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SPONTANEOUS FISSION OF SOME HEAVY ISOTOPES




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# Ernest O. Lawrence Radiation Laboratory



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# SPONTANEOUS FISSION OF SOME HEAVY ISOTOPES

Reinhard Brandt

(Ph.D. Thesis)

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## Contents

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ABSTRACT

A back-to-back semiconductor counter system was used to study the energy and mass distributions of the fragments in the spontaneous fission of $Fm^{254}$, $E^{253}$, $Cf^{254}$, $Cf^{250}$, and $Cm^{248}$. The results are compared with the spontaneous-fission properties of $Cf^{252}$, which is used as a standard.

All distributions (including those of the odd-mass isotope $E^{253}$) are rather similar, but not identical with the standard. The total kinetic energy of the fragments increases with Z. The asymmetry of the mass distribution shows only small differences between the isotopes. The variances (widths) of all distributions increase with Z and seem to increase with A.

A new fermium isotope with a spontaneous-fission half life of $(11^{+10}_{-6})$ days has been observed. The mass is most likely 258 or 257. The spontaneous-fission half life of $(1.0^{+0.6}_{-0.3}) \cdot 10^4$ y has been observed for $Fm^{255}$. The $Cf^{254}$, spontaneous-fission half life has been redetermined and is 60.5 ± 0.2 days. The alpha/fission ratio in the decay of $Cf^{250}$ has been redetermined and is (1330±45). The redetermined alpha-decay half life of $Fm^{255}$ is (19.9±0.3) h.

# I. INTRODUCTION

Shortly after the discovery of slow-neutron induced fission of $U^{235}$ in 1938 by Hahn and Strassmann[1] it was reported in 1940 by Petrzhak and Flerov[2] that $U^{238}$ fissions spontaneously into two medium-light nuclei. Since 1942 a total of 32 spontaneously fissioning nuclei have been discovered. The most important of those at present is $Cf^{252}$ which has been investigated very extensively (a recent summary of our knowledge about spontaneous fission has been given by Hyde[3]).

Some general, experimentally observed features of spontaneous fission are as follows:

(a) The half-lives range from $\sim 10^{17}$ y for $U^{235}$ down to $\sim 6$ sec for the isotope $102^{254}$. The half-lives decrease rapidly with increasing Z.

(b) The mass and energy distribution in spontaneous fission resembles very closely the mass and energy distribution of slow-neutron induced fission. The complementary fission fragments are predominantly unequal in mass. There is a fairly wide distribution of masses about the two most probable masses and there is also a wide distribution of kinetic energies for any mass group.

(c) The kinetic energies of the single fission fragments range from ~50 MeV to ~120 MeV and the total kinetic energy per fission is roughly 180 MeV.

(d) The moving fragments emit neutrons and prompt gamma rays.

(e) Occasionally fission occurs into 3 charged fragments one of which is usually an alpha particle.

A serious limitation in obtaining experimental data exists due to the fact that elements which are available in rather large quantities, such as uranium or plutonium, show extremely low specific activity rates for spontaneous fission. (For elements with $Z < 92$ the specific activity is so low as to be unobserved so far.) The specific activity for spontaneous fission increases considerably for elements near californium, but isotopes of these elements are generally available only in minute quantities, and their half lives are relatively short.

Spontaneous fission is a rather complex process when considered from a theoretical point of view. To illustrate this fact we refer to Fig. 1 which shows the original nucleus having a certain deformation and a certain potential energy $E_o$. When the nucleus is deformed slightly the potential energy of the system increases. (This accounts incidentally, for the fact that the nucleus does not undergo fission instantaneously.) When the deformation of the nucleus is increased further the potential energy of the system increases at first, but finally decreases. The point with the highest possible potential energy is called the saddle point. Increasing deformation leads finally to the separation of the two fragments at the so-called "scission point". Since the original nucleus has no excitation energy in spontaneous fission it has to penetrate the potential energy barrier.

A complete theoretical investigation has to describe this barrier penetration problem, in particular it has to describe:

(a) The potential energy curve and the corresponding deformation (this is a static problem).

(b) The dynamics have to be considered, since the nucleus is rapidly changing its shape during the deformation process.

The nuclear forces are however too complicated to allow an exact mathematical solution of this problem. Therefore some simplified models have to be found to describe the spontaneous fission process.

Since this is a problem of nuclear physics, it is obviously necessary to use the models developed in nuclear physics. One such model is the liquid drop model which assumes that the nucleus is uniformly charged and incompressible. But of course it is also possible to assume that the particles move independently of each other as it is done in the single particle model. A combination of the collective effects and single particle effects are used in the unified model (These models and their relevance to fission are discussed by Hyde[3]).

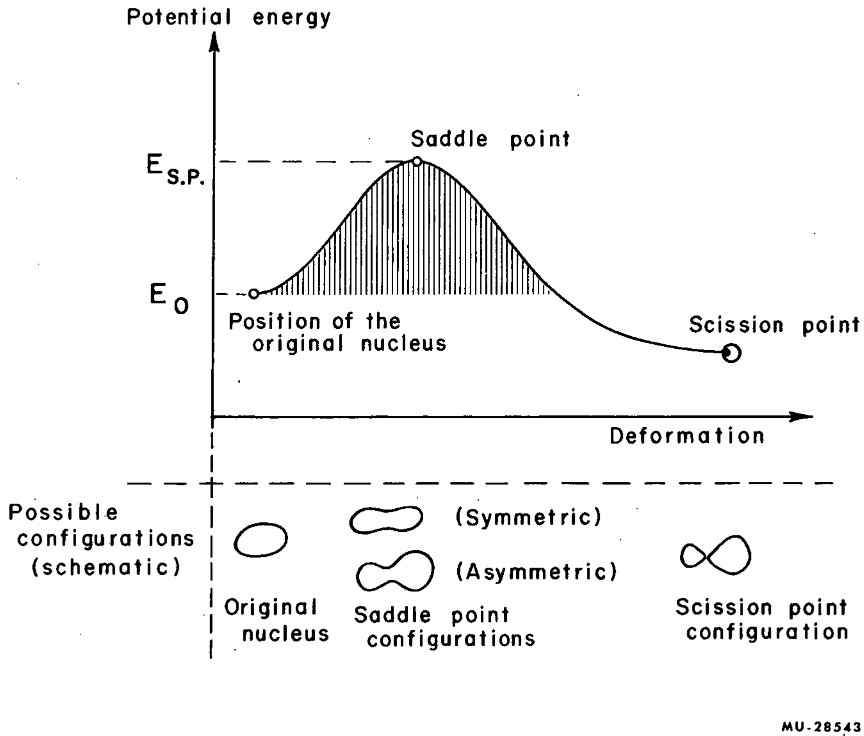


Fig. 1. Spontaneous fission as a barrier penetration problem. The nucleus has to penetrate the shaded area.

Historically, the first nuclear model applied to the interpretation of the fission process was the liquid-drop model. (Bohr and Wheeler.[4]) They introduced in particular the so-called "fissionability parameter X" of a nucleus. The nuclear attractive forces can be idealized as the surface tension of the nucleus, which is proportional to $A^{2/3}$ according to the liquid-drop model. The nuclear disruptive forces are due to the Coulomb repulsion forces between the protons and this can be idealized as proportional to $Z^2/A^{1/3}$. Bohr and Wheeler suggested that the ratio between the nuclear disruptive forces and nuclear attracting forces would be a good measure of the "fissionability" X of the nucleus.

$$X \sim \frac{Z^2/A^{1/3}}{A^{2/3}} = \frac{Z^2}{A}. \qquad (1)$$

Since that time the parameter X or its equivalent $Z^2/A$ is widely used in the discussion of fission properties.

Later, the liquid-drop model was worked out in considerably more detail by Swiatecki. He and his co-workers calculated in particular the potential energy of a charged liquid-drop at various deformations (see for example reference 5, where one may find references about other authors in this field). The results of these calculations seem to be useful for the interpretation of some of the results of induced fission in the region of elements below bismuth. In this region the nuclei have a comparably smaller charge (a low X-value) and the calculated saddle point configurations show a rather small "neck". The difference in shape between the saddle point and the scission point is also rather small. Thus the calculated potential energy maps might be [illegible] sufficient to describe some features of the fission process in this region. In particular, these liquid-drop calculations allow the interpretation of the predominance of symmetric fission in the region of low X.

The liquid-drop model however has so far been unable to explain the results of spontaneous fission. The spontaneously fissioning nuclei exhibit a rather high X-value. In this region the calculations of

Swiatecki also show that the saddle point configurations are symmetric and are rather close to the shape of the original nucleus. In this case the "neck" is thick and the "distance" to be travelled by the nucleus between the saddle point and the scission point is rather large. Here it seems that the dynamics of the process may become very important. However, dynamical processes have so far not been included in the calculations. The liquid drop model as further developed by Swiatecki is therefore so far unable to explain the following main features of spontaneous fission.

(a) The asymmetric mass distribution.

(b) The total kinetic energy release.

(c) The widths of the mass and energy distributions.

(Recently Viola et al. have studied the total kinetic energy release for many cases of heavy ion induced fission including the region of Z = 92 and higher.[6] Induced fission gives predominantly symmetric mass distributions and the results for the total kinetic energy release can be interpreted on the assumption that two spheroids of equal size are close to each other at the moment of scission. In spontaneous fission, however, the two fragments are of unequal size at the moment of scission.)

Several authors have suggested that the liquid drop model is much too oversimplified and have proposed a "more realistic model" to interpret the asymmetric mass distribution in spontaneous fission. It seems useful in particular to describe the attempt of Johansson to interpret the asymmetry in fission.[7] He uses the collective model for which Nilsson was able to calculate the energy levels of the single nucleons at various deformations of the total nucleus.[8] Johansson showed that the interaction between levels of opposite parity lower the potential energy when the nucleus is asymmetrically deformed. This implies that the nucleus is asymmetrically deformed at the saddle point and that this asymmetric deformation is responsible for the final asymmetric mass split. Johansson then derived a relation between the asymmetric mass split and the fissionability of the nucleus, which will be discussed later.[9] (Swiatecki in one of his older works also assumes an asymmetric saddle point deformation and derives a rather similar relation between the asymmetry and $Z^2/A$.[10]) Other models have also been proposed to explain the asymmetric mass distribution in fission. One example is the work of Fong[13] which was based on the statistical model. Another is the work of Brunner, et al.[14] which puts heavy emphasis on the nuclear attractive forces at scission.

However, a discussion of the variation of the asymmetry of the mass distributions with $Z^2/A$ is limited because the mass-yield curves are known experimentally only for five spontaneous fissioning isotopes. (The summary of those results is given by Hyde.[3]) It seemed useful, therefore, to measure the mass-yield curve for more spontaneously fissioning nuclei. The mass-yield curves can be determined in two ways. The first method involves the measurement of the amount of each fission fragment mass by means of radiochemical techniques. This procedure has the disadvantage - besides its tediousness - that it requires larger amounts of fissioning material than are available at this time for many heavy nuclei. The second method consists of the measurement of the energies of the two fission fragments for each fission event. From those energies the masses can be deduced on the basis of momentum conservation as shown in a following section. The latter method has several advantages. It requires much less material for the determination of a rather accurate mass-yield curve than the radiochemical method. Secondly, it gives not only information about the mass-distribution, but also, about the kinetic energy distribution of the fragments. In addition it gives the total kinetic energy distribution. This is the basis of the experiments which are reported in the first part of this thesis and which consist in the measurement of fission fragment energies of five heavy nuclei.

The method used in this work for the determination of the mass and energy distributions has the disadvantage that it is not extremely accurate since the neutrons which are emitted after the separation of the fission fragments introduce uncertainties. Recently Bowman, et al, made a careful investigation of the details of neutron emission in the spontaneous fission of $Cf^{252}$. They found that the numbers and energies of the neutrons which are emitted by the fragments show a very non-uniform behavior, and vary strongly with the masses and the kinetic energies of the fragments.[15,16] This implies that no simple corrections can be made in most cases for the emission of the neutrons. Further details are discussed in Appendix D.

In the second part of the thesis, some spontaneous fission half-lives are investigated. These spontaneous fission half-lives are interesting from an intrinsic point of view. The interpretation of half-lives from a theoretical point of view, however, is rather difficult due to the problems mentioned previously and it is not surprising that the logarithm of the spontaneous half life log $\tau_{1/2}$ does not vary smoothly with $Z^2/A$ as predicted by the liquid drop model. (Recently it was shown that an even more complex form of the fissionability parameter X did not account for the non-smoothness in the decrease of the logarithm of the half-life with X.[17])

There are, however, two models which attempt to explain the deviations from a smooth dependence of log $\tau_{1/2}$ with $Z^2/A$. One was proposed by Swiatecki.[18] It states that if the ground state masses ($E_o$ in Fig. 1) could be calculated on the basis of the liquid drop model alone, we would observe the smooth dependence of log $\tau_{1/2}$ with $Z^2/A$ in the way the theory predicts. However, it is known experimentally that single particle effects introduce a deviation in the ground state mass, $E_o$, relative to the pure liquid drop mass calculations. This results in a variation in the barrier which the nucleus has to penetrate (See Fig. 1) since the saddle point position is assumingly not affected by this kind of single particle effect.

A second model was proposed by Johansson to interpret the above mentioned deviation from a smooth decrease on the basis of the collective model.[19] According to this model the top pair of nucleons move in an energy level as calculated by Nilsson.[8] During the deformation this top pair of nucleons has to change the energy level in which it is moving and since the different "Nilsson levels" do not have the same slope with varying deformation the total energy of the system is not as smoothly varying with deformation as shown in Fig. 1. This deviation from a smooth curve changes the barrier which has to be penetrated and hence the half-life.

Both models proposed so far account in about the same way for the deviation of log $\tau_{1/2}$ from smoothly varying with $Z^2/A$ and no decision can be made so far which one of the two models is better for the description of the actual behavior.

Finally it may be interesting to consider the methods by which the heavy isotopes can be produced. One way is the bombardment of heavy elements with charged particles of high energy (for example $Cm^{244}$ ($\alpha$, 2n) $Cf^{246}$). Another way is the neutron bombardment of curium isotopes on a "slow" time scale, i.e. in a nuclear reactor. In this way isotopes as heavy as $Fm^{257}$ or $Fm^{258}$ have been produced.[20,21] The third way is the bombardment of uranium with neutrons on a "fast" time scale, i.e. in a thermonuclear explosion[(22)] which also leads to the production of elements at least as heavy as $Fm^{255}$. All three methods mentioned so far are probably responsible for the production of all nuclei out of hydrogen in stars in our Universe (Burbidge, et al.[23]).

The process of neutron capture on a "fast" time scale is of particular interest here. Burbidge, et al. suggested that a Type-I supernovae explosion produces a neutron flux similar to a neutron flux in certain thermonuclear explosions here on Earth. Therefore, heavy isotopes, especially $Cf^{254}$, which are produced in those thermonuclear explosions might also be produced in Type-I supernovae explosions.

The light-curve of Type-I supernovae decreases exponentially with a half-life of (55±1) day. Burbidge, et al. suggested that the energy released in spontaneous fission of $Cf^{254}$ might be responsible for the light output, since $Cf^{254}$ decays with approximately the same half life.[27] It seemed, therefore, interesting to remeasure the $Cf^{254}$ spontaneous fission half-life.

# II. ENERGY AND MASS DISTRIBUTIONS IN THE SPONTANEOUS FISSION OF $Fm^{254}$, $E^{253}$, $Cf^{254}$, $Cf^{250}$, and $Cm^{248}$

## A. Experiments

### 1. General Procedures

The production of comparatively large amounts of transcurium isotopes occurred recently as a result of the long-time irradiation of curium isotopes with neutrons, and their transmutation into elements as heavy as fermium.[28] The heavy isotopes investigated here were all produced in this way. Their production path is shown schematically in Table I. The actinides were separated by the use of ion-exchange columns.[20] The contamination of one isotope with spontaneous-fission events from another isotope was determined with standard techniques. Special care was taken to remove all contaminants to such a level that their influence could be neglected in the final results. The isolated and purified samples were electroplated on 5-μin-thick Ni foils. (For details see Appendix A.) The foils were placed between two back-to-back phosphorus-diffused silicon semiconductor counters, which allowed measurement of the energies of both fragments of each fission event. Each fission fragment produced an electronic pulse proportional to its energy in the semiconductor. This electronic pulse was linearly amplified and its binary equivalent stored on paper tape. The data recorded on paper tape were then transferred to magnetic tape in a form that retained the identity of each fission event and was directly acceptable by the IBM 7090 computer.

