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EUROMET Project No. 221 Final Report.

Bilateral intercomparison of the neutron emission rate of a ^{252}Cf spontaneous fission source between the National Physical Laboratory, Teddington, UK. and the Physikalisch-Technische Bundesanstalt, Braunschweig, Germany.

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ABSTRACT

The neutron emission rate of a small ^{252}Cf fission source, incorporated into a fission counter, has been measured by two different methods. The fission rate of the source was measured at PTB, the neutron emission rate being obtained by means of the neutron multiplicity factor $\bar{\nu}$. The manganese sulphate bath method was used at NPL to determine the total neutron emission from the chamber assembly. Agreement between the two independent methods was found to be within 1%.

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National Physical Laboratory
Teddington, Middlesex, UK, TW11 0LW

Approved on behalf of Chief Executive, NPL,
by Dr P Christmas, Head, Division of Radiation Science and Acoustics.

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1.0 INTRODUCTION

EUROMET collaboration project Reference Number 221 was set up between the National Physical Laboratory (NPL), Teddington, UK and the Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, Germany with the aim of strengthening traceability within Europe, to appropriate primary standards, for the determination of the absolute neutron emission rate from radionuclide neutron sources.

It is important that there is within Europe a unified system of such measurements which is traceable to the appropriate national standards. The basis of such a unified system is the intercomparison of measurements between national laboratories such as that for the measurement of neutron emission rate organised by BIPM in the early 1980's. No similar intercomparison is planned for the next decade.

The present project was intended to facilitate a bilateral comparison of neutron source emission rates between PTB and NPL in which each laboratory would measure the neutron emission rate of a specially constructed ^{252}Cf source, which is routinely used for the precise calibration of NE213 liquid scintillation detectors as part of the PTB time-of-flight spectrometry system.

For the specially designed ^{252}Cf neutron source, fabricated by Harwell Laboratories, UK, and incorporated in an ionisation chamber used as a fission fragment detector, the rate of disintegration by fission would be precisely determined at PTB [1]. The neutron emission rate would then be obtained by multiplying the fission rate by the well known neutron multiplicity $\bar{\nu}$ of 3.7676 for ^{252}Cf [2].

At NPL the neutron emission rate of the device would be measured by the manganese sulphate bath method [3], a well established technique, and for which a routine service has been operated by NPL for the past 30 years. The effect of the low mass counter device, which encloses the ^{252}Cf source, on the measured neutron emission rate would be calculated at NPL.

The determination of the primary neutron source emission rate will thus be made by these two completely independent methods.

It is anticipated that following the successful completion of this intercomparison, PTB will devolve radionuclide neutron source emission rate measurements to NPL. This will lead to a saving of resources by PTB in maintaining a suitable measuring system and will allow NPL to make more efficient use of it's established facility.

2.0 THE CALIFORNIUM SOURCE.

2.1 The fission chamber for the PTB measurement.

The design of the fission chamber as used at PTB to determine the fission rate of the ^{252}Cf source is shown in Figure 1(a). The use of such a chamber to calibrate a time-of-flight system requires minimal disturbance of the emitted neutron spectrum and imposes constraints on the construction of the chamber which have been previously discussed by Böttger et al[1].

2.2 The fission source for the NPL measurement.

The fission chamber as used at PTB was too large, due to the gas and high voltage connectors, to fit into the cavity used for the manganese bath measurement of emission rate. The chamber was therefore modified by replacing the rear cover, which carried the gas and electrical connections, with a plain cover similar to that used on the front of the chamber. The construction of the device was otherwise unchanged and is shown in Figure 1(b).

3.0 THE NPL MEASUREMENT OF THE NEUTRON EMISSION RATE.

The neutron emission rate from the ^{252}Cf source was determined by the manganese sulphate bath technique, with the same measurement regime as used for routine source measurements.

3.1 The irradiations.

The source assembly was mounted on a hemispherical block of expanded polystyrene foam which was enclosed in a thin walled spherical polystyrene container 88 mm in diameter with a wall thickness of approximately 1.5 mm. This was placed at the centre of a spherical stainless steel bath 0.98 m in diameter containing a concentrated aqueous solution of pure manganese sulphate ($MnSO_4$) which had a hydrogen to manganese atom ratio of 33.156. The solution was stirred and, at the same time, circulated by means of pumps through a shielded assembly which contained two sodium iodide (NaI) scintillation detectors (CH1 & CH2) as described by Axton et al[3].

