

amount of energy available makes it appear unlikely that this level would be populated from Ga^{72} in any case. Either a 1^- or 2^- assignment seems appropriate for this level. The intensities of the gamma-ray transitions from the 2.91- and 3.74-Mev levels to the first excited state relative to the ground-state transitions are in good agreement with the relative intensities expected for single-particle transitions,²² although presumably the transitions are not of this simple nature.

Our results seem to indicate only a weak feeding of the 3.34-Mev level in Ge^{72} from As^{72} . If this would imply a spin 4^- for this level, one would find disagreement with the observed 3.34-Mev ground-state transition in Ge^{72} .¹

²² V. F. Weisskopf, *Phys. Rev.* **83**, 1073 (1951).

VIII. ACKNOWLEDGMENTS

We wish to thank Me. L. Piette for performing the chemical separations and Mr. H. Enderton for help in the analysis of the data.

APPENDIX A

In Fig. 3 is shown the collimated gamma-ray spectrum of our second source two weeks after the time of irradiation. The following known lines are in evidence: 0.053 Mev (As^{73}), 0.175 Mev (As^{71}), 0.59 and 0.63 Mev (As^{74}), and 0.835 Mev (As^{72}). In addition to these, there are lines at 0.310 and 1.10 Mev which have been assigned to As^{71} since they decay with the 60-hour half-life characteristic of As^{71} . The intensities of these two lines relative to the 0.175-Mev gamma ray are 5 and 9%, respectively.

Spontaneous Fission Kinetics of $\text{Cf}^{252}\dagger$

A. B. SMITH, A. M. FRIEDMAN, AND P. R. FIELDS
Argonne National Laboratory, Lemont, Illinois

(Received January 6, 1956)

A double ionization chamber is used to determine the spontaneous fission modes of Cf^{252} [$\tau_{1/2}(\text{fission}) = 66$ years]. The most probable energies of the light and heavy fragments are measured to be 107 and 79 Mev, respectively. The asymmetry of the process is found to agree with chemical mass-yield determinations. The results of the experiment are compared with the kinetics of other fission processes.

INTRODUCTION

THE kinetics of particle-induced fission have been studied in detail.¹⁻⁵ It has been theoretically predicted^{6,7} and experimentally verified^{5,8} that heavy, naturally occurring isotopes spontaneously fission with long half-lives. The low probability of the process makes experimental measurements exceedingly difficult. However, the availability of transuranic elements with short fission lifetimes provides an excellent opportunity for the study of the spontaneous process. These nuclei, existing in relatively high states of excitation, should exhibit the same fission characteristics as stable nuclei excited by relatively energetic particles. This is partially born out by the experimental mass-yield measurements and kinetic energy determinations^{5,9,10} in Pu^{240} and Cm^{242} .

[†] This work performed under the auspices of the U. S. Atomic Energy Commission.

¹ M. Deutsch, Manhattan District Declassified Report MDDC 945 (unpublished).

² D. Brunton and G. Hanna, *Can. J. Research* **28A**, 190 (1950).

³ D. Brunton and W. Thompson, *Can. J. Research* **28A**, 498 (1950).

⁴ R. B. Leachman, *Phys. Rev.* **87**, 444 (1952).

⁵ E. Segrè and C. Wiegand, *Phys. Rev.* **94**, 157 (1954).

⁶ N. Bohr and J. Wheeler, *Phys. Rev.* **56**, 426 (1939).

⁷ D. Hill and J. Wheeler, *Phys. Rev.* **89**, 1102 (1953).

⁸ W. J. Whitehouse and J. Galbraith, *Phil. Mag.* **41**, 429 (1950).

⁹ L. Glendenin and E. Steinberg, *Phys. Rev.* **95**, 431 (1954).

¹⁰ R. L. Shurey, University of California Radiation Laboratory Report UCRL-793 (unpublished).

