PHYSICAL REVIEW VOLUME 101, NUMBER 3 EBRUARY 1, 1956

Multiplicities of Fission Neutrons[*]

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(Received September 6, 1955)

A large liquid scintillator with approximately 80 percent efficiency for detection of neutrons has been used to obtain data on numbers of neutrons emitted per fission for several fissioning nuclides. Reported here are the average number of neutrons per fission, $\bar{\nu}$, and the respective probabilities of 0, 1, 2,$\cdots$ neutrons per fission, for spontaneous fission of Pu^{240}, Cm^{244}, and Cf^{252}, and for fission induced in U^{233}, U^{235}, and Pu^{239} by 80-kev neutrons. The values of $\bar{\nu}$ for these cases are, respectively, 2.257 ± 0.045, 2.810 ± 0.059, 3.869 ± 0.078, 2.585 ± 0.062, 2.47 ± 0.03, and 3.048 ± 0.079. The thermal neutron value for U^{235} (2.46 ± 0.03), obtained by other methods, was used here as a standard for calibrating the detector efficiency. The efficiency was also measured by another method, involving scattering neutrons into the scintillator, with results in good agreement. The probabilities of 0, 1, 2,$\cdots$ neutrons per fission approach closely a binomial type distribution, with the maximum number of neutrons equal to 5, 6, or 7, depending on $\bar{\nu}$.

I. INTRODUCTION

THE development of a highly efficient neutron detector[1] at the Los Alamos Scientific Laboratory has made possible a much more detailed study of the multiplicities of fission neutrons than has been previously possible. Using low-efficiency detectors, many measurements[2–6] have been made of the average number of neutrons, $\bar{\nu}$, emitted per fission. References to much of this work may be found in the compilation by Hughes and Harvey[7] and in the paper by Leachman.[8] The high-efficiency neutron detector has now made it possible to obtain not only the average number of neutrons emitted but also information on the probabilities P_ν for the emission of various numbers of neutrons per fission up to five or more with fair precision. Several groups are working on this problem as applied to spontaneous fission.[9,10] This paper describes a set of experiments designed to study the multiplicities of fission neutrons from neutron-induced fission and also from spontaneous fission.

II. EXPERIMENTAL PROCEDURE

A. General

The general arrangement of experimental equipment is shown in Fig. 1. For spontaneous-fission measurements the neutron source and collimator are superfluous. In the case of neutron-induced fission, thermalized neutrons from a Pu$-$Be source or 80-kev neutrons from the T(p,n)He^3 reaction in the target of an electrostatic accelerator pass through the aperture of a collimator. These neutrons proceed down a cylindrical opening along the axis of a large liquid scintillator and pass through one or more foils of fissionable material in a counter located at the center. When a fission occurs, the pulse produced by the fission fragments in the counter triggers an oscilloscope sweep 30 μsec long on which are displayed the pulses from the liquid scintillator. Neutrons produced by the fission enter the scintillator, and in their first few collisions cause proton recoils which, along with the fission gamma rays, produce a prompt scintillator pulse. Nearly all the fission neutrons are slowed down by hydrogen nuclei and are finally captured by cadmium, present as a salt dissolved in the scintillator solution. As each neutron is captured in the cadmium, a pulse is produced in the scintillator by the capture gamma rays. These pulses appear on the oscilloscope sweep after the prompt pulse, separated in time except for accidental coincidences. The sweep is photographed, the film in the camera is advanced, a background sweep is triggered and photographed, and the film again advanced, after which the equipment is ready to record another fission.