Knowing the energies $E_1$ and $E_2$ of both fragments in a fission event, one can compute the masses $M_1$ and $M_2$ and the total kinetic energy ET for this event according to

$$\frac{M_1}{M_2} = \frac{E_2}{E_1} \qquad (2)$$

and

$$E_1 + E_2 = ET. \qquad (3)$$

Table I. Production and activity rates for $Fm^{254}$, $E^{253}$, $Cf^{254}$, $Cf^{250}$, and $Cm^{248}$

| Isotope | Production path | Sample activity at start (Fission/min) | Events recorded | Average contamination with other fission activities (fission 1%) | Total half life |
|---|---|---|---|---|---|
| $Fm^{254}$ | $E^{253}(n,\gamma)E^{254m}$<br>$E^{254m} \xrightarrow{\beta} Fm^{254}$ | ~4000 | 81 900 | < 0.1% | 3.2 h |
| $E^{253}$ | $Cf^{253} \xrightarrow{\beta} E^{253}$ | ~4 | 11 800 | ⩽ 6% $Cf^{252}$<br>⩽ 6% $E^{254}$ | 20.0 d |
| $Cf^{254}$ | $E^{253}(n,\gamma)E^{254m}$<br>$E^{254m} \xrightarrow{E.C.} Cf^{254}$ | ~40 | 83 800 | 4% $Cf^{250}$<br>2% $Cf^{252}$ | 60.5 d |
| $Cf^{250}$ | $E^{254} \xrightarrow{\alpha}$<br>$Bk^{250} \xrightarrow{\beta} Cf^{250}$ | ~5 | 12 100 | 1.5% $E^{254}$<br>< 4% $Cf^{252}$ | 13 y |
| $Cm^{248}$ | $Cf^{252} \xrightarrow{\alpha} Cm^{248}$ | ~400 | 70 000 | (5 ± 1)% $Cm^{244}$ | $4.7 \cdot 10^5$y |

The electronic equipment is described in detail in Appendix B. Appendix C contains the details of the procedures used for the computation of the final results.

The fission-fragment energies of $Cf^{252}$ were used for energy calibrations of the semiconductor fission-fragment detectors, since the absolute value of the fragment energy could not be measured with those detectors. Previously the spontaneous fission of $Cf^{252}$ had been studied by using time-of-flight techniques, which gave the absolute value for the energy distributions.[29] The most probable light- and heavy-fragment energies gave the two calibration points for the linear energy calibration of the solid-state detector. This investigation is only a comparative study between $Cf^{252}$ and the other isotopes. It was assumed that the "pulse-height-defect" as described recently by several authors (for example Haines[30]) was the same for all the isotopes investigated. The details of the energy calibration are given in Appendix D. In order to show how the experiments were done, it seems useful to describe below the $Fm^{254}$ experiment in some detail. The other experiments are then briefly mentioned.

## 2. The $Fm^{254}$ Experiment

The experiment was started by putting a $Cf^{252}$ standard between the two semiconductor counters. In one counter the distribution of fission fragments according to their energy was registered as a distribution in different channels (Fig. 2a). The peak positions of the light and heavy fragments were found to be in channels 75.5 and 51.0. These two channel numbers were associated with the most probable light and heavy fragment energies of 102.8 and 78.8 MeV. The conversion of a channel distribution into an energy distribution was accomplished with the aid of a computer as seen in Fig. 2b. (A refined conversion of the channel distribution into an energy distribution was carried out later. It used the mean values of the distributions and is described in Appendix D.) Both counters were calibrated in this way with about 10 000 events. The $Cf^{252}$ standard was then replaced by the $Fm^{254}$ source, and the fission fragments of this isotope were registered. $Fm^{254}$

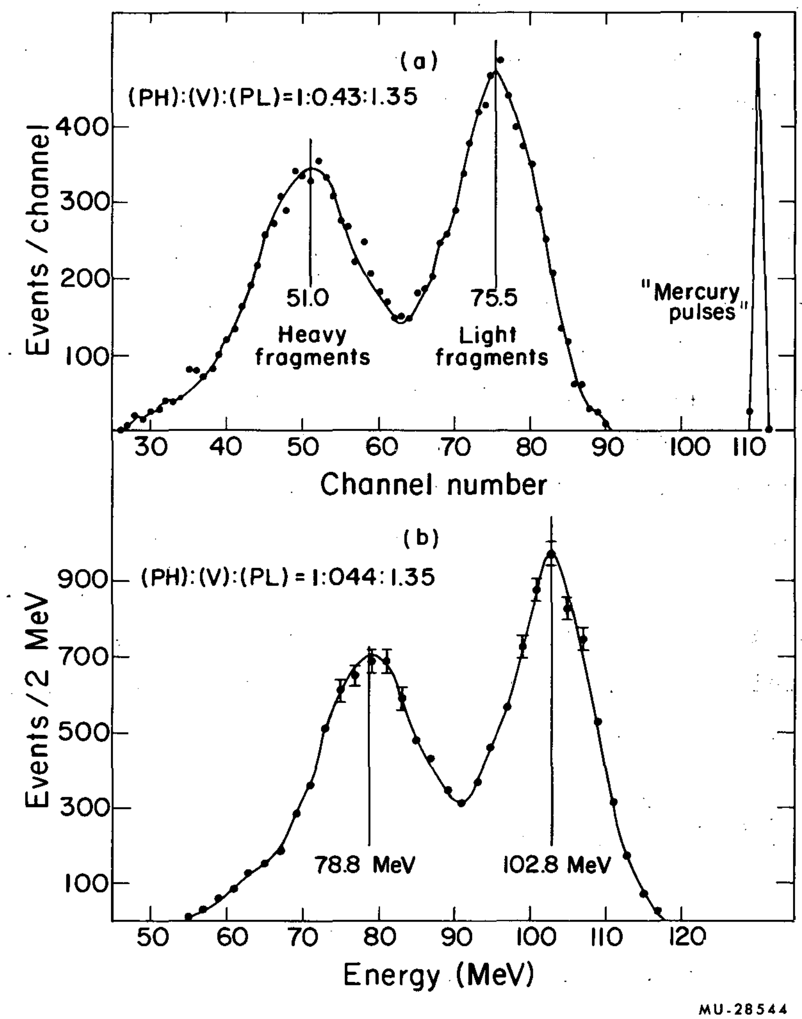


Fig. 2. Channel distribution (a) and corresponding energy distribution (b) of single fission fragments for $Cf^{252}$. (PH):(V):(PL) is the ratio of the number of events at the peak of the heavy fragment, the valley between both peaks, and the peak of the light fragment.

has a total half life of ~3 h; so the source decayed to insignificance within 1 day. A post-calibration was carried out with a $Cf^{252}$ source.

The stability of the system was checked in a twofold way. At first the position of the most probable light fragment in the precalibration and post calibration was determined (it shifted usually less than 0.5 channels/day). Thus the position of the 102.7 MeV point in the channel distribution of the $Fm^{254}$ run could be determined. In addition, the stability of the electronic equipment during the experiment was checked continously with a pulser. Corrections could also be made for any instabilities in the electronic system. The $Fm^{254}$ investigation was completed with two 1-d experiments.

3. Experiments with Other Isotopes

The $Cf^{254}$ experiment was done in essentially the same way as the $Fm^{254}$ investigation. Here several different sets of detectors were used, which all gave similar results.

The $Cf^{250}$ experiment was carried out with a rather small source of 5 fissions/min as compared to 4000 fissions/min used at the start of the $Fm^{254}$ experiment. The solid-state detectors used for this experiment were also relatively poor. This accounts for the large uncertainties in the results for this isotope as shown in a following section.

In the case of $Cm^{248}$ a source of 400 fissions/min was available for the study of fission fragment energies. It was investigated by two methods using different electronic techniques. The first was similar to those used previously. The second method attempted to minimize the effect of the ~$10^4$ alpha particles/sec which came from the decay of $Cm^{244}$ also present on the foil. The electric pulse coming from the preamplifier was shortened in its width from ~5 to ~0.4 μs, thus reducing considerably the chance that an accidental alpha particle could add its energy to the measured fission-fragment energy and distort the results. (For more details see Appendix B.) Both methods used for $Cm^{248}$ gave the same results. Thus far $E^{253}$ is the first odd-mass isotope investigated with respect to its spontaneous-fission fragment energies. It exhibits a very high alpha-to-fission ratio. This means that a very strong alpha-activity must be present to give a reasonable fission counting rate. Therefore the above-mentioned method employing pulse shortening was used to measure the fission-fragment energies. The high alpha-counting rate shifted the base-line in the electronic system, and this was measured as a shift in position of the pulser pulses which enabled a correction for the shift to be made. The accuracy of this correction was adequate, since the same energy spectra were obtained for different clipping systems (giving different base-line shifts) and different activity rates in the fission counters.

## B. Results

Having recorded 70 000 to 80 000 spontaneous-fission events of $Fm^{254}$, $Cf^{254}$, and $Cm^{248}$ and ~12 000 events of $Cf^{250}$ and $E^{253}$, one can compute the following distributions:

(a) the single-fragment energy distribution
(b) the energy distribution of the heavy fragment, EH
(c) the energy distribution of the light fragment, EL
(d) the total kinetic-energy distribution
(e) the mass-yield curve
(f) the variation with mass in the mean of the total energy released

The most compact, complete, and direct form of representing the data is given in the $E_1$ vs $E_2$ contour diagram or in its equivalent, the mass vs ET contour diagram. However, these contour diagrams have the practical disadvantage that it is difficult to visualize the results.

In addition to the above-mentioned distributions, it seemed useful to compute the mean values and the variances of the distributions, thus expressing quantitatively some essential features of these distributions.

It would occupy too much space in this paper to show five one-dimensional and two two-dimensional distributions for all five different isotopes with their respective calibrations exhibiting rather similar properties. Therefore only the results for $Fm^{254}$ are completely given in Figs. 3 through 8. The single fission-fragment energy spectra for the other isotopes with their respective calibrations are shown in Figs. 9 through 12. The mass vs yield curves are shown in Figs. 13 through 16. Figure 17 shows the mean total kinetic energy as a function of the mass fraction for $E^{253}$ and $Cm^{248}$. (In the case of $Cf^{254}$ and $Cf^{250}$ these distributions show no significant difference from the behavior of $Cf^{252}$.)

In the ET vs MF contour map (Fig. 4) as well as in the figures showing the mean-total kinetic-energy releases versus the mass fraction, the mass M of the fission fragments is expressed in dimensionless units of mass fractions MF:

$$MF = \frac{M}{A}, \qquad (5)$$

where A is the mass of the original nucleus.

The essential results are summarized in Tables II through IV. They show the computed mean values in the energy and mass distributions and the variances $\sigma^2$ for these distributions, with

$$\sigma^2 (E) = \langle E^2 \rangle - \langle E \rangle^2. \qquad (6)$$

(The variance and the full-width-at-half-maximum (FWHM) are connected for a Gaussian distribution by: (FWHM) = 2.35 $\sigma$)

The mean values for the $Cf^{252}$ standards always agree with each other because of the calibration method. The variances of the $Cf^{252}$ calibrations do not agree with each other, since different semiconductor detectors were used which vary in their behavior. In order to compare the variances of all isotopes with each other, they were normalized to one arbitrary value of the variance for $Cf^{252}$.

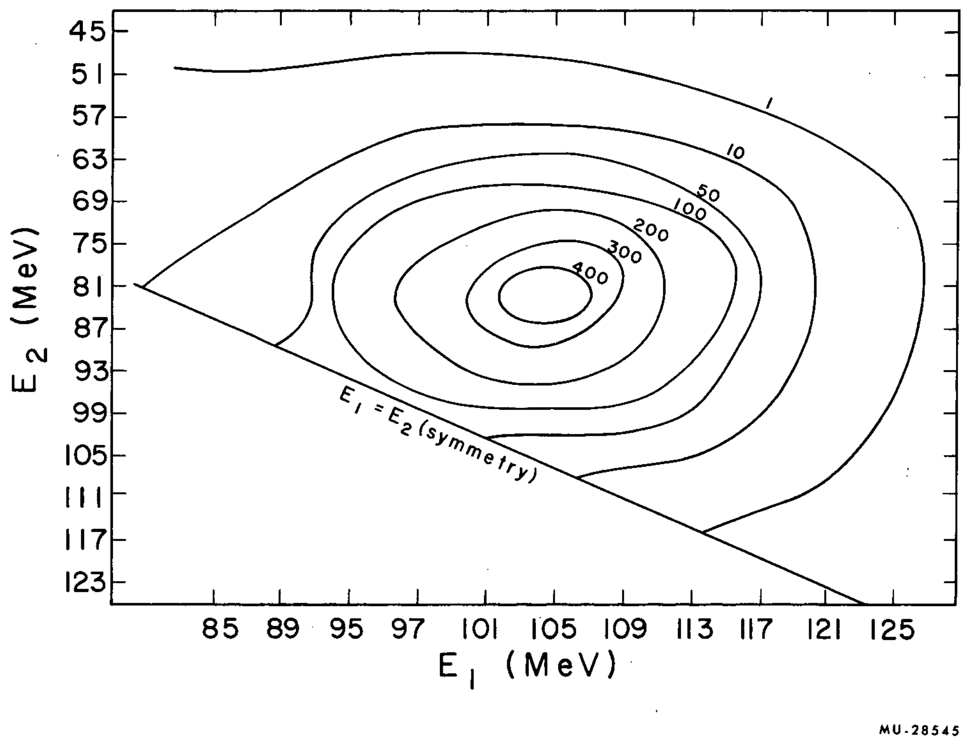


Fig. 3. Contour map in the coordinates $E_1$ and $E_2$ for $Fm^{254}$. The contours are lines of constant $N(E_1, E_2)$.

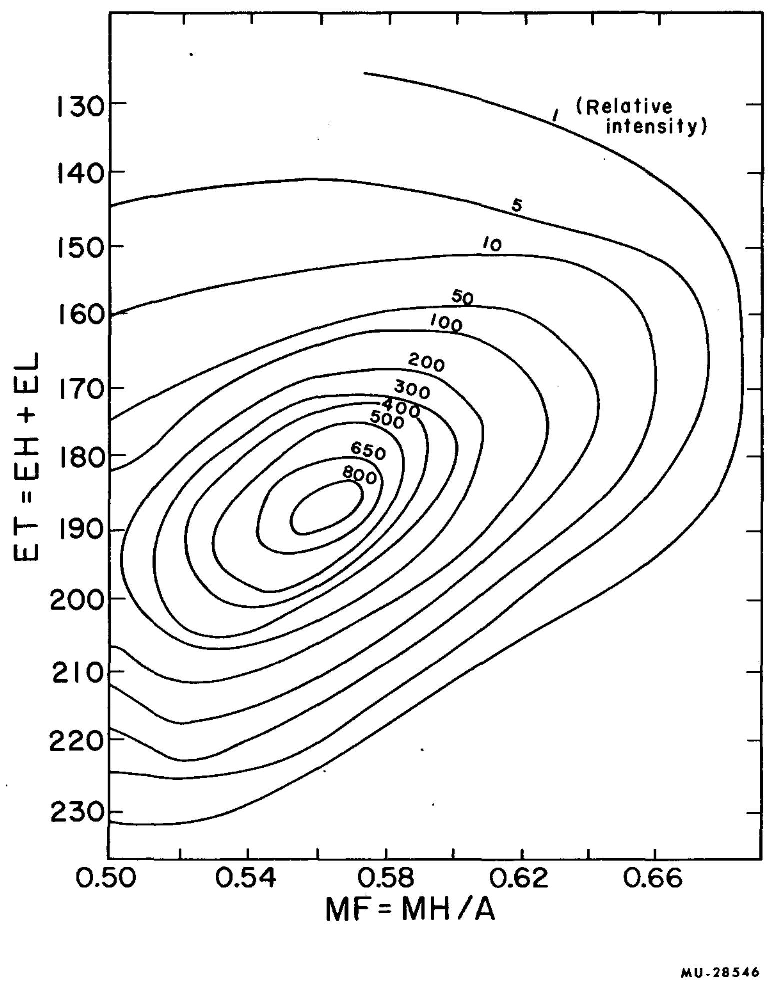


Fig. 4. Contour map in the coordinates MF (mass-fraction) and ET (total kinetic energy) for $Fm^{254}$. The contours are lines of constant N(ET, MF).

