of a three-phonon state. That is,

$$\frac{B(E2\colon (3,4)+\to 4+)}{B(E2\colon (3,4)+\to 2'+)}=2.3\pm 0.4,$$

$$\frac{B(E2\colon (3,4)+\to 2'+)}{B(E2\colon (3,4)+\to 2+)}=69.8\pm 6.8.$$

The level at 2112 keV could be the 6+ member of the quintet.

The level structure of ^{132}Xe has been compared[3] with the levels predicted by various nuclear models[26–31] that have been developed to account for the properties of the low-lying excited states. These models only show some qualitative agreement. In later comparison with the model of Mallman,[31] Suarez and De Arsenberg[32] note that the agreement is so poor that even-even nuclei probably behave neither as a rigid rotor nor as a liquid-drop-plus-β vibration nor anything in between. Perhaps a better understanding of spherical even-even nuclei can be obtained if additional information such as presented here concerning the higher-energy states can be obtained. This information can be used to establish accurate and detailed nuclear systematics. With the large-volume high-resolution Ge(Li) detectors now available, it is possible for γ-γ directional-correlation experiments to be performed to determine additional information such as the spins of the high-energy excited states and mixing ratios of the deexciting transitions that can be compared with nuclear models. Such information would be very valuable to a more general understanding of the level structure of even-even nuclei. Indeed, this information is vital to determine which, if any, systematic trends such as explored by Sheline[33] and Sakai[34] exist in all mass ranges.

ACKNOWLEDGMENTS

We wish to thank Dr. R. Henck for private communication of his results and for helpful comments. We also wish to thank Dr. N. R. Johnson and Dr. A. V. Ramayya for helpful comments.

---

[26] G. Scharff-Goldhaber and J. Weneser, Phys. Rev. **98**, 212 (1955).

[27] L. Wilets and M. Jean, Phys. Rev. **102**, 788 (1956).

[28] A. S. Davydov and A. A. Chaban, Nucl. Phys. **20**, 499 (1960).

[29] A. S. Davydov and G. F. Filippov, Nucl. Phys. **8**, 237 (1958).

[30] B. J. Raz, Phys. Rev. **114**, 1116 (1959).

[31] C. A. Mallman, Nucl. Phys. **24**, 535 (1961).

[32] J. F. Suarez and E. Y. De Arsenberg, Nucl. Phys. **A90**, 449 (1967).

[33] R. K. Sheline, Rev. Mod. Phys. **32**, 1 (1960).

[34] M. Sakai, Nucl. Phys. **104**, 301 (1967).

PHYSICAL REVIEW C VOLUME 1, NUMBER 2 FEBRUARY 1970

Neutron Yield of ^{252}Cf Based on Absolute Measurements of the Neutron Rate and Fission Rate*

A. De Volpi and K. G. Porges
Argonne National Laboratory, Argonne, Illinois 60439

(Received 2 June 1969)

The need for accurate measurements of the total yield of neutrons (ν) in spontaneous fission of ^{252}Cf has continued. Such a measurement, incorporating advances over previous work, has been accomplished with a high degree of authentication. The yield was determined by separate direct evaluations of the neutron rate and fission rate of three fission-fragment counters plated with ^{252}Cf. The neutron emission-rate factor in the yield was found with the aid of a manganese-bath facility independently calibrated with reference to absolute β-γ coincidence counting of ^{56}Mn. Special measurements were also made of other subsidiary parameters, while precise support in the accuracy of neutron assay was obtained from comparisons with international standard sources. The fission rate was derived primarily from absolute fission-neutron coincidence data obtained from the fission counters, with special attention to angular-correlation effects. The resulting value of $\nu(^{252}\mathrm{Cf})=3.725\pm 0.015$ (total yield) is generally independent of cross sections, neutron spectrum assumptions, counting statistics, and fission-fragment self-absorption. The error assigned is a combination of statistical and systematic effects at a confidence level of 68%. Two unverified correction factors have been isolated.

I. INTRODUCTION

CALIFORNIUM-252 is a relatively long-lived isotope with significant spontaneous-fission specific activity (about two-thirds of a million fissions per second per microgram). These properties of high specific activity and long life—coupled with increasing availability—have allowed this isotope to become a standard with regard to measurements of the average number of total neutrons emitted per fission (ν).

That such a standard is necessary at present can be

---

* Work performed under the auspices of the U.S. Atomic Energy Commission.

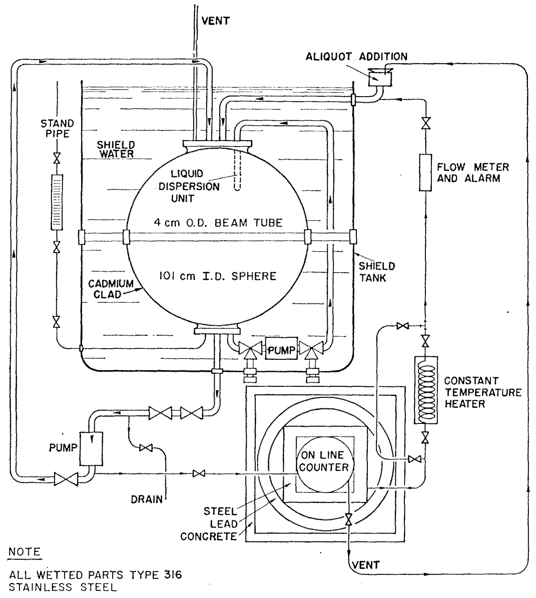


FIG. 1. Schematic of circulation system employed to mix and move the bath solution into the on-line counter.

deduced from the large spread of ν values[1] determined for fissile materials in a reactor- or accelerator-generated background environment.

But even $\nu(^{252}\mathrm{Cf})$ has proven to be difficult to determine accurately. The reasons for this impedance appear to lie with the rather complex steps required to make absolute measurements of high-energy neutron rates and perhaps also in problems associated with accurate fission-rate determinations.

The various experimental techniques directed towards absolute measurements of $\nu(^{252}\mathrm{Cf})$ were outlined at the Second Conference on Cross Sections and Technology.[2] Essentially, there have been two types of measurements: those classified as "gated" measurements in which the mean product $\langle\nu_p\epsilon_n\rangle_{\mathrm{av}}$ is found in a coincidence experiment (the neutron detector efficiency ϵ_n being independently evaluated), and "ungated" experiments in which there is a separate determination of the fission rate and the neutron rate.

Some of the difficulties connected with both techniques were also discussed at the Washington Conference.[2] There has been a prevailing gulf of about 2% which so far has defied coherent explanation.

In this article we report a direct ungated measurement of $\nu(^{252}\mathrm{Cf})$, making use of absolute neutron detection with the manganese bath,[3] and absolute fission-counter calibration by a coincidence method. The essentials of both of these features have already been published[4]; so, aside from an outline of the methods, we shall emphasize primarily changes or supplementary techniques.

