Radiochemical Investigation of the Spontaneous Fission of Cm^{242}†

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Spontaneous fission of Cm^{242} has been investigated by radiochemical determination of the yields of 21 fission products ranging in mass number from 91 to 140. Fission is observed to be asymmetric. The yield-mass curve exhibits somewhat narrower peaks than observed in induced fission and a peak-to-trough ratio of greater than 700 to 1. Prominent fine structure is observed in the mass distribution, and an analysis of this phenomenon is made. The distribution of nuclear charge in spontaneous fission appears to be essentially the same as that in low-energy induced fission.

I. INTRODUCTION

THE phenomenon of spontaneous fission was considered theoretically by Bohr and Wheeler[1] and first observed in uranium by Petrzhak and Flerov.[2] Later studies on spontaneous fission, recently summarized by Segrè,[3] established the decay constants for this process in a number of heavy nuclides, and, in the case of U^{238}, a value for the average number of neutrons emitted per fission. It is of great interest to compare the characteristics of the mass distribution resulting from this process with that of induced fission. Extensive radiochemical investigations of the latter have shown a pronounced asymmetry in fission with the probabilities of symmetrical fission modes ("trough") and, perhaps, very asymmetric modes ("wings") increasing with excitation energy of the fissioning nucleus. It might, therefore, be expected that spontaneous fission, occurring in the ground state, would exhibit a lower trough and narrower distribution of mass than induced fission. Anomalous yields have been observed in induced fission in the mass region of the 82-neutron shell[4,5] and its complement.[6,7] This so-called "fine structure" is presumably associated with a preference for a closed-shell configuration in the fission act[5,6] and the excessive evaporation from primary fission fragments of the loosely bound neutrons just outside closed shells.[8,9] It is expected that this phenomenon will also be prominent in spontaneous fission. Ionization chamber studies have been made recently on the kinetic energy distribution of the fragments from the spontaneous fission of U^{238} [10] and Cm^{242}.[11,12] Predominantly asymmetric fission was indeed observed, not significantly different from that of low-energy neutron-induced fission. However, the rather poor resolution inherent in the ionization method precludes a significant comparison of the width of the mass distribution and frequency of symmetric fission modes in spontaneous and induced fission.

More detailed information on the mass distribution in spontaneous fission has been obtained by Thode and co-workers[13,14] and by Wetherill[15] using the highly sensitive mass spectrometric technique. These investigators examined the isotopic composition of the rare gases krypton and xenon produced in various minerals by spontaneous fission of U^{238} and Th^{232}. The observed abundances of the fission-produced isotopes of these elements indicate a narrower mass distribution with lower frequency of fission modes near symmetry as would be expected. Evidence was also presented for an abnormally high yield (fine structure) at mass 132, as might be expected from the closed-shell effects mentioned above. Although a rough yield-mass curve may be deduced from the relative isotopic abundances of the krypton and xenon alone under the reasonable assumption of identical yields at complementary masses, these mass spectrometric data do not provide a complete mass distribution.

The radiochemical method, which supplies detailed information on the distribution of mass and charge in fission, has not been applied previously to the study of spontaneous fission since the slow rate of decay by this process in available materials (e.g., U^{238} and Th^{232}) does not provide sufficient fission product activity for

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[1] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
[2] K. A. Petrzhak and G. N. Flerov, Compt. rend. acad. sci. (U.S.S.R.) **28**, 500 (1940); J. Phys. (U.S.S.R.) **3**, 275 (1940).
[3] E. Segrè, Phys. Rev. **86**, 21 (1952).
[4] H. G. Thode and R. L. Graham, Can. J. Research **A25**, 1 (1947); MacNamara, Collins, and Thode, Phys. Rev. **78**, 129 (1950).
[5] D. R. Wiles, M.Sc. thesis, Dept. of Chemistry, McMaster University, Hamilton, Ontario, August, 1950 (unpublished); Wiles, Smith, Horsley, and Thode, Can. J. Phys. **31**, 419 (1953).
[6] Glendenin, Steinberg, Inghram, and Hess, Phys. Rev. **84**, 860 (1951).
[7] D. R. Wiles, Ph.D. thesis, Department of Chemistry, Mass. Inst. Technol., Cambridge, Massachusetts, November, 1953 (unpublished).
[8] L. E. Glendenin, Laboratory for Nuclear Science and Engineering, Mass. Inst. Technol. Tech. Rept. No. 35, December, 1949 (unpublished).
[9] A. C. Pappas, Laboratory for Nuclear Science and Engineering, Mass. Inst. Technol. Tech. Rept. No. 63, September, 1953 (unpublished).
[10] W. J. Whitehouse and W. Galbraith, Phil. Mag. **41**, 429 (1950).
[11] R. L. Shuey, University of California Radiation Laboratory Report UCRL-959, October, 1950 (unpublished).
[12] Hanna, Harvey, Moss, and Tunnicliffe, Phys. Rev. **81**, 466 (1951).
[13] J. MacNamara and H. G. Thode, Phys. Rev. **80**, 471 (1950).
[14] W. H. Fleming and H. G. Thode, Phys. Rev. **92**, 378 (1953).
[15] G. W. Wetherill, Phys. Rev. **92**, 907 (1953).

accurate measurements. With the recent availability of milligram quantities of Cm^{242}, which has a highly favorable spontaneous fission rate (4.66×10^5 fissions/min mg),[12] it became feasible for the first time to investigate the spontaneous fission process in detail by the radiochemical method. The great advantage of Cm^{242} is illustrated in Table I, where the spontaneous fission rates[3,12] and the activity of a typical fission product (saturation disintegration rate for a 6 percent fission yield) for Th^{232}, U^{238}, and Cm^{242} are compared. It is apparent that the determination of yields in the region of symmetric spontaneous fission, which are expected to be even lower than those in neutron-induced fission of U^{235} (about 0.01 percent), would require formidable quantities of Th^{232} or U^{238}. For example, at least a ton of U^{238} would be required to obtain a measurable activity of a fission product with a yield of 0.01 percent, whereas only one milligram of Cm^{242} would suffice.

