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UCRL-3441

UNIVERSITY OF CALIFORNIA

Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

PROBABILITIES OF PROMPT-NEUTRON EMISSION FROM SPONTANEOUS FISSION

Donald A. Hicks

(Thesis)

June 13, 1956

Printed for the U. S. Atomic Energy Commission

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ABSTRACT

The neutron-number distributions from the spontaneous fission of eight isotopes have been measured by use of a fission chamber placed at the center of a cadmium-loaded liquid scintillation tank. The experimental distributions can be approximated by binomial distributions.

The average numbers of neutrons per spontaneous fission have been found to be 2.30 ± 0.19 for Pu^{236}, 2.33 ± 0.08 for Pu^{238}, 2.18 ± 0.09 for Pu^{242}, 2.65 ± 0.09 for Cm^{242}, 2.84 ± 0.09 for Cm^{244}, 3.82 ± 0.12 for Cf^{252}, and 4.05 ± 0.19 for Fm^{254}, all based on 2.257 ± 0.046 for Pu^{240}, which in turn is based on 2.46 ± 0.03 for the thermal-neutron-induced fission of U^{235}.

Introduction

Fission is the process in which a heavy complex nucleus breaks up into two complex nuclear fragments with the release of considerable energy. This energy is distributed between the kinetic energy and the excitation energy of the fission fragments.

Two different approaches have been used in attempts to understand the fission phenomena. Bohr and Wheeler,[1] with extensions by Present and Knipp,[2] Metropolis and Frankel,[3] and others, have used the liquid-drop model to explain and predict certain observable factors in fission. The liquid-drop model has been fairly successful in the explanation of certain general features, but because of the complexities involved it has failed to explain, or has disagreed with, some empirical results. One of the most notable disagreements is the theoretical prediction of symmetric fission, whereas asymmetric fission is observed.

The second method, by considering the experimentally determined distribution of charge, mass, and energy of the two fragments and the distribution of the energy between kinetic and excitation energies, attempts to develop some consistent empirical theory that will explain the intricacies of the fission process. Analyses of this type have been made by Way and Wigner,[4] Present,[5] Coryell, Glendenin and Edwards,[6] Brunton,[7] Leachman,[8] Fong,[9] and others.

The work reported in this paper, concerning the multiplicity distribution of the neutrons from fission, provides information on the distribution of the energy of fission.

The discovery of fission resulted from experiments conducted by Fermi and his collaborators[10, 11] in an attempt to produce transuranic elements. It was thought that the bombardment of uranium (element 92) by neutrons would produce isotopes of the uranium not found in nature, and that by negative beta decay atoms of atomic charge 93, or higher, would be formed. Several artificial radioactive periods were discovered, and after showing that these activities were not ascribable to isotopes of any elements from radon (element 86) to uranium, Fermi concluded that they were due to transuranic elements.

These experiments stimulated Curie and Savitch, and Hahn, Meitner, and Strassmann to investigate these "transuranic elements" during the next few years. In 1939 Hahn and Strassmann,[12] after some exhaustive tests, showed that an isotope and its daughter that resulted from the neutron bombardment of thorium (element 90), and that had been identified as radium (element 88) and actinium (element 89), were in reality barium (element 56) and lanthanum (element 57). They suggested that the thorium nucleus had split into two approximately equal fragments after the capture of a neutron.

This hypothesis was soon confirmed. It was realized by Meitner and Frisch[13] that a large amount of energy (approximately 200 Mev) would be released in fission because of the mass difference between the original excited nucleus and the sum of the two fission fragments. Since most of this energy would be in the form of kinetic energy of the fragments resulting from the Coulomb repulsion between the two fragments, a large amount of ionization would be produced by a fission fragment in the gas of an ionization chamber. (If a fission fragment and an alpha particle give up all of their energy in the chamber, the fission fragment will give a pulse approximately ten to twenty times as large as that of the alpha particle.) Frisch was the first to observe these large pulses. Jentschke and Prankl[14] found that the pulse heights from the fission fragments could be divided roughly into two groups at about 60 Mev and 100 Mev. This was the first indication that fission was actually asymmetric, which was at variance with the theoretical conclusion by Bohr and Wheeler[1] that fission was most probably symmetric.

That transuranic elements were also formed when uranium was bombarded with neutrons was shown by McMillan,[15] who bombarded thin uranium foils with neutrons. Because of their high kinetic energy most of the fission fragments escaped, and the remaining activity of the foils was due primarily to the decay of the newly formed transuranic elements. He observed activities with half lives of 25 minutes and 2 days.

It was soon determined that fission of heavy elements could be produced by fast protons, deuterons, alpha particles, and γ-rays, in addition to neutrons. Following a theoretical prediction by Bohr and

Wheeler,[1] spontaneous fission, for which no incident particle is required, was observed experimentally by Flerov and Petrzhak.[16] In the experiment to be reported here, all the isotopes used undergo fission spontaneously and contain even numbers of neutrons and protons. Isotopes containing odd numbers of neutrons or protons were not used because of their very long spontaneous-fission half lives. (The reason for this difference in the spontaneous fission rate between even-even and odd-A isotopes has been discussed by Wheeler.[17])

Regardless of how the nucleus divides in fission, the fragments have too many neutrons for stability. This condition can be corrected by negative beta decay or the emission of neutrons. Neutron emission was shown to occur in experiments conducted by Von Halban, Joliot, and Kowarski[18] and by Anderson, Fermi, and Hanstein.[19] The energy of these neutrons has been investigated by many groups, and the published data have been summarized by Boyer and Tettle.[20] The energy distribution has the form

$$N(E)dE = \exp(-E)\sinh\sqrt{2E}\ dE.$$

Soon after the discovery of the neutrons it was determined by Anderson, Fermi, and Szilard that sufficient neutrons were emitted to allow a chain reaction to take place in natural uranium (which is composed almost entirely of the isotopes 235 and 238 in the ratio 1:140). The basic problem involved in creating this chain reaction is the arrangement of the fissionable material with some moderating substance, such as carbon, that will so reduce the energy of the fission neutrons that neutron resonance capture in U^{238} becomes improbable, with the result that any neutron capture is more likely to lead to another fission event. In order for the reaction to continue, on the average at least one of the neutrons emitted must cause another fission. The fact that such a chain reaction can take place has led to the many nuclear reactors and nuclear weapons now in existence.