Briefly, our experimental procedure is as follows: A fission counter is fabricated with a californium sample yielding close to 10^6 neutrons/sec deposited on one electrode. The neutron emission rate is found by inserting the counter in the manganese bath. The bath has been cross-correlated with other absolute neutron standards, and extensive work has been done to verify its accuracy. Next, the counter is placed in a position where the fission-fragment rate can be counted while the neutron-emission rate is observed by a prompt-neutron detector. Coincidences between these two detectors are recorded as a function of angle between the fission counter normal and the neutron detector axis. From this information the fission rate is deduced. These bilateral procedures have been carried out for three fission counters.

A major goal of this series of experiments has been high accuracy—approaching $\frac{1}{2}\%$. To achieve this in terms of combined accuracy and precision, it has been necessary first to reduce effects which produce random errors and second to authenticate nearly every aspect which is known to lead to systematic error.

Particular emphasis has been reliance on absolute techniques: that is, procedures which do not directly depend on cross-section values, precise knowledge of neutron spectra, or detailed understanding of self-absorption corrections.

II. NEUTRON RATE

The manganese-bath method is the one technique for measurement of neutron rate which, so far, has been extensively evaluated and is subject to convenient application, stable operation, and cross checking by exchange of sources. Present-day versions of the bath, such as the one used in this experiment, consist of (1) a sphere filled with an aqueous solution of manganous sulfate; the radius of the tank is sufficient to contain nearly all high-energy neutrons originating from the source; (2) an on-line counter through which the activated solution is continuously circulated, and (3) related equipment for absolute calibration of the system by coincidence counting.

Figure 1 schematically indicates the circulation system employed to mix and move the liquid into the on-line counter. Figure 2 is a cutaway view of the on-line counter. The detection element is an annular sodium-iodide crystal viewed by a bialkali photomultiplier tube attached to one end.

Relatively high efficiency and stable operation are attained by choosing a bias level just above noise—equivalent to perhaps 50 keV of deposited energy from

---

[1] C. H. Westcott *et al.*, At. Energy Rev. **3**, 3 (1965); G. C. Hanna *et al.*, *ibid.* **8**, 3 (1969).

[2] A. De Volpi, in Second Conference on Neutron Cross Sections and Technology, Nat. Bur. Std. Special Publ. **299**, 183 (1968).

[3] K. W. Geiger, Metrologia **4**, 8 (1968).

[4] A. De Volpi and K. G. Porges, in *Proceedings of Conference on Nuclear Data for Reactors* (International Atomic Energy Agency, Vienna, 1967), Vol. I, p. 297.

TABLE I. Summary of neutron source emission rates.

Source	Type	ANL weighted mean[a]	Statistical error (%)	Systematic error (%)	Total error (%)	Reference date
1. NBS II	Ra Be(γ, n)	1.1776	0.09	0.28	0.37	1/1/65
2. ANL	Ra Be(γ, n)	1.1623	0.07	0.28	0.35	8/29/68
3. ^{252}Cf No. 0	Spontaneous fission	1.7929[b]	0.07	0.28	0.35	3/10/64
4. ANL	Ra Be(α, n)	5.398	0.06	0.29	0.35	3/10/64
5. NRC	Ra Be(α, n)	3.223	0.15	0.29	0.44	1/1/62
6. NPL	Am Be(α, n)	2.569	0.10	0.38	0.48	1/1/68
7. ^{252}Cf No. 1[c]	Spontaneous fission	0.5050[b]	0.20	0.28	0.48	7/27/65
8. ^{252}Cf No. 2[c]	Spontaneous fission	0.7220[b]	0.12	0.28	0.40	10/15/65
9. ^{252}Cf No. 3[c]	Spontaneous fission	0.6786[b]	0.10	0.28	0.38	4/6/67

[a] 10^6 neutrons/sec.

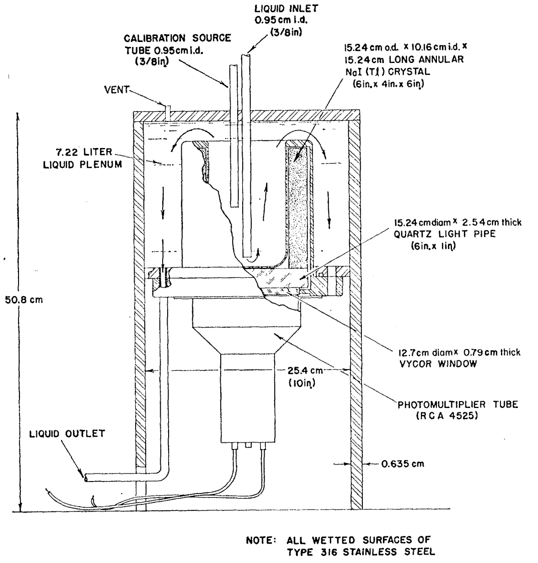
[b] These numbers are 0.4% less than reported in Ref. 9 because of recent (unpublished) integral experiments.

[c] Fission counter.

the ^{56}Mn γ rays. Through this mode of continuous monitoring, counting statistics are no longer a limiting problem in the calibration of neutron sources which have emission rates exceeding 10^4/sec. Additional information concerning the manganese bath apparatus may be found elsewhere.[5]

Independent absolute calibration of the manganese bath is contingent upon capability for accurate assessment of ^{56}Mn activity. This is accomplished at our laboratory by β-γ coincidence detection of aliquots introduced into liquid scintillators.[6] Since the β-detection efficiency is in the order of 98%, a correction of only 0.2% is needed for conversion of the analyzed data to the absolute-disintegration rates.[7]

In principle, calibration of the bath can be accomplished by borrowing a reference neutron source which has been calibrated on an international scale. However, for the accuracy required in the entire measurement of $\nu(^{252}\text{Cf})$, the quoted international average of 0.9%[8] has been inadequate. Moreover, there were indications of possible systematic errors in source calibration.



NOTE: ALL WETTED SURFACES OF TYPE 316 STAINLESS STEEL

FIG. 2. Cutaway view of the on-line counter.

Consequently, we recently completed a thorough program of neutron-source evaluation and comparison, the results of which are being published separately.[9] By gauging the neutron outputs of four sources that have a wide range of emission spectra, some plausible causes of the existing world-wide discrepancies have been uncovered. We have concluded, on the basis of the data and on the basis of comparisons with the international standard source, that a combined accuracy and precision of less than 0.4% can be claimed for our calibrations of neutron-emission rates.

Pertinent results from the neutron-source calibration program are repeated here. Table I contains the summary of all neutron-emission rates; the evaluation of these source strengths is based on a large number of calibrations spread over a wide range of manganese-sulfate concentrations. For example, the three fission counters which have been used to determine $\nu(^{252}\text{Cf})$ have been tested for neutron rate a combined total of 33 times. Figure 3 indicates the extent to which our measurement of the international standard Ra-Be(α, n) source N-200-1 agrees with world-wide measurements.

Hidden in the apparent superficial agreement, however, is a clear systematic difference in an important correction for high-energy neutron absorption in the sulfur and oxygen contained in the bath. The corrections we have deduced from experiment are about 1.2% lower for the NRC standard and about 0.6% lower for a ^{252}Cf source than those commonly applied. If the various international sources are normalized to our correction factors, then the less satisfying picture shown in Fig. 4 is obtained. Table II lists the results we have found and compares them with other measurements, particularly the evaluations at the National Physical Laboratory of the United Kingdom.