Although the fission rate of Cm^{242} is adequate for a convenient radiochemical study of spontaneous fission, there are other factors which must be considered. Most important, of course, is the absence of appreciable fission product activity from any source other than spontaneous fission. Neutrons arising from spontaneous fission and from the (α,n) reaction, particularly in light elements, could induce fission, for example, in Am^{242} (thermal neutron fission cross section = 6000 barns[16]) which is likely to be present in the Cm^{242} source. Since induced fission may produce fission products in the region of the trough in yields perhaps as high as 0.1 percent (much higher than those expected for spontaneous fission), an induced fission rate equal to 10 percent of that of spontaneous fission would result in apparent observed spontaneous fission yields in this region of about 0.01 percent. This value is just at the limit of detection for spontaneous fission yields from a one-milligram source of Cm^{242}. It is shown by the following calculation that the contribution of neutron-induced fission is at most only 2 percent and probably much lower, and is hence a negligible source of error.

The production of 1.6 neutrons per spontaneous fission by the α, n reaction in an aqueous solution of Cm^{242} is estimated by taking a thick target yield in oxygen of one neutron per 10^7 alpha particles of about 6 Mev energy.[17] No other light elements with appreciable (α,n) yields are present in solution with the possible exception of slight boron impurities from Pyrex glass containers. As a conservative approximation, the total neutron yield from (α,n) reactions is taken as three neutrons per spontaneous fission (equivalent to allowing boron to be present in the very unlikely concentration of about 0.2 molar). In addition, the emission of three neutrons accompanies each spontaneous fission (see Sec. III), giving a total upper estimate of about six neutrons produced in the solution per spontaneous fission. An upper limit to the effect of these neutrons in inducing fission in a 10-ml aqueous solution containing about one milligram each of americium and curium (see Sec. IIA) may be estimated by assuming that all of the neutrons are thermalized and absorbed. The probability of a neutron colliding with an atom of americum or curium and inducing fission during the approximately 30 collisions required for thermalization is about 2×10^{-3}. The probability of inducing fission after thermalization is equal to the ratio of the total thermal neutron fission cross section of americium and curium to the total thermal neutron absorption cross section of the solution and is about 10^{-3}. Thus, for six neutrons per spontaneous fission, the ratio of induced to spontaneous fissions is the order of 2 percent. Since a majority of the neutrons will certainly escape the solution after a few collisions, the above result is at least an order of magnitude high.[18] Other causes of induced fission such as gamma rays, cosmic rays, etc., are much less important.

The intense alpha radioactivity of Cm^{242} (7.37×10^{12} alphas/min mg)[19] also presents some problems in radiochemical operations. Facilities and techniques for handling such sources have been developed by the Nuclear Chemistry and Health Chemistry groups at the University of California Radiation Laboratory, and these were made available for this investigation. In general, decontamination factors of the order of 10^{10} would be required to reduce the alpha activity to a reasonably low level in an isolated fission product sample. The standard radiochemical procedures for isolation of fission products[20] are easily adapted to this need. Gas evolution and the slight temperature rise in solutions of such intense alpha radioactivity are minor problems.

The foregoing considerations indicate the feasibility of a radiochemical study of the spontaneous fission process in Cm^{242} with only minor modifications from the well-established techniques employed in the studies of induced fission. The present investigation was therefore undertaken to establish, within the limitations of

TABLE I. Comparison of spontaneous fission rates.

Nuclide	Fissions/min mg	Saturation activity of Ba^{139} (dis/min)
1.4×10^{10}-yr Th^{232}	2.5×10^{-6}	1.5×10^{-7}
4.5×10^{9}-yr U^{238}	4.14×10^{-4}	2.5×10^{-5}
162.5-day Cm^{242}	4.66×10^{5}	2.8×10^{4}

[16] Street, Ghiorso, and Thompson, Phys. Rev. **85**, 135 (1952).

[17] H. L. Anderson, "Neutron from Alpha Emitters" (National Research Council Committee on Nuclear Science, December, 1948) Prelim. Rept. No. 3 of Nuclear Science Series (unpublished).

[18] We are indebted to Dr. B. I. Spinrad of the Physics Division, Argonne National Laboratory and Prof. A. Turkevich of the Inst. for Nuclear Studies, Univ. of Chicago, for helpful discussions on this subject.

[19] Hanna, Harvey, and Moss, Phys. Rev. **78**, 472 (1950).

[20] *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Div. IV, Vol. 9.

time and amount of Cm^{242} available, as complete a mass distribution as possible with particular attention to the frequency of symmetrical fission modes, the presence of fine structure, and charge distribution.

II. EXPERIMENTAL

A. General Procedure

The Cm^{242} source used in this work was isolated by Dr. S. G. Thompson and Dr. G. H. Higgins and personnel of the Health Chemistry group of the University of California Radiation Laboratory from americium which had been irradiated in the Canadian pile at Chalk River. Cation and anion exchange techniques were employed for the separation of the curium (along with some of the americium) from the intense fission product rare earth activities produced mainly by fission of Am^{242}. The final sample prepared for our use consisted of approximately 1 mg each of curium (90 percent Cm^{242}, 5 percent Cm^{243}, and 5 percent Cm^{244}) and americium (75 percent Am^{241}, 2 percent Am^{242}, and 23 percent Am^{243}), with Cm^{242} accounting for more than 99 percent of the alpha activity and about 97 percent of the spontaneous fissions (3 percent being due to Cm^{244}).

All radiochemical operations were carried out in enclosed, vented glove-boxes ("Berkeley box") commonly employed for the safe handling of intense sources of alpha activity.[21] Since the curium source continuously produces spontaneous fission product activities and may contain additional fission product activities produced during the pile irradiation of the Am^{241} and incompletely removed in the purification of the curium, it was necessary to remove all activities of the element of interest before the determination of spontaneous fission yields. In general, this was accomplished by successive quantitative separations of added carrier from the curium solution, the last removal establishing the zero time for growth of activities of the element from spontaneous fission. A known weight of carrier was then added, and the fission rate of the solution was determined by alpha counting a small aliquot and applying the known alpha-to-fission ratios for Cm^{242} and Cm^{244}. Periods of growth varying from about one day to two weeks were employed to suit the half-life ranges of the nuclides under investigation. The fission rate was corrected for decay of Cm^{242} during the growth period where necessary.