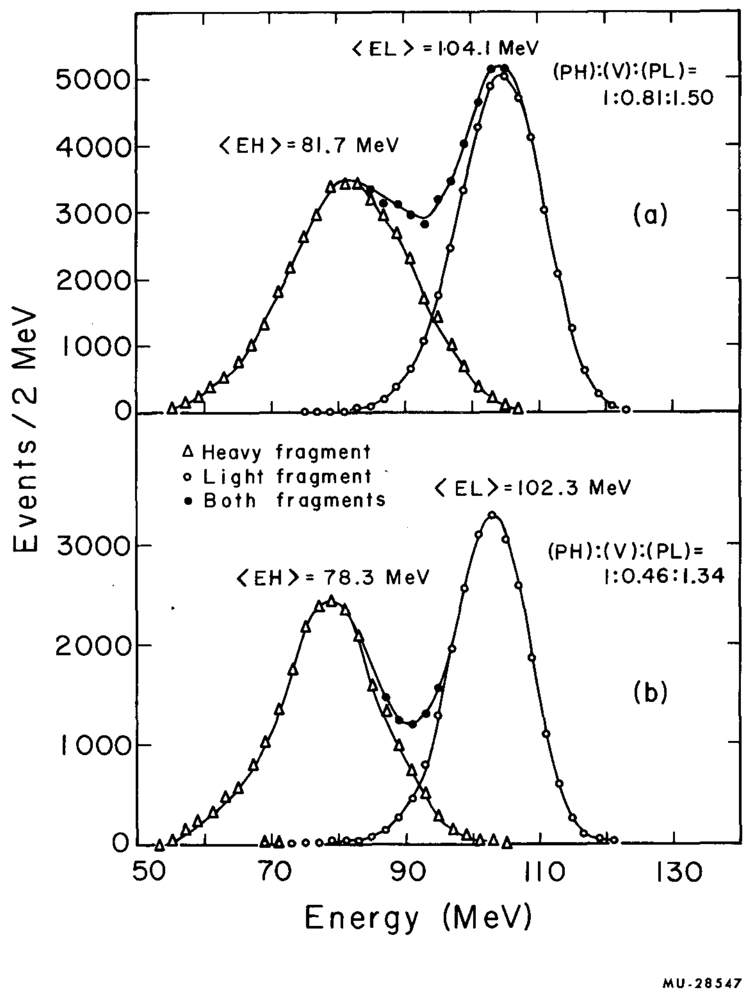


Fig. 5. (a) Single fission-fragment energy spectrum of $Fm^{254}$, subdivided into light- and heavy-fragment spectra; (b) the corresponding spectrum for $Cf^{252}$ used as a standard. (PH):(V):(PL) is defined in the legend of Fig. 2.

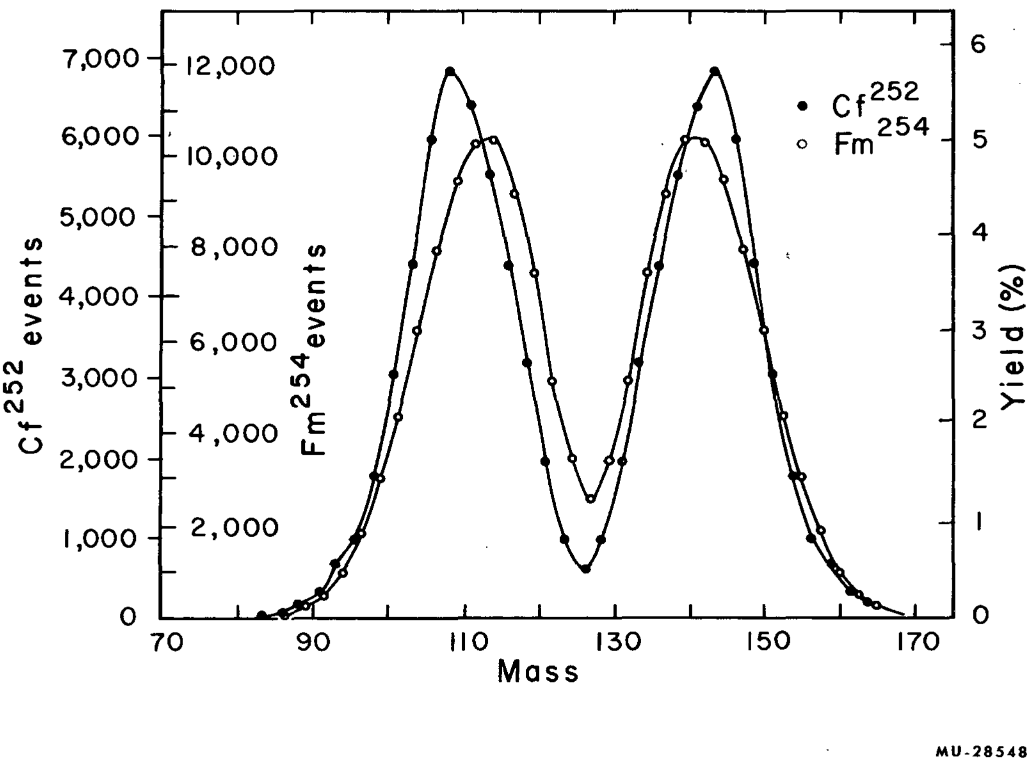


Fig. 6. Mass-yield curve of $Fm^{254}$ and of the standard $Cf^{252}$.

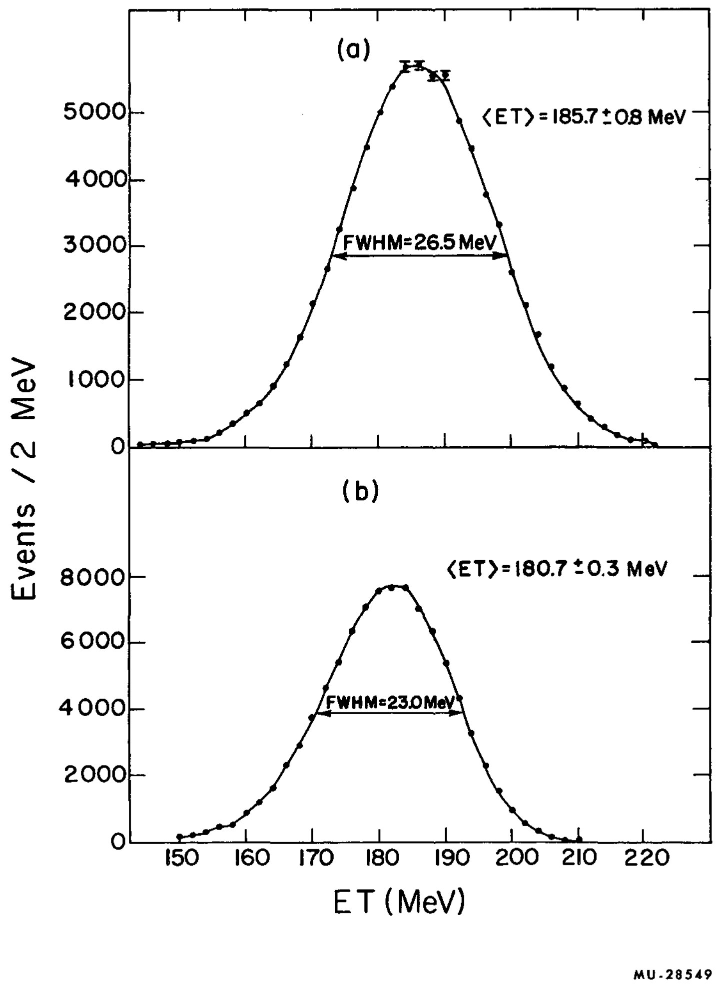


Fig. 7. Total kinetic-energy distribution for $Fm^{254}$ (a) together with the standard $Cf^{252}$ (b); FWHM is the "full width at half-maximum".

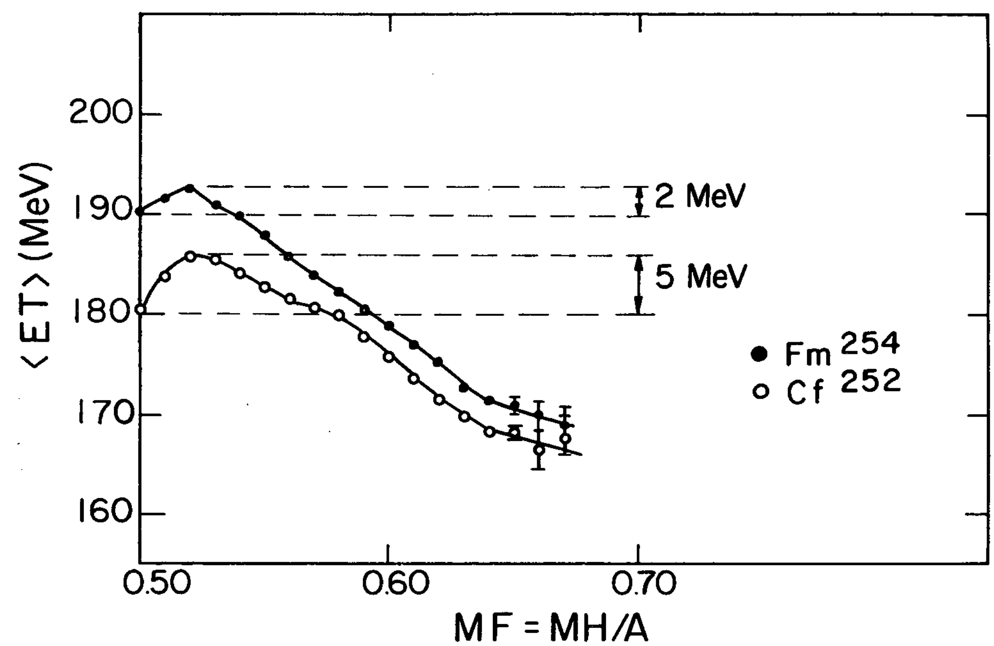


Fig. 8. Mean total kinetic energy ⟨ET⟩ as a function of the mass-fraction MF for $Fm^{254}$ and the standard $Cf^{252}$.

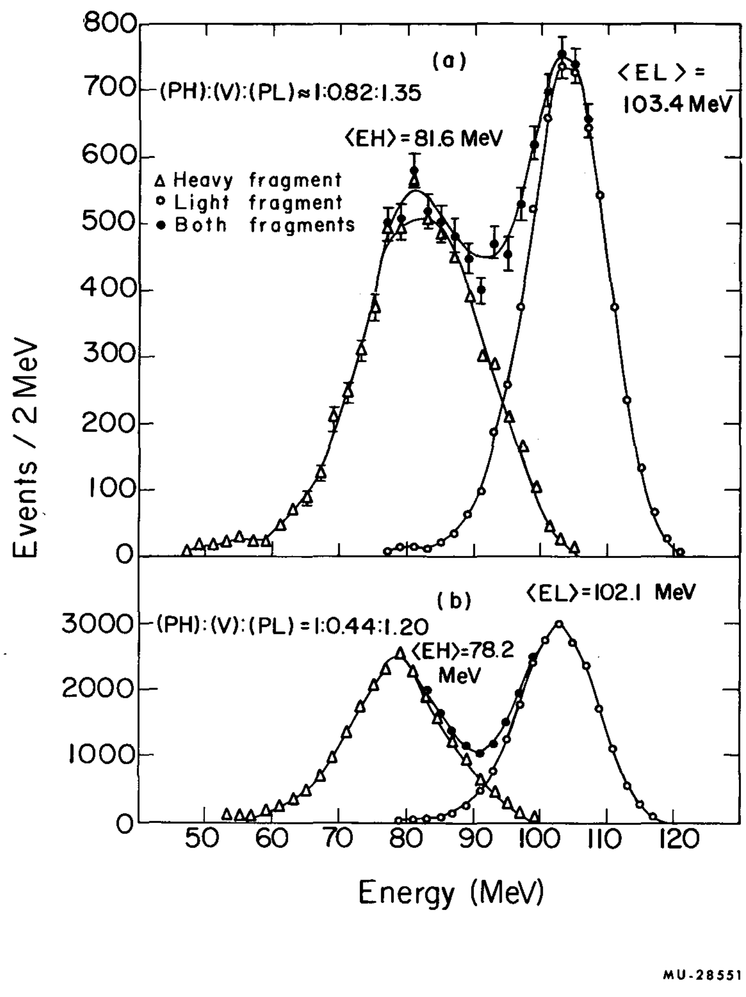


Fig. 9. (a) Single fission-fragment energy spectrum of $E^{253}$ subdivided into light- and heavy-fragment spectra; (b) the corresponding spectrum for $Cf^{252}$ used as a standard. (PH):(V):(PL) is defined in the legend of Fig. 2.

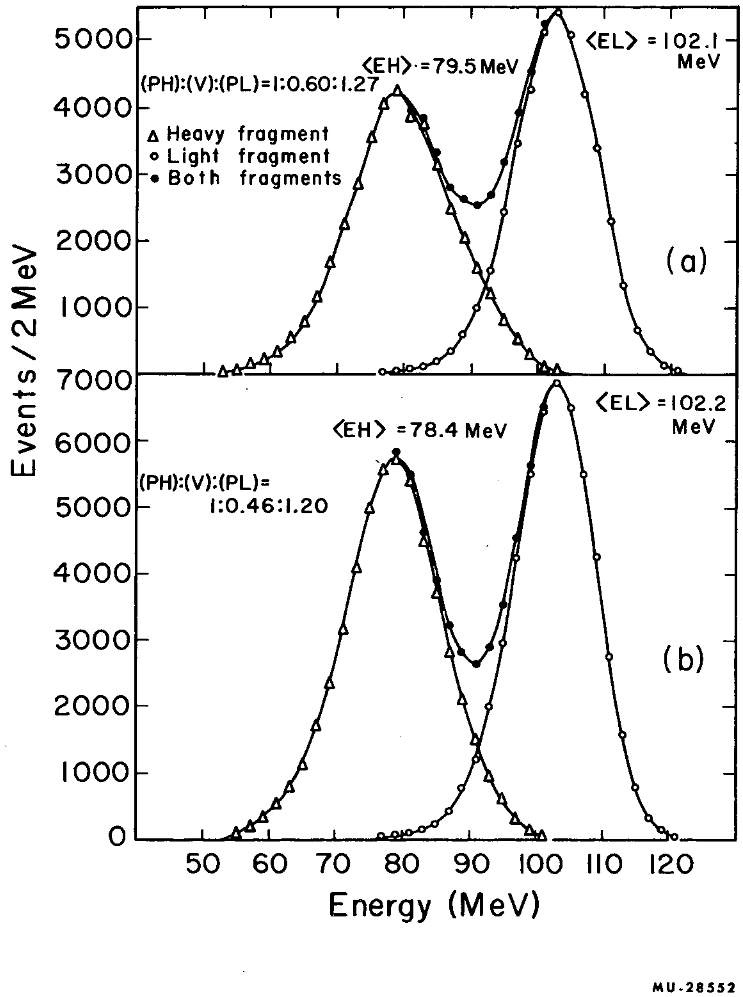


Fig. 10. (a) Single fission-fragment energy spectrum of $Cf^{254}$, subdivided into light- and heavy-fragment spectra; (b) the corresponding spectrum for $Cf^{252}$ used as a standard. (PH):(V):(PL) is defined in the legend of Fig. 2.