---

[5] A. De Volpi and K. G. Porges, in Second Conference on Neutron Cross Sections and Technology, Natl. Bur. Std. Special Publ. **299**, 213 (1968).

[6] A. De Volpi, K. G. Porges, and R. Armani, in *Proceedings of Symposium on Standardization of Radionuclides* (International Atomic Energy Agency, Vienna, 1967), p. 717.

[7] A. P. Baerg, Metrologia **2**, 23 (1966).

[8] V. Naggiar, Metrologia **3**, 51 (1967).

[9] A. De Volpi and K. G. Porges, Metrologia **5**, 128 (1969).

TABLE II. Comparison of results[a] from sources jointly calibrated at ANL and NPL.

Source	Type	ANL	±	NPL	±	Other values	±	Reference date
NBS II	Ra Be(γ, n)	1.1776	0.0043	1.172	0.006	1.177[b]	0.012	1/1/65
NRC	Ra Be(α, n)	3.223	0.014	3.197	0.030	3.230[c]	0.030	1/1/62
NPL	Am Be(α, n)	2.569	0.012	2.529[d]	0.018			1/1/68

[a] 10^6 neutrons/sec.

[b] NBS Reports [V. Spiegel, National Bureau of Standards (private communication)].

[c] International average (Ref. 8).

[d] Unpublished value (not final) [E. J. Axton, National Physical Laboratory (private communication)].

Note: The ANL values would be 1.2% higher for the NRC source if the relatively high fast-neutron absorption correction used at NPL were applied to our data as well.

It should be mentioned at this point that not only have we directly determined the controversial correction factors from concentration experiments with four sources of varying emission spectra, but our results for high-energy neutron absorption are in good agreement with calculated values based on the latest cross sections for sulfur and oxygen[10] as well as unpublished transport-code computations conducted at Argonne National Laboratory. In any event, there is a direct connection between our neutron-source emission rates and those of the world standard with which we are in concord, although by a different combination of corrections.

Another outcome of the neutron source determinations has been a determination of the ^{252}Cf half-life by neutron counting of a sealed source. We find a half-life of 2.621±0.006 yr,[11,12] which is 1% less than a measurement done by fission-fragment counting.[13] Although errors are much larger, the half-lives by neutron counting of the fission chambers are in agreement with the sealed source. This verification also tends to exclude the possibility of a significant contamination from some isotope with a neutron yield markedly different from that of ^{252}Cf. Nearly simultaneous evaluation of the fission and neutron-emission rates eliminates the need for precise knowledge of the disintegration constant.

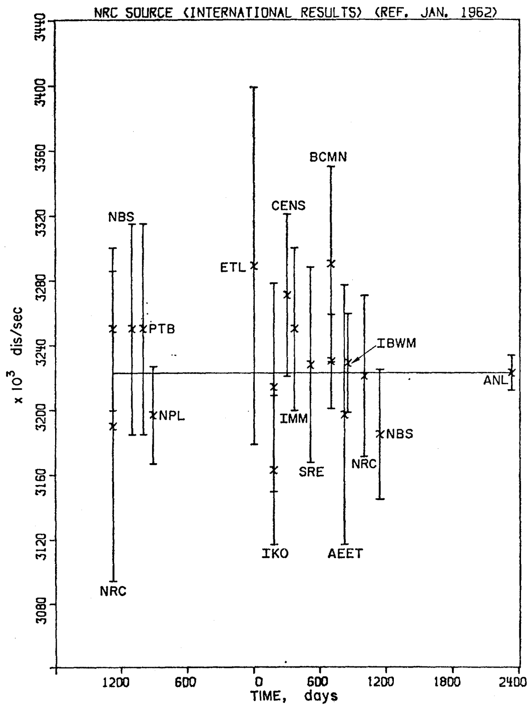


FIG. 3. Independent calibrations of the NRC International Standard Source.

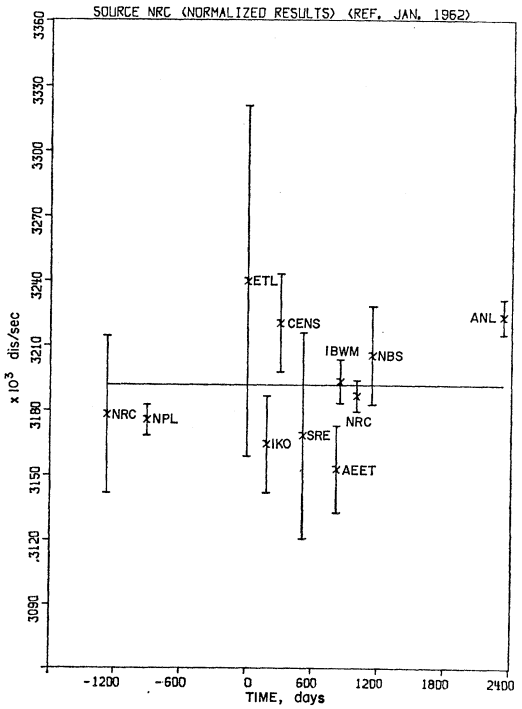


FIG. 4. Calibrations of the NRC source normalized to a "low" set of fast-neutron corrections.

---

[10] P. W. F. Louwrier, thesis, University of Amsterdam, 1966 (unpublished).

[11] A. De Volpi and K. G. Porges, Inorg. Nucl. Chem. Letters **5**, 111 (1969).

[12] A. De Volpi and K. G. Porges, Inorg. Nucl. Chem. Letters **5**, 699 (1969).

[13] D. Metta *et al.*, J. Inorg. Nucl. Chem. **27**, 33 (1965).

III. FISSION RATE

A. Fission Fragment Detectors

The evaluation of the spontaneous-neutron rate from three fission counters has been outlined; now we turn to the problems associated with measurement of their contemporary fission rate. The rather long and well-known total half-life of ^{252}Cf allows timewise separation of these two objectives without the intercession of a monitor or any necessity for stabilization of the electronic chain that processes fission pulses.

Each of our californium sources has been deposited on one electrode of a 2π fission-fragment counter. Two of these counters were described in connection with an earlier report on their neutron yield.[4]

All three counters have the following characteristics in common: The active region of the counter is at one end of a sealed tube over 100 cm long, which permits insertion of the entire counter into the center of the manganese-bath spheres. Each counter operates in the current-pulse mode,[14] requiring plate spacings in the order of 1–2 mm and a methane filling of about 2 atm.

The counters differ in the following respects: No. 1 has a solution-evaporated deposit placed on a disk mounted perpendicular to the axis of the counter; its efficiency is rather poor, being about 60%. Counter No. 2 has hemispherical electrodes, with an efficiency approaching 90%; however, the coating—also evaporated from solution—is not uniform, but appears to have accumulated primarily to one side of the hemisphere. Our newest counter, No. 3, has a vacuum-deposited foil yielding over 99% fission-fragment efficiency; its plane electrodes have been arranged parallel to the fission-counter axis.

B. Coincidence Calibration of Fission Rate

Our primary approach for appraisal of the absolute fission rate is based on the coincidence method.[15] Considering the various forms in which energy is released in fission, the nearly simultaneous emission of fragments and neutrons offers the best opportunity for unambiguous satisfaction of the requirements for absolute coincidence counting, namely, high efficiency for at least one detector and penetrating radiation viewed by the second detector—coupled with a one-to-one relationship in "decay" scheme.