The growth period was ended by separation of the carrier from the curium. After making sure that no more than 0.1 percent of the curium accompanied the carrier, the latter was transferred to a secondary Berkeley box so that final decontamination from curium and purification from other fission products was made in an environment free from the bulk of the intense alpha activity. Before removal from the secondary box for final precipitation and mounting, the fission product sample was always assayed for alpha activity to verify the adequacy of decontamination from curium. The final precipitate in each case was collected on a small circle of filter paper, weighted to determine the chemical yield, mounted at the center of a 2.5×3.25 in. cardboard support, covered with thin Cellophane, and its counting rate measured with an end-window Geiger counter. A ring of anticoincidence counters was used to reduce background. The radiochemical purity of each fission product was determined by its absorption and decay characteristics.

The observed counting rates were corrected for chemical yield, decay, fractional saturation during the growth period, and the presence of other activities due to genetically related or isotopic species. Empirical self-absorption and self-scattering factors[22] were used to correct for the effects of sample weight (usually about 20 mg), and an exponential absorption correction was made for absorption in the Cellophane covering over the sample, in the air layer between the source and the counter, and in the counter window. A geometry factor of 24.0 percent was determined by the RaDEF calibration method[23] for the source position used.

B. Individual Determinations

Strontium (9.7-hr Sr^{91} *and* 2.7-hr Sr^{92})

Precipitations of $Sr(NO_3)_2$ with fuming HNO_3 were made to remove all strontium activities from the curium solution. After a growth period of 65.6 hr, strontium was again removed as nitrate and purified[24] from contaminating fission products and curium by $Fe(OH)_3$ and $BaCrO_4$ removals, and finally precipitated as $SrCO_3$ for mounting and counting. The last $Fe(OH)_3$ precipitation was made about two hours after the end of the growth period, removing the yttrium activities (17-min Y^{94} and 11-hr Y^{93}) resulting from the complete decay of the 2-min Sr^{94} and 7-min Sr^{93} and establishing a zero time for the growth of the 51-min Y^{91m} and 3.5-hr Y^{92}.

The determination of 9.7-hr Sr^{91} and 2.7-hr Sr^{92} was made by analyses of decay and absorption curves taking into account the activities of the 51-min Y^{91m} and 3.5-hr Y^{92} daughters, and a long-lived component of low intensity probably due to 53-day Sr^{89} and 61-day Y^{91}.

Molybdenum (67-hr Mo^{99})

Precipitations of molybdenum as sulfide were employed to start growth periods of 72.0 and 89.5 hr. A modified alpha-benzoinoxime procedure[25] was then used for the isolation of molybdenum. An initial sulfide separation was followed with a specific tellurium removal by reduction to metal with SO_2. The molybdenum was then precipitated with alpha-benzoinoxime, purified

[21] See, e.g., B. B. Cunningham, Nucleonics **5**, No. 5, 83 (1949).

[22] Engelkemeir, Seiler, Steinberg, and Winsberg, reference 20, Paper 4.

[23] T. B. Novey, Rev. Sci. Instr. **21**, 280 (1950).

[24] L. E. Glendenin, reference 20, Paper 236.

[25] N. E. Ballou, reference 20, Paper 257.

by repeated precipitations of $Fe(OH)_3$, and finally isolated as $PbMoO_4$.

Since complete decay of the shorter-lived molybdenum activities occurred before final isolation and counting of the samples, only the 67-hr Mo^{99} was observed. The molybdenum samples were counted through 12.5 mg/cm^2 of aluminum to filter out the low-energy conversion electrons from 5.9-hr Tc^{99m}. Allowance for this additional absorber was made in the absorption correction.

Ruthenium (40-*day* Ru^{103}, 4.5-hr Ru^{105}, *and* 1.0-yr Ru^{106})

Ruthenium activities were removed from the curium solution by sulfide precipitations in preparation for growth periods of one and two days. The ruthenium was again precipitated as sulfide to end these growth periods and was purified[26] by distillation into NaOH from acidic $KMnO_4$, removal of $Fe(OH)_3$, and reduction to metal with magnesium. A longer period of 6.8 days was employed for the growth of the 40-day Ru^{103} and 1.0-yr Ru^{106} and was started and ended by distillation of ruthenium from $HClO_4$ solution.[26]

The 4.5-hr Ru^{105} was determined in the three runs by analyses of decay and absorption curves. Corrections were made for the presence of 45-sec Rh^{105m} and 36.5-hr Rh^{105} daughters. After complete decay of the 4.5-hr Ru^{105}, rhodium was removed from the ruthenium sample, the ruthenium remounted, and the relative contributions of 40-day Ru^{103} and 1.0-yr Ru^{106} (in equilibrium with 30-sec Rh^{106}) determined by analysis of an absorption curve.

Palladium (13.1-hr Pd^{109} *and* 21-hr Pd^{112})

Palladium was isolated by the dimethylglyoxime procedure[27] after initial removal from the curium by sulfide precipitation. During the first day of a 67-hr growth period it was observed that the palladium carrier was reduced to metal (presumably by decomposition products formed by the action of the intense alpha radiation on the solution). The metallic palladium was dissolved to insure exchange with fission-produced species and reprecipitated as the sulfide at the end of the growth period. After purification by dimethylglyoxime precipitations and $Fe(OH)_3$ removals, a AgCl precipitation was made to establish zero time for the growth of 3.2-hr Ag^{112}, and the palladium was finally precipitated with dimethylglyoxime.

The activities due to 13.1-hr Pd^{109} and 21-hr Pd^{112} (through its 3.2-hr Ag^{112} daughter) were determined by analyses of decay and absorption curves.

[26] L. E. Glendenin, reference 20, Paper 260.

[27] J. A. Seiler, reference 20, Paper 264; L. E. Glendenin, reference 20, Paper 265.