B. Scintillator

The scintillator used is the "large detector" described by Reines *et al.*,[1] modified to carry out these experiments. The scintillator tank is a cylinder 30 in. long and 28½ in. in diameter with ¼-in. thick steel walls, coated on the inside with light-reflecting paint. The photocathodes of 90 Du Mont 6292 and 1177 photomultiplier tubes are flush with the inner surface of the cylinder. The ends of the cylinder are disks of steel about 42 in. in diameter. These disks rest on a set of rollers, thus making it possible to rotate a defective phototube to the top and replace it without emptying the solution. A steel tube of 2.6-in. inner diameter forms a cylindrical opening along the axis of the tank down which the

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

[†] Herbert C. Martin met an untimely death on July 3, 1955, while mountain-climbing in Colorado.

[1] Reines, Cowan, Harrison, and Carter, Rev. Sci. Instr. **25**, 1061 (1954).

[2] Barclay, Galbraith, and Whitehouse, Proc. Phys. Soc. (London) **A65**, 73 (1952).

[3] F. R. Barclay and W. J. Whitehouse, Proc. Phys. Soc. (London) **A66**, 447 (1953).

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[6] E. Segrè, Phys. Rev. **86**, 21 (1952).

[7] D. J. Hughes and J. A. Harvey, *Neutron Cross Sections*, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).

[8] R. B. Leachman, preceding paper [Phys. Rev. **101**, 1005 (1956)].

[9] J. E. Hammel and J. F. Kephart, Phys. Rev. **100**, 190 (1955).

[10] Hicks, Ise, and Pyle, Phys. Rev. **97**, 564 (1955); **98**, 1521 (1955); following paper [Phys. Rev. **101**, 1016 (1956)].

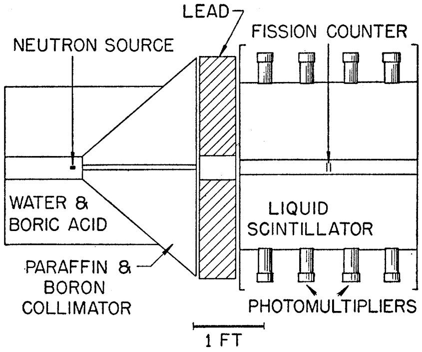

FIG. 1. Schematic diagram of the experimental equipment as used with T(p,n)He^3 neutrons. A collimator with a larger aperture was used for measurements with thermalized neutrons from a Pu—Be source.

neutron beam can pass without striking the scintillator. The solution in the scintillator is similar to that described by Reines *et al.*[1] for a cadmium-to-hydrogen atomic ratio of 0.003 except that the wavelength shifter used is "POPOP"[11,12] rather than αNPO. The other ingredients are toluene, terphenyl, methanol, and cadmium propionate.

The collectors of the 90 phototubes are connected in parallel; the photocathodes are grounded. The 1700-volt positive high voltage is connected to the resistor stacks of the individual tubes through trimmer resistors adjusted to give equal gains in all tubes. Pulses from the collectors go through a blocking condenser to the preamplifier of a modified Los Alamos Model 503 amplifier. The output pulses from this system have an over-all width of about 0.2 μsec and are delayed by 3 μsec.

C. Measurements

At the start of every run, pulses produced in the scintillator by cosmic rays are used to determine the over-all gain of the system. By exposing one frame of film to a large number of sweeps triggered by these pulses, the group produced by charged particles traversing the detector near minimum ionization is clearly defined. A 10-channel pulse-height analyzer is used to select a bias for the fission counter which is low enough to accept nearly all fission events but high enough to discriminate against alpha particles.

About 1500 pairs of fission-triggered and background sweeps are then recorded photographically on a roll of film. A typical pair of oscilloscope traces is reproduced in Fig. 2. The developed film is placed in an enlarger, and the magnification of the enlarger is adjusted so that the calibrating group of cosmic-ray pulses has a standard height on the projection table. The minimum acceptable height of neutron pulses is also marked on the table. In order to avoid sweeps triggered by events other than fission, only those traces are read which have a properly located prompt pulse due to fission gammas and proton recoils; at least 99 percent of the fission events produce such a pulse. The number of acceptable pulses after the prompt-pulse time from each fission and background trace are then recorded. Such measurements have been made for fission induced by thermal neutrons and by 80-kev neutrons in U^{233}, U^{235}, and Pu^{239} and for spontaneous fission of Pu^{240}, Cm^{244}, and Cf^{252}.