The probabilities of prompt neutron emission from fission--i.e., the fraction of fissions producing 0, 1, 2, . . . neutrons--are of interest both in theoretical considerations and in reactor design. The many earlier studies[21-31] have been limited by the low neutron-detection

efficiency of the experimental apparatus to measurements of the first and second moments of the multiplicity distributions. However, the cadmium-loaded liquid scintillation tank[32] with its high neutron-detection efficiency permits an accurate determination of the prompt-neutron emission probabilities. In addition to measurements at this laboratory[33] of the multiplicity distributions from the spontaneous fission of eight isotopes similar measurements have been made by groups at the Los Alamos Scientific Laboratory[34, 35] on isotopes undergoing either neutron-induced or spontaneous fission. Before discussing the experimental details, we will consider the theoretical treatment of the multiplicity distributions as first presented by R. B. Leachman.[36]

Theory

Leachman uses the experimentally determined total kinetic energy distributions of the fission fragment pairs[37, 38, 39] together with the mass difference between these fragment pairs and the original nucleus to estimate the distributions in the excitation energies of the fission fragments. These excitation energies are used with neutron evaporation theory to determine the probabilities of neutron emission.

The mass equation of binary fission, which holds just after the fission has taken place and before the emission of neutrons,* is given by

$$M(A, \delta, Z) + E_n + B = M(A^L, \delta^L, Z^L) + M(A^H, \delta^H, Z^H) + E_k + E_x, \quad (1)$$

where the atomic masses M are functions of the atomic number A, the charge Z, and the even-odd parameter δ, of the semiempirical mass formula. The superscripts L and H refer to the light and heavy fragments respectively. E_n, the kinetic energy of the incident neutron, and B, the neutron binding energy, need be considered only in neutron-induced fission. E_k is the total kinetic energy and E_x is the total excitation energy of the two fragments. The relations $A^H + A^L = A$ and $Z^H + Z^L = Z$, of course, are required to hold.

The mass of the fissioning nucleus, $M(A, \delta, Z)$, can be determined experimentally by using alpha- and beta-particle energies combined with mass of the Pb^{208} isotope, which has been found by mass spectrographic means.[44-47] However, the ground-state masses of the two fission fragments, $M(A^L, \delta^L, Z^L)$ and $M(A^H, \delta^H, Z^H)$, must be approximated by an extension of the semiempirical mass surface. Leachman assumes that this region can be constructed by extending the parabolic form, the constants of which have been determined by Coryell,[48] of a given isobar. The mass value of the valley of the mass surface can be

* Studies[40-43] of the angular correlations between the direction of emission of the neutrons and the direction of the fission fragment are consistent with the neutrons' being emitted isotropically from the moving fragment in its center-of-mass system.

ascertained by using the mass spectrographic results of Duckworth et al.[49] and the Minnesota group.[50] The values of A^H, A^L, Z^H, Z^L, to be used in finding the masses of the two fragments from the assumed mass surface, are determined by considering a given $R_A = A^H/A^L$ and by using the charge displacements of the fission fragments from the hypothetically most stable charges (which are in general noninteger, since they are located at the apex of a given isobaric parabola). These charge displacements have been determined by the experimental observations of Glendenin et al.[51] and by Pappas.[52] Since the fragments observed by Glendenin and Pappas have emitted some neutrons, a correction was made by adding neutrons to the light and heavy fragments. The ratio of neutrons emitted by the light and heavy fragments as determined by Fraser,[53] together with the average number of neutrons per fission for the fissioning isotope considered, was used in the correction.

In order to simplify his calculations, Leachman used only three mass ratios in his determination of the multiplicity distribution for a given fissioning isotope. The most probable noninteger charges, Z^L and Z^H, were used and were determined by R_A. Although a very narrow charge distribution of about one charge full width at half maximum actually exists,[51] it can be neglected because the variation in the total energy released resulting from this distribution can be shown to be small.

From the mass equation of fission, when a given mass ratio R_A is considered, E_x can immediately be determined from E_k. However, for a given R_A, a distribution in the total kinetic energy $K(E_k)$ exists. This distribution cannot be directly observed experimentally, because several effects tend to broaden and distort it. The observed distribution is a composition of at least three effects: (a) the inherent distribution of the process $K(E_k)$, which we seek (and which is caused primarily by the charge distribution of the fragments), (b) momentum distribution due to the recoil of the fission fragment when prompt neutrons are

emitted, (c) dispersion caused by the rather poor energy resolution of fission chambers and by ionization defect.* Effect (c) can be eliminated by replacing a fission chamber with a "back-to-back" velocity selector.[56]

The observed distribution $I(E_I + \Delta, R_A)$ experimentally determined for the thermal-neutron-induced fission of Pu^{239} by Brunton and Thompson,[38] but with the energy scale corrected for ionization defect, is shown in Fig. 1. E_I is the energy reported in these measurements and Δ is the ionization defect. The three energy distributions, $I(E_I + \Delta)$, used by Leachman as discussed above are also shown.

The effect (b), listed above, can be neglected in our consideration because it is small. Using a comparison between fission chamber measurements and chemical fission-product mass data, Leachman[8] has attempted to determine the dispersion in $I(E_I + \Delta)$ caused by the energy resolution of the fission chamber and the ionization defect of the fission fragments. It was assumed for simplicity that the dispersion caused by

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* It has been shown experimentally[54] that the fission fragment is less efficient in ionizing a gas for a given energy loss than an alpha particle. This phenomenon is known as ionization defect and has been studied theoretically by Knipp and Ling.[55] It is thought that when the fission fragment loses enough energy so that its velocity is approximately that of the orbital electrons of the gas atom, it is unable to give its energy to these electrons and must resort to direct nculear collisions with the gas atoms. These recoiling gas atoms themselves have a reduced efficiency for producing ionization, because they in turn produce recoil atoms, and so on. Now the fission fragments, which are massive compared with an alpha particle, contain several Mev of energy at this critical velocity. However, an alpha particle has practically no energy at this velocity and so its ionization defect is negligible. The ionization defect then is a function of the mass of the fission fragment. Since chambers are calibrated by observing the ionization produced by an alpha particle of known energy, we see that a correction must be made for the ionization defect of the fission fragment, if our energy scale for fission fragments is to be corrected.