Four separate measurements were made of the source emission rate, on 26 February and 1, 5 and 8 March 1990. Each measurement consisted of a 24-hour irradiation of the solution during which the observed detector counts were recorded every 1000 s, followed by approximately 11 hours measurement of the decaying activity in the solution after removal of the source.

The observed detector count rates were corrected for deadtime, background, decay of the ^{56}Mn during transit to the detector assembly from the bath, and for the decay of the source during the counting and the irradiation periods. The results of each measurement were

corrected to a common reference time of 1200 h on 26 February 1990 using a half-life of 2.645 years [4].

3.2 Neutron emission rate.

The neutron emission rate of the source is given by:

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

where A is the corrected saturation count rate from the detectors, ε_{Mn} is the counting efficiency defined as the count 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 captured in the source and it's mounting assembly, O is the fraction of neutrons captured in neutron induced charged particle reactions in oxygen and sulphur in the solution. The solution is specified as having the composition:

$$MnSO_4 + \left(\frac{N_{Im}}{N_{Mn}}\right) ImSO_4 + \left(\frac{N_H}{2N_{Mn}}\right) H_2O \qquad (2)$$

and f is the fraction of the remaining neutrons which are captured in manganese, given by:

$$\frac{1}{f} = 1 + \frac{N_{Im}}{N_{Mn}} \frac{\sigma_{Im}\,(1 + \bar{r}s)_{Im}}{\sigma_{Mn}(1 + G\bar{r}s)_{Mn}} + \left[1 + \frac{N_{Im}}{N_{Mn}}\right] \frac{\sigma_S\,(1 + \bar{r}s)_S}{\sigma_{Mn}(1 + G\bar{r}s)_{Mn}} + \frac{N_H}{N_{Mn}} \frac{\sigma_H}{\sigma_{Mn}(1 + G\bar{r}s)_{Mn}} \qquad (3)$$

where N_H/N_{Mn} etc. are the atom ratios of the constituents of the solution. Im is an imaginary impurity element which when present as $ImSO_4$ accounts for the weight of dry impurities in the determination of N_H/N_{Mn} (Axton and Bardell) [5]. The capture cross section σ_{Im} and the resonance capture integral parameter s_{Im} are chosen to give the correct neutron absorption properties to the solution. G is a resonance self shielding factor, and $\bar{r}$ is the Westcott [6] epithermal flux parameter averaged over the system. The values of these constants are given in Table 1, together with the other cross sections and half-lives used.

3.3 Corrections.

3.3.1 Neutron leakage.

The number of neutrons escaping from the boundaries of the bath was measured with a De Pangher precision long counter placed with it's front face tangential to the surface of the bath.

The resulting count rates were corrected for dead time and background effects and integrated over the surface of the bath. The long counter efficiency was obtained from a least squares fit to the data from a dilution experiment carried out to determine the σ_H/σ_{Mn} cross-section ratio (Axton et al)[7]. The neutron leakage per unit source is given in Table 2.

3.3.2 Neutron capture in the source and holder.

Details of the source and encapsulation materials were obtained from PTB. The construction of the source is shown in Figure 1b. It was originally thought that the incorporation of a high capture cross section indium metal seal in the source assembly would complicate the calculation of the source capture term. In the event it was found that the small amount of indium present had only a very minor effect on the correction. This calculation was verified by an experimental measurement of another small ^{252}Cf source with and without a similar quantity of indium. The calculated masses of the components and their cross-sections are given in Table 2. The correction factor for thermal neutron capture in the source and it's holder was calculated by a combination of Monte Carlo and diffusion methods using the fission spectrum described by Grundl and Eisenhauer[8]. The values of the macroscopic capture cross section of the source, the thermal neutron fluence at the cavity boundary, and the corrections for self shielding and flux depression derived by these methods are also given in Table 2.