Cadmium (53-hr Cd^{115})

Cadmium was separated from the curium as the sulfide to start and end the growth period. Purification[28] was achieved by several removals of basic iron acetate from buffered solution (pH 5) and PdS from 6N HCl solution, and cadmium finally isolated as the sulfide. A final precipitation of basic iron acetate to remove indium was made near the end of the procedure to establish zero time for the growth of 4.5-hr In^{115m} and 2-hr In^{117}. Since the fission yields of cadmium isotopes were expected to be low, a relatively long growth period of 8 days was used to obtain essentially saturation activities.

The activity due to 53-hr Cd^{115} was determined from an analysis of a growth and decay curve taking into account the contributions of the 4.5-hr In^{115m} daughter and a long-lived activity, presumably 43-day Cd^{115m}, present in low intensity.

Antimony (93-hr Sb^{127} *and* 4.2-hr Sb^{129})

Antimony was oxidized with Br_2 to Sb(V) and separated from the curium by sulfide precipitation from 1N HCl solution to start and end a 7.1-day growth period. Purification was achieved by precipitations of molybdenum alpha-benzoinoxime, tellurium (by reduction with SO_2), and $Fe(OH)_3$. Separation from tin was made by sulfide precipitations of Sb(III) in the presence of HF. A removal of tellurium as sulfide from 6N HCl solution was made near the end of the procedure to establish zero time for the growth of tellurium daughter activities, and the antimony was isolated for mounting as Sb_2S_3.

Analyses of decay and absorption data were made to obtain the relative contributions of 93-hr Sb^{127} and 4.2-hr Sb^{129} to the total activity. The beta-decay scheme of Sb^{129} has not been reported. Absorption data obtained in this work indicate it to be complex with an initial half-thickness in aluminum of about 36 mg/cm^2, and this value was used for the absorption corrections. Branching ratios for the decay of 93-hr Sb^{127} to 9.3-hr Te^{127} and of 4.2-hr Sb^{129} to 70-min Te^{129} were taken as 84 percent and 80 percent, respectively.

Tellurium (30-hr Te^{131m} *and* 77-hr Te^{132})

Tellurium was removed from the curium solution as sulfide to start a 69-hr growth period. After ending the growth period by precipitation as sulfide, the tellurium was reprecipitated by reduction to the elementary state with SO_2 and set aside to allow the 9.3-hr Te^{127} to decay to a negligible intensity. The tellurium was then purified[29] by $Fe(OH)_3$ removals and finally precipitated by reduction with SO_2 to establish zero time for the growth of 2.4-hr I^{132}.

[28] R. P. Metcalf, reference 20, Paper 268; L. E. Glendenin, reference 20, Paper 265.

[29] T. B. Novey, reference 20, Paper 273; L. E. Glendenin, reference 20, Paper 274.

Analyses of decay and absorption data were made to determine the contributions of the chains, 30-hr $Te^{131m}\rightarrow$ 25-min $Te^{131}\rightarrow$8.0-day I^{131} and 77-hr $Te^{132}\rightarrow$2.4-hr I^{132}. A low intensity background activity probably due to 32-day Te^{129m} was also observed.

Iodine (8.0-day I^{131}, 21-hr I^{133}, 52.5-min I^{134}, *and* 6.7-hr I^{135})

Iodine and tellurium activities were removed from the curium solution to start the growth period. The curium solution was made just basic with NaOH and a known amount of I^- carrier added. After a growth period of 20.33 hr the solution was acidified to dissolve the $Cm(OH)_3$ and any incorporated fission-produced elements, made basic, and boiled to remove H_2O_2. The basic hypochlorite procedure[30] was then employed for the isolation of radiochemically pure iodine activities.

Since the 21-hr I^{133} and 6.7-hr I^{135} decay to radioactive xenon daughters which complicate the analysis of the total iodine decay curve, it is customary to prepare an iodine stock solution from which aliquots are withdrawn periodically, the xenon activities boiled out, and the iodine activity determined. The total iodine activity available in the present investigation was insufficient for the application of this technique. Therefore, after the initial decay of the 52.5-min I^{134}, the iodine sample (as AgI) was periodically dissolved with KCN, the xenon activities boiled out, and the iodine remounted as AgI for counting. In this way it was possible to obtain from a single sample a total iodine decay curve which could be analyzed for the various iodine isotopes. The activity contributed by 2.4-hr I^{132} formed directly in fission and from 77-hr Te^{132} was taken into account. The observed 8.0-day I^{131} activity was corrected for the fraction (about 20 percent) produced from 30-hr Te^{131m} during the iodine growth period to obtain the fission yield of I^{131} formed independently of the 30-hr Te^{131m}.

Cesium (13.7-day Cs^{136})

The separation of cesium activities from the curium solution in preparation for the growth period was accomplished by $Cm(OH)_3$ precipitation from hot NaOH solution. After a period of 14 days $Cm(OH)_3$ was again precipitated to end the growth. Cesium was purified[31] by a silicotungstic acid precipitation, several perchlorate precipitations, exhaustive $Fe(OH)_3$ removals, and finally isolated as $CsClO_4$ for mounting and counting. Decay and absorption data indicated the presence of a single activity due to 13.7-day Cs^{136}. The absence of detectable 33-yr Cs^{137} activity is consistent with the growth period used.

[30] Glendenin, Metcalf, Novey, and Coryell, reference 20, Papers 278 and 279.

[31] L. E. Glendenin and C. M. Nelson, reference 20, Paper 283.

Barium (85-min Ba^{139} *and* 12.8-day Ba^{140})

Precipitations of $Ba(NO_3)_2$ with fuming HNO_3 were made to remove all barium activities from the curium solution. After growth periods of 17.5 hours and 65.5 hours, barium was again removed as nitrate and purified[32] by precipitations as $Ba(NO_3)_2$ and $BaCl_2$, removals of $Fe(OH)_3$, and isolation as $BaCO_3$ for mounting and counting. The last $Fe(OH)_3$ removal established zero time for the growth of La^{140}.

Since sufficient time was allowed for decay of the short-lived species prior to the final isolation of barium, the only significant activities observed were 85-min Ba^{139}, 12.8-day Ba^{140}, and 40-hr La^{140}. These were determined by analyses of decay curves.