Among the transuranic elements, $^{98}\text{Cf}^{252}$ is ideally suited to an investigation of spontaneous fission. It has a fission half-life¹¹ of 66 years and a relatively long-lived alpha activity (2.2 years), making good measurements possible in a short period of time. A careful study of the alpha decay¹² has shown this isotope to follow the systematics of even-even alpha emitters. Glendenin and Steinberg¹³ have determined chemically the spontaneous fission mass yields. They obtain a most probable mass ratio of ~ 1.34 along with evidence of typical mass-yield fine structure. The most probable number of neutrons released in the fission of Cf^{252} is 3.54–4.06.¹⁴

In the present experiment the total fragment energy in the spontaneous fission of Cf^{252} is measured. The relative probability of the various modes of fission is obtained and a comparison made with other fission processes.

SOURCE

The californium was taken from a sample that had been made by neutron irradiation of Pu^{239} in the materials testing reactor.¹¹ It was a radiochemically pure sample of californium consisting, by mass, of 50% Cf^{252} , 10% Cf^{251} , and 40% Cf^{250} so that essentially

¹¹ L. B. Magnussen *et al.*, *Phys. Rev.* **96**, 1576 (1954).

¹² F. Asaro *et al.*, *Phys. Rev.* **100**, 137 (1955).

¹³ L. Glendenin and E. Steinberg, *J. Inorg. and Nuclear Chem.* **1**, 45 (1955).

¹⁴ Hicks, Ise, and Pyle, *Phys. Rev.* **98**, 1521 (1955).

all the observable fission activity was due to the Cf^{252} .

The source was prepared by volatilization of the californium onto a $5\text{--}10\ \mu\text{g}/\text{cm}^2$ Formvar film. Previous work had shown that the amount of inert material accompanying the californium had to be reduced to the microgram range in order to keep the volatilized atoms from breaking the collecting film.¹⁵ Therefore, the californium was carefully purified before volatilization by elution from a series of cation and anion resin columns with 12M hydrochloric acid and finally by extraction into a benzene solution of thenoyltrifluoroacetone (TTA). The californium was re-extracted from the TTA solution into a single drop of 0.1M hydrochloric acid which was then evaporated on the tungsten filament. The more volatile impurities were removed from the sample by heating the filament to about $800\text{--}900^\circ\text{C}$. The californium was then volatilized at about 1700° onto the film through a collimating mask which served as a radiation shield between the hot filament and the collecting film.

APPARATUS

The detector is a double-sided ionization chamber of conventional design.^{16,17} A thin source is placed between the two halves, and the fragments from binary fission are measured in time coincidence. The amplifier band width is adjusted to permit electron collection only, and a grid structure within the chamber shields the collectors from the source.

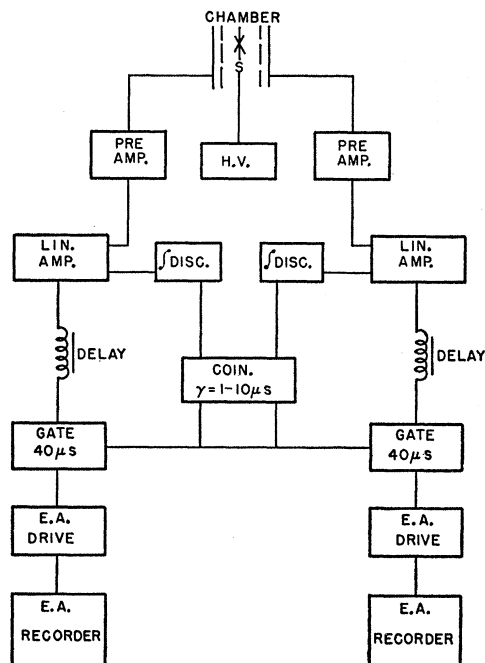


FIG. 1. Block diagram of circuitry used in the measurement of fragment energies.

¹⁵ F. Wagner (private communication).

¹⁶ D. H. Wilkinson, *Ionization Chambers and Counters* (Cambridge University Press, New York, 1950).