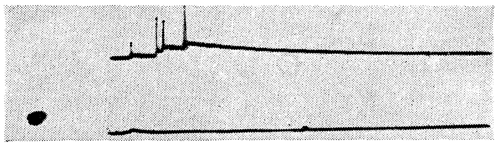

FIG. 2. Typical pair of oscilloscope traces showing pulses from the liquid scintillator. The upper trace was triggered by a pulse from the fission counter. The first pulse on the left is the prompt pulse, due mainly to fission gamma rays; the three pulses following are due to capture of three neutrons by cadmium. The second trace is a background trace which is triggered a fixed time after the fission event and is identified by the dot of light to the left.

To deduce the emission probabilities, P_ν, from the various numbers of pulses per sweep, it is necessary to know the efficiency of the detector for fission neutrons. Several effects contribute to a reduction of the efficiency from 100 percent. One such effect is leakage of neutrons out of the detector. A Monte Carlo calculation has been made which shows that when neutrons originate from an isotropic source at the center of the detector, the fraction of neutrons captured by cadmium in the solution is 99.6 percent for thermal neutrons, 98.4 percent for 1-Mev neutrons, 92.1 percent for 3.5-Mev neutrons, 85.0 percent for 5-Mev neutrons, and 74.0 percent for 8-Mev neutrons. From this and knowledge of the fission spectrum, one can determine that about 95 percent of fission neutrons are captured in the detector. A second and larger reduction of efficiency is due to the loss of small pulses. The distribution of heights of capture pulses is shown in Fig. 3. To discriminate against the large number of small background pulses, only pulses whose heights are greater than the value indicated by the dotted line are chosen. In addition, some pulses are lost because of the finite sweep length, about 2 percent of the neutrons being captured after the end of the sweep.

The efficiency was obtained experimentally in two ways. The more accurate method was to observe pulses from thermal and 80-kev fission of U^{235}, and to normalize to the value of $\bar{\nu}$ as measured[7] by other methods. The measured value used for thermal energy was 2.46±0.03 and the 80-kev value was assumed to be 2.47±0.03. The efficiency was then the mean number of pulses per fission-triggered sweep less the mean number per background sweep, divided by the value of $\bar{\nu}$. With this method the efficiency was found to be 81.4±1.5 percent; this figure was used in the correction of data.

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[11] Obtained from the Arapahoe Chemical Company, Denver, Colorado.

[12] Hays, Rogers, and Ott, J. Am. Chem. Soc. **77**, 1950 (1955).

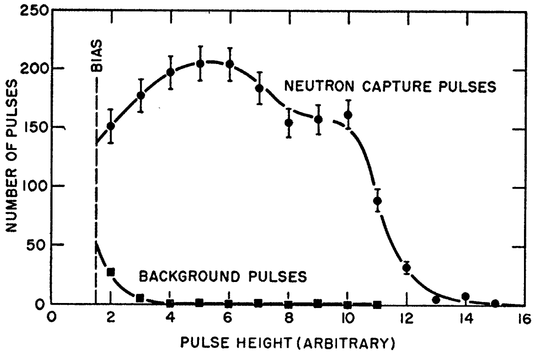


FIG. 3. Distribution in height of pulses resulting from capture of neutrons in cadmium. Background has been subtracted.

