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ABSOLUTE MEASUREMENTS OF ^{252}Cf TOTAL NU-BAR BY THE MANGENSE BATH METHOD

made at
the V.G. Khlopin Radium Institute

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ABSOLUTE MEASUREMENTS OF ^{252}Cf TOTAL NU-BAR BY THE MANGENSE BATH METHOD

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English translation of the papers presented at the
1975 and 1980 Kiev Conferences:

1. DETERMINATION OF THE AVERAGE NUMBER OF NEUTRONS PER FISSION EVENT FOR CALIFORNIUM-252

B.M. Aleksandrov, L.M. Belov, L.V. Drapchinskij, Ya.M. Kramarovskij, G.E. Lozhkomoev, V.G. Matyukhanov, K.A. Petrzhak, A.G. Prusakov, A.V. Sorokina, Eh.A. Shlyamin, O.A. Migun'kov, G.M. Stukov, V.T. Shchebolev and I.A. Yaritsyna

V.G. Khlopin Radium Institute and D.I. Mendeleev All-Union Scientific Research Institute of Metrology

Proceedings of the 1975 Kiev Conference, 5 (1976) 166-169

2. ABSOLUTE MEASUREMENTS OF $\bar{\nu}(^{252}Cf)$ BY THE MANGANESE BATH METHOD

B.M. Aleksandrov, E.V. Korolev, Ya.M. Kramarovskij, G.E. Lozhkomoev, V.G. Matyukhanov, K.A. Petrzhak, A.G. Prusakov, A.V. Sorikina and Eh.A. Shlyamin

V.G. Khlopin Radium Institute

Proceedings of the 1980 Kiev Conference, 4 (1981) 119-123

Editor's note: All nu-bar values quoted include delayed neutrons.

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DETERMINATION OF THE AVERAGE NUMBER OF NEUTRONS PER FISSION EVENT FOR CALIFORNIUM-252

B.M. Aleksandrov, L.M. Belov, L.V. Drapchinskij, Ya.M. Kramarovskij, G.E. Lozhkomoev, V.G. Matyukhanov, K.A. Petrzhak, A.G. Prusakov, A.V. Sorokina, Eh.A. Shlyamin, O.A. Migun'kov, G.M. Stukov, V.T. Shchebolev and I.A. Yaritsyna
V.G. Khlopin Radium Institute and D.I. Mendeleev All-Union Scientific Research Institute of Metrology

Proceedings of the 1975 Kiev Conference, 5 (1976) 166-169

ABSTRACT

By means of a separate determination of neutron yields and fission event rates, the value of $\bar{\nu}(^{252}Cf)$ has been measured for a series of new high-purity sources. The improved quality of the source active layers has reduced the error in determining the fission rate to 0.35%. The value obtained for $\bar{\nu}(^{252}Cf)$ is 3.747 ± 0.036. A description is given of the design and the parameters of a spherical manganese bath in which the work on refining the value of $\bar{\nu}(^{252}Cf)$ will be continued.

Work on the measurement of $\bar{\nu}(^{252}Cf)$ was started by the V.G. Khlopin Radium Institute and the D.I. Mendeleev All-Union Scientific Research Institute of Metrology in 1972 [1]. The californium sources were prepared and the fission rates were measured at the Radium Institute - the small-solid-angle method being used. The neutron yields from the same sources were measured at the Mendeleev Institute by various techniques. The average weighted error in the determination of the neutron yields was 0.9%. The error in the measurement of the fission rate was 0.7%. The preliminary value obtained for $\bar{\nu}(^{252}Cf)$ was 3.770 ± 0.045.

A further reduction in the error associated with the $\bar{\nu}$ measurement was accomplished in two ways: (a) by the preparation of new high-quality sources made of highly-purified californium and subsequent measurement of the fission rate by a small-solid-angle chamber; (b) by the measurement of neutron yields with the aid of a magnesium flow bath constructed at the Radium Institute.

The californium sources were prepared from a hydrochloric-acid solution of californium after coarse and fine chromatographic purification. Since the main requirement for the sources was that the active layer should be uniform and have sufficient strength, a thermal sputtering method was used in their preparation.