The fission-fragment ionization detector, operating with relatively low deadtime (300 nsec), delivers a fast signal to one leg of the coincidence gate. The other signal is developed in a Hornyak-button[16] fast-neutron detector. The bias level for the neutron channel is almost unavoidably above 1.5 MeV in this type of detection. Since a negligible number of delayed neutrons have energies greater than 1 MeV, only prompt neutrons are observed in the count, a fact which has been directly verified in two ways. With a neutron source having a 1-MeV upper neutron limit, no significant increase in background was noted. Also, the coincidence-timing resolution was used to deduce the maximum time of flight of neutrons allowed to produce pulses above the electronic bias, thereby providing a measure of energy cutoff.

Consequently, we may write in simplified form the fission-channel count rate F (after corrections for deadtime) as

$$F=\epsilon_f S, \tag{1}$$

where ϵ_f is the fragment-detection efficiency, and S is the actual fission rate.

The corrected neutron-channel count rate N is based on the sum over the multiplicity distribution:

$$N=\sum_{\nu}\left[1-(1-\epsilon_\nu)_\nu\right]P(\nu)\,S \tag{2}$$

for ν neutrons with probability distribution $P(\nu)$ and detection efficiency per neutron of ϵ_ν.

If the efficiency of the detector is small and approximately uniform over the prompt-neutron energy range ($\epsilon_\nu\equiv\epsilon_n$), then the limiting value of the neutron-count rate rapidly approaches

$$N=\nu_p\epsilon_n S, \tag{3}$$

provided only prompt neutrons are detected.

The coincidence rate, after proper corrections for deadtimes and accidentals, becomes under these conditions

$$C=\nu_p\epsilon_n\epsilon_f S, \tag{4}$$

and the fission-source rate develops as

$$S=NF/C. \tag{5}$$

Computations of the fission rate are accomplished through the program COINC[17] which is fashioned after derivations by Gandy.[18]

C. Electronic Elements of Fission Coincidence Calibration

Specific features of the electronic system used for coincidence calibration may be explained with the aid of Fig. 5.

The fission counter, operated in the current-pulse mode, produces narrow pulses of about 20 nsec width. The amplitude of these pulses may be maximized by choosing an E/p (electric-field/gas-pressure) value corresponding to the peak-drift velocity for methane. Figure 6, in fact, is the drift-velocity distribution ob-

---

[14] A. De Volpi, K. G. Porges, and C. Rush, Bull. Am. Phys. Soc. **9**, 46 (1964).

[15] K. G. Porges and A. De Volpi, in *Proceedings of Symposium on Standization of Radionuclides* (International Atomic Energy Agency, Vienna, 1967), p. 693.

[16] W. F. Hornyak, Rev. Sci. Instr. **23**, 264 (1962).

[17] A. De Volpi, K. G. Porges, and G. Jensen, Intern. J. Appl. Radiation Isotopes **17**, 277 (1966).

[18] A. Gandy, Intern. J. Appl. Radiation Isotopes **13**, 501 (1962).

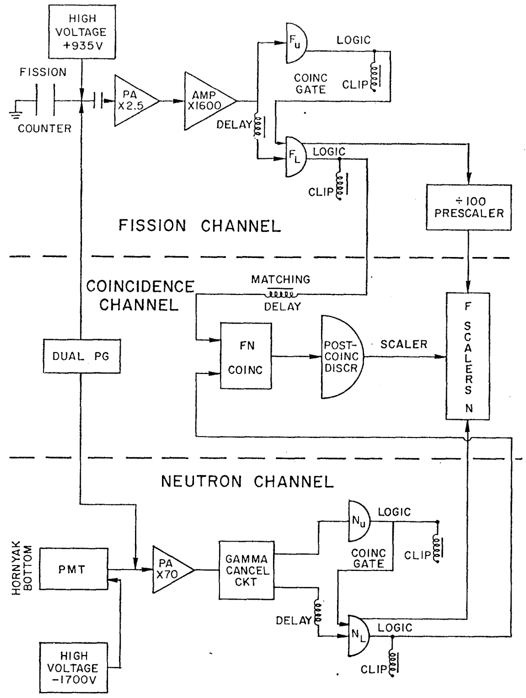


FIG. 5. Fission-counter calibration system.

tained using fission fragments in an ionization counter in which both the interelectrode spacing and gas pressure could be varied. At 2 atm, less than 1000 V is necessary to get good pulse-height distribution for a plate separation of about 2 mm.

Because of the small spacing and low pressure, α pileup is not observed above the intrinsic noise level of the preamplifier. In fact, in the current-pulse mode of counting, very high α rates can be tolerated without interference to fission fragments.

A low-noise grounded-grid hybrid preamplifier, in a configuration analogous to that developed by Rush,[19] conveys the current pulse (with a gain of 2.5) from the parallel-plate fission counter to the main amplifier. This preamplifier uses a ceramic triode for the first active element. Input load of the tube is about 25 Ω, which satisfies the low impedance requirements of current pulses. The input-noise level of the vacuum tube is substantially lower than that obtainable from grounded-base junction transistors. In addition, the tube is quite immune to voltage discharges at the input.

A current pulse amplifier designed by Rush[19] increases the signal level to an amplitude sufficient to trigger the fast discriminators, which require at least 50 mV into 50 Ω. Even though the rise time at this point is about 3 nsec, the time jitter in triggering level of the tunnel-diode discriminator is large enough (over 10 nsec) to require some compensation. This is accomplished by splitting the input signal and feeding two discriminators. The upper unit (marked F_U) triggers at the desired pulse height, but the lower unit (F_L) is set at a lower bias level. By gating F_L with the output of F_U, the natural time walk of F_U is bypassed. (The output pulse from F_L is a narrow pulse delayed to fit within the wider span of the F_U pulse.) Outputs from this discriminator pair go to the fission-fragment scaler and, after clipping and delay, to the fission-neutron (FN) coincidence analyzer. (All triggering units used are adjustable paralysis discriminators (APD)[20] that have versatile provisions for fast and slow outputs, adjustable pulse width, precise paralysis, and external gating.)

On the neutron side, fast neutrons from fission are detected in a 12.5-cm diameter "Hornyak button"[16] coupled to a bialkali photomultiplier tube. The electrical signals thus generated are fed to a current-mode preamplifier to obtain a nonsaturated output ranging up to 10 V. These linear pulses go into a γ-ray cancellation circuit[21] that reduces the amplitude of narrow γ-ray and noise pulses while preserving the amplitude and risetime of neutron-scintillation events. The output of the cancellation circuit is also split so as to drive a pair of APD discriminators in antiwalk arrangement.

Coincidence resolution in the order of 50–200 nsec has been utilized for various phases of the measurement. A post-coincidence discriminator reshapes the overlap pulse in order to drive the following scaler. With the aid of a dual-pulse generator, the inherent coincidence resolution can be measured.