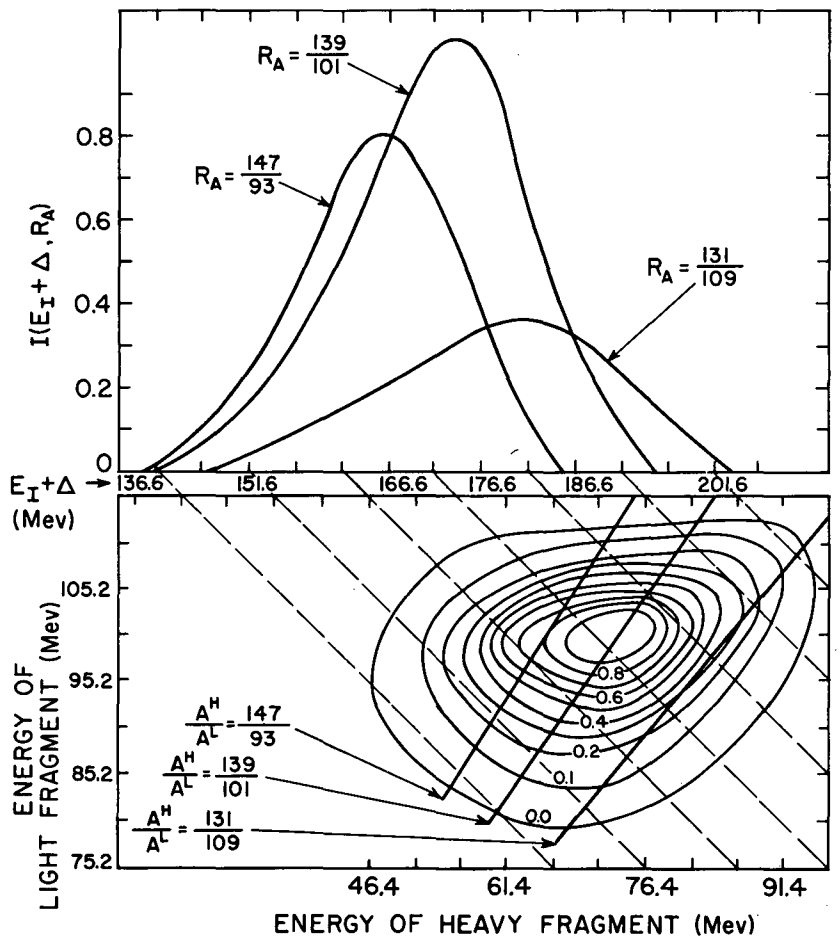

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Fig. 1. The experimental distribution of the total kinetic energy of the fission fragment pairs from the thermal-neutron-induced fission of Pu^{239} as determined by Brunton and Thompson[38] with the energy scales corrected for ionization defect. The values on the contours give the relative magnitude of $I(E_I + \Delta, R_A)$ for a given $E_I + \Delta$ and R_A. The distributions $I(E_I + \Delta, R_A)$ for $R_A = 147/93$, $139/101$, and $131/109$ were used by Leachman in his theoretical determination of the neutron multiplicity distribution.

the chamber is the gaussian distribution

$$D(E_k, E_I + \Delta) = Ce^{-\left(\frac{E_I + \Delta - E_k}{u}\right)^2}, \tag{2}$$

where $u = 7.2$ Mev as determined from Leachman's analysis[8] is used. Some assumption is required since only the width and not the shape of the distribution can be determined experimentally. (Some recent work by Pyle, Ise, and the author seems to indicate a low-energy tail on the dispersion function. The over-all effect of such an asymmetric distribution on the results discussed herein is probably slight.) We then have the convolution

$$I(E_I + \Delta, R_A) = \int_{-\infty}^{\infty} dE_k \, D(E_k, E_I + \Delta) \, K(E_k, R_A), \tag{3}$$

and, since we know $I(E_I + \Delta, R_A)$ and have semiempirically determined $D(E_k, E_I + \Delta)$, we can find $K(E_k, R_A)$. The excitation energy distribution, $X(E_x, R_A)$, then follows immediately from the mass equation of fission.

Since the experimental quantity $I(E_I + \Delta, R_A)$ had not been determined accurately for the isotopes that spontaneously fission, Leachman used the results for the thermal-neutron-induced fission of Pu^{239} shown in Fig. 1 in all these cases. The mass distribution of the heavy fragment was considered fixed as the different fissioning isotopes were considered, which is in agreement with the available data.

Since it is not known how the excitation energy is distributed between the two fragments, it was assumed that the excitation probability was the same for both the light and heavy fragment. Then we have

$$X(\delta^H, \delta^L, E_x, R_A) = \int_{-\infty}^{\infty} dE_x \, {}^L X^L(\delta^H \delta^L, E_x{}^L, R_A) X^H(\delta^H, \delta^L, E_x - E_x^L, R_A), \tag{4}$$

where both X^L and X^H can now be determined by extensive calculations and where X^L and X^H are represented by a summation of gaussian functions.

If we then assume that these excited fragments lose their energies through neutron emission as long as it is energetically possible, we can relate the probability for the emission of a given number of neutrons to a given excitation energy. In order to do this we must know the neutron binding energies, which we obtain from the extension of the semiempirical mass surface, and the probability that the neutron will carry off an amount of kinetic energy ϵ. This is determined from the neutron-evaporation relation[57] for the emission probability $N(\epsilon)$ of neutrons of energy ϵ,

$$N(\epsilon) \approx \epsilon e^{-\epsilon/\theta}, \tag{5}$$

where θ is the nuclear temperature, which is considered constant for simplicity. The best value for θ in the energy region considered was determined from (n, 2n) excitation function measurements[58-60] to be 1.4 Mev.

These probabilities $N^L(\delta^L, E_x^L, \nu^L, R_A)$ and $N^H(\delta^H, E_x^H, \nu^H, R_A)$ for the emission of ν^L and ν^H neutrons for a given E_x^L and E_x^H by the light and heavy fragments, respectively, are combined then with the excitation energy distributions X^L and X^H. The resulting probability for emitting ν^L neutrons from the light fragment is

$$P^L(\delta^H, \delta^L, \nu^L, R_A) = \int_{-\infty}^{\infty} dE_x^L X^L(\delta^H, \delta^L, E_x^L, R_A) N^L(\delta^L, E_x^L, \nu^L, R_A), \tag{6}$$

with a similar expression holding for the heavy fragment.

These probabilities must be combined if they are to be compared with experiment:

$$P(\nu, R_A) = \sum_{\eta=0}^{\nu} P^L(\eta^L, R_A) P^H(\nu - \eta^H, R_A). \tag{7}$$

The probabilities for the three mass ratios considered in the calculations for a given isotope are then combined after proper weighting to give $P(\nu)$.

The Δ term, contained in Eqs. (2) and (3), rather than merely correcting for ionization defect, is used to fit the average number of neutrons per fission, $\bar{\nu}$, found from the theoretically calculated $P(\nu)$,

to that observed experimentally. This is primarily necessary because of the errors in the determination of the fission fragment masses. As an example Leachman found it necessary to use a Δ of 17.2 Mev for Pu^{239} rather than an experimentally determined Δ of 11.6 Mev.

The comparison of the calculated $P(\nu)$ with the empirically determined $P(\nu)$ is given in the Discussion.

Experimental Apparatus

The nuclide to be investigated was mounted as a very thin sample upon a platinum foil, which served as the cathode in a 3-inch-diameter parallel-plate fission chamber. The fission chamber was positioned at the center of the scintillator tank by a 3-inch-diameter well along the axis of the tank. The neutron detector is nearly identical to that built at Los Alamos.[32*] The tank is 30 inches long and 30 inches in diameter, with 1/4-inch steel walls. The inside surfaces have been sprayed with molten aluminum as a reflecting and protective coating. Although the reflectivity of aluminum is less than that of some other types of coatings, it was sufficient for our purposes and has shown no tendency to deteriorate over long periods of time.