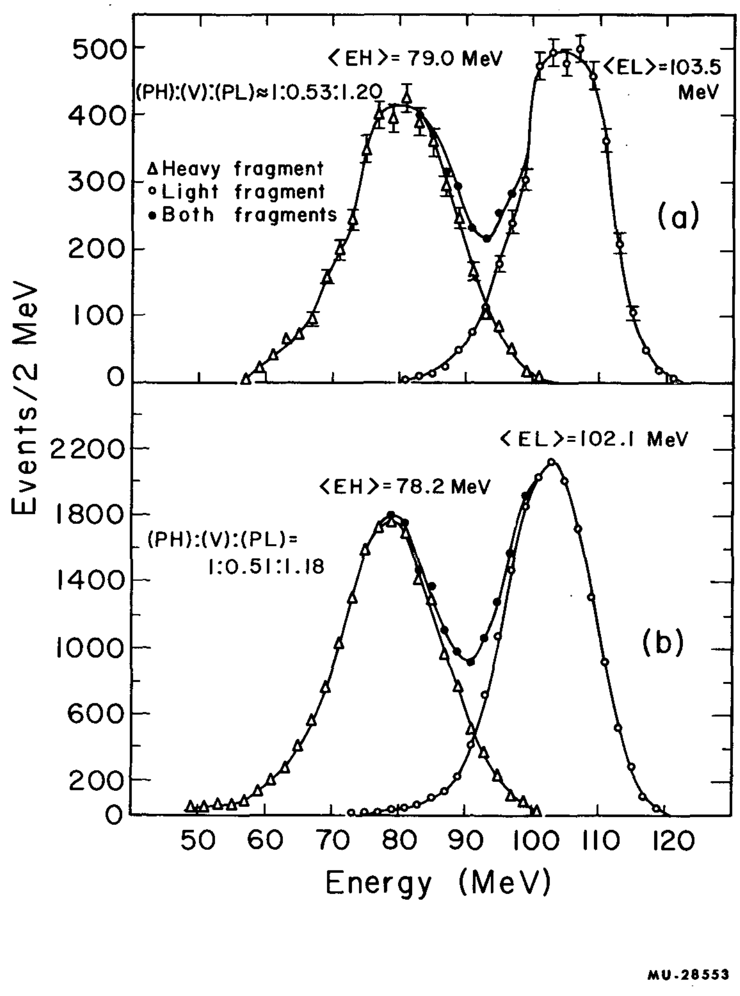


Fig. 11. (a) Single fission-fragment energy spectrum of $Cf^{250}$, subdivided into light- and heavy-fragment spectra; (b) the corresponding spectrum for $Cf^{252}$ used as a standard. (PH):(V):(PL) is defined in the legend of Fig. 2.

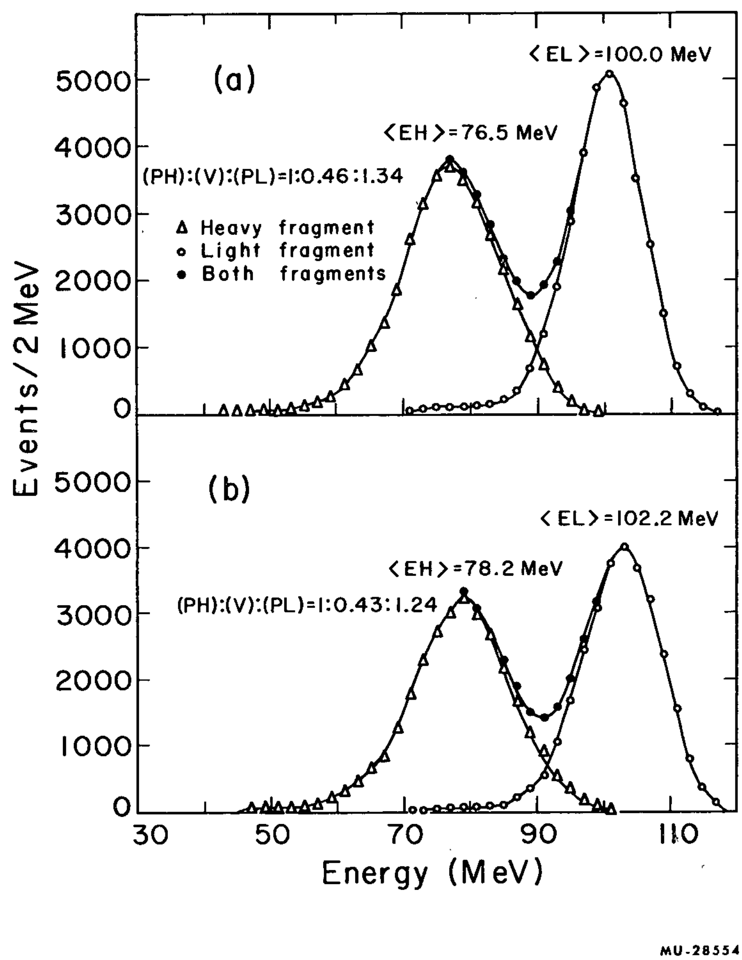


Fig. 12. (a) Single fission-fragment energy spectrum of $Cm^{248}$, subdivided into light- and heavy-fragment spectra; (b) the corresponding spectrum for $Cf^{252}$ used as a standard. (PH):(V):(PL) is defined in the legend of Fig. 2.

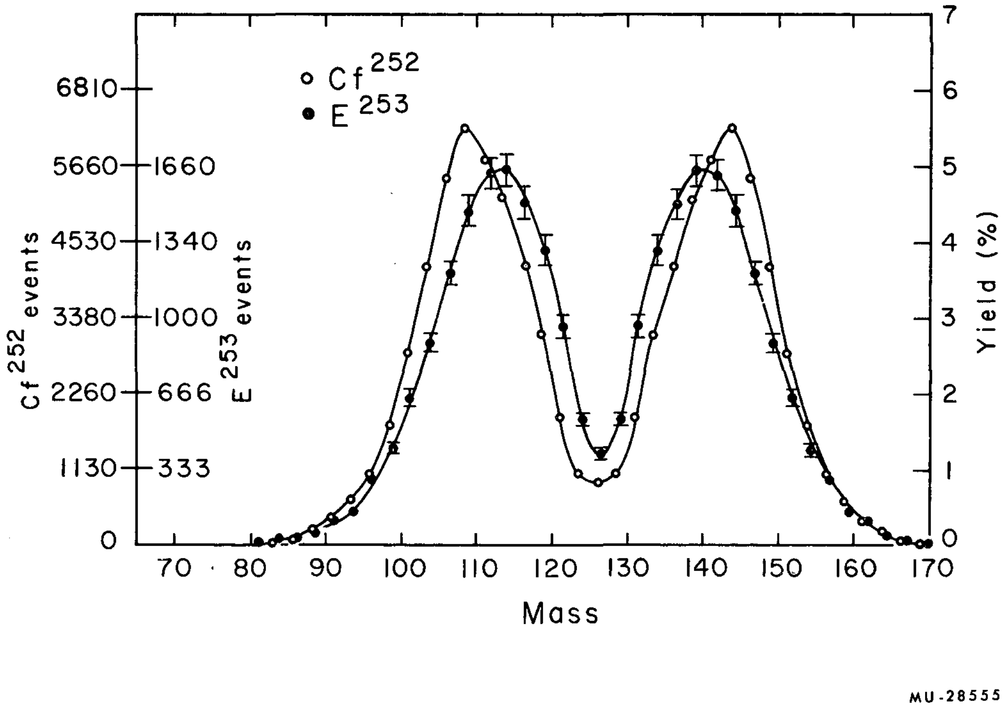


Fig. 13. Mass-yield curve of $E^{253}$ and for the standard $Cf^{252}$.

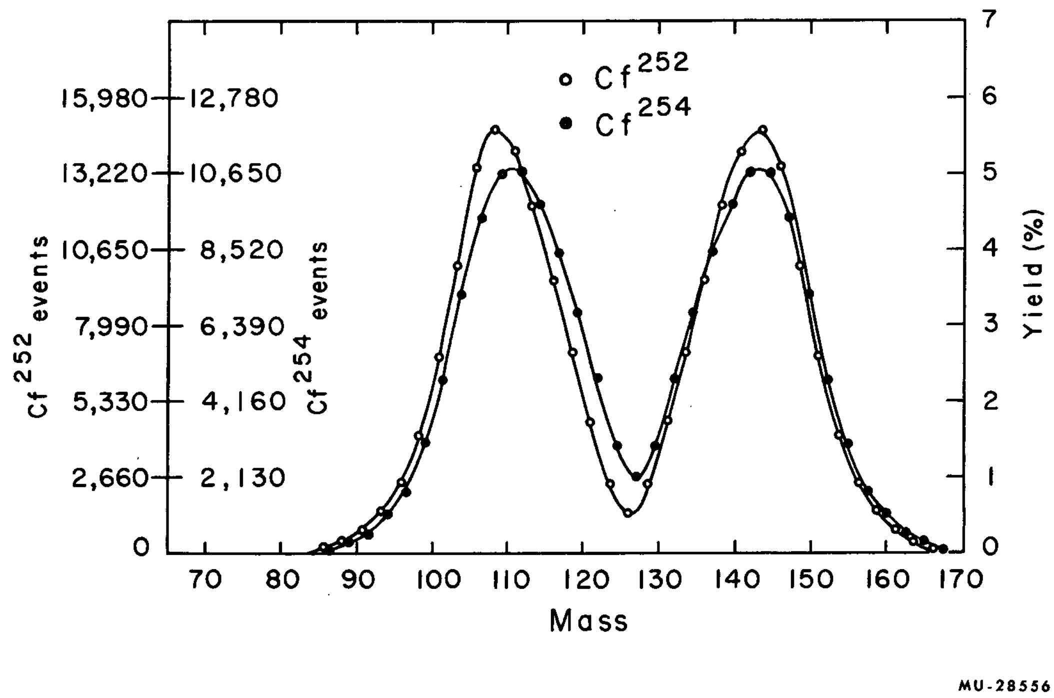


Fig. 14. Mass-yield curve of $Cf^{254}$ and for the standard $Cf^{252}$.

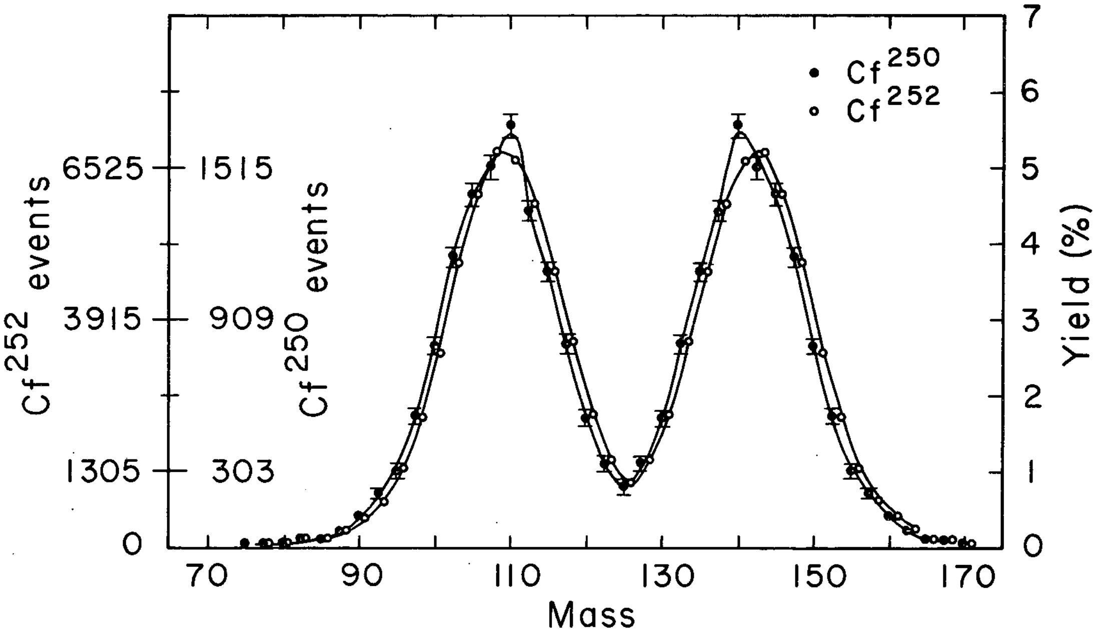


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Fig. 15. Mass-yield curve of $Cf^{250}$ and for the standard $Cf^{252}$.

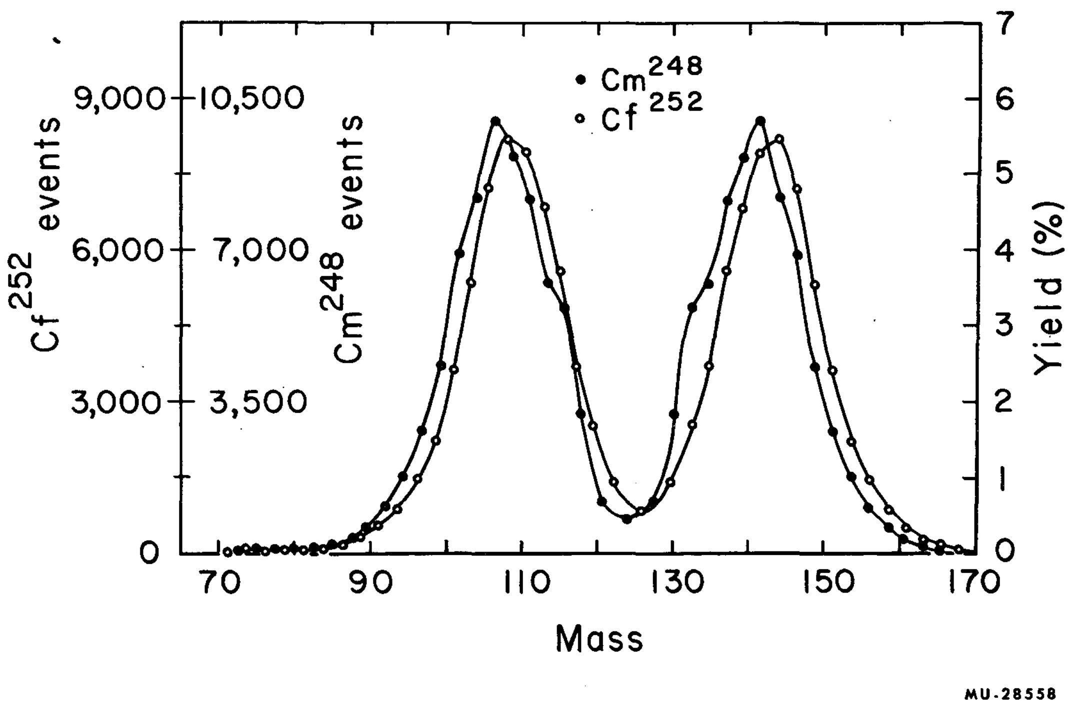


Fig. 16. Mass-yield curve of $Cm^{248}$ and for the standard $Cf^{252}$.

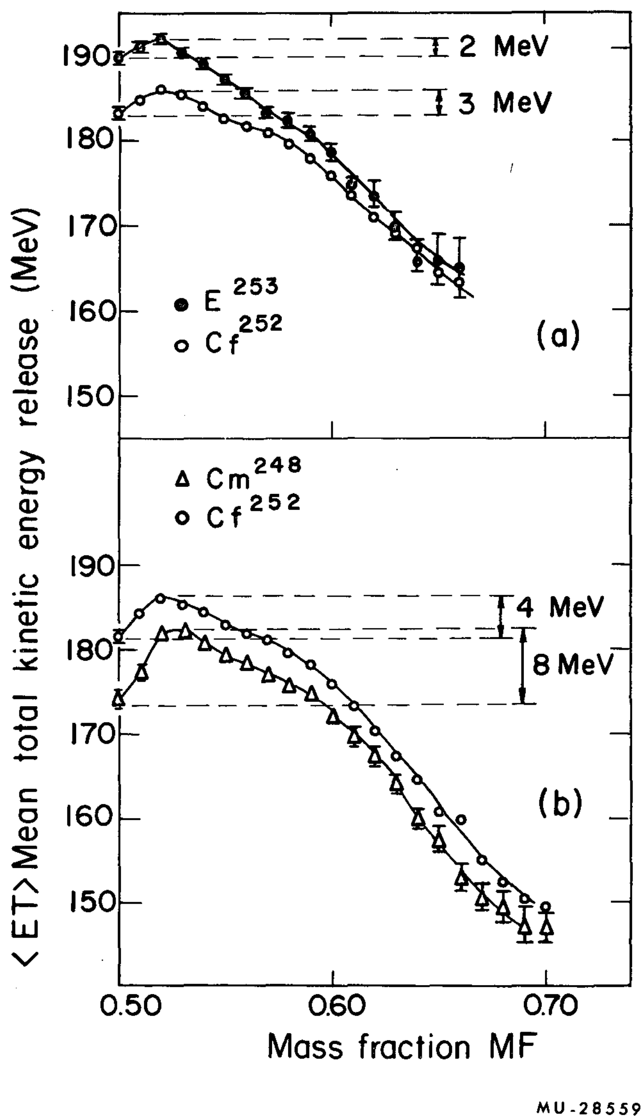


Fig. 17. Mean total kinetic energy release ⟨ET⟩ as a function of the mass-fraction MF for $E^{253}$ and $Cm^{248}$ as compared to $Cf^{252}$.