Cerium

An attempt to determine the yields of cerium fission products was unsuccessful due to major contamination by 282-day Ce^{144} produced during the pile irradiation and incompletely separated from the curium. Time was not available for the necessary purification of curium from the rare earths.

III. RESULTS AND DISCUSSION

The fission yield of a nuclide may be defined as the percentage of fissions which lead to its formation. Thus,

$$Y_N=(D_N/F)100, \qquad (1)$$

where Y_N is the fission yield of nuclide N, D_N is its saturation disintegration rate (equal to the rate of formation), and F is the fission rate. In the case of induced fission, F is usually determined by fission counting during the irradiation. Alternatively, an arbitrary value may be assumed for the yield of a particular nuclide, others determined relative to it, and the sum of all fission yields then normalized to 200 percent. In the present investigation, the alpha-to-fission ratios for Cm^{242} and Cm^{244} are known[33] and F is readily obtained from an accurate alpha assay of the source. Appropriate corrections applied to the observed beta counting rates give D_N and hence, absolute fission yields.

The results of the determination of the yields of 21 nuclides produced in the spontaneous fission of Cm^{242} are presented in Table II, column 2, and plotted (circles) against mass number in Fig. 1. The estimates of reliability given in the table are based on the relative complexity of the absorption and decay data, the difficulty of calculating beta disintegration rates from observed counting rates, and the number of determinations. Since the limitations of time prevented replicate determinations in all but a few cases (Mo^{99}, Ru^{105}, Ba^{139}, and Ba^{140}), the estimated errors have been increased substantially over those normally accepted as representative of the radiochemical method. In determining

[32] L. E. Glendenin, reference 20, Papers 236 and 288.

[33] Hollander, Perlman, and Seaborg, *Table of Isotopes*, Revs. Modern Phys. **25**, 469 (1953).

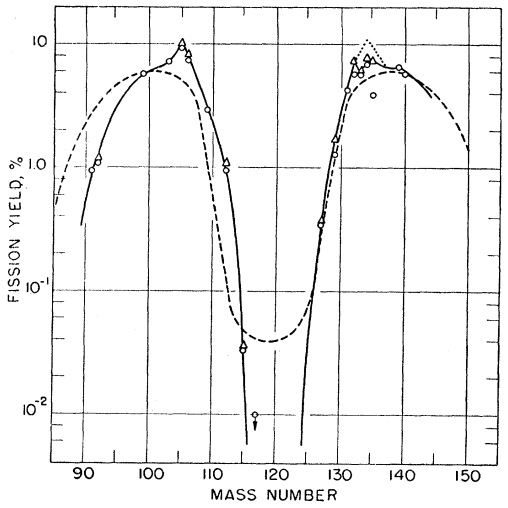

FIG. 1. Yield-mass curves for spontaneous fission of Cm^{242} (solid line) and pile-neutron induced fission of Pu^{239} (dashed line). Circles represent observed yields and triangles estimated total chain yields. The dotted line is the predicted shape of the heavy group. (See discussion of Table IV.)

the total fission yield of a chain of given mass number, an attempt is usually made to choose for analysis a member of the chain near stability where any independent formation of nuclides beyond that member would be negligible. This was not always feasible in the present investigation, and it was therefore necessary in a number of cases to calculate these expected independent yields to obtain total chain yields. The calculated independent yields are given in column 3 of Table II and are based on the charge distribution postulate of equal charge displacement from stability[8,9,34] for complementary products, and a value of 3.0 for the average number of neutrons emitted per fission (see discussion below and Table III). Total chain yields (plotted as triangles in Fig. 1) are given in column 4 with the estimated reliabilities increased somewhat from those of column 2 to reflect the uncertainty in the calculated independent yields.

A few of the values in Table II deserve special comment—namely, Cd^{115m}, Cd^{117}, Te^{131m}, I^{131}, and Cs^{136}. In the cadmium determination the sample contained about 30 counts per minute of an activity with the growth and decay characteristics of the chain 53-hr $Cd^{115}\rightarrow$4.5-hr In^{115m} and a few counts per minute of an unidentified long-lived component. Since the latter may have been due in part to contamination and was in too low an intensity to be adequately characterized, no fission yield determination for 43-day Cd^{115m} was attempted. An assumed value of 0.003 percent is listed in Table II based on a fission yield ratio of 43-day Cd^{115m} to 53-hr Cd^{115} of about 10 percent as has been observed in pile neutron fission[35,36] of U^{233}, U^{235}, and Pu^{239} as well as in 14-Mev neutron fission[36] of U^{235}. Although no activity attributable to the chain 3.0-hr $Cd^{117m}\rightarrow$50-min $Cd^{117}\rightarrow$2-hr In^{117}, was observed in the decay curve, an estimate of the maximum activity which could have escaped detection (10 counts per minute) was used to set an upper limit of 0.01 percent for the fission yield of Cd^{117m}.

The observed yield of 30-hr Te^{131m} represents about one-half of the total chain yield—the remainder presumably proceeding through the isomeric ground state (25-min Te^{131}) to 8.0-day I^{131}. It is interesting to note that an analysis of the expected yields of primary fission products in this region (Table III) predicts a value for mass 131 at tellurium (2.2 percent) close to that observed for Te^{131m} (2.3±0.5 percent). This is also true in the case of slow neutron fission of U^{235}.[9,34] These data may be interpreted as indicating a very small branch in the decay of 23-min Sb^{131} to 30-hr Te^{131m} (not inconsistent with observation[9,37]) and the apparently exclusive formation of the excited state as a primary fission product. The spins of Te^{131m} and Te^{131} are 11/2 and 3/2, respectively, and the favored formation of the higher spin state is in accord with the observations of Biller[38] on the yields of primary products of bismuth fission. It was found that the higher spin state of isomeric pairs was always formed in higher yield, often to the apparent exclusion of the lower spin state. Levy[39] studied this phenomenon further and proposed a reasonable mechanism based on the preferred formation of higher spin states at high excitation and the tendency toward only small spin changes during de-excitation.