The scintillator consists of toluene, cadmium propionate, p-terphenyl, methanol, and alpha-NPO, and is mixed as described by Reines et al.[32] The cadmium-to-hydrogen atom ratio used is 0.0019, resulting in a mean capture time for the neutrons, after thermalization, of approximately 10 microseconds (Fig. 2).

The curved surface of the tank has ninety 0.25-in. glass windows of 2-1/8 in. diameter, sealed with neoprene O-rings. A Dumont 6292 photomultiplier tube, mounted in a soft steel collar, is placed against the outside of each window, with the cathode end immersed in mineral oil to give good optical contact. The photomultiplier tubes are wired in parallel in two banks of 45 tubes each, both banks observing all portions of the scintillator. A copper shield is placed around the entire photomultiplier tube assembly, increasing the assembly diameter to 48 inches. The entire apparatus is surrounded by 2 in. of lead and 0.5 in. of boric acid powder, the boric acid powder external to the lead. In order to keep the background low and constant, the apparatus was gated off during all cyclotron and linear accelerator beam pulses.

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* The use of a loaded scintillator tank as an efficient neutron detector at this laboratory was conceived by Dr. Walter E. Crandall for use on another project. While the Crandall tank was still in the early stages of development, information was received about the Los Alamos tank, and its design was utilized.

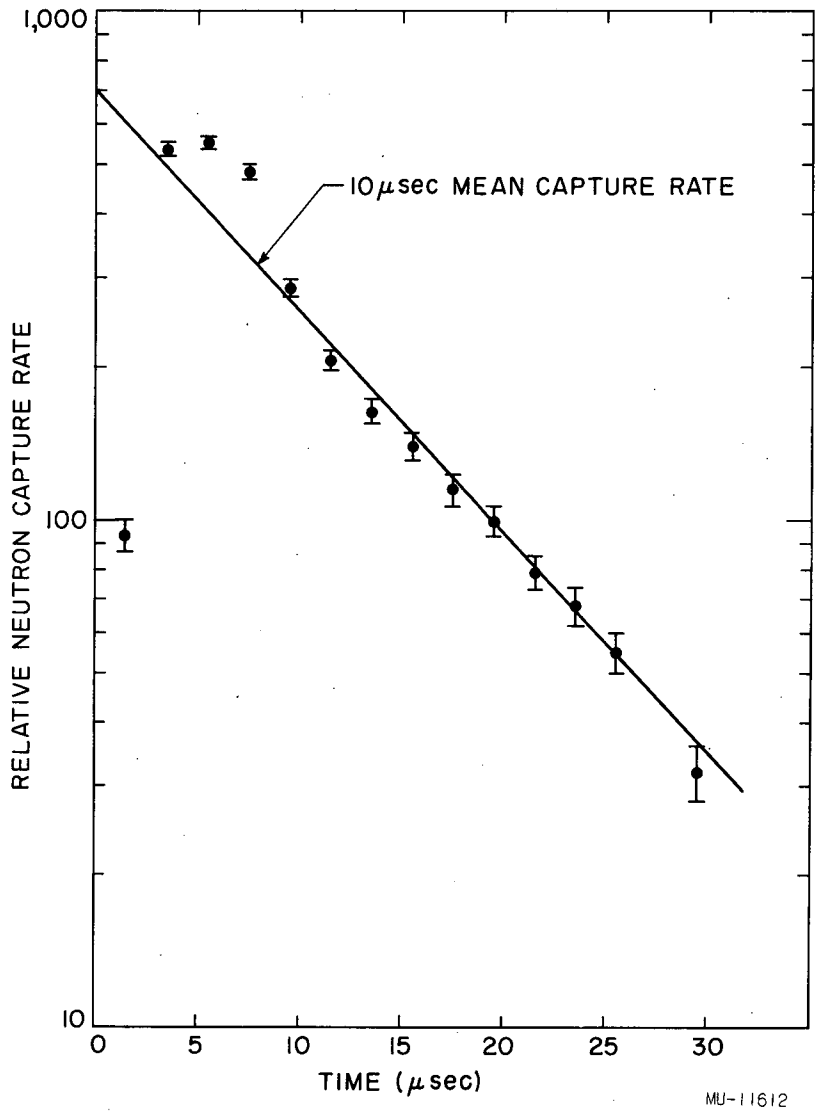


Fig. 2. Experimental capture rates of fission neutrons in the liquid scintillator tank.

The external wiring diagram is shown in Fig. 3. The phototube linear amplifiers have a low-noise, wide-band 950-ohm triode input, matching the 950-ohm 1-microsecond delay line. The fission chamber amplifier and preamplifier are of a standard design. The pulses from the fission chamber and photomultiplier tubes are displayed on the sweep of a Tektronix Model 517 oscilloscope, the sweep speed being exponential, i.e.,

$$x = x_0(1 - e^{-t/\tau}),$$

where x is the distance that the sweep has progressed in time t. The sweep duration is set at 30 microseconds and the mean time τ is set at 10 microseconds. A Dumont Model 304 oscillographic record camera was used to record the data. The data were read after projection with a Recordak film projector.

The isotopes studied were Pu^{236}, Pu^{238}, Pu^{240}, Pu^{242}, Cm^{242}, Cm^{244}, Cf^{252}, and Fm^{254}. The method of production of these isotopes is discussed in a review article by Hyde and Seaborg,[61] and is shown in Table I. The isotopes are created primarily by (n, γ) reactions and β decay.