Delayed coincidence curves taken with the ^{252}Cf fission counters in position near the Hornyak-button detector demonstrate the requisite matching delay. By observing the extent to which rounded edges and a tail develop as a function of neutron bias, it is possible

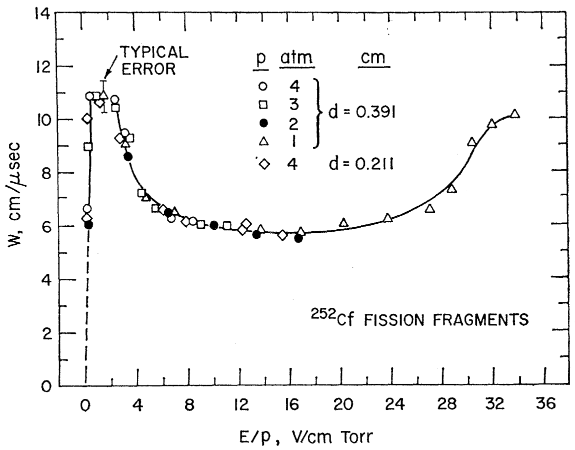


FIG. 6. Drift velocity in methane determined as a function of plate spacing, pressure, and voltage applied to a parallel-plate ionization counter using ^{252}Cf fission fragments.

---

[19] C. Rush, Rev. Sci. Instr. **35**, 149 (1964).

[20] A. De Volpi *et al.*, Rev. Sci. Instr. **37**, 1592 (1966).

[21] K. G. Proges, A. De Volpi, and P. Polinski, Rev. Sci. Instr. **35**, 1602 (1964).

to determine the neutron-discrimination level required to exclude delayed, slow, and scattered neutrons from the neutron (and coincidence) count.

Various tests have been made regularly of coincidence efficiency, scaler response to known pulse rates, coincidence discrimination against single-channel feedthrough, background, and other relevant factors.

For the fission channel, a deadtime in the order of 300 nsec has been sufficient. However, in the neutron channel, to prevent afterpulses from triggering the discriminator, a 17-μsec deadtime has been applied. All deadtimes are common to both the scaler and coincidence-circuit inputs. Recovery of circuit elements is possible immediately after cessation of the deadtime because a dominating paralysis that exceeds the actual driving-pulse width is applied to each channel.

D. Analysis of Fission-Coincidence Angular Distribution

Several approaches have been tried in analyzing the angular distribution which results from recording the coincidence rate with a neutron detector having less than 4π solid-angle and a fission-fragment detector not having 100% efficiency.

A simplified analytical method,[15] based on Legendre expansions up to the P_4 term, has been our basis for evaluation of the true spontaneous-fission rate. Despite some difficulties in application of the analysis to the actual measurement conditions, this has been the primary procedure for estimation of the average fission rate. Upon drawing a smooth curve through the observed distribution, the average value is taken by summing the fission rates at four nodes (±37.4° and 79.2°) and dividing by four. Sensitivity to the absolute angle is small. The averaging process has the largest uncertainty for the fission counters with the lowest efficiency; accordingly, the relevant error estimates reflect both the imprecision and the likelihood of systematic error.

A more rigorous analysis of the data taken for fission counter No. 3 will be published separately. The design of this fission counter and the quality of its coating have permitted an extensive evaluation of the full angular distribution at two neutron-detector solid angles, as well as the expected front/back asymmetry produced by range differences between light and heavy fragments.

Some real features of the angular traverses are difficult to treat analytically without artificial assumptions. The same limitations are true of a digital-computer simulation of the experiment, but the pragmatic assumptions can be of a different kind. Such a simulation was conducted, resulting in production of the distributions shown in Fig. 7. Only two empirical bounds were placed on the distribution: that the fission-fragment efficiency matched that determined for the fission counter and that the anisotropy range (maximum less minimum) of the angular distribution was the same as that observed experimentally.

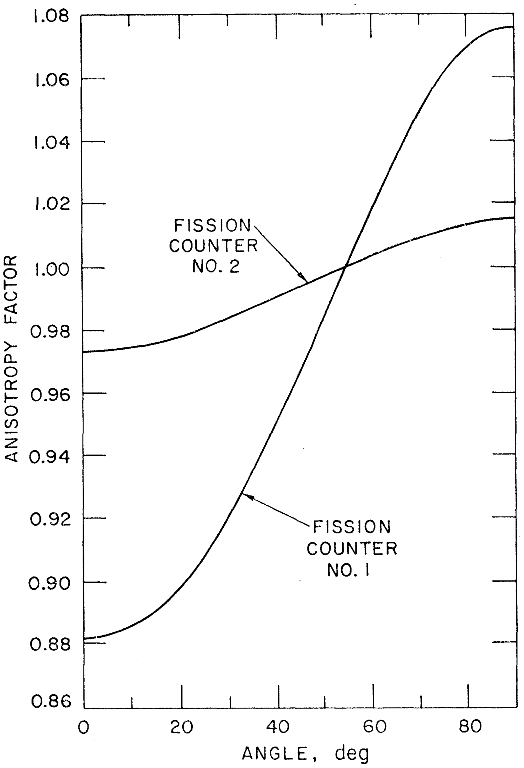


FIG. 7. Fission-rate anisotropy (computer simulation).

Fragments were allowed to have all orientations, and all angles between the neutrons and their fragment precursors were permitted. The Legendre expansion was truncated at the P_4 term, with the ratio of coefficients based on published data[22]; as mentioned, the magnitude of the coefficients was adjusted according to the observed distribution. Departure from unity of the average values determined at the four nodes was taken to be the measure of anisotropy.

The computer simulation verified suspicions that, for a fission counter with low efficiency, it is difficult to analytically treat a nonideal mechanical situation. The simulation offered a correction factor of 0.65% for the analytical method applied to fission counter No. 1 (which has 59% efficiency and an anisotropy range of 23%). For fission counter No. 2 (with 90% efficiency and only 4.4% anisotropy), the correction factor already vanishes, so fission counter No. 3 (99.5% efficiency, 1% anisotropy) can be considered well within the bounds of the analytical treatment.

E. Accumulated Data

Each of the three fission counters has been independently calibrated, applying more than one method where possible. A major advantage of the coincidence aspect of calibration is that by constraining unavoidable recoil aggregates to the general vicinity of the ionization region, the resulting change in geometry should

---

[22] H. R. Bowman *et al.*, Phys. Rev. **129**, 2133 (1963).

TABLE III. Fission rate angular distributions.

Counter	Date	Distance (cm)	Fission rate[a]				Average[b]	Estimated random error	Estimated systematic error
			−37.4°	+37.4°	−79.2°	+79.2°			
1	1966	5.1	1.230	1.350	1.480	1.440	1.377	±0.002	
	1967	5.1	1.245	1.380	1.460	1.400	1.376	±0.002	
	Average of both data sets:						1.377	±0.002	±0.011
	Adjustment for anisotropy:						1.007		±0.005
	Net fission rate:						1.367	±0.002	±0.012
2	1966	4.8	1.940	1.963	1.915	1.923	1.936	±0.002	
	1967	4.8	1.956	1.917	1.914	1.910	1.930	±0.002	
	Average of both data sets:						1.933	±0.004	±0.004
	Adjustment for anisotropy:						1.000		±0.001
	Net fission rate:						1.933	±0.004	±0.004
	Fission rate extrapolated from solid angle data[c]:						1.947	±0.020	±0.010
	Weighted mean:						1.934	±0.004	±0.004
3	1967	3.2	1.8213	1.8164	1.8258	1.8258	1.8223		
		5.2	1.8162	1.8150	1.8279	1.8263	1.8213		
	Average of both distances:						1.8218		

[a] 10^5 fissions/sec.