3.3.3 Oxygen and sulphur reactions.

The losses due to neutron induced charged particle reactions in oxygen and sulphur were calculated by the same Monte Carlo method used in 3.3.2 and are given in Table 2.

3.3.4 The manganese fraction.

The manganese capture fraction (f) depends on the atom ratios of the constituents of the solution as shown in equation (2), and their thermal neutron capture cross-section ratios as given in equation (3). The manganese sulphate used to make up the solution is of "Analar" quality and has been the subject of extensive chemical analysis for impurities as reported in Reference 5.

The hydrogen to manganese atom ratio was determined gravimetrically by evaporating a weighed sample of the solution to dryness and baking in a furnace at 300°C to drive off the water of crystallization. The cross-sections and their ratios are those derived by Axton[9] from a least-squares evaluation of the thermal neutron absorption cross-sections of Mn and S.

3.3.5 ^{56}Mn detection efficiency.

The detection efficiency of the system (ε_{Mn}) was determined on 15 February and 15 March 1990 by the addition of known amounts of ^{56}Mn to the inactive system. The specific activity

of the added active solution was determined by measurement in an ionisation chamber which had been calibrated via solutions standardised by $4\pi\beta$-γ coincidence counting. Values of the measured efficiencies are given in Table 1. Efficiency values for the dates of measurement of the source were obtained by interpolation, assuming a linear change with time.

4.0 THE PTB FISSION RATE MEASUREMENT

The measurements were performed using the fission chamber in the time-of-flight (TOF) arrangement to determine the counting efficiency, as described by Böttger et al[1].

4.1 Neutron Emission Rate

Assuming the well known neutron multiplicity $\bar{\nu}$ for the spontaneous fission of ^{252}Cf[2], the neutron source strength Q [s^{-1}] is determined by

$$Q = A \times \frac{1}{(1 - A \times \tau)} \times \frac{1}{\varepsilon_{Cf}} \times \bar{\nu} \qquad (4)$$

Where A [s^{-1}] is the measured fission event rate, τ is the nonextending dead time in the fission detector chain, and ε_{Cf} is the efficiency for the detection of one of the two fission products.

While the dead time τ can easily be measured by means of a time interval generator, the precise determination of the efficiency ε_{Cf} requires considerable additional effort[1]. The efficiency of this type of fission chamber is typically in the range 0.95 - 0.99, depending on the device specific parameters.

4.2 Determination of the Fission Detector Efficiency

To determine the efficiency of the ionisation chamber for detection of fission fragments emitted from the ^{252}Cf layer, the correlation between prompt neutrons and fission fragments was studied. A 3.81 cm × 3.81 cm NE213 liquid scintillation counter, well shielded against background radiation, was used to detect prompt neutrons in a time-of-flight arrangement with the fission detector (see Reference 1). Because of the lower count rate from the scintillator a fast signal from this detector was used as a START signal for a time-to-amplitude converter (TAC). The STOP signal was derived from the fission counter. Neutron identification, based on a two-dimensional display of pulse amplitude against pulse shape, was used for the NE213 scintillator to discriminate against photons, and the total number N_N of neutron induced events above a threshold of 1.8 MeV was recorded.

A subgroup of N_N are those neutrons N_{NF} for which a correlated fission fragment was identified with the constraint that an event was observed in the TOF spectrum within the

limits T_{MIN} and T_{MAX} (see Figure 2). The time chosen between the zero point Z of the TAC and the coincidence peak in the TOF spectrum was larger than the dead time, as can be seen in Figure 2. Then the difference between N_N and N_{NF} corresponds to the undetectable fission fragments which are emitted nearly parallel to the surface of the backing such that their signal does not exceed the threshold set by the discriminator to avoid electronic noise or pile-up from alpha particles. The efficiency,

$$\varepsilon_{Cf} = \frac{N_{NF}}{N_N} \tag{5}$$

determined in such a coincidence experiment, depends on the neutron detector threshold, the angle between the normal to the front plate of the fission detector and the neutron detector axis, and on mechanical properties of the backing for the Cf layer. As discussed in detail in Reference 1, the average efficiency ε_{Cf}, which is the quantity of interest here for the neutron source strength determination, can experimentally be determined at $\theta = 60°$, at which angle ε_{Cf} is independent of the neutron detector threshold.