Before the deposition of the californium on the substrate, the dish containing the dry californium residue was subjected to preliminary heating to remove the resin and volatile impurities remaining after the fine purification cycle. The heating was continued up to a temperature of 1200°C and no significant sputtering of the californium was observed (this usually takes place at a temperature of 1800°C). When the sputtering was carried out at a distance of 15 mm from the substrate, the radial non-uniformity in the active spot was as much as 10%. In an attempt to improve the uniformity, we used dishes of different shapes, including one with a directed nozzle of diameter 1 mm and height 3 mm. Experience showed, however, that it was not possible to achieve any noticeable improvement. Further sputtering was therefore carried out at a distance of 30 mm, which ensured uniformity of the layer to within 3%. The radial uniformity in the thickness of the active spot was tested by recording the α-particles with a stopped-down ($\emptyset$ = 0.5 mm) surface-barrier detector.

When preparing the californium sources by thermal sputtering, separation of the transuranium elements was found to occur. For example, the ratio of the curium and californium alpha-activities in the initial solution was 2.5, whereas in the target it was equal to 0.25. This effect is most probably due to the different volatilities of the curium and californium oxides and can be utilized for additional purification of the californium directly in the source preparation process. In all we prepared four sources with ^{252}Cf weights in the range 0.129-0.424 μg.

Because of the good layer uniformity, the accurate geometry of the active spot and the reduced amount of uncertainty in the self-sputtering, the error in the measurement of the fission rate in these sources by the small-solid-angle method was reduced to 0.35%.

To improve the accuracy of the neutron flux measurements for the californium targets, a device using a spherical manganese flow bath was constructed at the Radium Institute in 1974.

Flow conditions for the measurement of the induced ^{56}Mn activity have the advantage over static conditions that the measurement time and hence the statistical data accumulated were not in any way limited, so that the random error in the induced activity measurement can be reduced almost to zero. By contrast, the counting statistics for activity measurements by an immersed counter are determined by the area of the ^{56}Mn decay curve. Two variants are possible with the flow method: (a) direct measurements with an immersed neutron source; (b) measurements to determine saturation of the bath activity after the source is removed. In both cases, the saturation activity of the bath is calculated from the results of gamma-gamma flow counter measurements with the use of the appropriate equations from Ref. [2].

The bath is in the form of a sphere, welded from stainless steel sheeting, with two flanges along a vertical diameter, to which the tubes are fixed. An aqueous solution of manganese sulphate is pumped through two closed loops. One loop serves only for mixing the solution and includes two ND400/16 metering pumps with a total discharge of 8 L/h; the other loop has two ND100/10 metering pumps with a 200 L/h discharge and a gamma-gamma flow counter.

The gamma-gamma counter is a stainless steel chamber (t = 0.5 m) containing two NaI(Tl) crystals of 150 x 100 mm with an FEhU-4913 photomultiplier tube. To increase the registration efficiency, the counter was designed so that the active solution flowed not only over the ends of the crystals but also over their side surface.

To record the induced ^{56}Mn activity, we constructed a high stability gamma-gamma coincidence unit. Prolonged control measurements with ^{88}Y and ^{54}Mn reference sources showed that the fluctuations in the counting channels and the coincidence channel were determined only by the measurement statistics.

The bath was filled with a 25% aqueous solution of manganese sulphate of an especially high degree of purity - the capture of thermal neutrons by impurity elements did not exceed in total 0.1% of the neutron capture by the manganese.

A high-concentration solution was prepared to allow for subsequent dilution and operation of the bath at different solution strengths. This makes it possible to exclude from the resultant error in the determination of the neutron flux that part associated with determining the neutron capture cross-sections of manganese and hydrogen.

During the time that the manganese bath was being tried out and adjusted, neutron yield measurements on the new californium sources were being carried out at the Mendeleev Institute by the methods described in Ref. [3]. The value of $\nu(^{252}Cf)$ obtained was 3.747 ± 0.036.

Further refinement of the $\nu(^{252}Cf)$ value will be possible with the aid of the Radium Institute manganese bath.

REFERENCES

[1] Neutron Physics [in Russian], IV, Obninsk (1974) 76.

[2] J. Nucl. Energ. 19 (1965) 409.

[3] YARITSYNA, I.A., IVANOV, Yu.V., ANDREEV, O.D., Neutron Measurements [in Russian], Moscow (1973).

