CUMULATIVE YIELDS OF THE HEAVY FRAGMENTS IN U^{235} THERMAL NEUTRON FISSION

By

HARRY FARRAR, B.A., M.Sc.

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AUTHOR: Harry Farrar, B.A. (University of Toronto)

M.Sc. (McMaster University)

SUPERVISOR: Professor R. H. Tomlinson

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SCOPE AND CONTENTS: Isotopic abundances of the elements barium, cerium, neodymium, samarium and europium formed in the thermal neutron fission of U^{235} have been studied with a mass spectrometer. Suitable analyses of very young and old fission products have made possible the normalization of the isotopic abundances of each of these elements through isobaric nuclides. In this manner the relative cumulative fission yield of every fission chain from mass 137 to 154 has been obtained. These yields, together with previously reported relative yields of masses 131 to 137, when normalized to total 100%, give the absolute cumulative yield of all the significant heavy fragment chains. The origin of the fine structure found in this mass yield distribution is discussed. It has been found that all fine structure can be explained in terms of a smooth primary mass-yield curve and a simple neutron distribution both of which are compared with experimental data from other laboratories.

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GENERAL INTRODUCTION

A. The Fission Process

Nuclear fission is the break-up of a heavy nucleus into two or more medium-heavy fragments with the release of a large amount of energy. The process is accompanied by the emission of neutrons and more rarely by alpha particles and possibly other light fragments. Although many of the heaviest elements undergo fission spontaneously, the probability of the process is considerably enhanced by the addition of some excitation energy. Bombardment by low energy or "thermal" neutrons can result in the fission of several isotopes of thorium, protoactinium, uranium, neptunium and isotopes of all the transuranic elements. Fission has also been induced in some nuclides (notably U^{235}, U^{238} and Th^{232}) by neutrons, protons, deuterons, helium ions, mesons and gamma rays of moderate energies. With higher bombarding energies (50 to 450 MeV), lighter elements such as bismuth, lead, gold, tantalum and some rare earths have been shown also to undergo fission. By far the most important and most studied of these reactions are the thermal neutron fission of U^{233}, U^{235} and Pu^{239}; and the fast neutron fission of Th^{232} and U^{238}. This is because these nuclides are either found in nature or are produced easily by nuclear reactions.

All nuclides with more than 82 protons and 126 neutrons are unstable, decaying through a complicated series of beta and alpha emissions to either lead or bismuth. Spontaneous fission becomes increaseingly important for elements of high atomic number where the observed half-lives for this mode of decay vary from more than 10^{20} years for Th^{232}

to 3 hours for Fm^{256}. In the latter case it is the principle mode of decay and spontaneous fission will be a limiting factor in the discovery of further transuranic elements.

The mass distribution of fission products varies with the fissioning nuclide and with the energy of the bombarding particle. It has been found in low energy fission that the most common occurrence is a splitting into two fragments of unequal mass. It has also been observed that as the excitation of the fissioning nucleus is increased, a greater percentage of the fragments has equal mass. The distribution of fission fragments varies from a double-hump curve representing principally asymmetric fission at low excitation energy, through an intermediate triple-hump curve, to a single broad curve representing a predominance of symmetric fission at high energies. In low energy and spontaneous fission, as the mass of the fissioning nuclide is raised, the mass distribution of the light fragments rises proportionally while the heavy fragment distribution remains approximately fixed.

The analogy between a nucleus and a liquid drop that had been suggested by Bohr (1936) to account for various nuclear reactions was extended by Bohr and Wheeler (1939) to explain fission in a qualitative way. Although they did not explain the asymmetry of the mass split, they showed that there is a certain critical size for a nucleus, depending on Z^2/A above which the force of electrostatic repulsion should be greater than the forces holding the nucleus together. This critical size occurs for Z near 120 (Hyde, 1960), and it is therefore reasonable that for a nucleus that is only slightly below this limit, a small excitation could be sufficient to induce break-up into two or more fragments. Swiatecki

(1958) has shown that the energy released in the division of an idealized charged drop into more than two fragments is greater than for binary fission, as the critical size is approached - a phenomenon that has not yet been observed in fission. Because nuclei of medium mass have much higher binding energies per nucleon than have the heaviest elements, the fission process is accompanied by a very large energy release; approximately 200 MeV. Although many theoretical developments since 1939 have been put forward, no adequate theory of fission has yet been developed. The great variety of observations on this highly complex nuclear phenomenon present a formidable task to the theoretician.

