where O is the fraction of neutrons lost to O(n,p), S(n,p), and S(n,α) reactions. A typical value for the uncertainty in O is ± 20%, with a rectangular distribution assumed, which is then multiplied by c_O to give the propagated uncertainty in the emission rate. The value of the uncertainty is based on the uncertainty analysis in reference 13.

5.1.5   Neutron capture in the source and source container [S]

This correction depends on the source spectrum, source material, source capsule, and whether the large or small cavity sphere was used. The value of the sensitivity coefficient c_S has to be considered for each calibration. Partial differentiation of equation 1 yields:

$$c_S = \frac{S}{1-S} \qquad (4)$$

where S is the fraction of neutrons captured by the source and source container. The uncertainty in S is typically ± 5% with a rectangular distribution assumed. The value of the uncertainty is based on the uncertainty analysis in reference 13.

5.1.6 Neutron leakage from the boundaries of the bath [L]

This correction is solution concentration and source type dependent. Partial differentiation of equation 1 yields:

$$c_L = \frac{L}{1-L} \tag{5}$$

where L is the fraction of neutrons that escape from the boundaries of the bath. The uncertainty in L is typically $\pm$ 10%. The value of the uncertainty is based on the uncertainty analysis in reference 13.

5.1.7 Source transfer timing

This is to allow for uncertainty in the extra irradiation time experienced by the bath when the source is being transferred to, and from, the source cell. A value of $\pm$ 0.05% is typically assigned, with a rectangular distribution assumed.

5.1.8 Solution mixing effects

The uncertainty is estimated from the pipe-time measurements, and is typically $\pm$ 0.2%, with a rectangular distribution assumed.

5.1.9 Solution concentration

Uncertainty in the gravimetric determination of the hydrogen to manganese atom ratio (N_H/N_{Mn}) will propagate via the manganese capture fraction through to the calculated emission rate. The program SOURCEIMP calculates an extrapolated value for the concentration from linear fit parameters obtained using ORIGIN. A statistical estimate of the uncertainty in the value is therefore possible and is typically $\pm$ 0.1% at the 67% confidence level. The distribution is assumed to be normal.

5.1.10 Background [B]

The background correction has an inherent uncertainty of typically $\pm$ 10% at the 67% confidence level. However the effect of this uncertainty is very dependent on the emission rate of the source. To allow for this, a sensitivity coefficient c_B is applied:

$$c_B = \frac{B}{R} \tag{6}$$

where B is the mean background rate (s^{-1}) in the channels used, and R is the maximum count rate (s^{-1}) in the channels used. The uncertainty in the background is determined statistically from measurements made prior to each source measurement, and is therefore assumed to have a normal distribution.

5.1.11 Dead-time [D]

As mentioned in section 2.4, the 1999 measurement used dead-times from the two-source method. These values were averaged to give the values used in the program MNBATH6. A

statistical evaluation of the uncertainty was performed with a normal distribution assumed. Large variations were observed in the values so a typical value for the uncertainty from this method was ± 15% at the 67% confidence level.

The values of the dead-time from the decaying source method are still averaged to give the values used in the program MNBATH6, and a statistical evaluation of the uncertainty is performed with a normal distribution assumed. The spread of values is much less than for the two-source method and so the uncertainty is typically ± 5% from this method at the 67% confidence level.

The effect of this uncertainty on the combined uncertainty is very dependent on the count rates observed in the detector channels. To allow for this a sensitivity coefficient c_D is applied:

$$c_D = D \times R \tag{7}$$

where D is the mean dead-time in the channels used (s), and R is the maximum count rate (s^{-1}) in the channels used.

5.1.12 Half-life of source

The half-life of the source, τ, is not normally included in the manganese bath uncertainty budget as the longest decay correction would be 2 weeks and therefore any small uncertainty in the half-life would have a negligible effect on the overall uncertainty. The half-life uncertainty component has therefore not been included in the uncertainty in the measurement when corrected to the reference date of the measurement. However, it has been included for the result corrected to the comparison reference date of 1st January 2000.

As the effect of the half-life uncertainty on the emission rate uncertainty depends on the length of time in days, t, over which the decay correction is made, a sensitivity coefficient, c_τ, was applied:

$$c_\tau = \frac{|t| \times \ln(2)}{\tau} \tag{8}$$

The value of the half-life of ^{241}Am was specified in the protocol for the intercomparison as 1.5785×10^5 days. The uncertainty in the half-life was taken as 0.0024×10^5 days (with a rectangular distribution) from IAEA TECDOC-619[17].

5.1.13 Correlations

The main source of correlations with the uncertainties of other participants is likely to be the use of the same cross-section data. A H:Mn capture cross section ratio of 0.024799 ± 0.00005, and a S:Mn capture cross section ratio of 0.03990 ± 0.0005 were used in equation 2 which both come from reference 9. The three correction factors O, S, and L, were calculated using MCNP with mostly ENDF/B-VI cross sections. There will therefore be some degree of correlation with other participants who use the same method to determine the correction factors, however the actual degree will depend on other factors like the concentrations of their solutions, and the sizes of their baths.

5.2   1999 measurement

The full uncertainty budget for 1999 measurement is shown below. The uncertainty components are added in quadrature to give the combined uncertainty in the emission rate of the source.