¹⁷ L. Hewig *et al.*, *Rev. Sci. Instr.* **26**, 929 (1955).

The stopping gas is 90% argon and 10% nitrogen at a pressure of one atmosphere. This gas is continuously circulated through a calcium purifier. The voltages within the chamber are chosen to give saturation of the fission-fragment ionization pulses. The alpha-particle resolution of the instrument is 1.5% (peak width at one-half maximum). It is felt that the limit of resolution is set by the recording equipment rather than the chambers.

The circuitry is composed of conventional units. The block diagram of Fig. 1 shows the completed assembly. Events occurring in either side of the chamber must produce more than twice the ionization of the alpha particles from the source before they are accepted by the discriminator circuits. After a pulse has fulfilled the amplitude criteria, it is mixed in a coincidence circuit with similar pulses from the other side of the chamber. Unless simultaneous events occur, the recorders are not actuated. The amplitudes of the respective pulses appear directly on the Esterline-Angus recording milliamper meters. These two units are run at the same speed permitting a one to one time correspondence between events from either side of the chamber. The coincidence resolving time of $3\ \mu\text{sec}$ is short enough to reduce pulse pile up to a negligible amount. The recording circuits have dead times of $\frac{1}{2}$ second which is sufficient for the phenomena under study. The stability of the entire system is periodically checked by introducing a precision test pulse onto the collecting electrodes of the chamber. The reliability of the apparatus is excellent.

EXPERIMENTAL MEASUREMENTS

Four coincident determinations of the fragment energy distribution are made. Each analysis involves some 5000 events spread over several days time. The absolute calibration of the fragment energy loss due to gas ionization is made by comparison with the known

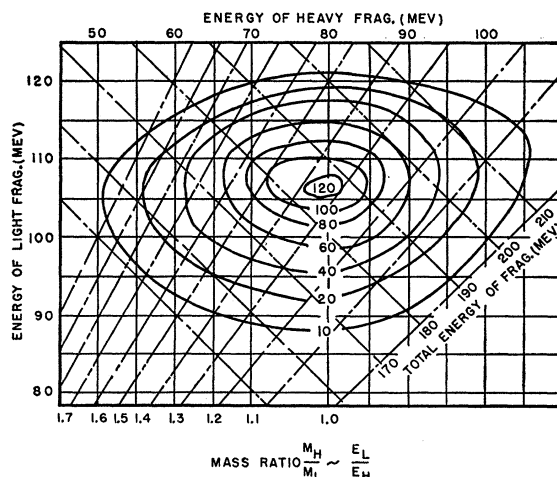


FIG. 2. Relative probabilities of Cf^{252} spontaneous fission modes.

TABLE I. Fission fragment energies.

Element	Reference	Energy light fragment Mev	Energy heavy fragment Mev	Total fragment energy Mev	Z^2/A	Mass ratio
$^{233}\text{U} + n$	2	99.6	60.6	160.2	36.17	1.64
$^{235}\text{U} + n$	2	101.2	64.4	165.6	35.86	1.57
$^{238}\text{U} + n$	8	~100.0	~65.0	~165.0	35.56	~1.55
$^{239}\text{Pu} + n$	3	101.2	69.8	171.0	36.81	1.44
^{240}Pu	5	~101.0	~70.0	~171.0	36.81	~1.45
^{242}Cm (Berkeley)	10	112.4	83.5	195.9	38.08	1.34
^{242}Cm (Chalk River)	20	~101.0	~70.0	~171.0	38.08	~1.44
^{252}Cf Present exper.		107.0	79.0	186.0	38.11	1.36

ionization produced by Cf^{252} alpha particles and U^{235} fission fragments. A U^{235} source is placed directly behind the Cf^{252} sample so both sources see the same collimator and source backing material. Comparison of the fragment distributions with and without thermal neutrons present gives an accurate energy comparison. A separate measurement shows the thermal-neutron-induced fission of the Cf^{252} sample to be negligible under the experimental conditions existing during calibration.