4.3 Corrections

4.3.1 Background

The background from other neutron sources was determined separately with the ^{252}Cf source well shielded against the scintillation detector by a water tank but without changing other experimental parameters. Randomly distributed events in the TOF spectrum indicate that there are no other correlations between fission fragments and detected neutrons. The background amounts to 0.25% for N_N. For N_{NF}, however, the background to be considered is much smaller (in this case by a factor of 12) and can thus be neglected. The background correction changes the result for $(1-\varepsilon_{Cf})$ by 10%.

4.3.2 Non-correlated events

For N_{NF} it was assumed that it comprises only events with a neutron starting and the correlated fission fragment stopping the time measurement of the TAC. However, a neutron from an undetected fission fragment can also start the time measurement which may then be stopped with a probability

$$P = (T_{max} - T_{min})_{TAC} \times A \tag{6}$$

by a non-correlated fission fragment, where T_{max} and T_{min} are the upper and lower limits of the TAC range and A is the rate of disintegration by fission. This correction reduces N_{NF} in this investigation by about 0.2%. Depending on the chosen experimental conditions, this correction may just compensate with the background correction.

4.3.3 TOF zero point

Another correction to be considered, which applies only for N_{NF}, is that part of the TOF spectrum between the zero point (TRUE-START) and the experimentally chosen beginning of the spectrum. It is taken into account by extrapolation from higher channel numbers. This correction enlarges N_{NF} by about 0.2%.

4.4 The measured fission detection efficiency

With the electronic set up used in previous investigations and leading edge timing a neutron energy independent efficiency for fission fragment detection at 60° of 97.00% ± 0.35% was determined with a measuring time of 21 hours in total. In agreement with previous investigations, the efficiency at 0° was found significantly higher (≥98.3%) and threshold dependent.

5.0 RESULTS.

5.1 The NPL neutron emission rate measurement

The neutron emission rate of the ^{252}Cf source at 12.00 hours on 26 February 1990 was measured as $(4.972 \pm 0.025) \times 10^5$ s^{-1}. In order to compare the two measurements this value was corrected for decay back to the reference date used by PTB, using a value of 2.645 y for the ^{252}Cf half-life, giving a neutron emission rate of $(5.066 \pm 0.025) \times 10^5$ s^{-1} on 31 January 1990.

The random uncertainty associated with this measurement is estimated as ± 0.1% (standard error of the mean). The limits of uncertainties systematic to the measurement are estimated as follows:

Cross section ratios (σ_H : σ_{Mn})	± 0.3%
Detection efficiency (ε_{Mn})	± 0.3%
Fast neutron capture by oxygen and sulphur	± 0.1%
Neutron escape from the boundaries of the bath	± 0.1%
Neutron capture in the source and holder	± 0.1%
Overall uncertainty by addition of variances	± 0.5%

All uncertainties are expressed at 1σ (68% confidence level).

5.2 The PTB fission rate measurement

With an average efficiency ε_{Cf} of 0.970, a dead time of 507 ± 10 ns and a total neutron multiplicity of 3.7676 ± 0.0049 [2], a neutron source emission rate Q inside the chamber of (5.117 ± 0.020) × 10^5 s^{-1} at 31 January 1990 was determined.
The uncertainty of the neutron emission rate of ± 0.4% is dominated by the contribution from the efficiency determination, where the fraction of undetected fission events is determined with an uncertainty of about 10%, as can be seen from the list of uncertainties below.

Statistics of measured fission rate	± 0.05%
Efficiency for fission detection (ε_{Cf})	± 0.35%
Dead time correction (τ)	± 0.10%
Total neutron multiplicity ($\bar{\nu}$)	± 0.13%
Correction for activity to reference date	± 0.06%
Overall uncertainty by addition of variances	± 0.40%

All uncertainties are expressed at 1σ (68% confidence level).