[b] Adjusted to $\lambda = 7.23 \times 10^{-5}$ day^{-1}.

[c] Reference 15.

not affect, to a good approximation, the distintegration rate determined by coincidence counting.

Counter No. 1

This is our earliest fission counter and the one with the poorest efficiency. In addition, the rate of aggregate recoil loss was sufficient to reduce the fragment-counting efficiency from an initial value of nearly 70% to a final value of under 60% in the $1\frac{1}{2}$ yr over which the counter was calibrated.

The first calibration, previously reported,[15] is based on the selection of two nodes from the angular distribution obtained by sampling about the axis of the fission counter, each sample being an average over axial rotation of the fission counter. The latter rotation compensates for a nonuniform coating on the plane of the fission foil, while data taken at different azimuthal angles with respect to the foil normal provides a basis for integration over the neutron-emission anisotropy. Figure 8 contains the resulting distribution.

A second calibration was done approximately a year later, with results also depicted in Fig. 8. Again the nodes at ±37.4 and ±79.2° were chosen as a basis for evaluation of the fission rate. A summary of the values read from the graphs is given in Table III, along with a correction for anisotropy derived from a computer simulation of the measurement.

The failure of the minima and maxima to correspond to the mechanical orientation of the foil and Hornyak button, as well as their symmetry reflection in the two measurements, is a feature that we do not understand. Accordingly, it has been necessary to downgrade the fission-rate calibration of counter No. 1 by associating a relatively large systematic error.

Counter No. 2

Although the second counter has hemispherical electrodes intended to provide a physical integration of the anisotropy, there remains a 4% effect due to the unevenness of the deposit. Efficiency for this counter began at about 90%, eventually deteriorating to almost 80% during two years of observation.

The primary calibrations of this fission counter come from two angular traverses, shown in Fig. 8. It has been necessary to allow a systematic error for uncertainty in application of the analysis to this geometry. The derived averages are listed in Table III.

A second approach to calibration, based on increasing separation of the neutron detector from the fission foil, is included in Table III. Difficulties arise in this type of measurement because of competition between the need for narrow coincidence gates to keep a low accidental level versus wide coincidence gates required to exceed the finite time of flight of the neutrons at large distances. In addition, the limitations of determining the proper solid angle for a spatially finite source and a somewhat indeterminate detector also detract from the confidence that can be attributed to this type of calibration.

Because of the relatively small angular extremes of the traversal data, the computer simulation indicates (Fig. 7) that the correction for anisotropy is less than 0.1% for the second counter.

Counter No. 3

Fission counter No. 3 is the primary basis for our results for $\nu(^{252}\text{Cf})$. Although the neutron-emission rates for each counter have been similar, with relatively small attendant errors, the quality of the fission-rate determination varies strongly for the three counters. The efficiency of counter No. 3 is over 99%, and this fact greatly minimizes possibilities for systematic error. In addition, three types of calibration were carried out for this counter, although almost exclusive weight is associated with the angular traverses, as with the other fission counters.

The apparently carrier-free deposit obtained from vacuum evaporation was initially subjected to defined small solid-angle counting. Unfortunately, insufficient (only 0.4%) counting statistics were obtained before the foil was sealed in its ionization counter. The total fission rate was determined to be $1.813 \pm 0.007 \times 10^5$ dis/sec using a geometric efficiency of nearly 10^{-5}. An α/fission ratio of 31.9 ± 0.8 was found that was in general agreement with the value of 31.0 ± 0.5 by Horrocks.[23] The α-emission rate from ^{250}Cf was observed to be less than 2% and that from ^{254}Cf to be negligible.

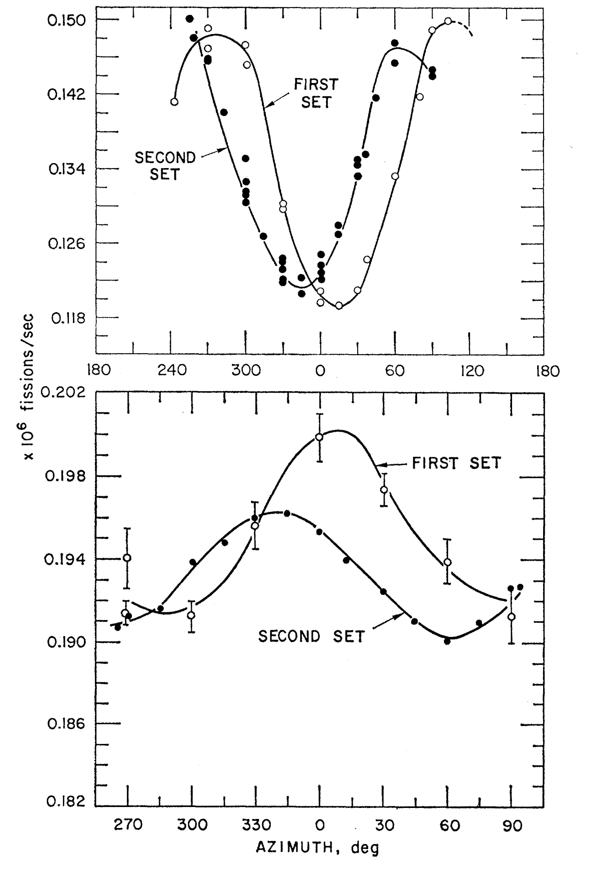


FIG. 8. Fission-rate angular distributions for Counters Nos. 1 and 2. Each data set was taken about a year apart.

Angular traverses for counter No. 3 were performed in a way different than for the other two chambers. Because the foil was mounted parallel to the counter-tube axis, it became possible to measure the front and back asymmetry which arises from differences in range between light and heavy fragments. Figure 9 portrays, and Table III records, data taken for two distances between the foil and the Hornyak-button detector. Since the extreme fission rates only differ by about 1% over the entire angular span, there is a high degree of confidence in the ultimate averaging process.

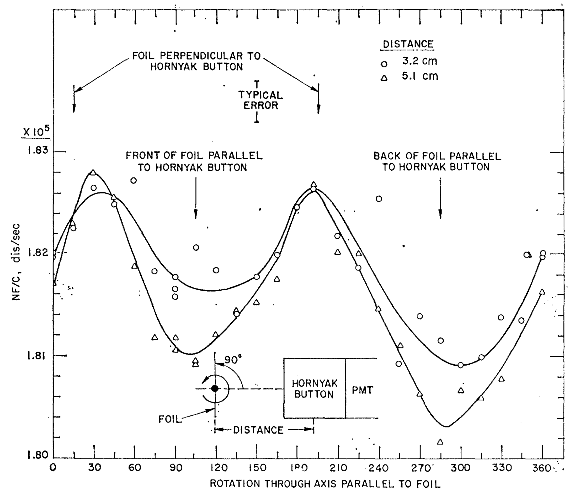


FIG. 9. Fission rate angular distributions for Counter No. 3.