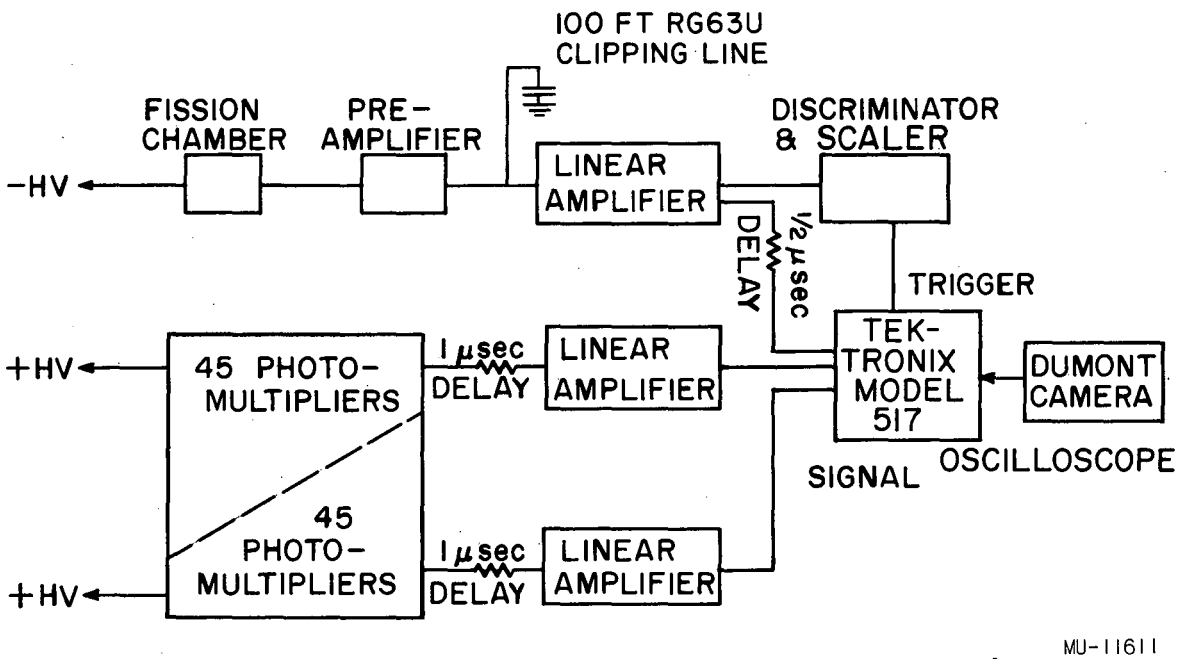
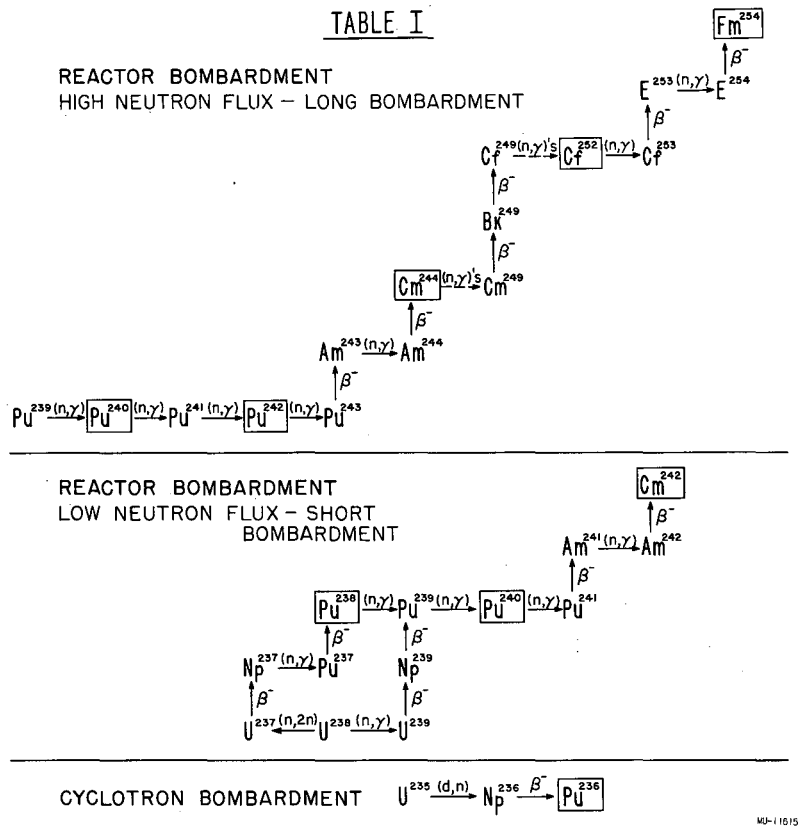


Fig. 3. Wiring schematic.

Table I. Method of production of some transuranic isotopes. The isotopes used in this experiment are indicated by the boxes.



Procedure

The spontaneous fission event occurring in the fission chamber is accompanied by the release of prompt neutrons and γ rays. The neutrons transmit practically all of their energies to recoil protons in a time much shorter than a microsecond, although not all the neutrons are completely thermalized until about 4 microseconds after the event. These recoil protons and any of the converted γ rays from the fission appear as one prompt pulse from the liquid scintillator tank. The thermalized neutrons then are captured exponentially in time by the cadmium-113 (or the hydrogen) in the solution. The Cd^{113} radiative capture immediately gives a cascade decay with a total energy of 9.2 Mev,[62,63] some fraction of which is converted in the tank and gives a pulse indicating the neutron capture. Approximately 15% of the pulses observed resulted from neturon capture in hydrogen.

A fission-chamber discriminator plateau was obtained for each isotope except Pu^{236}, and the discriminator bias was set so that all fissions and no alpha pile-up pulses were observed. In all but 0.3% of the events the fission pulse was accompanied by a prompt tank pulse. With Pu^{236} and Fm^{234}, although the samples were so thin that good discriminator plateaus could in principle have been obtained, the activity was so low as to render this experimentally impractical. In these cases the discriminator bias was set so low that some alpha pile-up pulses were observed, but these were readily distinguishable from the fission pulses by the lack of prompt tank pulse.

When a fission occurs the oscilloscope is triggered and the fission pulse is displayed at the beginning of the sweep. The tank pulses are delayed one microsecond and also displayed on the sweep. Both the fission pulse and the prompt tank pulse were required before any trace was counted as a fission event. This eliminated any chance of mistaking pile-up alpha pulses for fission pulses (Fig. 4).

Californium-252 was run periodically as a calibration for every isotope investigated; this procedure reveals immediately any changes in the over-all efficiency of the apparatus. Experience has shown that after several months the cadmium salt begins to come out of solution,

MU-10453

Fig. 4. Sweep triggered by fission-chamber pulse. Left to right: fission pulse, prompt tank pulse from fission γ rays and recoil protons, and four neutron-capture pulses.

and since appreciable periods of time elapsed between the availability dates of the isotopes, this method of obtaining the relative tank efficiency was the most satisfactory one. In the recent runs the tank efficiency was actually a few percent lower than in the earlier runs. Background pulses with the fission chamber in place were also recorded several times during each run by random triggering of the oscilloscope, using a rate generator.

The absolute tank efficiency for detection of fission neutrons is based on $\bar{\nu}$ (the mean number of prompt neutrons emitted per fission) for Pu^{240} obtained by Diven et al.;[64] this value was measured relative to $\bar{\nu}$ for the fission of U^{235} by thermal neutrons.[30] The absolute tank efficiency at the time the Pu^{240} was run was 76.9 ± 2.2%.