The second method of determining efficiency was to trigger the oscilloscope sweep with pulses from protons which recoiled in *n-p* scattering events within an anthracene crystal and scattered neutrons into the large detector. The neutrons, which were produced by the $T(p,n)He^3$ and $D(d,n)He^3$ reactions, passed through a narrow collimating system between the gas target of a 2.5-Mev electrostatic accelerator and the opening through the scintillator. The crystal, mounted on a photomultiplier tube, was placed in the same position that the fission counter normally occupied. The efficiency was given by the probability of observing a pulse on a sweep triggered by a proton recoil, minus the probability of a pulse on a background sweep. A single-channel analyzer was used to select a narrow range of recoil-proton pulse heights to trigger the sweep. This procedure defined the energy of the scattered neutrons and their recoil angles with respect to the axis of the scintillator. Since there is some possibility that events other than proton recoils will trigger the sweep, a similar set of data was taken with a helium-filled target, and from this a small correction was made to the recoil-proton data. The efficiency was thus determined for a fixed neutron energy and angle, and a set of such measurements allowed the efficiency for fission-spectrum neutrons to be calculated. The experimental efficiencies obtained were 87±2 percent and 85±3 percent, respectively, for neutrons with energies of 0 to 100 kev and 2 Mev. These numbers have been corrected very slightly to apply to isotropic neutron sources, with the aid of the Monte Carlo calculations mentioned earlier.

TABLE I. Effect of corrections on typical data (for Cf^{252}): n (or ν) is the number of pulses (or neutrons). F_n' and B_n' are observed probabilities of n pulses on fission and background traces, completely uncorrected. C_n are probabilities of n capture pulses, corrected for coincidence and background. P_ν are the final data, corrected for efficiency.

n (or ν)	F_n'	B_n'	C_n	P_ν
0	0.0121	0.9202	0.0132	0.0057
1	0.0719	0.0767	0.0723	0.0042
2	0.2314	0.0029	0.2310	0.1376
3	0.3076	0.0000	0.3013	0.2227
4	0.2425	0.0002	0.2466	0.3566
5	0.0983	···	0.0973	0.1751
6	0.0293	···	0.0305	0.0710
7	0.0062	···	0.0070	0.0220
8	0.0007	···	0.0009	0.0059
9	···	···	−0.0001	−0.0008

If it is assumed that neutron leakage is the primary reason for change in efficiency with energy, the Monte Carlo calculations may be used to plot a smooth curve of efficiency *versus* energy passing through the two experimental points. For fission-spectrum neutrons the average efficiency thus determined is 83±2 percent. No differences in the capture pulse-height spectrum have been observed in data taken with fission-spectrum neutrons and those of 0- to 100-kev and 2-Mev energy.

III. ANALYSIS AND RESULTS

To convert the observed probabilities of detecting various numbers of pulses per sweep into probabilities of various numbers of neutrons being emitted in fission, a series of three calculations was made. The symbols used were defined as follows: τ = resolving time (0.15 ±0.05 μsec by counting loss measurement); $f(t)$ = normalized time distribution of capture pulses; $k=2\tau\int f^2(t)dt$ = probability of coincidence between any two capture pulses (experimentally, $k=0.02$); F_n' = probability of observing n pulses per fission sweep, uncorrected for coincidence loss; B_n' = probability of observing n background pulses per sweep, uncorrected for coincidence loss; F_n = probability of observing n pulses per fission sweep, corrected for coincidence loss; B_n = probability of observing n background pulses per sweep, corrected for coincidence loss; C_n = probability of observing n fission neutron pulses per fission sweep; P_ν = probability of emission of ν neutrons per fission; and ϵ = detector efficiency.

First, a small correction was made to the fission data for loss due to accidental coincidence of capture pulses. This correction made use of the equation

$$F_n\left\{1-\left[\frac{n!}{2!(n-2)!}\right]k\right\}=F_n'-\left[\frac{(n+1)!}{2!(n-1)!}\right]kF_{n+1}.$$