Table II. Energies in spontaneous fission (in MeV)

| Isotopes | ⟨EH⟩[a] | ⟨EL⟩[b] | ⟨ET⟩[c] | ⟨EK⟩[d] |
|---|---|---|---|---|
| $Fm^{254}$ | 81.7±0.3 | 104.0±0.4 | 185.7±0.8 | 189±2 |
| ($Cf^{252}$) | (78.4±0.1) | (102.3±0.1) | (180.7±0.3) | |
| $E^{253}$ | 81.6±1.0 | 103.4±1.0 | 185.0±1.5 | 188±3 |
| ($Cf^{252}$) | (78.2±0.1) | (102.1±0.1) | (180.4±0.4) | |
| $Cf^{254}$ | 79.5±0.5 | 102.1±0.8 | 181.7±1.0 | 185±2 |
| ($Cf^{252}$) | (78.4±0.1) | (102.1±0.1) | (180.6±0.4) | |
| $Cf^{250}$ | 79.0±1.0 | 103.5±1.0 | 182.5±1.5 | 185±3 |
| ($Cf^{252}$) | (78.1±0.3) | (102.1±0.2) | (180.0±0.5) | |
| $Cm^{248}$ | 76.5±0.6 | 100.0±0.8 | 176.5±1.0 | 179±2 |
| ($Cf^{252}$) | (78.2±0.2) | (102.2±0.2) | (180.4±0.2) | |

[a] ⟨EH⟩ is the mean heavy-fragment energy.

[b] ⟨EL⟩ is the mean light-fragment energy.

[c] ⟨ET⟩ is the mean in the sum of the two measured kinetic energies.

[d] ⟨EK⟩ is the mean in the total kinetic energy release (neutron corrected) as compared to a corresponding value for $Cf^{252}$ of 183.0±0.5 MeV. The uncertainties quoted here include corrections for different foil thicknesses.

Table III. Variance $\sigma^2$ in the energy distribution in spontaneous fission (in $MeV^2$)

| | Directly observed | | | Normalized to one set of values for $Cf^{252}$ | | |
|---|---|---|---|---|---|---|
| Isotope | $\sigma^2(EH)$ [a] | $\sigma^2(EL)$ [b] | $\sigma^2(ET)$ [c] | $\sigma^2(EH)$ | $\sigma^2(EL)$ | $\sigma^2(ET)$ |
| $Fm^{254}$ | 85±2 | 43±2 | 138±4 | 85±3 | 43±3 | 138±5 |
| ($Cf^{252}$) | (64±2) | (39±2) | (110±3) | | | |
| $E^{253}$ | 94±5 | 49±3 | 165±8 | 87±7 | 43±4 | 145±10 |
| ($Cf^{252}$) | (71±4) | (45±2) | (130±4) | | | |
| $Cf^{254}$ | 71±3 | 46±2 | 126±6 | 74±4 | 40±3 | 124±8 |
| ($Cf^{252}$) | (61±2) | (45±2) | (112±4) | | | |
| $Cf^{252}$ | | | | 64 | 39 | 110 |
| $Cf^{250}$ | 81±5 | 55±4 | 146±8 | 64±7 | 39±6 | 110±10 |
| ($Cf^{252}$) | (81±5) | (55±4) | (146±5) | | | |
| $Cm^{248}$ | 64±3 | 42±2 | 132±4 | 60±5 | 38±3 | 109±5 |
| ($Cf^{252}$) | (68±3) | (43±2) | (133±3) | | | |

[a] $\sigma^2(EH)$ is the variance in the heavy-fragment energy distribution.

[b] $\sigma^2(EL)$ is the variance in the light-fragment energy distribution.

[c] $\sigma^2(ET)$ is the variance in the total fragment energy distribution.

Table IV. The mean and variance in the mass distribution for spontaneous fission

| Isotope | ⟨ML⟩ | ⟨MH⟩ | $\sigma^2(M)$[a] | $\sigma^2(M)$[b] |
|---|---|---|---|---|
| $Fm^{254}$ | 111.5±0.3 | 142.5±0.3 | 61±2 | 52±3 |
| ($Cf^{252}$) | (109.1±0.1) | (142.8±0.1) | (52±2) | |
| $E^{253}$ | 111.3±0.5 | 141.7±0.5 | 66±4 | 52±5 |
| ($Cf^{252}$) | (109.2±0.1) | (142.8±0.1) | (57±3) | |
| $Cf^{254}$ | 110.9±0.4 | 143.0±0.4 | 58±3 | 48±4 |
| ($Cf^{252}$) | (109.2±0.2) | (142.8±0.2) | (53±2) | |
| $Cf^{250}$ | 108.0±0.4 | 141.9±0.4 | 64±4 | 41±5 |
| ($Cf^{252}$) | (108.9±0.3) | (143.0±0.3) | (66±2) | |
| $Cm^{248}$ | 107.3±0.2 | 140.7±0.2 | 58±2 | 41±3 |
| ($Cf^{252}$) | (109.0±0.1) | (142.9±0.1) | (60±2) | |

[a] $\sigma(M)$ is the variance of the light- (heavy-) fragment branch of the mass distribution.

[b] Normalized to $Cf^{252}$, with $\sigma^2(M) = 43$.

## C. Uncertainties in the Results

All results are uncorrected for experimental dispersions and the effect of neutrons on the measurements. Since most neutrons are emitted after the scission moment,[15] the kinetic energies of the moving fragments are lowered and thus an uncertainty in the calculated mass and energy distribution is introduced. Only a small number of neutrons are emitted compared to the mass of the fragment, therefore the effects are small. The neutron-emission process is very complex, and this introduces a small but very complicated uncertainty into the measurements.[15,16,31]

At present, no reliable way exists to correct for the dispersions in the energy and mass distributions caused by neutron emission when the fission-fragment energies are determined with semiconductor counters. (For a more detailed discussion see Appendix D.) This is largely because the time-of-flight measurements of fission fragments from $Cf^{252}$ contain rather large uncertainties. The properties of the prompt energy and mass distributions for $Cf^{252}$ are therefore uncertain. Consequently this paper is only a comparative study between the spontaneous fission of $Cf^{252}$ and other isotopes. The experimental uncertainties due to different detectors, electronic noise, etc., are the same for $Cf^{252}$ and the isotopes which were investigated.

A correction was made for the differences in the foil-thickness. The thickness of the foil was determined at the end of the experiment by the energy degradation of 5.8 MeV alpha-particles passing through the foil and it varied between 130 and 240 $\mu gr/cm^2$. (According to Whaling the energy loss in Ni is 400 keV $\cdot$ $cm^2/mg$.[32]) The differences in the foil thickness between the standard and the isotope to be investigated were determined and these differences amounted to $< 40\ \mu gr/cm^2$ in each case. Then the difference in the energy loss of both fission fragments in the foils on which they were deposited was estimated according to Bowman, et.al.,[15] and this was found to be ~1 MeV.

One correction for the neutron emission can be made easily; the mean prompt kinetic energy release $\langle EK \rangle$ is related to the measured mean total kinetic energy $\langle ET \rangle$ by*

$$\langle EK \rangle = \langle ET \rangle \left( 1 + \frac{\bar{\nu}}{A} \right). \qquad (7)$$

An estimated value of $\bar{\nu}$ (average number of neutrons emitted from one fission event) is used for $Cf^{250}$, $Cm^{248}$, and $E^{253}$, on the assumption that $\bar{\nu}$ increases linearly with A.[3]

The mass-yield curves as reported in this paper are not exactly the distributions of the masses of fission fragments before neutron emission (the so-called primary mass-yield curve). The emission of neutrons introduces an uncertainty in the measured energies. The mass-yield curves as determined with methods described in this paper are distorted primary mass-yield curves. They are not directly related to the mass-yield curves as determined by radiochemical methods, since the "radiochemical" mass-yield curve gives exactly the amount of each isotope produced in fission. When the details of the neutron-emission process are known exactly the radiochemical mass-yield curve can be calculated from the primary mass-yield curve. (Terrell[31])

* ( $\langle EK \rangle$ is the total kinetic energy of both fragments before neutron emission; $\langle ET \rangle$ on the other hand is the measured total kinetic energy of both fragments after the emission of neutrons.)

## D. Discussion

The most noticeable feature of the results is that the different isotopes show rather similar energy and mass distributions in their spontaneous fission. However, there are some small and distinct differences between them. The following discussion is restricted to the differences in the distributions and a comparison of the experimental results with some theoretical considerations concerning spontaneous fission.

### 1. Mean Prompt Kinetic Energy Release

The results of Table II can be described by the following statements:

(a) The mean prompt kinetic energy $\langle EK \rangle$ of fission fragments increases with Z.

(b) The mean prompt kinetic energy $\langle EK \rangle$ seems to be independent of A for a given Z within the limits of error. Since the number of fission neutrons $\bar{\nu}$ varies with A there may be small differences in the measured total kinetic energy $\langle ET \rangle$ for different isotopes of one element.

It might be interesting to compare the mean total kinetic energy release $\langle EK \rangle$ as measured in this work with $\langle EK \rangle$ values for other fissioning nuclei. Such a study has been carried out recently by Viola, et al. which, however, included data for fission induced by heavy ions.[6] They show that $\langle EK \rangle$ varies linearly with $Z^2/A^{1/3}$ and that a least-square fit through all observed points yields the following equation:

$$\langle EK \rangle = 0.1065 \, Z^2/A^{1/3} + 20.1 \, . \quad (8)$$

The observed values for $\langle EK \rangle$ are consistent with shapes of the fission fragments at the moment of separation (scission shapes) corresponding to spheroids separated by a small distance.[5,6]

The term $Z^2/A^{1/3}$ in equation (8) can be interpreted qualitatively as representing the Coulomb energy of the original nucleus. Fig. 18 shows the values for the mean total energy release $\langle EK \rangle$ as found in this work together with the straight line (equation 8) in an $\langle EK \rangle$ versus $Z^2/A^{1/3}$ diagram. It can be noted that the straight line describes fairly well the observed trend in $\langle EK \rangle$. It should be noted that the result for $\langle EK \rangle$ in the decay of $Fm^{254}$ does not agree with the results of an older experiment.[33]

The mean total kinetic energy release $\langle ET \rangle$ as a function of the mass fraction MF (Fig. 8 and 17) shows that the general trend for those functions is similar for all the isotopes studied here. However the decrease of $\langle ET \rangle$ for symmetric fission as compared to the maximum value of $\langle ET \rangle$ tends to become less for increasing Z. It should be emphasized however, that in Fig. 7 and 16 no experimental dispersions are considered.

## 2. Mean Values of the Mass Distribution

The overall properties of the mass distribution are as follows (see Table IV):

(a) All mass-yield curves show a strong asymmetric mass distribution.

(b) The mean heavy-fragment mass is always around 141 ± 1.

(c) The mean light-fragment mass values are between 107 and 111.

(d) The odd-mass isotope $E^{253}$ shows a mass-yield curve rather similar to those of the neighboring even-even nuclei.

Again, there is no generally accepted theory that explains quantitatively the mass-yield curves. There are however several proposed models that attempt to explain the asymmetry in spontaneous fission.

The two most important models and their physical basis are mentioned in the introduction. One model that tries to describe the

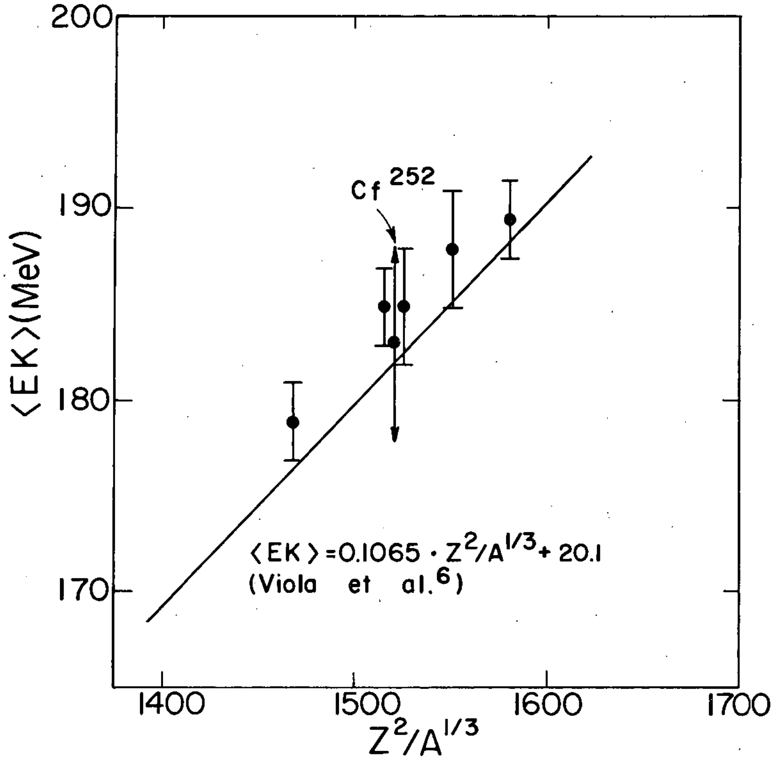


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Fig. 18. Mean total kinetic-energy release ⟨EK⟩ as varying with $Z^2/A^{1/3}$. The systematic uncertainty is shown for the standard $Cf^{252}$. For the other cases only the relative errors as compared to $Cf^{252}$ (⟨EK⟩ = 183 MeV) are given.

variation of the asymmetry in fission on the basis of the liquid drop model was proposed by Swiatecki.[10] On this basis the square of the asymmetry should decrease linearly with $Z^2/A$, the fissionability of the nucleus. The definition of the asymmetry in the mass-yield curve is arbitrary. Swiatecki used the most probable values of the radiochemical mass-yield curve. Milton suggested that the mean in the primary mass distribution is more adequate to express the overall picture of fission.[34] Accordingly the asymmetry AS in the fission mass-yield curve can be defined as:

$$AS = \frac{\langle MH \rangle - \langle ML \rangle}{A}, \qquad (9)$$

where $\langle MH \rangle$ and $\langle ML \rangle$ are the mean heavy and mean light fragments, respectively, and A is the initial mass. Figure 19 shows the results. The mass-yield curves for the slow-neutron-induced fission of $U^{233}$, $U^{235}$, and $Pu^{239}$ are used as described by Milton et al.[35] The other data are taken from Hyde.[3] The trend of the decrease of $AS^2$ with $Z^2/A$ is crudely described by a straight line. However, it seems useful to represent the experimental data in this summarized form. Recently Johansson proposed that the mass ration $\langle MH \rangle / \langle ML \rangle$ should decrease approximately linearly with $Z^2/A$.[9] He derived this as a property of the single-particle levels at the saddle point, as mentioned in the introduction. Figure 20 shows that on this basis the systematics are as good as those by Swiatecki.

The mean values of the single-fragment energy distributions are directly related to the mean values of the mass distribution and total kinetic-energy distribution. Therefore it is not necessary to treat them separately.

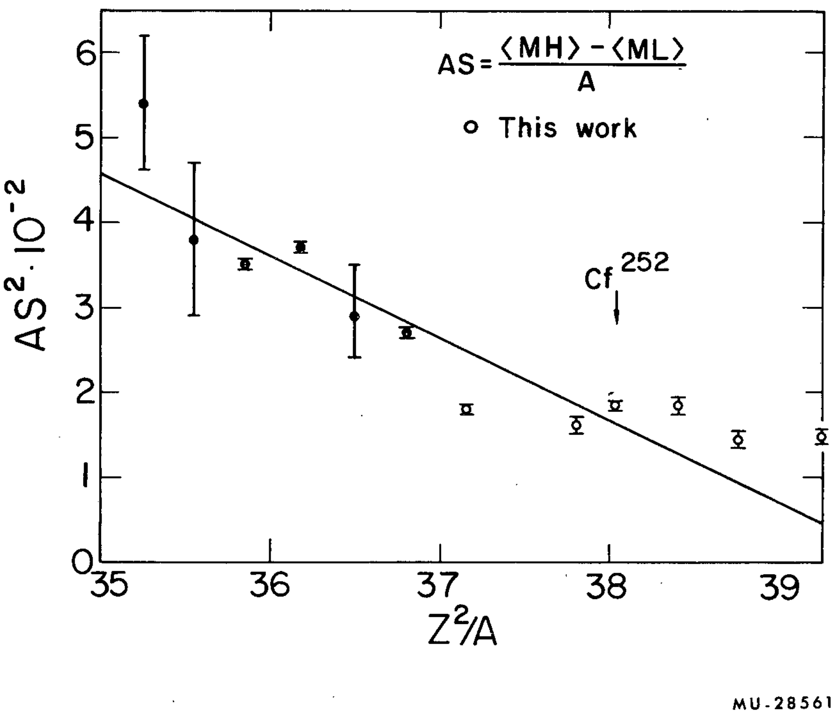


Fig. 19. Asymmetry AS of the mass-yield curve as a function of $Z^2/A$. The open circles are points taken from this work. The points with large errors represent cases where only radiochemical data is available.