The nuclide Cs^{136} is "shielded" by stable Xe^{136} from formation through beta decay so that the observed fission yield is only that fraction of the total chain yield formed directly in fission at Cs^{136}. Such independent yields have been utilized in developing an empirical hypothesis for the distribution of nuclear charge in fission[34] consistent with observations in slow neutron-induced fission of U^{235} and Pu^{239} and also apparently applicable[40] to 14-Mev neutron-induced fission of U^{235}. This hypothesis may be applied to the spontaneous fission of Cm^{242} to estimate the value of $\bar{\nu}$, the average number of neutrons emitted per fission, in the following manner. The observed yield for Cs^{136} (0.8±0.12 percent) represents 11±2 percent of the assumed total chain yield (7.0±1.0 percent). From the charge distribution curve[34] this value corresponds to a $Z-Z_p$ difference of 1.5, or a value of 53.5±0.1 for Z_p, the most probable charge of the primary fission product of

[34] Glendenin, Coryell, and Edwards, reference 20, Paper 52.
[35] Reference 20, Appendix *B*.
[36] A. C. Wahl and N. A. Bonner, Phys. Rev. **85**, 570 (1952).
[37] G. B. Cook, Atomic Energy Research Establishment (Harwell, England) Report AERE-C/R-729, June, 1951 (unpublished).
[38] W. F. Biller, University of California Radiation Laboratory Report UCRL-2067, January, 1953 (unpublished).
[39] H. G. Levy, University of California Radiation Laboratory Report UCRL-2305, August, 1953 (unpublished).
[40] G. P. Ford, U. S. Atomic Energy Commission Document AECD-3597, November, 1953 (unpublished).

mass 136. By definition,[34]

$$Z_p = Z_A - \tfrac{1}{2}(Z_A + Z_A^* - Z_F), \quad (2)$$

where Z_A is the most stable charge for mass A, Z_F is the charge of the fissioning nucleus, and Z_A^* is the most stable charge for the complementary mass A^*; and

$$A^* = A_F - A - \bar{\nu}, \quad (3)$$

where A_F is mass number of the fissioning nucleus. Using a value[34] of 56.2 for Z_A at mass 136, Eq. (2) gives 45.2 ± 0.2 for Z_A^*, corresponding to 103.0 ± 0.5 for A^*, and a value of 3.0 ± 0.5 for $\bar{\nu}$ from Eq. (3). This is in good agreement with a recently reported value of 3.0 ± 0.4 determined by direct measurement[41] and lends support to the application of the charge distribution hypothesis to spontaneous fission. A value for ν may also be obtained from the difference in mass of the fissioning nucleus and the sum of complementary masses. From Fig. 1 it may be seen that the value of ν in the mass region near symmetry is smaller than $\bar{\nu}$, as has also been observed in the case of neutron-induced fission of U^{235}.[42] The above calculations may be reversed to obtain independent yields. Such calculations were made, using $\bar{\nu}=3.0$, to estimate the independent yields of nuclides of a chain beyond the member chosen for analysis (Table II, column 3).

A comparison of the yield-mass curves for spontaneous fission of Cm^{242} and pile neutron-induced fission of Pu^{239} is presented in Fig. 1. Spontaneous fission of Cm^{242} is seen to be asymmetric, but with somewhat higher and narrower peaks. Although the amount of Cm^{242} available was not sufficient to produce a measurable activity of fission products in the region of symmetrical fission (mass numbers 116–124), the upper limit set at mass 117 indicates a peak-to-trough ratio at least as large as that observed in the thermal-neutron-induced fission of U^{235} (about 680), and perhaps considerably larger. A decrease in the peak-to-trough ratio and a broadening of the mass distribution have been observed with increasing energy of excitation in induced fission,[43] and therefore it is to be expected that spontaneous fission (no excitation) would exhibit a larger peak-to-trough ratio and a narrower mass distribution. It has also been observed that the light group distribution shifts toward higher masses with increasing mass of the fissioning nucleus while the heavy group remains relatively constant, with perhaps a slight shift in the direction of smaller masses.[44] It is seen that this trend continues with Cm^{242}, the heaviest nuclide for which fission yields have been determined.

An interesting feature of the yield-mass curve for spontaneous fission of Cm^{242} is the occurrence of pronounced fine structure as indicated by the abnormally high yields in the regions around masses 105 and 134. This phenomenon appears to be associated with a nuclear structure preference in fission for fragments containing 82 neutrons[5,6] and excessive evaporation from primary fission fragments of the neutrons with abnormally low binding energy just outside closed shells.[8,9] The effect was first observed in mass spectrometric investigations[4,6] and subsequently in radiochemical studies of U^{235} fission,[7,9,45] although it was not apparent in the earlier radiochemical studies of U^{235} or Pu^{239} fission.[46]

A comparison of the yield-mass curves for spontaneous fission of Cm^{242} and U^{238} is given in Fig. 2. The data for U^{238} are the relative yields of the stable krypton and xenon isotopes produced by spontaneous fission of U^{238} in uranium minerals.[15] A dashed curve has been drawn through the data assuming symmetrical reflection about mass 118 (i.e., two neutron emitted per fission) and normalized to 200 percent yield summation. Although the region of symmetrical fission was not investigated, the upper limit at mass 129 again indicates a large peak-to-trough ratio for spontaneous fission. The apparently high yield at mass 132 also indicates fine structure in the mass region of the 82 neutron shell.[14,15] Similar results[15] were obtained for the krypton and xenon yields from spontaneous fission of Th^{232}.

The mass distribution of the light group in the spontaneous fission of Cm^{242} is plotted in Fig. 3 with that for pile neutron-induced fission of U^{235} [6,42] shown for com-

TABLE II. Fission yields in spontaneous fission of Cm^{242}.