5.3 Comparison

The results of the two measurements, corrected to a common reference date of 31 January 1990 are:

NPL 5.066 × 10^5 s^{-1} ± 0.5%
PTB 5.117 × 10^5 s^{-1} ± 0.4%
Ratio PTB:NPL 1.010 ± 0.007

The apparent difference of approximately 1% is possibly due to the fact that the two measurement methods do not measure the same quantity. The NPL system measures the total neutron emission outside the source encapsulation, the PTB measurement is of the neutron production in the fission layer, that is fission rate × $\bar{\nu}$, inside the encapsulation.

Neutron scattering in the capsule does not change the total neutron emission rate between inside and outside. Furthermore, those reactions which do attenuate the fluence may be compensated by (n,2n) reactions in the capsule. For this reason a simulation was performed by means of a general purpose neutron and photon transport code (MCNP [10]) taking into account the detailed model of the modified fission chamber as shown in Fig.1b. Integrating over all energies the calculated emission rate outside differs from the one inside the chamber by only -0.024%.

Another possible reason for the discrepancy, arising from the exchange of the chamber's rear cover, was also investigated. It is well known from previous experiments, that after two years of operation about 10% of the Cf-deposit can be found on the front plate ('creep effect'), but up to now no contamination larger than 10^{-4} of the total amount of Cf was detectable at the rear part of the chamber including the gas inlet and outlet.

6.0 CONCLUSION

The neutron emission rate of a ^{252}Cf spontaneous fission source was measured with two different methods at NPL and PTB . The difference of the two emission rates determined amounts to 1.0% and just exceeds the sum of the evaluated standard deviations of 0.9%. There is, however, no serious reason to doubt the evaluation procedure of either method or the parameters used.

Because of the good agreement obtained between the PTB and NPL measurements, the PTB propose to generally devolve official neutron source measurements from PTB to NPL. Future official requests for such measurements will be transferred from PTB to NPL.

The PTB will continue to perform neutron source emission rate measurements only within the framework of scientific collaboration, or in the case of special requests which are outside the scope of routine work.

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Table 1.

Manganese bath data.

N_H/N_{Mn}	33.156	
N_{Im}/N_{Mn}	3.100×10^{-4}	
σ_H/σ_{Mn}	2.4799×10^{-2}	
σ_{Mn}	13.408 b	
σ_S	0.535 b	
σ_{Im}	3.320 b	
$(1 + \bar{rs})_{Mn}$	1.0159	
$(1 + \bar{rs})_S$	1.0005	
$(1 + \bar{rs})_{Im}$	1.0071	
$T_{½}$ ^{252}Cf	2.645 y	
$T_{½}$ ^{56}Mn	2.580 h	
ε_{Mn} (15-2-90)	CH1 45.68	CH2 45.62
ε_{Mn} (15-3-90)	CH1 45.56	CH2 45.52

Table 2.

Source composition.

Source material	Evaporated Californium
Source backing	Platinum
Front and rear covers	Stainless steel
Clamping rings	Aluminium
Sealing gasket	Indium
Bolts	Stainless steel

Thermal neutron capture cross sections per gram of material.

Component	Mass[1] (g)	Σ b g^{-1}
Source and backing	0.6	1.707
Covers	4.26	0.033
Clamping ring	4.83	0.0051
Gasket	0.54	1.016
Bolts	0.43	0.033