Throughout the measurements careful consideration is given to collimator and sample losses. Because of the coincident nature of the experiment, physical collimation of one fragment effectively collimates the other. Thus a comparison of the response of the chamber's two sides gives a measure of collimator and source losses. The measured losses agree with those calculated from the experimental data of Lassen.¹⁸ It is felt that the most probable energy values for the light and heavy fragment groups are accurate to ± 3 Mev. It should be emphasized that the measured quantities are ionization energy losses and must be corrected for non-ionization loss to give the true fragment energies.

RESULTS AND DISCUSSION

The experimental results are presented as a topological plot in Fig. 2. Fragment mass distribution, energy distribution, and asymmetry can be obtained directly from this diagram. A comparison of the fission energetics of Cf^{252} with other known fission processes is given in Table I. All the energy measurements have been corrected to include non-ionizing energy loss by the fragments using the ionization defects measured by Leachman.⁴ This correction amounts to a 7% increase over the measured ionization energy. The most probable value of the fragment mass ratio is measured to be 1.36 in good agreement with the chemical yield determination. As expected, no fine structure is observed in the energy distributions owing to the statistical spread in the ionizing power of the fragments.

In the absence of a completely satisfactory theory of fission an empirical evaluation of the experimental

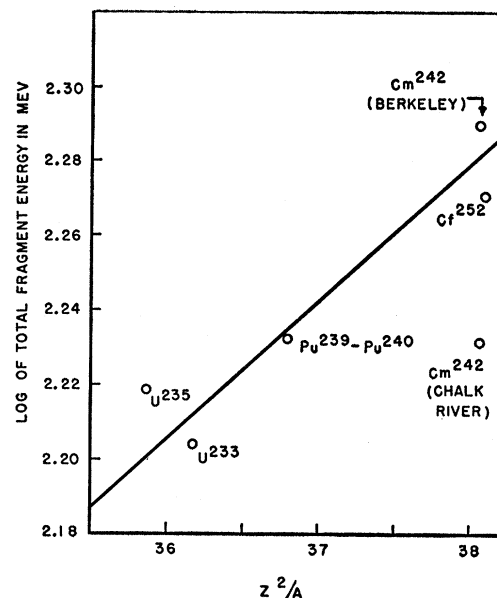


FIG. 3. Total fragment energies from the fission of even-even nuclei.

measurements must be resorted to. It has been shown that the spontaneous fission half-lives of even-even nuclei are simply related by $\ln \tau_{1/2} = a - bZ^2/A$.¹⁹ Qualitatively, this sort of dependence is to be expected as the lifetimes will be a decreasing function of the Coulomb-surface energy differences. In correlating fission energies we assume thermal-neutron-induced fission to be equivalent to the spontaneous process in a nucleus containing an additional neutron and having an excitation energy equivalent to the neutron binding energy. Further, we assume the difference between the fission energy and the fragment energy to be essentially constant for all fission processes. With these two assumptions, we plot in Fig. 3 the energy of fission of even-even nuclei as a function of the fission parameter Z^2/A . With the exception of the preliminary Cm^{242} results,²⁰ the experimental energy values indicate a logarithmic dependence of fission energy on Z^2/A . One thus concludes that the energetics of fission are governed largely by Coulomb-surface forces. That other forces do influence spontaneous fission is evidenced by the fission half-lives of even-even isotopic chains. It is believed that neutrons beyond closed subshells tend to couple with the collective motion of the nucleus to produce distortions and thus shorten fission half-lives.¹⁹ Before the effect (if any) of such forces on fission energetics can be determined, more experimental data must become available.

¹⁹ J. R. Huizenga, Geneva Conference Paper No. 836 (to be published).

²⁰ G. C. Hanna *et al.*, Phys. Rev. 81, 466 (1951).

¹⁸ N. Lassen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 25, 11 (1949).