There are various reasons for the tank efficiency to be less than 100%. Some of the neutrons escape from the tank without being captured. Some are captured but give a capture gamma-ray pulse too small to be observed since an insufficient amount of the gamma-ray energy is converted to electron kinetic energy. Because the background counting rate increases rapidly as smaller pulses are accepted, a lower pulse-height limit was set, and this caused the loss of all neutron pulses smaller than this lower limit. The differential pulse-height distribution of the pulses on fission traces is given in Fig. 5, which also shows the lower acceptance limit. Since the oscilloscope sweep length was only 30 microseconds, approximately 7% of the neutrons captured in the tank were captured after the sweep had ended.

Because a possibility exists that the number of neutrons per fission event is correlated with the neutron kinetic energy, the neutron-capture efficiency of the scintillator tank as a function of the neutron energy must be considered. Monte Carlo calculations performed at the University of California Livermore Laboratory show that the efficiency is about 99% for 1-Mev neutrons, 95% for 3-Mev neutrons, 89% for 5-Mev neutrons, and 84% for 7-Mev neutrons, when the neutrons are emitted isotropically at the center of the tank. If it is assumed that the energy spectra of the neutrons from all the isotopes approximate that from the fission of U^{235} by thermal neutrons, an over-all efficiency for capturing a fission neutron in the scintillator is calculated to be

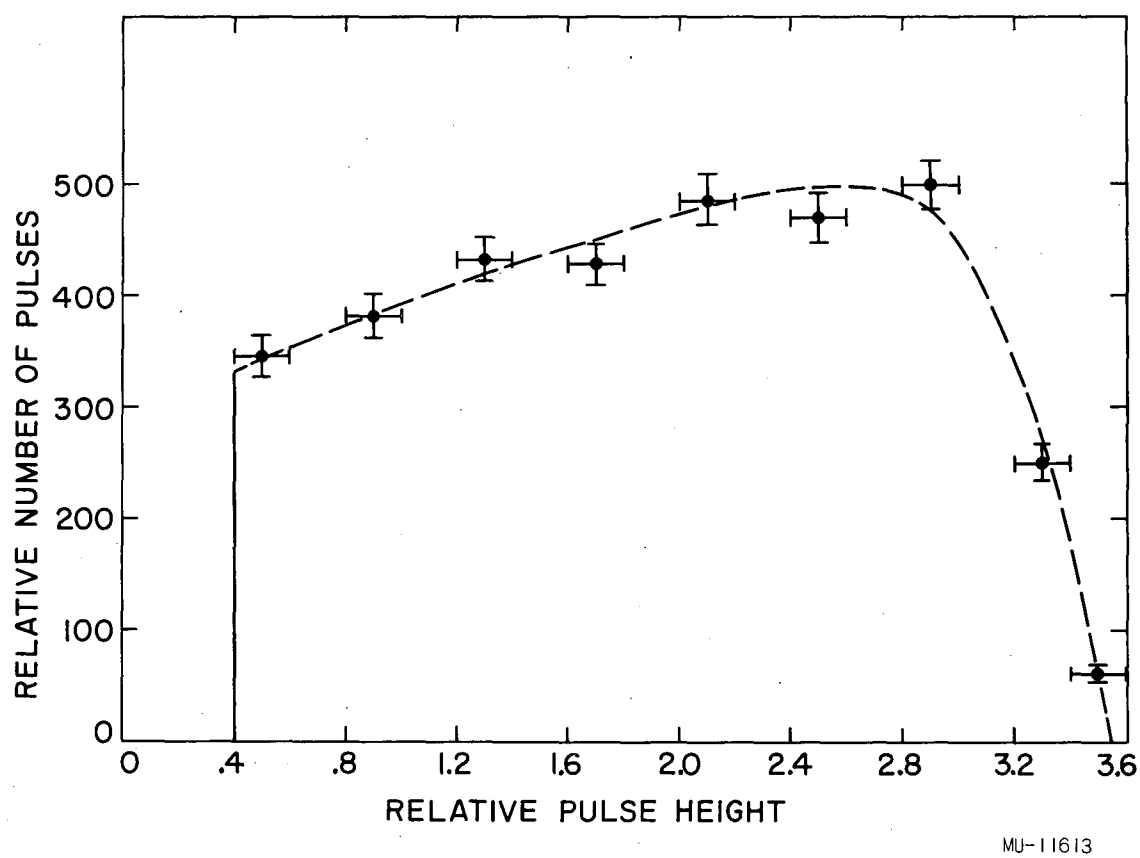


Fig. 5. Differential pulse-height distribution of the neutron-capture pulses on fission traces. Pulse heights less than 0.4 were not read because of rapidly increasing background in this region.

about 95%. Theoretical considerations by R. B. Leachman indicate that the above assumption is quite accurate. In addition, preliminary results of an experiment being performed at the Nobel Institute[65] indicate that the peak of the energy spectrum of the neutrons from the spontaneous fission of Cf^{252} is only slightly higher than the peak energy for U^{235} fission by thermal neutrons.

If all the high-energy neutrons originated from fissions giving only a single neutron, some bias would be expected in the data. Hammel and Kephart,[35] however, working with a 16-in.-diameter tank, have obtained emission probabilities of neutrons from Pu^{240} fission that are in good agreement with those obtained in this work, and since their capture efficiencies for high-energy neutrons are considerably less than for the larger tank, it can be concluded that no very marked relation exists between neutron energy and number of neutrons per fission.

Analysis of Data

The observed numbers of fissions giving 0, 1, 2, . . ., pulses per sweep are given in Table II. Two corrections are made before the true multiplicity distribution is calculated.

(a) "Resolving time." Because of the exponential sweep the neutron-capture pulses are uniformly distributed distancewise along the sweep trace, so it is better to use the "resolving distance" ΔL. The probability that two pulses fall within a cell ΔL long is

$$2\Delta L \int_0^L \frac{1}{L^2}\, dx = 2\, \Delta L/L,$$

where L is the total sweep length. If n pulses are observed on a trace it is possible that there were n pulses, all of them resolved, or that there were n + 1 pulses, two of which are not resolved. Higher-order corrections have been neglected. If $F''(n)$ is the number of fissions observed with n pulses per fission, and $F'(n)$ the corresponding number for $\Delta L = 0$, then we have

$$F''(n) = F'(n) \left[1 - \frac{n!}{(n-2)!\, 2!} \frac{2\Delta L}{L} \right] + F'(n+1) \frac{(n+1)!}{(n-1)!\, 2!} \frac{2\Delta L}{L} .$$

The $F'(n)$ were then calculated, with $2\Delta L/L = 0.0030 \pm 0.0015$, and with the value for ΔL estimated from the appearance of the sweeps. The separation of two pulses was measured for a number of cases, at all positions along the sweep, where two pulses could just be identified as separate pulses. It was possible to achieve the accuracy indicated above, despite the fact that the two pulses could not be completely resolved, because a bright spot and a break in the rise of a pulse were observed when another pulse occurred.