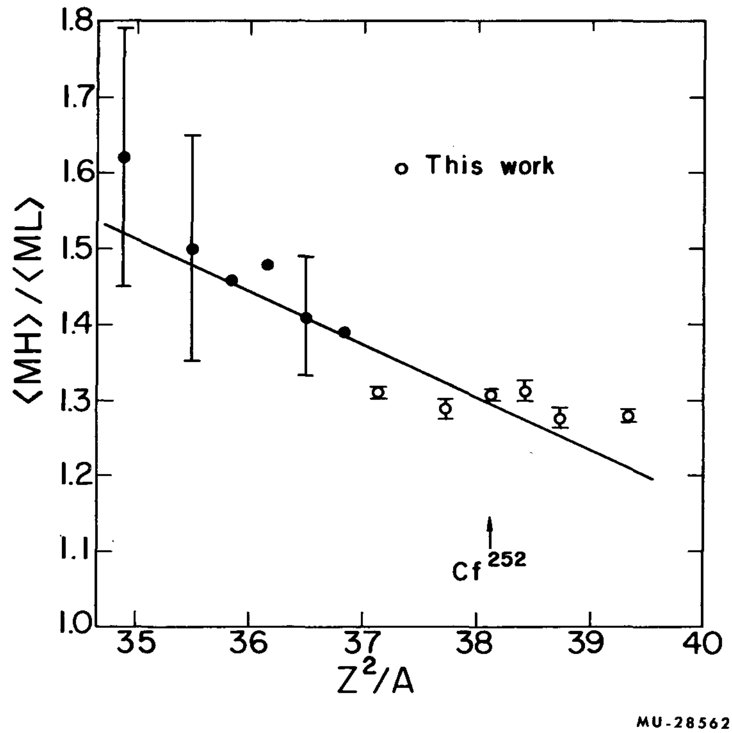


Fig. 20. Ratio of the mean heavy-fragment mass ⟨MH⟩ to the mean light-fragment mass ⟨ML⟩ as a function of $Z^2/A$ (see also the figure legend for Fig. 19).

### 3. Variances of the Distributions

Several sets of variances are listed in Tables III and IV. Their properties can be described as follows:

(a) The variance of the total-energy distribution $\sigma^2$ (ET) increases with Z.

(b) The variance in the energy distribution of the light fragment $\sigma^2$ (EL) is essentially constant.

(c) The variance in the energy distribution of the heavy fragment $\sigma^2$ (EH) increases with Z and seems to increase with A for a given Z.

(d) The variance in the light (or heavy) branch of the mass-yield curve $\sigma^2$ (M) increases with Z and seems to increase with A for a given Z.

Figure 21a shows $\sigma^2$ (M) as a function of $Z^2/A$. The values are normalized to the variance in the primary mass-yield curve of $Cf^{252}$ with $\sigma^2$ (M) = 43 as derived from time-of-flight measurements of Milton et al.[29] Figure 21b shows $\sigma^2$ (EH) and $\sigma^2$ (EL) as functions of $Z^2/A$. The values are uncorrected for neutron-emission, but normalized to one (arbitrary) set of values for $Cf^{252}$. (Figures 21a and 21b show also the values for the respective variances for the slow-neutron induced fission in $U^{233}$, $U^{235}$, $Pu^{239}$ as measured recently by Milton et al.[35] This inclusion, however, does not mean that the variances in spontaneous fission and slow-neutron induced fission could be compared directly from a theoretical viewpoint.) An increase in the variance with $Z^2/A$ can be observed for the mass and heavy-fragment energy distributions. It is also possible to draw a straight line through all these points. This correlation is as good (or bad) as all others in which experimental data such as the spontaneous-fission half-life or asymmetry in the mass distribution is plotted against $Z^2/A$. Nevertheless, it seemed useful to introduce for the first time these correlations with the variances as a function of $Z^2/A$ into the systematics of fission-fragment properties.

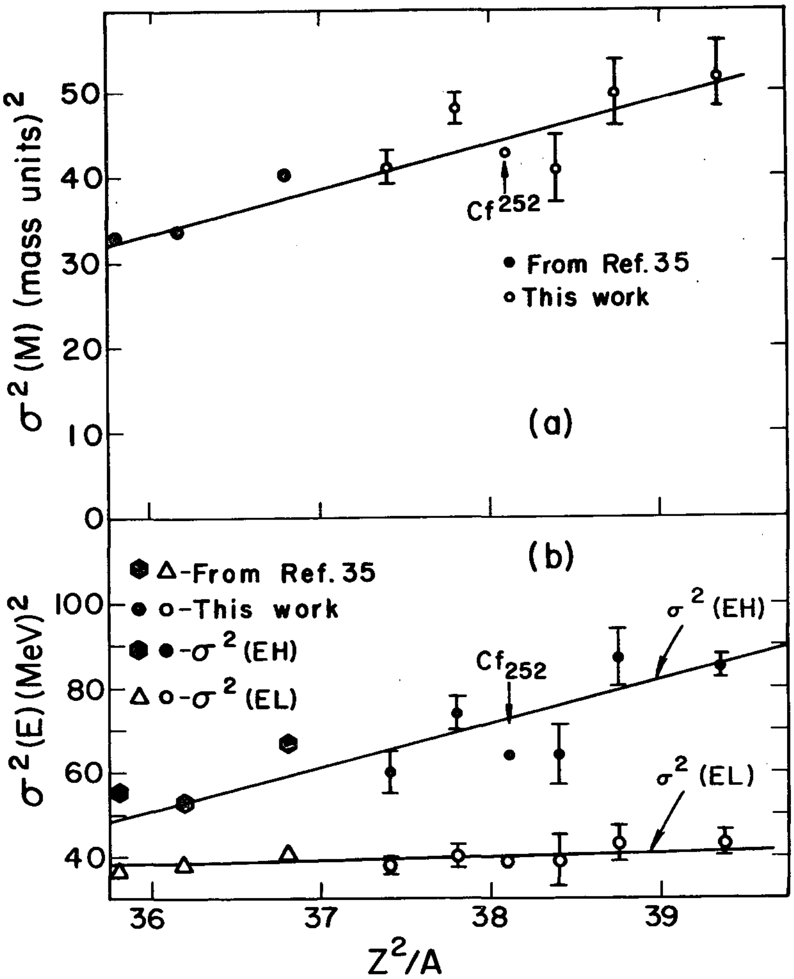


Fig. 21. (a) The variance $\sigma^2(M)$ in one branch of the mass-yield curve as a function of $Z^2/A$. The points are normalized to $\sigma^2(M) = 43$ for $Cf^{252}$. (b) The variance of the heavy-fission-fragment energy spectrum $\sigma^2(EH)$ and of the light-fission-fragment energy spectrum $\sigma^2(EL)$ as a function of $Z^2/A$. The values are uncorrected for neutron emission, but normalized to one (arbitrary) set of values for $Cf^{252}$.

Slow-neutron induced fission data of $U^{233}$, $U^{235}$, and $Pu^{239}$ are included (Milton et al.[35]).

Since comparably simple features of the fission process, such as the total kinetic-energy release and the asymmetry in the mass distribution, can scarcely be interpreted from a theoretical point of view, it is not astonishing that no successful model is known to interpret the variances in the distributions. Therefore only one proposal is mentioned which interprets the trends in the variances of the mass distribution on a qualitative basis.

Figure 22a shows the asymmetry AS as a function of the variance $\sigma^2$ (M) in one branch of the mass distribution. A large asymmetry is usually accompanied by a small variance and vice versa. This might be connected with the interpretation of the fission process given by Johansson.[7,9]

He suggests that the asymmetry of the mass distribution is connected with an asymmetric deformation of the nucleus at the saddle point. He also shows that for large asymmetries the valley in the potential-energy surface is "sharp" as compared to the small asymmetries where this valley is rather "shallow", as shown on Fig. 22b.

The possibility exists that a sharp valley is connected to a narrow mass distribution and this results in a small variance. A shallow valley might be connected with a wide mass distribution and a large variance $\sigma^2$ (M). This would account for the general trend in the decrease of AS with increasing $\sigma^2$ (M).

## E. Conclusion

Some essential results of this study of the kinetic energy and mass-distributions in the spontaneous fission of some heavy nuclei are again summarized below:

(a) The energy and mass distributions are rather similar (but not identical) for all the isotopes investigated here.

(b) The mean prompt kinetic energy release of fission fragments increases with Z. This agrees with the findings of Viola et al.[6]

(c) All mass-yield curves show a strong asymmetric mass-distribution. In agreement with previously observed trends (see Hyde[3])

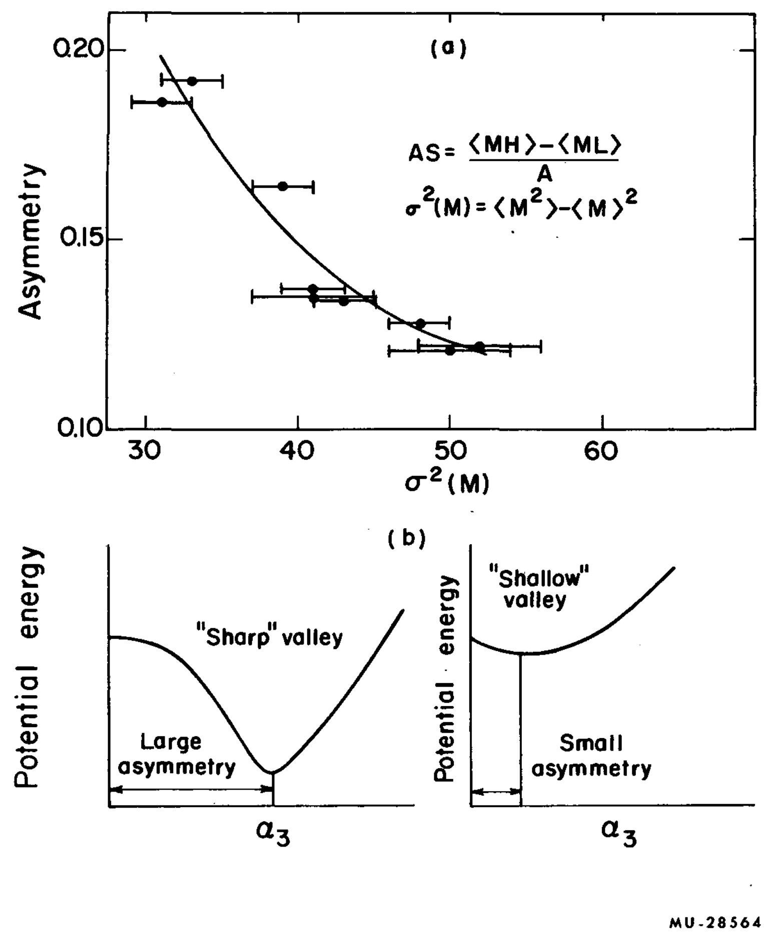


Fig. 22. (a) Asymmetry AS as a function of the variance $\sigma^2(M)$ in one branch of the mass-yield curve. The values are the same as those used for Fig. 19.

(b) Qualitative interpretation of Fig. 22 (a) as suggested by Johansson.[9] Potential energy as a function of asymmetric deformation $\alpha_3$ of the nucleus at the saddle point.

the mean heavy fragment mass is always around 142 ± 1, whereas the light fragment mass shows more variation.

(d) In addition to several previously known correlations a new semi-empirical correlation is proposed, in which the variances of some mass and energy distributions are shown as a function of $Z^2/A$. The variance in the energy distribution of the light fission fragment $\sigma^2$ (EL) is essentially constant. The variance in the energy distribution of the heavy fragment $\sigma^2$ (EH) as well as the variance in one branch of the mass-yield curve $\sigma^2$ (M) increases with Z and seems to increase with A for a given Z.

(e) The behavior of the fission fragments in the spontaneous fission of $E^{253}$ resembles very closely those of the neighboring even-even nuclei. (It should be noted that this is the first work in which the mass and energy distributions in the spontaneous fission of an odd-mass isotope have been investigated.)

Several models exist which interpret certain aspects of spontaneous fission qualitatively. However, further theoretical work is needed to give an accurate interpretation of the various aspects of fission, in particular concerning the mean values of the mass and energy distributions and the variances of those distributions.

The total energy balance in fission of the isotopes studied here is considered in the last Appendix. In order to make more precise studies of the energy balance the following empirical and theoretical investigations might be considered as a continuation of this work:

(a) More accurate data concerning the primary fission fragments of $Cf^{252}$ might make it possible to calculate the primary mass-yield curve for the isotopes investigated in this work. Then it would be possible also to calculate the radiochemical mass-yield curves for those isotopes according to Terrell's method.[31] In some cases it might be possible to compare those calculated radiochemical mass-yields with experimental data.

(b) It seems to be necessary to measure the average number of neutrons emitted in fission, $\bar{\nu}$ for more isotopes.

(c) The mean energy emitted by gamma-rays in fission ⟨EG⟩ should be studied for more isotopes.

(d) The details of the neutron emission process should be studied for spontaneously fissioning nuclei other than $Cf^{252}$.

(e) The accuracy of Cameron's mass equation and other mass formulae might be studied further.

(f) Not only a total energy balance, but also the energy balance in single fission events should be investigated.

(g) The mass and energy distributions of other isotopes should be determined using the methods employed in this work.

# III. SOME SPONTANEOUS-FISSION HALF LIVES

## A. Discovery of a New Fermium Isotope(21)

### 1. Introduction

It is known that the irradiation of curium isotopes at the Materials Testing Reactor, Idaho Falls, Idaho (MTR) produces transcurium isotopes as heavy as $Fm^{256}$.[20] In recent years the amounts of transcurium isotopes have increased significantly. A study of decay systematics suggested that $Fm^{257}$ might have a half-life of the order of days or longer. Therefore, it seemed interesting to search for this isotope.

### 2. Chemical Procedures

One hundred twenty milligrams of mixed curium isotopes were subject to an integrated flux of $2.4 \cdot 10^{22}$ neutrons/$cm^2$ at the MTR. The irradiated curium target material was purified by a standard chemical procedure.[28] The fermium-fraction separations and purification were completed about 12 days after the curium irradiation in the MTR. A separation factor of $\sim 10^9$ was obtained between fermium and einsteinium. The fermium sample used for the activity measurements contained ~100 alpha decays per minute of $E^{253}$. This source was prepared by electroplating on a platinum disc, and its alpha and spontaneous-fission activity were studied over a period of several weeks.

### 3. Activity Measurements

Results of the decay of the 11-day spontaneous-fission activity in the Fm fraction are presented in Fig. 23. Decay of the spontaneous-fission activity in the fermium fraction was followed with two

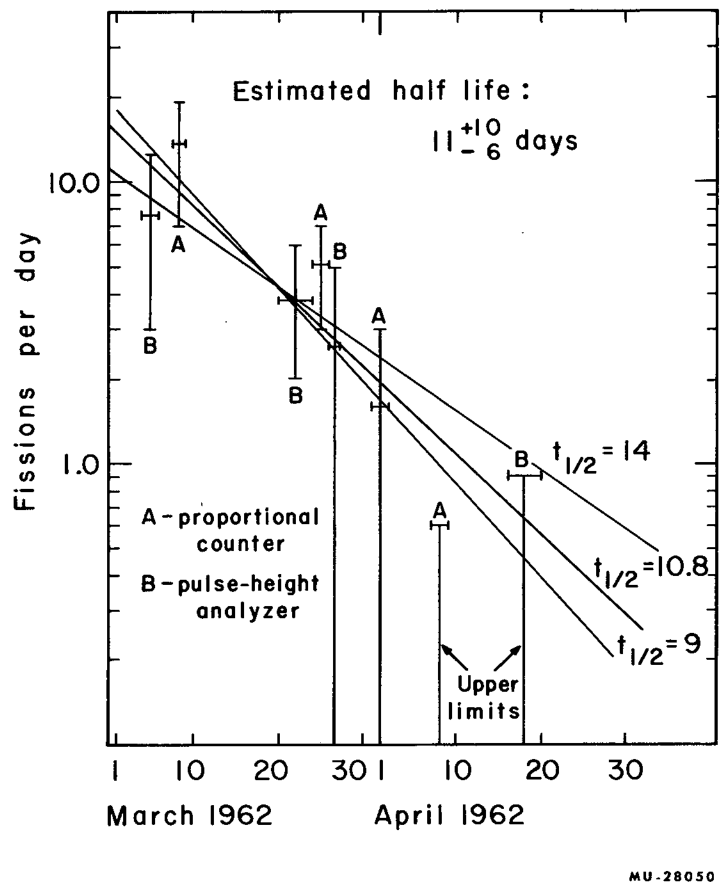


Fig. 23. Decay of a new Fm isotope. Approximately 50 events have been directly observed. The activity is calculated from the observed events and the geometry factor. Two different sets of counters were used. The background is subtracted.

independent counting systems. The first system involved a windowless proportional counter. These results are denoted by (A) in Fig. 23. In the second system, spontaneous-fission events were detected by a 1-in-diam phosphorus-diffused, solid-state detector and a multichannel pulse-height analyzer. These results are denoted by (B) in Fig. 23. A least-squares analysis of the data yielded a half life of 10.8 days with a standard deviation of $\left(^{+2.2}_{-1.8}\right)$ days. The reported half life is $\left(11\ ^{+10}_{-\ 6}\right)$ days. Because of the presence of 100 alpha decays per min of $E^{253}$ in the Fm fraction, it was impossible to observe Fm alpha particles which might have had energies less than 6.7 MeV.