Additional tests have been made to prove the independence of the fission rate on the fission-counter bias (Fig. 10) and on the neutron discrimination level (Fig. 10). Dependence on neutron bias could arise from delayed neutrons, from improper coincidence centering, and from γ sensitivity of the neutron detector. Measurements made as a function of distance between the fission foil and the neutron detector also extrapolate to a

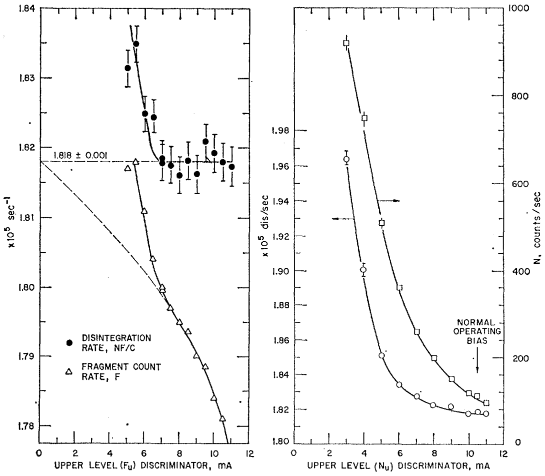


FIG. 10. Dependence on fission fragment and neutron bias (^{252}Cf Fission Counter No. 3).

---

[23] D. Horrocks, Phys. Rev. **134,** B1219 (1964).

TABLE IV. ^{252}Cf No. 3—Summary of fission rate determinations.

Method	Orientation	Value	Adjusted value	Estimated total error
Angular distribution[a]	all	1.8218	1.8218	0.0024
Function of solid angle[b]	90° and 270°	1.828	1.828	0.0040
Defined solid angle counting[c]	90°	1.813	1.813	0.00070
Function of fragment bias[d]	90°	1.818	1.822	0.0050
Fission fragment extrapolation[e]	90°	1.818	1.822	0.0500
Selected value:			1.8218	0.0024

[a] Table III.
[b] Figure 4.
[c] Text.
[d] Figure 10.
[e] Figure 11.

consistent fission rate. In Fig. 11, the neutron rate was decreased by moving the neutron detector away from the fission counter. All fission-rate determinations for californium fission counter No. 3 are summarized in Table IV. The combined results for the three fission samples may be found in Table V.

IV. SYSTEMATIC ERRORS

Systematic errors may be considered to enter the data in three ways: with the neutron assay, with the fission-rate calibration, or as errors common to both phases of the measurement. (Errors attributed to the neutron measurement have already been examined in a separate paper.[9])

Estimates of systematic errors in the fission-counter calibration (Table VI) have been devised according to the influence of the various correction factors upon the measured rate and the known uncertainty connected with these parameters. None of the correction factors exceed 1%.

The most difficult errors to estimate are those connected with integration of the angular traverses. Two features of those measurements produce unaccountable lattitude. The first of these is the correction for anisotropy which was appended to counter No. 1. Knowledge of the correct neutron correlation is rather limited; in addition, application of this knowledge through the truncated Legendre expansion cannot be given too much confidence. The magnitude of the anisotropy correction is thus a basis for estimation of this error. The other element of the calibration, which lacks adequate explanation, is the failure of the calibrations to be symmetric about the mechanical axis of fission counters 1 and 2. While some allowance must be made for machining tolerances, the reflection shift of the two distributions, when data was rerun in one-year intervals, warns of some lack of understanding of the underlying principles. An error was assigned to bridge this eccentricity.

Although the effect of self-transfer of aggregates is likely to be minor if contained in a small region within the purview of the neutron detector, we cannot be certain of this constraint. From the magnitude of the loss and from the time which lapsed in calibration, an error range was estimated.

The systematic errors were assigned to each fission counter and linearly added to the random error that was generally based on the degree of internal consistency obtained in the calibration. To some extent there is already some conservatism in error assignment because the imprecision associated with different methods of calibration, especially for fission counter No. 3, already includes some measure of systematic effects.

Each of the fission counters has been considered as an independent entity in terms of averaging to obtain the weighted mean for ν. The selection of errors common to the three quasi-independent measurements of ν is limited to recognized potential effects, which, upon examination, should influence the final result by no more than 0.1%.

A. Speculation on Systematic Corrections

At the time of writing, there are three known factors which could conceivably be subjected to a systematic adjustment in the future. The first of these is the sulfur-oxygen fast-neutron absorption correction. Our independent evaluation of this quantity results in a value of the neutron-emission rate that is about $\frac{2}{3}\%$ less than would have been reported on the basis of the correction factor taken from the older literature. In addition, there are two independent calculations based on current cross sections which support our data.

There has been a recent unreported comparison of ^{56}Mn issued as a chloride. Preliminary results from the six participating laboratories indicate our results are

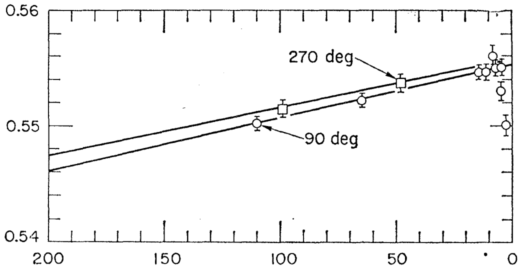


FIG. 11. Fission rate as a function of neutron-detector solid angle (Counter No. 3).

TABLE V. Total neutron yield for ^{252}Cf.[a]

Fission counter No.	Neutron rate[b] (10^5/sec)	± (%)	Fission rate[b,c]	± (%)	Fragment efficiency (%)	Anisotropy (%)	ν	±	
1	5.050	0.48	1.367	1.0	60	23	3.695	0.041	(1.1%)
2	7.220	0.40	1.934	0.40	88	4.4	3.733	0.021	(0.56%)
3	6.786	0.38	1.8218	0.13	99.5	1.0	3.725	0.015	(0.40%)
	Weighted mean:						3.725	0.012	(0.32%)
	Common systematic errors:							0.003	
	Result:						3.725	0.015	(0.40%)

[a] These results encompass and supersede our previous data (see Ref. 4).
[b] Corrected to respective reference dates.
[c] 10^5 fissions/sec.

0.2% above the average. However, there is a possible systematic difference between liquid-scintillation and proportional-counter data derived from the absolute coincidence counting. This factor is presently about 0.35%. If such a systematic effect were reproduced in future comparisons, our neutron-source rate could be subject to a maximum decrease of $\frac{1}{2}$%.

It should be noted that, with regard to systematic errors in technique and method of computation for coincidence measurements, common treatment is applied to both the fission-rate determination and the neutron-source calibration. Thus, some internal systematic defects will tend to cancel.

A third controversial correction concerns compensation for neutron leakage from the manganese bath sphere.[9] The correction we have applied for escape with a ^{252}Cf source is 0.4%, while some estimates derived from transport theory are as low as 0.2%. On the other hand, direct experimental evidence by Davy[24] concerning the manganese bath suggests that 1.2% of the neutrons escape from a bath of 48 cm effective radius. Our calculation arises from comparison of leakage by Am-Be(α, n), Ra-Be(α, n), and ^{252}Cf sources from a sphere of 28 cm radium and an equilateral cylinder of 40 cm radius.