(b) Background. The background count rate γ was about 0.01 pulse per sweep. The corrected number of fissions is calculated from

$$F(n) = \left[F'(n) - \gamma F(n-1) \right] / (1-\gamma).$$

Table II

Observed neutron multiplicity distributions from spontaneous fission and background pulses per sweep

	Pu^{236}	Pu^{238}	Pu^{240}	Pu^{242}	Cm^{242}	Cm^{244}	Cf^{252}	Fm^{254}
F_0''	42	642	368	471	569	999	384	—
F_1''	85	1532	1067	920	2495	4259	2519	—
F_2''	78	1450	1086	767	3584	5814	6218	—
F_3''	31	620	553	282	2312	3750	7687	—
F_4''	7	179	174	44	723	1174	5159	—
F_5''	1	25	20	10	159	188	2006	—
F_6''	0	0	1	0	13	16	525	—
F_7''	0	0	0	0	2	0	73	—
F_8''	0	0	0	0	0	0	8	—
Total	244	4448	3269	2494	9857	16200	24579	870
Background counts per sweep	0.0106	0.0108	0.0108	0.0119	0.0109	0.0081	0.0150	0.005

The number of background sweeps with two or more pulses was completely negligible. The background rates for the various isotopes are given in Table II.

If $P(\nu)$ is the true multiplicity distribution, $F(n)$ is given by

$$F(n) = \sum_{\nu=n}^{\nu=\nu_{max}} P(\nu)\ \epsilon^{n}\ (1-\epsilon)^{\nu-n}\ \frac{\nu!}{n!\ (\nu-n)!},$$

where ϵ is the absolute efficiency of the tank. It can be shown that inversion of the above set of equations leads to

$$P(\nu) = \sum_{n=\nu}^{n=n_{max}} F(n)\ \frac{n!}{\nu!\ (n-\nu)!}\ \epsilon^{-n}\ (\epsilon-1)^{n-\nu}.$$

The errors on the calculated $P(\nu)$ have five sources: (a) counting statistics, (b) uncertainty in the efficiency because of the uncertainty of $\overline{\nu}$ for Pu^{240}, (c) background fluctuations (which are negligible), (d) uncertainty in the value of ΔL used in the resolution corrections, and (e) reading errors. (There are small differences in the data obtained when one person reads the film twice, and similar fluctuations among different readers. All film has been read at least twice. The uncertainty is small compared with (a), (b), and (d).)

These calculations were made on an IBM Type 650 calculator at Livermore. The calculated values of $P(\nu)$ and their standard errors are given in Table III.

Table III

Calculated probabilities of emitting ν neutrons per spontaneous fission, $P(\nu)$, and the average number of neutrons per spontaneous fission, $\bar{\nu}$, based on $\bar{\nu} = 2.257 \pm 0.046$[16] for Pu^{240}

$P\nu$	Pu^{236}	Pu^{238}	Pu^{240}	Pu^{242}	Cm^{242}	Cm^{244}	Cf^{252}	Fm^{254}
P_0	0.062 ±0.035	0.044 ±0.009	0.041 ±0.009	0.063 ±0.013	0.011 ±0.005	0.001 ±0.004	0.001 ±0.001	
P_1	0.156 ±0.090	0.175 ±0.026	0.219 ±0.021	0.192 ±0.034	0.126 ±0.018	0.099 ±0.017	0.021 ±0.007	
P_2	0.38 ±0.13	0.384 ±0.026	0.351 ±0.021	0.351 ±0.041	0.323 ±0.018	0.281 ±0.022	0.111 ±0.019	
P_3	0.28 ±0.12	0.237 ±0.027	0.241 ±0.020	0.324 ±0.047	0.347 ±0.020	0.365 ±0.018	0.271 ±0.019	
P_4	0.096 ±0.086	0.124 ±0.021	0.127 ±0.018	0.033 ±0.026	0.139 ±0.013	0.198 ±0.220	0.326 ±0.018	
P_5	0.033 ±0.036	0.036 ±0.009	0.020 ±0.006	0.036 ±0.013	0.050 ±0.009	0.049 ±0.009	0.178 ±0.016	
P_6	—	—	0.001 ±0.002	—	0.004 ±0.002	0.007 ±0.002	0.077 ±0.013	
P_7	—	—	—	—	0.001 ±0.001	—	0.013 ±0.004	
P_8	—	—	—	—	—	—	0.003 ±0.001	
$\bar{\nu}$	2.30 ±0.19	2.33 ±0.08	2.257 ±0.046	2.18 ±0.09	2.65 ±0.09	2.84 ±0.09	3.82 ±0.12	4.05 ±0.19

Discussion

The multiplicity distributions of Pu^{240}, Cm^{242}, and Cf^{252} are shown in Fig. 6. The distributions of the Pu^{236}, Pu^{238}, Pu^{242}, and Cm^{244} are similar to those shown in the figure and are given in Table III. The distribution for Fm^{254} is not given because of the low number of fissions observed and because of a need to correct for a Cf^{252} contamination. Only $\bar{\nu}$ for Fm^{254} has any significance.

For certain practical applications it is advantageous to find a mathematical distribution that will approximate the observed results. Points obtained from the binomial distribution

$$B(\nu) = \frac{\nu_{max}!}{\nu!\,(\nu_{max}-\nu)!} \left(\frac{\bar{\nu}}{\nu_{max}}\right)^{\nu} \left(1 - \frac{\bar{\nu}}{\nu_{max}}\right)^{\nu_{max}-\nu}$$

also are shown in Fig. 6. It can be seen that this distribution gives a fair approximation to the experimental points.

A comparison between the experimental results and Leachman's theoretical calculations is shown in Fig. 7. It should be noted that Leachman's calculations were fitted to an approximate empirical distribution obtained before the final efficiency of the tank was determined, so that the number used for Δ (see page 13) is slightly in error. However, the correction is small and would have essentially no effect on the good agreement shown.

To summarize--we have considered the energy released because of the mass difference between the fissioning nucleus and the fission fragments for a given mass ratio. Subtracting the experimentally observed total kinetic energy distribution (corrected for instrumental dispersion) for this mass ratio, from the total energy available leads to distributions in the excitation energies of the fission fragments. Combined with simple neutron evaporation theory and grouped with similar calculations for other mass ratios, these excitation-energy distributions give the neutron-multiplicity distribution for a given fissioning isotope. A comparison with experiment indicates that this simple description is capable of representing this complicated aspect of the fission process.