## 4. Discussion

Assignment of the proton number Z = 100 to the 11-day fission activity is based on the following considerations. It was observed only in the fermium fraction. (The einsteinium fraction did not contain this activity--in any case, spontaneous-fission half life systematics also indicate that an 11-day half life is much too short for an einsteinium isotope.) The chemical separation of the mendelevium and fermium fractions, which was repeated, also rules out the possibility that the 11-day activity is a mendelevium isotope. The 11-day activity can not be the mendelevium daughter of a beta-unstable fermium parent, because the last fermium-mendelevium chemical separation was completed 12 days after the end of the MTR bombardment. If the half life of the beta-unstable fermium parent were less than 2 days, it would have decayed by this date. If the postulated half life of the beta-unstable fermium parent is longer than 2 days, we would have observed the growth of the 11-day daughter. This consideration is also supported by an examination of cross sections necessary for the formation of a fermium isotope of mass number 259, which is also predicted to be the first beta-unstable fermium isotope. It seems unlikely that the 11-day fission activity would be an isomer of a known Fm isotope--$Fm^{254}$, $Fm^{255}$, or $Fm^{256}$. Therefore, the mass assignment is most likely to be A = 257 or 258.

## B. Spontaneous Fission Half-Lives of $Cf^{254}$, $Fm^{255}$, and $Cf^{250}$ (36)

### 1. Introduction

Larger amounts of short-lived fermium, einsteinium, and californium isotopes became available recently when they were separated from a mixture of curium isotopes that had been reirradiated in the MTR. This has made it possible to remeasure the spontaneous fission half lives of $Cf^{254}$ and $Cf^{250}$ with greater precision.

In addition, the spontaneous-fission half life of $Fm^{255}$ was determined for the first time, and the $Fm^{255}$ alpha half life remeasured. The electron-capture branching ratio of metastable $E^{254}$ was also remeasured.

### 2. Experimental Procedures

The $Cf^{254}$ source, which contained only 1.3% $Cf^{250}$ fission activity at the beginning of the $Cf^{254}$ fission half life measurement was prepared by irradiating isotopically pure $E^{253}$ at the MTR. Standard chemical procedures were used to purify and prepare the $Cf^{254}$ sample.[3] The $Cf^{254}$ sample contained less than 0.05% of $Cf^{252}$ fission activity.

Neutron bombardment of $E^{253}$ produces $E^{254m}$, which then decays to produce the daughters $Fm^{254}$ and $Cf^{254}$, as shown below:

$$E^{253}(n,\gamma)E^{254m}\ \begin{array}{l} \xrightarrow[\beta^-]{\sim 100\%} \ Fm^{254} \xrightarrow[3\ h]{\alpha} Cf^{250} \\ \xrightarrow[E.C.\ \ 0.078\%]{37\text{-}h} \ Cf^{254} \xrightarrow[60.5\ d]{S.F.} \text{Fission products.} \end{array} \qquad (10)$$

The electron-capture branching ratio of $E^{254m}$ was measured by separating $Fm^{254}$ and $Cf^{254}$, which grew in from a pure sample of the $E^{254m}$ parent, on a column of Dowex 50X 12% cation-exchange resin. Ammonium α-hydroxy isobutyrate was used as the eluant. The rates of alpha emission and spontaneous fission in pure $Fm^{254}$ and $Cf^{254}$ fractions were measured in a windowless proportional counter.

The $Fm^{255}$ sample was obtained by separating it from its parent $E^{255}$, which had been chemically separated from other elements, then counted for alpha particles and fission fragments in two independent counting systems. The decay of the $Fm^{255}$ fission activity was observed for about 100 h (five half lives).

The $Cf^{250}$ sample was separated from an isotopically pure sample of $E^{254}$ ($t_{1/2}$ = 480 days). The $Cf^{250}$ daughter had been growing into the parent sample for nearly one year before separation. The $Cf^{250}$ alpha/fission ratio was measured in a windowless proportional counter.

## 3. Results

The measured $Fm^{255}$ fission/alpha ratio is $(2.4 {}^{+1.1}_{-0.9})\cdot 10^{-7}$. The remeasured $Fm^{255}$ alpha half life is 19.9 ± 0.3h. This alpha half life agrees well with a recent measurement by Asaro.[37] Our remeasured value for the $Fm^{255}$ spontaneous-fission half life of $Fm^{255}$ is $(1.0 {}^{+0.6}_{-0.3})\cdot 10^{4}$y. This value agrees well with Johansson's estimated value of $2\cdot 10^{4}$y.[38] It also fits smoothly into the spontaneous-fission systematics as developed by Swiatecki.[18]

The new value for the electron-capture/$\beta^{-}$ -decay branching ratio in $E^{254m}$ is (0.078 ± 0.006)%.

The experimental value for the $Cf^{250}$ alpha/fission ratio is (1330 ± 45). The $Cf^{250}$ spontaneous-fission half life is $(1.73 \pm 0.06)\cdot 10^{4}$y based on a 13-y alpha half life for $Cf^{250}$.[39]

A least-squares analysis of the data obtained by following the spontaneous-fission decay of $Cf^{254}$ over a period represented by four half lives yields a value of 60.5 ± 0.1 days for the $Cf^{254}$ half life (see Fig. 24). This result includes a correction for the spontaneous-fission activity (1.3%) from a small amount of $Cf^{250}$. The presence of $Cf^{250}$ in a sample of $Cf^{254}$ separated from $E^{254m}$ is unavoidable, as may be seen from Eq.(10). The correction involved measurement of the amount of $Cf^{250}$ present in the source by means of an alpha-particle pulse-height analyzer. From our value for the alpha/fission ratio of (1320 ± 40) we obtain a corrected least-squares value of 60.5 days for the $Cf^{254}$ spontaneous-fission half life. Because of the uncertainty in the $Cf^{250}$ correction, an uncertainty of ± 0.2 days is reported for the $Cf^{254}$ half life. This is twice the standard deviation of ± 0.1 day calculated directly from the least squares analysis. The older values for the decay constants of the isotopes investigated here have been reported by Strominger et al.[40]

It has been mentioned in the introduction that Burbidge et al. suggested that $Cf^{254}$ might have supplied the energy responsible for the exponential part of the light curves of Type-I supernovae which decays with a (55 ± 1)- day half life,[23] since the hitherto-reported half life for $Cf^{254}$ was (56.2 ± 0.7) days.[27] In a more recent study, Hoyle et al. suggested that this problem may be more complicated.[26] This suggestion is supported by the fact that the half life of $Cf^{254}$ does not agree with the above-mentioned decay of the light curves of Type-I supernovae.

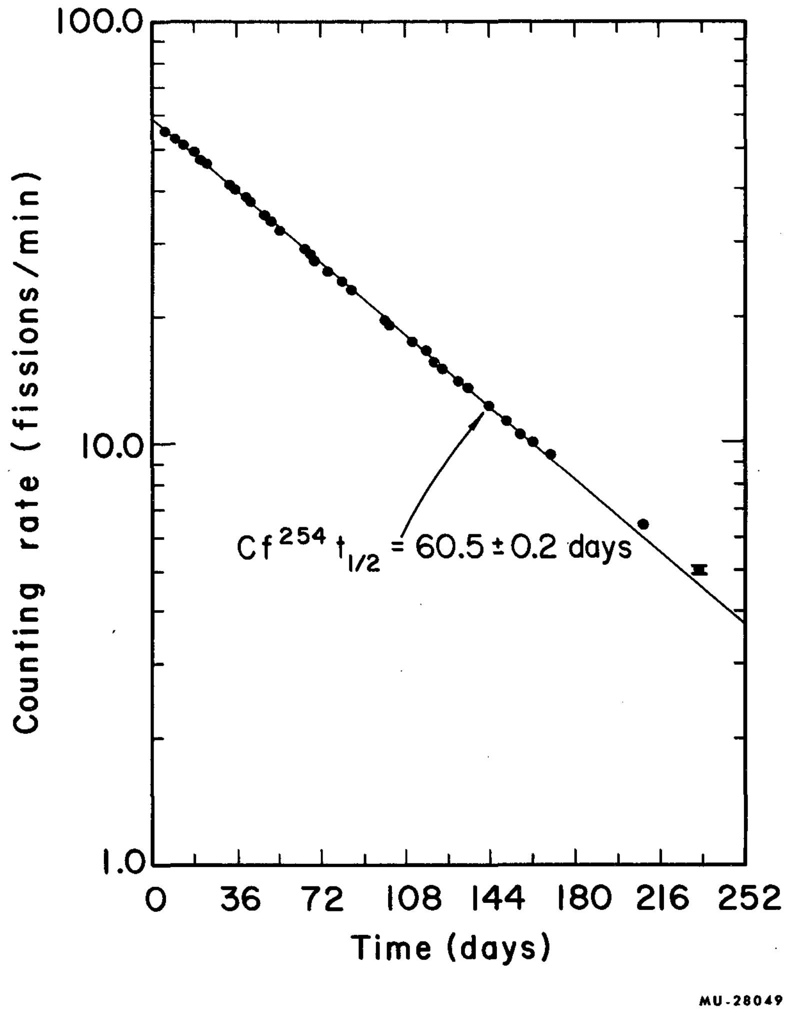


Fig. 24. Spontaneous fission half life of $Cf^{254}$.

# IV. APPENDIXES

## A. Chemical Procedures

Production of the isotopes is shown schematically in Table I. The actinides were separated on a Dowex 50X 12% cation-resin column. The eluant was 0.23M ammonium α-hydroxy isobutyrate.[12] This separation method is very effective, as it is possible to decrease the amount of einsteinium present in a fermium sample by $10^5$ with one separation.

The isolated and purified samples were electroplated on 5-μin-thick(110 μgr/$cm^2$) Ni foils. The Ni-foil was mounted between two steel rings with an open diameter of 1.0 cm. Two methods were used for the cathodic electrodeposition of the radioactivity. A drop of concentrated $NH_4Cl$ solution containing the actinide was placed together with a Pt-wire anode on the thin Ni foil and electroplated with 3 to 4 V for about 15 min. In the second method the steel ring holder of the foil was completely covered with a nonelectrolytic plastic film. It was immersed in conductivity water together with the Pt anode. The activity was added in a small amount of weak acidic solution and electroplated at 20 V for approximately 1 h. The electrodepositing yields of the two processes varied between 50 and 90% in both cases.

Contamination of the sample with fission events from other isotopes was determined in two ways. In the case of the 3-h $Fm^{254}$ and the 20-day $E^{253}$ this contamination was determined by following the decay of the fission activity. The fission impurities in the isotopes $Cf^{254}$, $Cf^{250}$, and $Cm^{248}$ were determined by alpha-pulse-height analysis using phosphorus-diffused silicon solid-state detectors. (The alpha/fission ratios necessary for these determinations were taken from Hyde[3] or Phillips et al.[36]) All samples were sufficiently clean so that the fission events from impurities did not significantly influence the results. This influence could therefore be neglected.

## B. Counting Equipment

The thin foil with the radioactivity was placed in the center of two parallel solid-state detectors 14 mm apart. The detectors were guard-ring types as described by Goulding et al.[25] To make them especially suitable for the measurement of the energy of fission fragments, care was taken that only a thin layer of phosphorus was diffused into 1700-Ω-cm p-type silicon. The applied bias voltage was approximately 90V and the effective diameter 10 mm. The detectors retained good resolution for several weeks. The overall counting efficiency of the system was approximately 10%. This comparably high counting efficiency was necessary to get sufficient information from rather small amounts of activity. Counting was carried out in vacuo. The pulses from the fission events in the solid-state detector were sent into a preamplifier, then into a linear pulse amplifier, and finally into a four-dimensional pulse-height analyzer (FDPHA) as described by Bowman et al.[15] The details of the standard electronic equipment are described in the Lawrence Radiation Laboratory Counting Handbook.[24] When two fission fragments entered the two detectors simultaneously, a slow-coincidence system opened the gate of the FDPHA, and the binary equivalents of the two linearly amplified fission pulses were punched onto paper tape.

The stability of the system (except the charge collection efficiency detectors) was checked with a mercury pulser.[24] It sent a pulse at a rate of one per minute through the amplification system and in addition directly into one channel of the FDPHA. Thus any instability of the electronic system could be detected and corrected. The stability of the detectors was checked with a post- and precalibration run of a $Cf^{252}$ source, which was prepared in the same way as the investigated isotope. The schematic of the electronic system is shown in Fig. 25 (a).

In the case of the $E^{253}$ experiment and the second part of the $Cm^{248}$ experiment more complicated equipment was used. Because of the high alpha/fission ratio of $8 \cdot 10^{6}$ in the decay of $E^{253}$, it was necessary to reduce the width of the pulse which came out of the preamplifier to about 1/10 of its original value. This reduced considerably the accidental chance that an alpha particle might distort the measurements by adding its energy to that of the fission fragment. The pulse was amplified linearly in a pulse amplifier, but its width was then too short to be accepted by the FDPHA. This width was therefore stretched in a biased amplifier to an acceptable length for the FDPHA. The bias was set to such a height that only pulses from a fission fragment were accepted by the biased amplifier [Fig. 25 (b)].

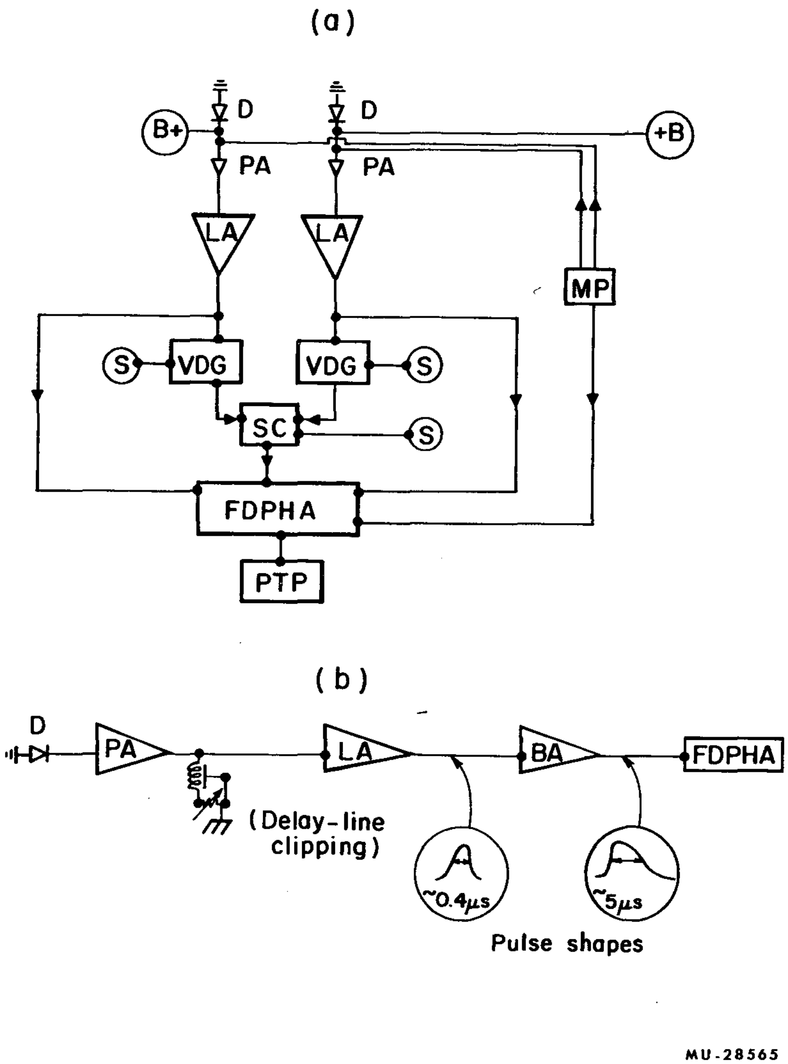


Fig. 25. (a) Electronic system. The components include (D) 1700-Ω-cm phosphorus-diffused silicon detectors,[25] (PA) preamplifiers, (LA) linear amplifiers, (VDG) variable-delay-and-gate units, (S) scalers, (MP) mercury pulser, (B) bias supplies, (FDPHA) four-dimensional pulse-height analyzer, (PTP) paper-tape puncher, and (BA) biased amplifiers.