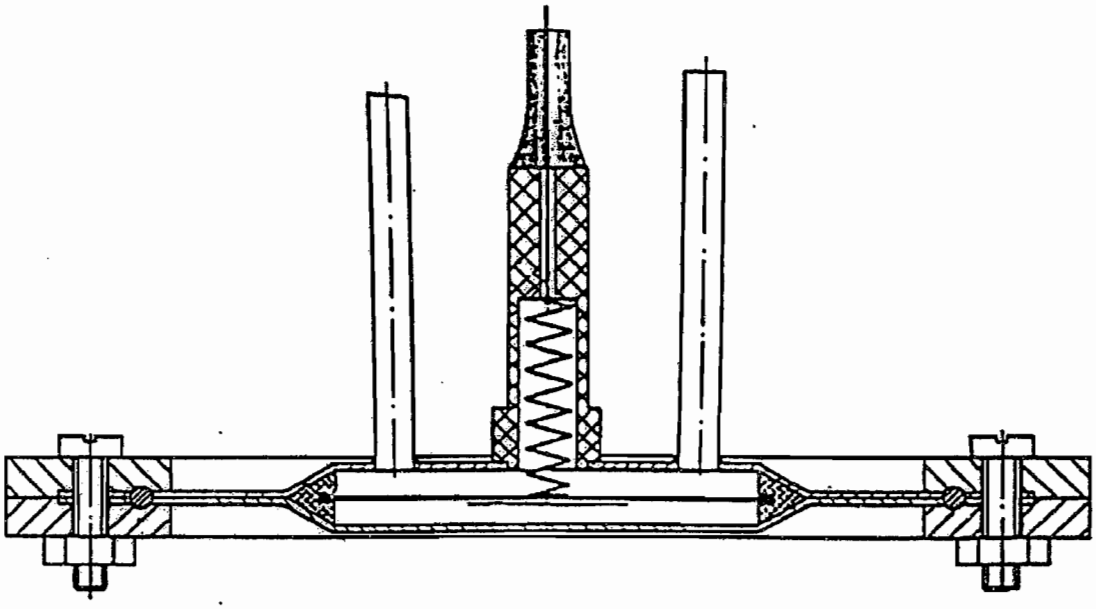
(1) Estimated from dimensions supplied on sketch of the source construction.

Fluences and reactions per unit source.

Effective source and cavity cross section	1.864 b
Cavity thermal neutron fluence	1.81×10^{-3} cm^{-2}
Depressed cavity fluence	1.73×10^{-3} cm^{-2}
Total thermal neutron capture	2.19×10^{-3}
Reactions in oxygen and sulphur	6.43×10^{-3}
Neutron escape from the bath	3.69×10^{-3}

Figure 1.

(a) The ^{252}Cf fission chamber used for the PTB measurement.



(b) The ^{252}Cf source for the NPL measurement.

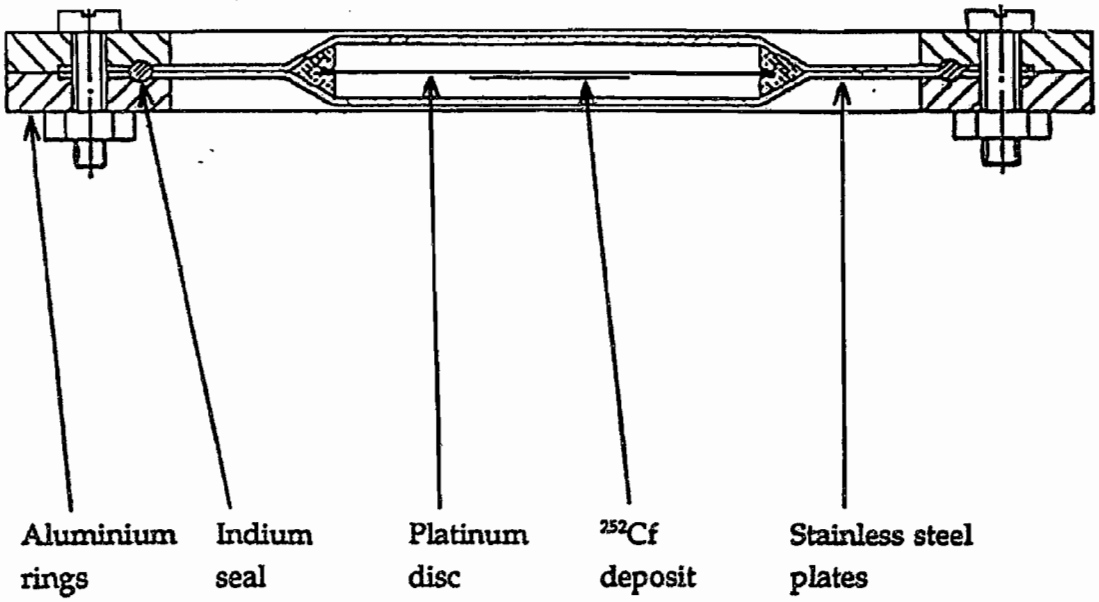


Figure 2.

Time of flight spectrum only for neutrons (1 ch = 0.93 ns).
Zero-point Z, TMIN, TMAX and dead time τ are indicated.