Nuclide	Observed fission yield (%)	Calculated independent fission yield of daughter (%)	Total fission yield of chain (%)
9.7-hr Sr^{91}	0.94 ±0.3	0.01	0.95 ±0.3
2.7-hr Sr^{92}	1.1 ±0.3	0.1	1.2 ±0.3
67 -hr Mo^{99}	5.7 ±0.7	0	5.7 ±0.7
40 -day Ru^{103}	7.2 ±1.5	0	7.2 ±1.5
4.5-hr Ru^{105}	9.5 ±0.9	0.4	9.9 ±1.0
1.0-yr Ru^{106}	7.4 ±0.8	1.0	8.4 ±1.0
13.1-hr Pd^{109}	2.9 ±0.4	0	2.9 ±0.4
21 -hr Pd^{112}	0.95 ±0.15	0.15	1.1 ±0.2
53 -hr Cd^{115}	0.033±0.01	0	
43 -day Cd^{115m}	(0.003)[a]	0	0.036±0.01
3.0-hr Cd^{117m}	<0.01	0	<0.01
93 -hr Sb^{127}	0.35 ±0.1	0.02	0.37 ±0.1
4.2-hr Sb^{129}	1.3 ±0.3	0.4	1.7 ±0.4
30 -hr Te^{131m}	2.3 ±0.5	...	
8.0-day I^{131}	2.0 ±0.4[b]	0	4.3 ±0.7
77 -hr Te^{132}	5.8 ±0.9	1.6	7.4 ±1.3
21 -hr I^{133}	5.7 ±0.8	0.3	6.0 ±0.9
52.5-min I^{134}	6.9 ±1.0	1.1	8.0 ±1.3
6.7-hr I^{135}	3.9 ±0.6	3.4	7.3 ±1.4
13.7-day Cs^{136}	0.80 ±0.12	...	...
85 -min Ba^{139}	6.6 ±0.7	0	6.6 ±0.7
12.8-day Ba^{140}	5.9 ±0.8	0	5.9 ±0.8

[a] Assumed yield from known branching ratio in induced fission.
[b] Yield independent of 30-hr Te^{131m}.

[41] F. R. Barclay and W. J. Whitehouse, Proc. Phys. Soc. (London) **A41**, 447 (1953).
[42] Glendenin, Steinberg, Hayden, Inghram, and Flynn (to be published).
[43] See, e.g., Turkevich, Niday, and Tompkins, Phys. Rev. **89**, 552 (1953), for discussion and references.
[44] E. P. Steinberg and M. S. Freedman, reference 20, Paper 219.
[45] Yaffe, Day, and Greer, Can. J. Chem. **31**, 48 (1953).
[46] Reference 20, pp. 537–542.

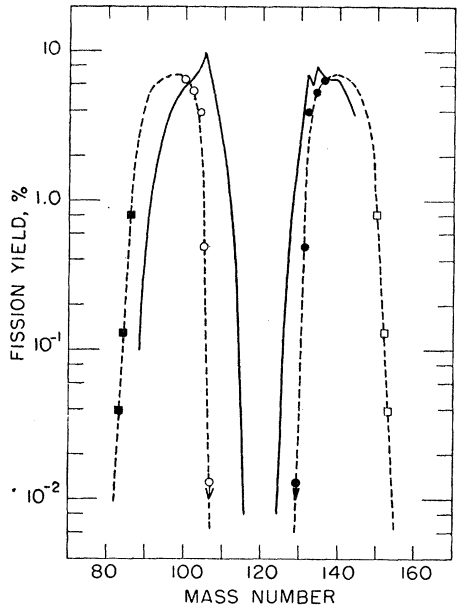

FIG. 2. Yield-mass curves for spontaneous fission of Cm^{242} (solid line) and U^{238} (dashed line). Solid squares and circles represent data for krypton and xenon isotopes, respectively; open squares and circles represent mirror points ($\nu=2$).

parison. Fine structure is seen to occur around mass 100 in the U^{235} curve and mass 105 in the case of Cm^{242}. The fact that these mass numbers are complementary in both cases to the mass region of the primary heavy fission products which contain 82 neutrons (about mass 134) and that there are no important nuclear shell closures in the mass region of the light group peaks are considered as evidence that the fine structure observed in the light group represents the effect of preference for an 82-neutron configuration in the fission act itself. The effect is seen to be more prominent in the case of Cm^{242} than in U^{235} indicating that the nuclear structure preference in fission may diminish with increasing excitation energy of the fissioning nucleus. It is reasonable that this selectivity in the fission process would diminish with increasing excitation energy as has been suggested by Wiles.[5]

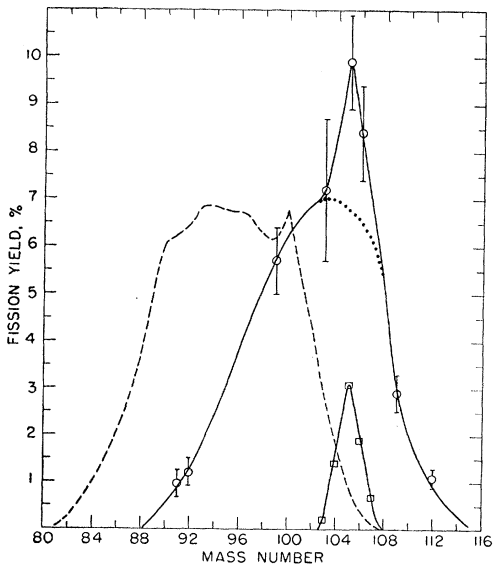

FIG. 3. Fission yields of the light groups in spontaneous fission of Cm^{242} (circles and solid line) and pile-neutron induced fission of U^{235} (dashed line). The dotted line (representing a hypothetical "smooth" yield curve) has been subtracted from the solid line to give the distribution of "excess" yields plotted as squares.

A hypothetical "smooth" curve (dotted line in Fig. 3) has been subtracted from the solid curve of Fig. 3 to give approximate "excess" yields (plotted as squares) due to the 82-neutron shell preference in the spontaneous fission of Cm^{242}. If it is assumed that for these fission modes the heavy fragment containing 82 neutrons emits one prompt neutron and its complement emits two (total $\nu=3$), it is possible to determine the identity of the primary fission products occurring in excess yield as presented in Table IV. The complementary fragments (before neutron emission) are given in columns 2 and 3, and the corresponding products (after neutron emission) in columns 1 and 4. The excess yields of these products obtained from Fig. 3 are listed in column 5. According to this analysis the excess yields due to the 82-neutron preference effect in fission amount to about 7 percent. A summation of the fission yields represented by the solid line in Fig. 3 gives 103 percent, in excellent agreement with the theoretical value of 100 percent.