A similar correction was made to the background data ($k_B=0.010$). Then the effect of background on the observed distribution was removed by means of the relation

$$F_n=C_0B_n+C_1B_{n-1}+\ldots+C_nB_0$$

and from the observed sets of F_n and B_n the values of C_n were calculated. Finally,

$$P_\nu=\sum_{n=\nu}^{n=\infty}\frac{n!}{\nu!(n-\nu)!}\left(1-\frac{1}{\epsilon}\right)^{n-\nu}\left(\frac{1}{\epsilon}\right)^{\nu}C_n,$$

from which the P_ν were obtained from the C_n. The first and second moments, $\bar{\nu}$ and $\langle\nu^2\rangle_{Av}$, as well as other

TABLE II. Summary of results: $\bar{\nu}$ and $\langle\nu^2\rangle_{\mathrm{Av}}$ are the average and the average square of the number of neutrons per fission; P_0, P_1, $P_2,\cdots$ are the respective probabilities of emission of 0, 1, $2,\cdots$ neutrons per fission. The quantity $[\langle\nu^2\rangle_{\mathrm{Av}}-\bar{\nu}]/\bar{\nu}^2$ is a measure of the relative width of the neutron multiplicity distribution. It would be equal to 1.0 for a Poisson distribution. The right-hand column gives results obtained by scattering neutrons from an anthracene crystal in the scintillator tank and triggering the oscilloscope sweep with proton-recoil pulses from the crystal. The efficiency used in this case was 0.872±0.015.

	Neutron-induced fission[a]			Spontaneous fission			Scattered neutrons
Nuclide	U^{233}	U^{235}	Pu^{239}	Pu^{240}	Cm^{244}	Cf^{252}	(0–100 kev)
Fissions analyzed	1632	10715	1376	8355	3301	4545	1470
$\bar{\nu}$	2.585 ± 0.062	2.47[b] ±0.03	3.048 ± 0.079	2.257 ± 0.045	2.810 ± 0.059	3.869 ± 0.078	1.000[b]
$\langle\nu^2\rangle_{\mathrm{Av}}$	7.84 ± 0.34	7.32 ± 0.15	10.62 ± 0.53	6.37 ± 0.21	9.20 ± 0.34	16.59 ± 0.62	1.007 ± 0.033
$[\langle\nu^2\rangle_{\mathrm{Av}}-\bar{\nu}]/\bar{\nu}^2$	0.786 ± 0.013	0.795 ± 0.007	0.815 ± 0.017	0.807 ± 0.008	0.810 ± 0.008	0.850 ± 0.006	0.007 ± 0.033
P_0	0.010 ± 0.008	0.027 ± 0.004	-0.01 ± 0.01	0.049 ± 0.006	0.009 ± 0.005	0.005 ± 0.002	0.001 ± 0.013
P_1	0.151 ± 0.024	0.158 ± 0.010	0.11 ± 0.03	0.214 ± 0.012	0.109 ± 0.016	0.004 ± 0.009	0.999 ± 0.028
P_2	0.326 ± 0.037	0.339 ± 0.014	0.13 ± 0.06	0.321 ± 0.014	0.292 ± 0.023	0.138 ± 0.019	0.000 ± 0.015
P_3	0.301 ± 0.044	0.305 ± 0.015	0.56 ± 0.08	0.282 ± 0.017	0.315 ± 0.027	0.223 ± 0.032	-0.002 ± 0.004
P_4	0.176 ± 0.041	0.133 ± 0.013	0.11 ± 0.08	0.112 ± 0.013	0.224 ± 0.027	0.356 ± 0.035	0.002 ± 0.002
P_5	0.042 ± 0.028	0.038 ± 0.009	0.06 ± 0.09	0.021 ± 0.008	0.030 ± 0.017	0.175 ± 0.034	0.000 ± 0.000
P_6	-0.010 ± 0.017	-0.001 ± 0.003	0.05 ± 0.08	0.001 ± 0.003	0.021 ± 0.010	0.071 ± 0.028	···
P_7	0.006 ± 0.009	0.001 ± 0.002	0.00 ± 0.06	0.000 ± 0.002	0.000 ± 0.003	0.022 ± 0.017	···
P_8	-0.002 ± 0.002	0.000 ± 0.000	-0.01 ± 0.03	0.000 ± 0.000	0.000 ± 0.000	0.006 ± 0.007	···

[a] Results given are for 80-kev neutrons.
[b] Normalizing value.

moments and the standard deviations, were also calculated. These calculations were made by means of I.B.M. 701 and card-programmed computers.