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ABSOLUTE MEASUREMENTS OF $\bar{\nu}(^{252}Cf)$ BY THE MANGANESE BATH METHOD

B.M. Aleksandrov, E.V. Korolev, Ya.M. Kramarovskij, G.E. Lozhkomoev, V.G. Matyukhanov, K.A. Petrzhak, A.G. Prusakov, A.V. Sorokina and Eh.A. Shlyamin
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Proceedings of the 1980 Kiev Conference, 4 (1981) 119-123

ABSTRACT

The manganese bath method has been used with fragment counting over a small solid angle to measure the value of $\bar{\nu}(^{252}Cf)$. The corrections for loss of neutrons from the bath and for absorption in the source and holder material were determined experimentally. The correction for fast-neutron capture by sulphur and oxygen was calculated by the Monte Carlo method. The value of $\bar{\nu}(^{252}Cf)$ obtained was 3.758 ± 0.015.

Measurements of the average number of neutrons per fission event in californium-252 have been carried out for almost 20 years. Two main methods have been used: a technique involving a large liquid scintillator, where the average number of prompt neutrons is measured during a "gate" time defined by the fragment burst, and the manganese bath technique. In the latter case, separate measurements are made of the neutron yield from the californium source by means of a manganese bath and of the fission rate by means of different fission fragment counting techniques.

The reason for the undiminished interested in the problem of establishing the most reliable value of $\bar{\nu}(^{252}Cf)$ is that the accuracy required by reactor designers is 0.25%, but there is a 3% scatter in the results of individual experiments - which are themselves carried out to an accuracy of 0.5%. Practically all the original values of $\bar{\nu}(^{252}Cf)$ have been changed by a process of repeated revision.

Table 1 (Ref. [1])

$\bar{\nu}(^{252}Cf)$

	1972	1979	Group average
Liquid scintillation method			
Spencer		3.792±0.011	
Boldeman	3.744±0.014	3.755±0.016	3.780±0.009
Asplund, Nilsson	3.778±0.060	3.792±0.040	
Hopkins, Diven	3.770±0.031	3.777±0.030	
Manganese bath method			
Axton	3.725±0.019	3.743±0.019	3.750±0.011
De Volpi	3.729±0.030	3.747±0.019	
Bozorgmanesh		3.744±0.023	
White, Axton	3.797±0.040	3.815±0.040	
Aleksandrov		3.747±0.036	
Boron pile method			
Colvin	3.713±0.015	3.739±0.021	
Weighted mean value		3.766±0.007	

It can be seen from Table 1 that there is good agreement between all the values obtained by the manganese bath method except for the results of White and Axton. It may be noted that the absolute fragment measurement techniques in these studies were different. Axton used fragment-fragment coincidence and De Volpi neutron-fragment coincidence counting, while Bozorgmanesh and Aleksandrov employed a small-solid-angle chamber.

An analysis of the data in Table 1 led Smith to the conclusion that there is a systematic error in the manganese bath method. As a possible cause of the fact that the manganese results are 0.5% lower than the weighted mean value of $\bar{\nu}(^{252}Cf)$, Smith considers the thermal neutron absoprtion cross-section of sulphur and suggests that the 30-year-old

measurements of this quantity should be repeated with an accuracy better than 1%. Such measurements will certainly help to eliminate the existing discrepancy in $\bar{\nu}(^{252}Cf)$, since changes in the sulphur cross-section affect the η-values, which in turn would lead to higher values of $\bar{\nu}(^{252}Cf)$, close to those obtained by the liquid-scintillator method.

The method used at the V.G. Khlopin Radium Institute for measuring $\bar{\nu}(^{252}Cf)$ involved separate determinations of the neutron flux and the fission rate of the californium sources. The neutron fluxes were measured with a manganese flow bath, which in 1977 received metrological certification as a working standard of unit flux density in accordance with the All-Union test procedure laid down in GOST 8.031.74.

As in Ref. [2], a small-solid-angle chamber fitted with a surface barrier detector was used to register the fission fragments. The californium sources were prepared by vacuum vaporization onto platinum substrates (t = 0.5 mm).

The fission rate of the californium sources was determined over different solid angles by varying the detector aperture and the distance from the detector to the source. The solid-angle calculations were checked by means of a ^{241}Am reference source, whose absolute activity was measured by an alpha-gamma-coincidence method to an accuracy of 0.1%.

To reduce the contribution to the final error made by self-sputtering of the californium, the sources were kept for several months in the chamber, following its construction, before the measurements were made.