(b) The linear amplification system used for $E^{253}$.

## C. Calculations

The calculations were performed on the Lawrence Radiation Laboratory IBM 7090 computer by using a FORTRAN-MONITOR compiled program.

The program itself is quite straightforward. The results of each experiment were divided into several files. For each file there exists a post- and precalibration with $Cf^{252}$. The two points necessary to define the linear calibration between energy and channel number for one file were fed into the computer. Then the energies, masses, and the total kinetic energy were calculated for each event separately and the results stored into the respective distributions and arrays. Results of several files could be combined.

Small shifts in the gain of the electronic system are reflected in the shift of the channel number in which the pulser is registered; this pulser shift is corrected for. The channel into which the pulser falls can be thought of as connected with a certain fission-fragment energy $E_p$. As an approximation it was assumed that the energy calibration line was shifted, as shown on Fig. 26 when the pulser was recorded in another channel.

So called "grid fluctuation" in the calculations was avoided by adding a random number betweeen -0.5 and +0.5 to the channel number into which each fission fragment was stored.

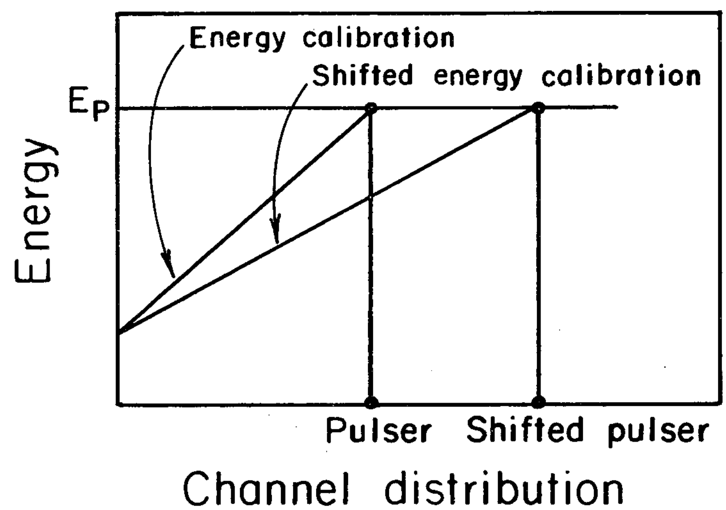


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Fig. 26. Correction for the shift in the linear amplification gain as measured with a mercury electronic pulser.

## D. Effects of the Neutron Emission

The purpose of this study is to obtain the energy and mass distributions for the immediate or primary fission fragments. However, emission of neutrons introduces a dispersion and uncertainty into all distributions.

It will be assumed that all neutrons are emitted from the moving fragments.[15] The kinetic energy E of a primary fragment is lowered by the emission of a number of neutrons $\nu$. This reduced energy $E^*$ is measured finally in the solid-state detectors. Equation (A-1) gives the average relation between E and $E^*$,

$$E^* = E\left[1 - \nu\left(\frac{M}{FM}\right)\right] , \qquad \text{(A-1)}$$

where M is the neutron mass and FM is the fragment mass.

The energy calibration of the solid-state detectors was carried out with $Cf^{252}$. Time-of-flight measurements of the fission fragments of $Cf^{252}$ show that the most probable light fission-fragment energy is 104.7 ± 1.0 MeV for the primary fragments.[29] The light fragment emits an average of 2.1 neutrons.[15,16] The most probable light-fragment energy measured with solid-state detectors is therefore 102.9 ± 1.0 MeV. The mean value of the light-fragment energies with this most probable energy is found to be 102.2 ± 1.0 MeV. It is easy to compute the mean values of the distribution. Therefore, the final calibration of the detector was carried out in such a way that the mean value for the light fragment energy was 102.2 ± 0.2 MeV.

In a similar way the most probable heavy-fragment energy was found to be 78.9 ± 1.0 MeV. The mean value used for the calibration was 78.2 ± 0.2 MeV.

The most serious and complicated influence of the neutron emission is found in the calculation of masses from kinetic energies. From the conservation of momentum, it follows that

$$\frac{EH}{EL} = \frac{ML}{MH} , \qquad \text{(A-2)}$$

where EH and EL are the primary kinetic energies of the heavy and light fragments with masses MH and ML. The sum of the fragment masses is connected to the mass of the fissioning nuclei MA by:

$$MH + ML = MA - \frac{EK}{c^2} , \qquad (A-3)$$

where c is the velocity of light. The last term in this equation arises from conversion of matter into energy and amounts to ~0.2 mass units. The primary energies for one event cannot be calculated. Therefore the masses must be calculated by using:

$$\frac{EH^*}{EL^*} = \frac{ML^*}{MH^*} , \qquad (A-4)$$

where $EH^*$, $EL^*$ are the measured fragment energies and $MH^*$, $ML^*$ the apparent masses.

Terrell has shown recently that the neutron emission introduces first a shift in the mass distribution and second a dispersion of the mass distribution when the fission-fragment masses are determined from the energies measured with solid-state detectors.[31] On the average this mass shift $\Delta M$ from the apparent mass $MH^*$ to the initial mass MH amounts to

$$\Delta M = MH^* - MH \approx (ML \cdot \nu_H - MH \cdot \nu_L)/MA, \qquad (A-5)$$

where $\nu_H$ and $\nu_L$ are the average number of neutrons emitted from the light and heavy fragments ML and MH. The mass shift is always small and varies between -1 and +1 mass units, but for its computation one needs the exact knowledge of the number of neutrons $\nu$ emitted from each fragment mass. This is known experimentally only for $Cf^{252}$.[15,16]

Neutron emission also causes a broadening of the mass-yield curve. This effect can be measured quantitatively as an increase $\Delta\sigma^2(M)$ in the variance of one branch of the mass yield-curve.

Terrell suggested an approximate correction method.[31] This increase in the variance of the mass-yield curve can be corrected for by folding into the observed mass-yield curve M(A) an undispersing function u(y) according to

$$P(A) = \int M(A-y) \cdot u(y)\, dy, \qquad \text{(A-6)}$$

where P(A) is the dispersion-corrected mass-yield curve. The only condition that must be satisfied by the function u(y) is that it has a variance of $[-\Delta\sigma^2 (M)]$.

The qualitative effects of neutron emission are clear. A reliable quantitative treatment is not possible so far. Figure 27(a) shows the light branch of the mass-yield curve for $Cf^{252}$ as determined in this paper. [Its variance: $\sigma^2(M) = 52$ (mass units)$^2$]. Figure 27(b) shows the same mass-yield curve, but corrected for the mass shift according to Equation (A-5). Its variance is $\sigma^2(M) = 46$. A primary mass-yield curve for $Cf^{252}$ as deduced from time-of-flight data is shown in Fig. 27(d).[31] It measures $\sigma^2(M) = (48 \pm 4)$ and contains no symmetric fission events, which means no events at mass 126 = 252/2. When the variance of the shift-corrected mass-yield curve [as shown on Fig. 27(b)] is reduced to 44 (mass-units)$^2$, it still contains symmetric fission events. Figure 26(c) shows the mass-yield curve obtained when the variance is reduced to the extent that there are no longer any symmetric fission events. Its variance is $\sigma^2(M) = 37$. This curve should approximate a primary mass-yield curve, but its variance is much smaller than the variance of the mass-yield curve deduced from time-of-flight measurements of the fission fragments of $Cf^{252}$. Therefore, the conclusion is that the primary mass-yield curve as deduced from time-of-flight data does not agree with the primary mass-yield curve deduced from this work.

One might obtain closer agreement when more precise time-of-flight measurements of the fission fragments of $Cf^{252}$ are available.

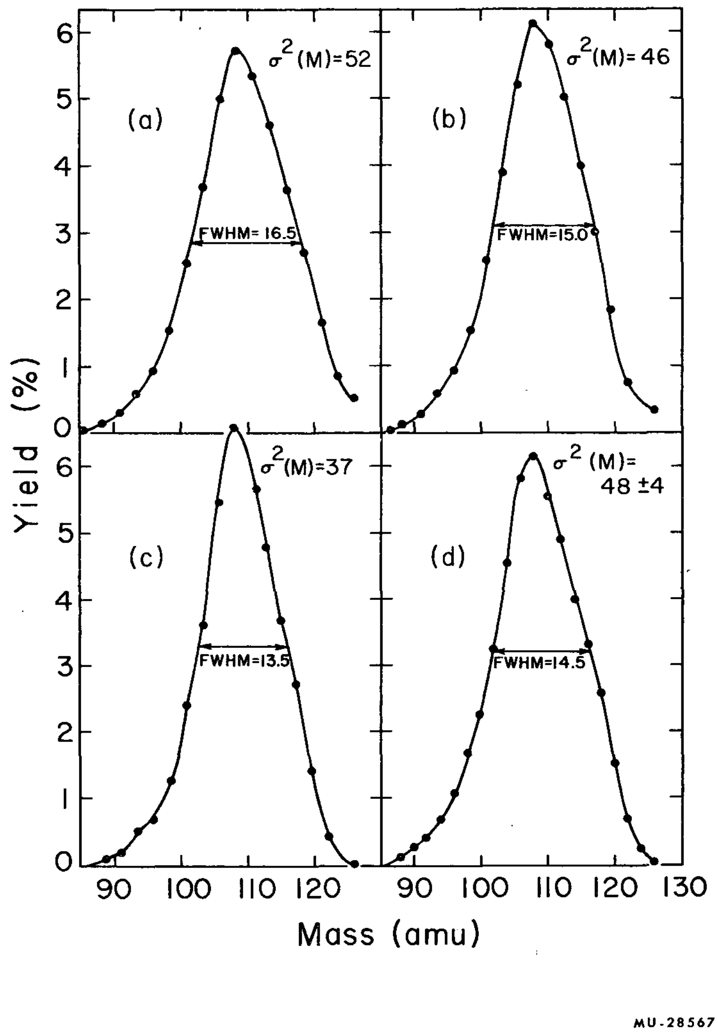


Fig. 27. Mass-yield curves for $Cf^{252}$; (a) Directly observed; (b) Corrected for mass shift; (c) Corrected for mass shift and dispersion; (d) Primary mass-yield curve derived from time-of-flight experiments.[29]

Such experiments are now in progress.[34] It should also be mentioned, that the mean values for the two branches in the mass distribution for $Cf^{252}$, as determined by time-of-flight techniques and solid-state measurements, agree within the limits of errors given for the most probable single fragment energies.

Since it is not possible to obtain in a reliable way the primary mass-yield curve for $Cf^{252}$, the computation of the primary mass-yield curve for the other isotopes is omitted. (There also enters the additional uncertainty that the variation of $\nu$ with A is known only for $Cf^{252}$.)

## E. Approximate Treatment of the Total Energy Balance

During fission the total energy released ETO is made up of

(a) The kinetic energy EK of the primary fragments.

(b) The internal excitation energy EX of both fragments.

If one assumes that all neutrons are emitted from the separated fragments, the total energy release of one fission event is

$$\mathrm{ETO} = \mathrm{EK} + \mathrm{EX}. \qquad (\text{A-7})$$

The excitation of the fragments is lowered by the emission of a number of neutrons $\nu$. Each neutron lowers the excitation energy by its binding energy and the kinetic energy it carries off in the center-of-mass system of the moving fission fragment. Gamma rays are also emitted. Denoting the average neutron binding energy by NBE, the average kinetic energy of the neutrons in the center-of-mass systems by KEN, and the energy carried off by $\gamma$-rays by EG, we have for one event,

$$\mathrm{EX} = \nu\,(\mathrm{NBE} + \mathrm{KEN}) + \mathrm{EG}. \qquad (\text{A-8})$$

Averaged in a weighted form over all possible fission modes, the total energy balance can be written as

$$\langle \mathrm{ETO} \rangle = \langle \mathrm{EK} \rangle + \bar{\nu}\,(\langle \mathrm{NBE} \rangle + \langle \mathrm{KEN} \rangle) + \langle \mathrm{EG} \rangle . \qquad (\text{A-9})$$

This allows calculation of $\bar{\nu}$ (the average number of neutrons in spontaneous fission) from energy data alone, according to

$$\bar{\nu} = \frac{\langle \mathrm{ETO} \rangle - \langle \mathrm{EK} \rangle - \langle \mathrm{EG} \rangle}{\langle \mathrm{NBE} \rangle + \langle \mathrm{KEN} \rangle} . \qquad (\text{A-10})$$

The average total energy release $\langle \mathrm{ETO} \rangle$ can now be calculated when the mass-yield curve and ETO for each pair of fragment masses is known. The latter value ETO has recently been calculated by Milton.[11] He used the mass formula of Cameron to compute the masses of the fragments and assumed that the charge distribution for a given mass was gaussian with $\sigma = 0.700$ charge units. He also calculated the average neutron binding energy NBE for one fragment mass. The average kinetic energy of the neutrons ⟨KEN⟩ in the center-of-mass system is given by Bowman et al.[15] The average gamma-energy ⟨EG⟩ was assumed to be 8.5 MeV for all investigated isotopes.[3] The mean prompt total kinetic energy and the mass-yield curve is taken from the present work. The results of these calculations are shown in Table V. The last column contains the difference D between ⟨ETO⟩ and ⟨ETC⟩, where ⟨ETC⟩ is the total kinetic-energy release calculated according to Eq. (A-9) from the experimental value for $\bar{\nu}$.

The difference D is essentially a measurement of the accuracy of this total kinetic-energy balance. It is interesting that D is only on the order of a few MeV, since the calculations involve the following approximations and uncertainties:

(a) The uncertainty in the mean prompt total kinetic-energy release of the standard $Cf^{252}$ is (±5) MeV.[29]

(b) The mass-yield curve used here is not identical with the primary mass-yield curve.

(c) It is not known how much the mean total gamma-ray energy ⟨EG⟩ and the mean kinetic energy of neutrons ⟨KEN⟩ vary for different spontaneously fissioning isotopes.

(d) The accuracy of Cameron's mass equation is not known for the isotopes of interest here.

(e) It is not known how good is the assumption that the number of neutrons $\bar{\nu}$ emitted in spontaneous fission depends only on the mass A of the fissioning nuclei.[3]

Table V. Energy balance in spontaneous fission (MeV)

| Isotope | ⟨ETO⟩[a] | ⟨EX⟩[b] | ⟨NBE⟩[c] | $\bar{\nu}$ exp.[d] | D = ⟨ETO⟩ - ⟨ETC⟩ |
|---|---|---|---|---|---|
| $Fm^{254}$ | 230.4 | 41.7 | 5.63 | 4.05 | + 5 |
| $E^{253}$ | 225.0 | 37.1 | 5.46 | 3.9[e] | + 2 |
| $Cf^{254}$ | 216.5 | 32.0 | 5.01 | 3.9 | - 2 |
| $Cf^{252}$ | 216.1 | 33.2 | 5.16 | 3.8 | - 1 |
| $Cf^{250}$ | 216.8 | 32.0 | 5.43 | 3.5[e] | - 1 |
| $Cm^{248}$ | 204.9 | 26.0 | 5.05 | 3.3[e] | - 4 |

[a] ⟨ETO⟩ is the mean total energy release.

[b] ⟨EX⟩ is the mean fragment excitation energies.

[c] ⟨NBE⟩ is the mean neutron binding energy.

[d] $\bar{\nu}$ the mean number of neutrons emitted.

[e] estimated value.

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