Table 9: Full uncertainty budget for 1999 measurement

Source	Uncertainty (%)	Distribution	Sensitivity	Uncertainty component (%)	Degrees of freedom
Counting	0.1	Normal	1	0.1	∞
Cross-section ratio	0.2	Normal	1	0.2	∞
Efficiency	0.4	Normal	1	0.4	∞
O & S losses	20	Rectangular	0.03445	0.4	∞
Cavity and source capture	5	Rectangular	0.01595	0.05	∞
Leakage	4	Rectangular	0.01430	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	0.00521	0.05	9
Dead-time	15	Normal	0.00216	0.03	9
Half-life of source	0.15	Rectangular	0.00032	0.00005	∞

5.2.1   Correcting to local reference time

Type A (random) uncertainty = ± 0.11%
Type B (non-random) uncertainty = ± 0.62%
Combined uncertainty (at 1σ level) = ± 0.63%
Degrees of freedom = 13231 (therefore coverage factor of k=2 can be used)

Expanded uncertainty (at 2σ level) = ± 1.26%

The emission rate of the source at 12 noon GMT on the 21st October 1999 was therefore (2.432 ± 0.031) × 10^6 s^{-1}. Uncertainty quoted at 95% confidence level.

5.2.2   Correcting to comparison reference time

Type A (random) uncertainty = ± 0.11%
Type B (non-random) uncertainty = ± 0.62%

Combined uncertainty (at 1σ level) = ± 0.63%
Degrees of freedom = 13231(therefore coverage factor of k=2 can be used)

Expanded uncertainty (at 2σ level) = ± 1.26%

The emission rate of the source at 12 noon GMT on the 1st January 2000 was therefore (2.431 ± 0.031) × 10^6 s^{-1}. Uncertainty quoted at 95% confidence level.

5.3   2004 measurement

Table 10: Full uncertainty budget for 2004 measurement

Source	Uncertainty (%)	Distribution	Sensitivity	Uncertainty component (%)	Degrees of freedom
Counting	0.05	Normal	1	0.05	∞
Cross-section ratio	0.2	Normal	1	0.2	∞
Efficiency	0.4	Normal	1	0.4	∞
O & S losses	20	Rectangular	0.03445	0.4	∞
Cavity and source capture	5	Rectangular	0.01595	0.05	∞
Leakage	4	Rectangular	0.01574	0.04	∞
Timing	0.05	Rectangular	1	0.03	∞
Mixing	0.2	Rectangular	1	0.1	∞
Solution concentration	0.1	Normal	1	0.1	9
Background	7	Normal	0.00519	0.04	9
Dead-time	5	Normal	0.00199	0.01	9
Half-life of source	0.15	Normal	0.00727	0.001	∞

5.3.1   Correcting to local reference time

Type A (random) uncertainty = ± 0.06%
Type B (non-random) uncertainty = ± 0.62%

Combined uncertainty (at 1σ level) = ± 0.62%
Degrees of freedom = 13420 (therefore coverage factor of k=2 can be used)

Expanded uncertainty (at 2σ level) = ± 1.25%

The emission rate of the source at 12 noon GMT on the 14th July 2004 was therefore (2.415 ± 0.030) × 10^6 s^{-1}. Uncertainty quoted at 95% confidence level.

5.3.2   Correcting to comparison reference time

Type A (random) uncertainty = ± 0.06%
Type B (non-random) uncertainty = ± 0.62%

Combined uncertainty (at 1σ level) = ± 0.62%
Degrees of freedom = 13420 (therefore coverage factor of k=2 can be used)

Expanded uncertainty (at 2σ level) = ± 1.25%

The emission rate of the source at 12 noon GMT on the 1st January 2000 was therefore (2.433 ± 0.030) × 10^6 s^{-1}. Uncertainty quoted at 95% confidence level.

6 CONCLUSION

The 1999 and 2004 NPL measurements of the neutron emission rate of the comparison source yielded values of $(2.431 \pm 0.031) \times 10^6 \ s^{-1}$ and $(2.433 \pm 0.030) \times 10^6 \ s^{-1}$ respectively, referenced to 12 noon GMT on 1st January 2000, with uncertainties quoted at the 95% confidence level.

The ratio of the decay-corrected 1999 to 2004 measurements is 0.9994 ± 0.0062. The uncertainty in the ratio comes from the components which are uncorrelated between the two measurements (i.e. it excludes the cross-section ratio, O and S losses, cavity and source capture, and leakage components) and is at the 67% confidence level. This indicates that there is excellent agreement between the two measurements and verifies that the source emission rate has remained consistent with the decay constant throughout the comparison exercise thus far.

Since both measurements have near equal uncertainties the best estimate of the emission rate is a simple mean. From Table 9 and Table 10 it can be seen that the majority of the uncertainties are either fully correlated or very largely correlated since they are due to uncertainties in corrections which are common to both measurements. The only truly uncorrelated uncertainties are the counting statistics and these are essentially negligible when added in quadrature with the other components.

The best estimate of the neutron emission rate of the comparison source, referenced to 12 noon GMT on 1st January 2000, with uncertainties quoted at the 95% confidence level values, is thus $(2.432 \pm 0.030) \times 10^6 \ s^{-1}$.

7 ACKNOWLEDGEMENTS

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