In the reduction of the fission rate results to a single instant of time, a half-life T = 2.638 years was used.

The various components of the error in the determination of the fission rate are given in Table 2.

Table 2

Error in fission rate measurement

Source of error	Random S_x,%	Systematic θ_i,%
1. Counting statistics	±0.05	
2. Linear extrapolation of the "tail" of the fragment energy distribution		±0.2
3. Self-sputtering of californium		±0.2
4. Determination of solid angle		±0.2

The neutron yields of the sources were measured in the manganese flow bath. In the measuring loop, two gamma-gamma flow counters were placed behind a 20-cm lead screen at a distance of 3 m from the bath.

In one of the counters, the ^{56}Mn gamma-photons were detected by means of an FEhU-82 photomultiplier tube with 70 x 70 NaI(Tl) crystals. The other counter had an FEhU-49B tube with 150 x 150 NaI(Tl) crystals. The housing of the manganese bath was temperature controlled. This increased the operational stability of the photomultiplier and electronic equipment and ensured that the hydrodynamic conditions of the pumping system remained constant.

In the calculation of the neutron flux, the following parameter values were used:

$\sigma_S = 0.523 \times 10^{-28}$ m^2 - thermal neutron capture cross-section for sulphur;

$\sigma_{Mn} = 13.35 \times 10^{-28}$ m^2 - thermal neutron capture cross-section for manganese;

$\lambda = 2.5774$ h^{-1} - decay constant of ^{56}Mn;

$\sigma_H/\sigma_{Mn} = 0.02486$ [1] - ratio of the hydrogen and manganese thermal neutron capture cross-sections;

$1 + \alpha = 1.0132$ [3] - correction for resonance neutron absorption by manganese;

$1 + m = 1.00756$ - correction for self-absorption of neutrons in the source cover.

This correction was determined experimentally from a measurement of the thermal neutron flux by means of gold foils in a complete sphere R = 5 cm with the source at the centre;

$1 + \ell = 1.00243$ - correction for neutron leakage from the bath.

The leakage correction was determined experimentally for the given californium source and the working strength of the manganese bath [4];

$1 + \kappa = 1.00414$ - correction for fast neutron capture by oxygen and sulphur nuclei.

This correction was calculated by the Monte Carlo method with the use of nuclear data in a 75-group representation ranging from thermal energies up to 15 MeV. The group cross-sections were obtained by averaging data from the ENDL library. For energies up to 2.5 MeV, averaging over a Fermi spectrum and from 2.5 MeV to 15 MeV over a Watt spectrum was employed. Two reactions were considered in modelling the neutron trajectory: elastic and inelastic scattering; others were allowed for by the introduction of statistical weights. The working strength of the manganese sulphate solution was C = 25.900%.

The errors in determining the neutron flux of the californium sources are given in Table 3.

Table 3: Errors in determining neutron fluxes

Source of error	Random S_x,%	Systematic θ_i,%
1. Measurement of saturated ^{56}Mn activity	±0.1	
2. Determination of coefficient F		±0.2
3. Efficiency of gamma flow counter		±0.11
4. Neutron leakage		±0.01
5. Fast-neutron capture by sulphur and oxygen		±0.08
6. Neutron absorption in source		±0.1
7. Uncertainty in manganese absorption resonance integral		±0.1
8. Chemical purity of bath		±0.1
9. Half-life of ^{252}Cf		±0.15

The total error in determining $\bar{\nu}(^{252}Cf)$ was calculated from the equation

$$S_\Sigma = \sqrt{S_x^2 + \left(\frac{1}{3}\Sigma\theta_i\right)^2}$$

i.e. the systematic errors were added arithmetically and the standard deviation of the combination of systematic errors was taken equal to 1/3 ΣQ_i; S_x is the standard deviation of the average value of the measured quantity.

The mean value obtained for the total number of neutrons per fission event in californium-252 was 3.758 ± 0.015.

REFERENCES

[1] Smith J.R.-EPRI. NP-1252 Research Project 704-4.

[2] ALEKSANDROV, B.M. et al., Neutron Physics [in Russian], Part 5 (1976) 166.

[3] Axton E.J.,Ryves T.B.-J.Nucl.Energy,1967,v.21,p.543.

[4] SOROKINA, A.V., SHLYAMIN, Eh.A., et al., At. Ehnerg. 45 6 (1978) 439.