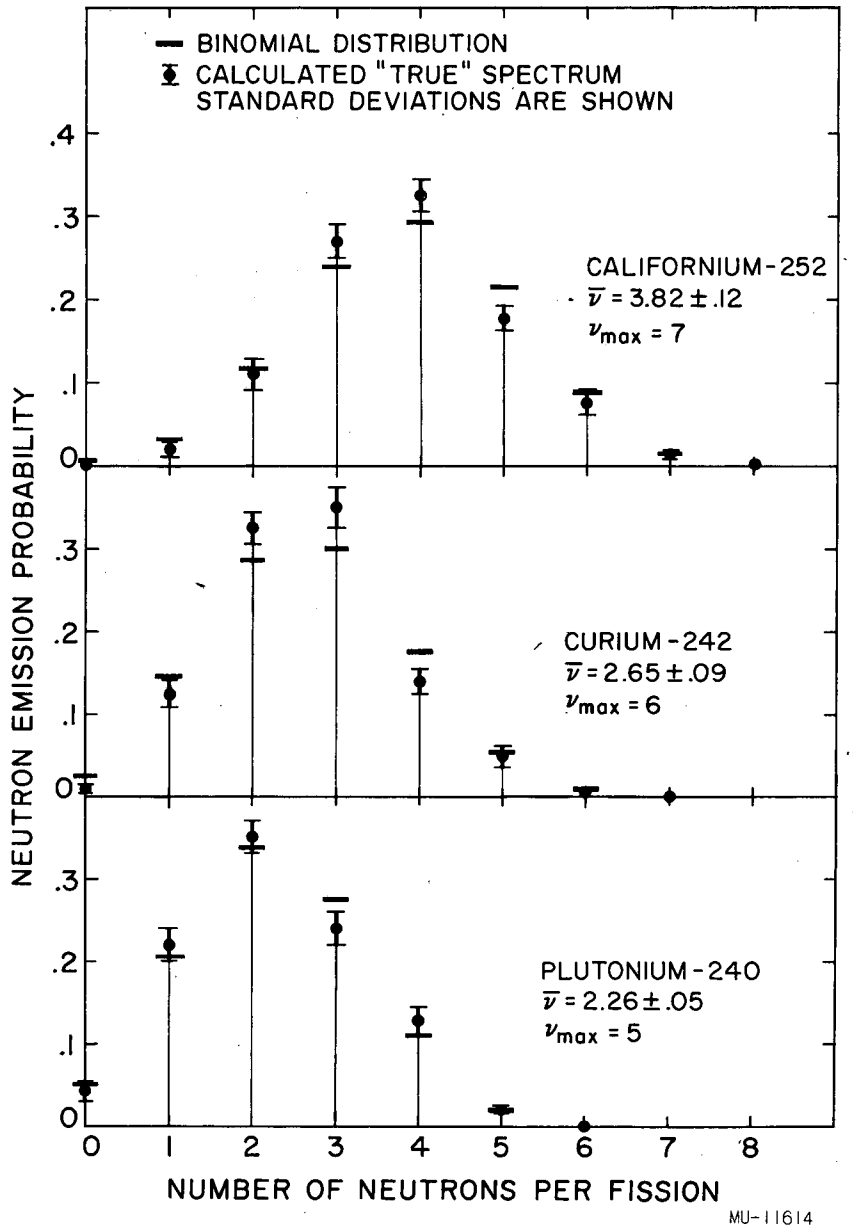


Fig. 6. The probabilities for prompt-neutron emission from the spontaneous fission of plutonium-240, curium-242, and californium-252. Binomial distributions approximating the emission probabilities are also shown.

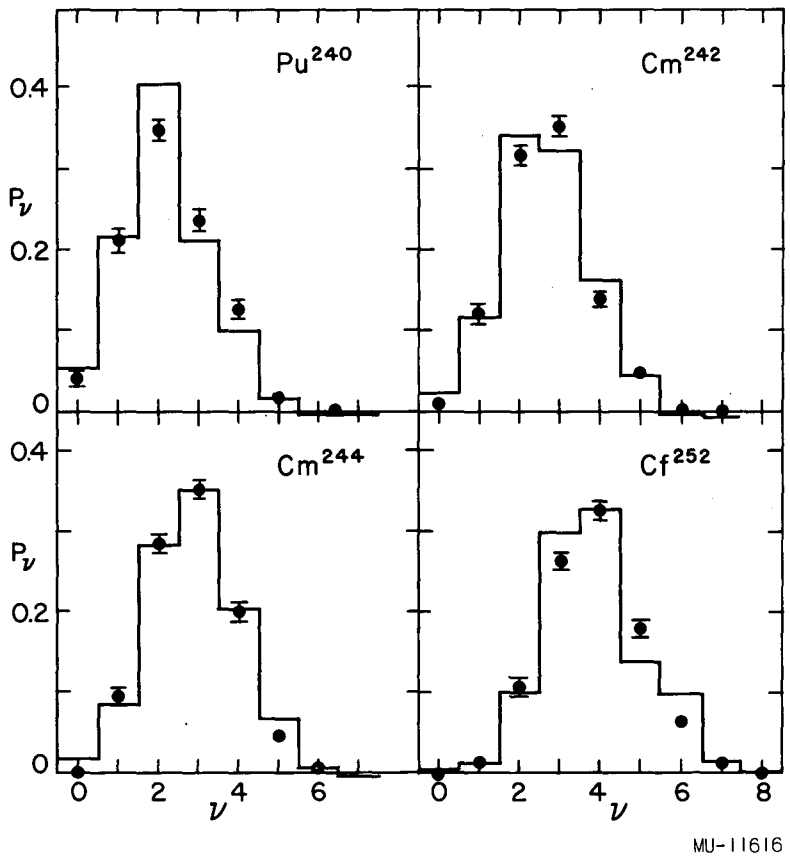


Fig. 7. Comparison between Leachman's theoretical determination of the neutron emission probability (shown as a histogram) and the experimental results. (See Discussion.)

Acknowledgments

The author wishes to express his appreciation to Dr. Robert V. Pyle and Dr. John Ise, Jr., his collaborators in this experiment. The interest and assistance of Professor Burton J. Moyer and Dr. Walter E. Crandall, and the continuing encouragement of Dr. C. M. Van Atta, are gratefully acknowledged.

We were particularly fortunate in having the cooperation of the University of California Radiation Laboratory Nuclear Chemistry Group, under the direction of Professor Glenn T. Seaborg, which supplied us with the eight spontaneously fissioning isotopes. In particular I wish to thank Gregory Choppin, Bernard Harvey, Albert Ghiorso, Stanley Thompson, Richard Glass, Gary Higgins, and William Crane, of this group.

Useful discussions were held with the Los Alamos group under Dr. B. C. Diven, and with George Millburn of this laboratory.

I am greatly indebted to William Goldsworthy for his aid with the electronics problems.

The film was read by David Johnson and the machine calculations were made by James Baker and Francis Halpern. The Monte Carlo calculations were performed under the direction of Edward Lechan.

This work was done under the auspices of the U. S. Atomic Energy Commission.

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