TABLE VI. Percent of systematic errors.[a]

	Counter No. 1	2	3
Fission rate			
Half-life	0.01	0.01	0.01
Delayed neutrons	0.05	0.05	0.05
γ sensitivity of neutron detector	0.02	0.02	0.02
Absolute counting	0.10	0.10	0.10
Anisotropy factor	0.50	0.10	0.02
Angular-distribution eccentricity	0.60	0.15	0.02
α pileup	0.05	0.05	0.05
Aggregate-recoil loss effect	0.30	0.10	0.05
Quadrature sum	0.85	0.24	0.13
Errors common to the three measurements			
Spontaneous-fission contaminants			0.02
(α, n) reactions			0.02
Neutron-detector efficiency dependence on ν			0.05
Fission-detector efficiency dependence on ν			0.05
Multiplicity averaging			0.05
Quadrature sum			0.09

[a] All errors pertain to correction factors which are calculated or measured to have less than 1% influence on ν.

From a local viewpoint, there is no *a priori* basis to allow for an error any larger than already applied to the separate phases of the experiment. It is possible, though, to speculate that three unrelated factors noted above, which have not yet been fully certified, could act on our evaluation of ν in a combination of increases or decreases amounting to 1%. Countermanding this speculation, however, is our significant, although superficial, agreement in neutron-source calibration for the NRC Ra-Be(α, n) neutron standard, a calibration which is sensitive to such effects.

V. DISCUSSION

Because of prominant discrepancies in neutron-yield measurements reported with high precision, there have already been a number of inquiries into potential systematic errors.[2,25–27] Accordingly, we shall concentrate our discussion on certain sensitive features of the present experiment with limited reference to other measurements.

It was noted that our results encompass and supersede an earlier average which we published for ν(^{252}Cf).[4] In the neutron-rate and fission-rate determinations, both the old and the new data have been included in the mean values. Of course, the weighting of the older data is somewhat small compared to the more precise results recently obtained from the manganese bath. The new fission-rate determinations for fission counters 1 and 2 have about the same result as the older measurements.

As a consequence of its high native fission-fragment efficiency, primary confidence has been associated with our third fission counter. Thus, while the first two counters have been independently reevaluated, they retain somewhat similar average values and associated errors. This in itself is some measure of the validity of the chosen error limits.

---

[24] D. R. Davy, J. Nucl. Energy **20**, 227 (1966).

[25] D. W. Colvin, M. G. Sowerby, and R. I. McDonald, in *Proceedings of Conference on Nuclear Data for Reactors* (International Atomic Energy Agency, Vienna, 1967), Vol. I, p. 307.

[26] H. Condé, IAEA Panel on Nuclear Standards, Brussels, 1967 (unpublished).

[27] A. De Volpi, Reactor Fuel Processing **10**, 271 (1967).

Of other reports in the literature concerning $\nu(^{252}\mathrm{Cf})$, none have measured the yield for more than one fission source, nor have efforts been made to the extent that we have made them to vary the conditions of measurement. As with Colvin and Sowerby,[25] we have compared our neutron efficiency with that of standard sources. In fact, our efforts have gone several steps further. Moreover, the conditions under which our comparisons have been done are closely allied to the neutron rate evaluation. With regard to a measurement reported by White and Axton,[28] comparable efforts have been made in neutron assay, although it is clear[9] that a systematic difference remains (Table II). With regard to the fission portion of the measurement, we have gone further in doing both the defined solid-angle counting and the coincidence assessment. It is our opinion that the absolute-coincidence technique is less likely to be subject to errors from range losses and from self-transfer of aggregates during the course of the experiment. If the systematic differences which we have with NPL in neutron calibration were applied to decrease the Axton and White result,[28] then their value of 3.796 would be from $\frac{1}{3}$ to 1% lower, and conversely.

Until some of these neutron counting issues can be resolved, it is necessary to continue to report measurements such as ours partially *in vaccuo*, that is, without ultimate verification of all pertinent factors.

There are, however, sufficient reasons to believe that this report represents an improvement in terms of authentication of results:

(1) Our evaluation of the international standard neutron source is in precise superficial agreement with the world average.

(2) We have made independent measurements of the major correction factors, specifically the effective hydrogen/manganese cross-section ratio, the fast-neutron parasitic absorption, neutron escape, and cavity absorption.

(3) The basis for *both* the neutron-rate determination and the fission-rate determination is absolute-coincidence counting under the best conditions, namely, one channel having efficiency approaching 100% and the other channel being sensitive to highly penetrating radiation.

(4) The various residual corrections for the absolute-coincidence counting have been extensively evaluated under a variety of conditions.

(5) Potential systematic errors of small order in coincidence counting caused by common features of method or analysis will tend to cancel since they appear directly as a ratio in the neutron/fission quotient.

(6) Counting statistics are negligible in all phases of the measurement.

(7) There is no prominent dependence on cross-section values for any aspect of the experiment; and in the case where some cross-section *ratios* appear, they have been directly measured with the existing apparatus.

(8) There is no important dependence of the experiment on knowledge of neutron spectrum.

(9) We have measured three quasi-independent values for three fission counters with different efficiency, geometry, and techniques of calibration, and the results are reasonably consistent.

Having thus bypassed most of the known weakness of previous experiments, we are in a position to report a value for the total yield which is intermediate to other measurements in the literature. In the absence of any established evidence for systematic discrepancies in the other experiments, this new result suggests that the other measurements are consistent with this value and that the discrepancies which have existed in the past have been of a random uncorrelated nature.

VI. SUMMARY

The total neutron yield from spontaneous fission of $^{252}\mathrm{Cf}$ is found to be 3.725 ± 0.015 neutrons/fission by the following method:

Three fission counters, containing from $\frac{1}{4}$ to $\frac{1}{2}$ μg of $^{252}\mathrm{Cf}$ on an electrode, were fabricated. The neutron-emission rate from the counter was measured with a manganese-bath system calibrated absolutely in an independent basis, and results have been supported by reference to some international standard neutron sources. The manganese-bath calibration is chiefly related to the effective manganese/hydrogen cross-section ratio, which was also independently evaluated, and to absolute β-γ-coincidence counting of $^{56}\mathrm{Mn}$. The fission rate of the californium sources was determined primarily by absolute neutron-fission coincidence counting of the fragments from $^{252}\mathrm{Cf}$, with special attention to effects of angular correlation.

The yields from the three fission counters are reasonably self-consistent, and the reported value is essentially independent of cross-section data, neutron-spectron-spectrum knowledge, counting statistics, and fission-fragment self-absorption. The quoted error is a combination of statistical and systematic effects at a 68% confidence level.

ACKNOWLEDGMENTS

We would like to note specifically that the high quality $^{252}\mathrm{Cf}$ coating for fission counter No. 3 was provided by R. Sjoblom and P. Fields. Except for R. Armani's determination of the fission rate by defined solid-angle counting, most of the data were collected and processed by F. H. Ozer.

---

[28] P. H. White and E. J. Axton, J. Nucl. Energy **22**, 73 (1968).