The effect of these corrections on the original data is illustrated in Table I, where the original data and final corrected neutron distributions are given for the case of Cf^{252}. Backgrounds were not a serious problem at any time; average backgrounds were usually of the order of one or two pulses in ten traces.

Final results are given in Table II. Thermal neutron data have been omitted because of the greater weight of the 80-kev data; there was no significant difference. The errors shown are standard deviations. Allowance has been made for counting statistics, uncertainty in the efficiency, and uncertainty in the coincidence correction. It should be pointed out that these results refer only to relatively prompt neutrons, emitted within a few microseconds after fission. In the last column, the results of a typical set of data obtained by detecting neutrons scattered from protons are shown. Since, in this case, ν is always equal to unity, the fact that all P_ν are near zero except P_1 indicates that spurious effects such as multiple pulsing of the phototubes are absent.

IV. DISCUSSION

For each fissioning nuclide examined, the distribution of emission probabilities P_ν as a function of ν is definitely too narrow to approach a Poisson distribution,[5] $P_\nu=[\bar{\nu}^\nu \exp(-\bar{\nu})]/\nu!$.

A binomial type distribution,[10] given by

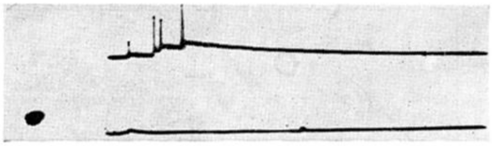
$$P_\nu=\frac{m!}{\nu!(m-\nu)!}\left(\frac{\bar{\nu}}{m}\right)^\nu\left(1-\frac{\bar{\nu}}{m}\right)^{m-\nu},$$

where m is the maximum number of neutrons emitted, is a close representation of the data, with $m=5$, 6, or 7. In some cases, a mixture of two binomial distributions gives a better fit to the data. The proper value (or values) of m may be determined from the observed second moment $\langle\nu^2\rangle_{\mathrm{Av}}$. For a binomial distribution, $\langle\nu^2\rangle_{\mathrm{Av}}=\bar{\nu}+(m-1)\bar{\nu}^2/m$; the Poisson case is obtained for $m=\infty$. The value of $[\langle\nu^2\rangle_{\mathrm{Av}}-\bar{\nu}]/\bar{\nu}^2$ is independent of counter efficiency and can be closely reproduced by the equation $[\langle\nu^2\rangle_{\mathrm{Av}}-\bar{\nu}]/\bar{\nu}^2=0.714+0.035\bar{\nu}$. This allows a close estimate of the second moment, if the first moment is known.

All of the data reported here agree well with the calculations of Leachman.[8]

V. ACKNOWLEDGMENTS

The authors wish to thank Dr. A. W. Schardt and Mr. Ralph Lewis for the testing and selection of all of the phototubes used in the detector. We are particularly indebted to Dr. A. R. Ronzio for the preparation of the scintillating solution. Dr. F. Reines and Dr. C. L. Cowan kindly furnished many of the parts for the detector and contributed valuable advice about its operation. Much of the film was read by W. J. Masilun. The computer calculations for data analysis were made by Max Goldstein, and the Monte Carlo calculations by Dr. C. J. Everett, Dr. E. Cashwell, and Dr. G. I. Bell. The foils of fissionable material were supplied to us by Mr. John Povelites, Dr. Robert Penneman and Dr. Charles Browne. The Argonne National Laboratory was the source of our Cm^{242} and Cf^{252}. We wish to thank Dr. M. Goldhaber for a suggestion which led to this program.



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