A complete analysis of the distribution of fission yields in the 82-neutron shell region (mass numbers 131–141) is presented in Table III. The independent yields (by direct formation) of fission products along the chains have been calculated using assumed "smooth" chain yields complementary (with $\nu=3$) to those of the light group (dotted line of Fig. 3) and the charge distribution based on equal charge displacement. These yields represent what might be considered as the "normal" distribution in fission in the absence of perturbations due to the closed shell of 82 neutrons. The excess yields (from Table IV) are given in parentheses at their respective chain positions and may be seen to approximate closely the corresponding "normal" yields in distribution and magnitude. It seems reasonable to interpret these parallel distributions as indicative of a fundamental charge distribution in fission for all modes with a superposed selectivity for an 82-neutron configuration.[47] The arrows in Table III represent the extra evaporation from primary fission fragments of the 83rd ($S+1$), 85th ($S+3$), and 87th ($S+5$) neutrons, i.e., the odd neutrons having abnormally low binding energies[9] due to their location just outside a closed shell.

The data in Table III may be used to calculate expected cumulative yields at any chain member as well

[47] This conclusion has been used as an assumption by D. R. Wiles in an independent and somewhat similar treatment of fine structure in induced fission of U^{235} and U^{238}. (See reference 7.)

TABLE III. Analysis of fission yields in the 82-neutron shell region.[a]

Mass No.	Z_p	Independent fission yield in chain (%) $_{50}$Sn	$_{51}$Sb	$_{52}$Te	$_{53}$I	$_{54}$Xe	$_{55}$Cs	$_{56}$Ba	$_{57}$La	$_{58}$Ce	"Smooth" chain yield (%)	Nuclide determined	Calculated yield (%)	Observed yield (%)
131	51.6	0.4	2.0	2.2	0.7						5.3	Te^{131m}	2.2	2.3 ±0.5
			(0.7)									I^{131}	3.1[b]	2.0 ±0.4[b]
132	52.0	0.06	1.5	3.0	1.5	0.06					6.1	Te^{132}	5.3	5.8 ±0.9
		↑S+1		(1.9)										
133	52.3	0.01	1.1	2.9	2.2	0.3					6.5	I^{133}	8.5	5.7 ±0.8
			↑S+1		(3.1)									
134	52.7		0.4	2.2	3.0	1.1	0.01				6.7	I^{134}	9.8	6.9 ±1.0
				↑S+1		(1.4)								
135	53.1		0.05	1.5	3.3	1.9	0.1				6.9	I^{135}	6.1	3.9 ±0.6
					↑S+1		(0.2)							
136	53.5			0.7	2.8	2.8	0.7				7.0	Cs^{136}	0.9	0.80 ±0.12
				↑S+3		↑S+1								
137	53.9			0.1	1.9	3.3	1.5	0.05			6.9	—	6.8[c]	—
					↑S+3		↑S+1							
138	54.3			0.01	1.1	2.9	2.2	0.4			6.6	—	6.5[c]	—
						↑S+3		↑S+1						
139	54.7				0.4	2.2	2.8	1.0	0.01		6.4	Ba^{139}	6.2	6.6 ±0.7
							↑S+3		↑S+1					
140	55.2				0.02	1.2	2.8	1.8	0.2		6.0	Ba^{140}	5.7	5.9 ±0.8
						↑S+5		↑S+3		↑S+1				
141	55.6					0.4	2.1	2.3	0.7	<0.01	5.5	—	—	—

[a] Parentheses denote excess yields due to 82-neutron shell preference in fission. Vertical arrows indicate yield branching by extra neutron emission from fragments containing 83(S+1), 85(S+3), and 87(S+5) neutrons.
[b] Yield of I^{131} independent of 30-hr Te^{131m}.
[c] Total chain yield.

as total chain yields by summation of the appropriate independent yields (both "normal" and "excess"), taking into account the losses and gains in yield indicated by the arrows. Good agreement between yields calculated in this manner and those observed for the nuclides indicated is apparent in the last two columns of the table, except for the yields of the iodine isotopes. The fact that the observed yields of the latter are all about 67 percent of the calculated values may be significant. Difficulties in exchange between carrier- and fission-produced species of iodine are well known,[30] and it is possible that the presence of H_2O_2 or other decomposition products from the action of the intense alpha radioactivity in the curium solution may have interfered with the achievement of complete interchange. Until this point can be clarified by further study of the iodine chemistry in such solutions or by the determination of the cumulative yields at the xenon isotopes, the calculated total chain yields for mass numbers 133–136 have been used to predict the shape of the fine structure in the heavy group (dotted line in Fig. 1).

Further experiments on spontaneous fission are planned using larger sources of Cm^{242} and Cm^{244}. In particular, it would be desirable to extend the study of charge distribution (by determination of independent yields along chains and yields of shielded nuclides) and of mass distribution in the regions of the trough, fine structure, and the rare earth elements.

TABLE IV. Distribution of "excess" yields due to 82-neutron shell preference in fission.

Light product	Light fragment	Heavy fragment (82 neutrons)	Heavy product	Approximate "excess" yield (%)
$_{41}Nb^{103}$	$_{41}Nb^{105}$	$_{55}Cs^{137}$	$_{55}Cs^{136}$	0.2
$_{42}Mo^{104}$	$_{42}Mo^{106}$	$_{54}Xe^{136}$	$_{54}Xe^{135}$	1.4
$_{43}Tc^{105}$	$_{43}Tc^{107}$	$_{53}I^{135}$	$_{53}I^{134}$	3.1
$_{44}Ru^{106}$	$_{44}Ru^{108}$	$_{52}Te^{134}$	$_{52}Te^{133}$	1.9
$_{45}Rh^{107}$	$_{45}Rh^{109}$	$_{51}Sb^{133}$	$_{51}Sb^{132}$	0.7
$_{46}Pd^{108}$	$_{46}Pd^{110}$	$_{50}Sn^{132}$	$_{50}Sn^{131}$	...

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