B. Thermal Neutron Fission of U^{235}

One of the simplest ways of exciting nuclei is by bombardment with neutrons, which are not repelled by the long-range coulomb forces of the nucleus. When the neutron penetrates the nucleus to form a compound nucleus, it interacts strongly with the nuclear field and imparts to the nucleus excitation energy equal to its binding energy. The excited compound nucleus, which is an isotope of the target element, can de-excite by gamma emission or by fission; the ratio of the two processes being about 1 to 5 for U^{235}. The most common occurrence in U^{235} fission is a splitting into two large fragments with mass numbers 96 and 138, with the accompaniment of an average of 2.43 neutrons (Hughes et al., 1960). The observed prompt mass distribution ranges from 72 to 161 but is symmetrical about the central minimum, since for every heavy fragment there corresponds a light fragment with the same percentage abundance. Because heavy elements have a greater percentage of neutrons than the stable elements in the fission product region, the fission fragments generally have an excess of neutrons. If the fission fragment has sufficient excitation energy, it emits one or more "prompt" neutrons. When there is insufficient energy for the emission of further neutrons, the excess remaining decay to protons with the emission of negatons and antineutrinos. Thus, the primary fission product decays to its stable isobar by several successive beta processes.

The primary fission yield of a nuclide is the percentage of fissions which result in the formation of that nuclide, prior to any beta decay but after the emission of the prompt neutrons. Although a number of nuclides may be formed with the same mass number, nearly all of them that are formed in an appreciable yield have charges three or four units from stability and have, therefore, short half-lives. These nuclides rapidly decay to a longer-lived or stable nuclide of the same mass number. Thus, in nearly every case, a measurement of the yield of this latter nuclide, a few hours after irradiation, will give the sum of all the primary fission yields for that mass number.

There are two processes, however, that can cause the measured yield to be different from the sum of the primary yields.

(i) A few nuclides, especially those with a few neutrons in excess of closed neutron shells, emit neutrons after beta decay has occurred. Because the characteristics of some of these delayed neutron emitters have been established, their influence on the observed fission yields may be estimated.

(ii) Neutron absorption by some members of a decay chain causes part of the yield of that chain to enter that of the next higher mass number. This effect becomes appreciable only for long or intense irradiations or for nuclides with high cross section. With a knowledge of the cross sections involved, data obtained can be corrected for this effect with adequate precision.

The "cumulative yield" of a particular mass chain is the sum of the yields of the fragments that finish in that chain, corrected for the effect of neutron capture. Since delayed neutron emission is a characteristic of the fission process itself and is not dependent on the irradiation conditions, the cumulative yields quoted in the literature are not usually corrected for this process.

C. Radiochemistry

The techniques of radiochemistry led to the discovery of nuclear fission and have contributed greatly to an elucidation of some of the main features of this reaction. In the radiochemical method, a measured amount of non-radioactive "carrier" of a given fission product element is added to a solution of uranium in which a known number of fission events has occurred. After adequate mixing of the radioactive isotopes with the isotopes of the carrier, the solution is subjected to an analytical procedure to separate this element in a state of chemical and radiochemical purity. The fission yield may be determined with counting techniques and with a knowledge of the fractional recovery of the inert carrier containing the fission product isotopes.

D. Mass Spectrometry

The mass spectrometer is particularly suitable for the study of fission products because it is a means of measuring ratios of stable and long-lived isotopes, usually with an accuracy of better than 1%. Another advantage is that since only ratios are measured, losses due to chemical separations (assuming that no isotopic fractionation occurs) do not influence the relative yield measurements. Great care must be taken not to contaminate the fission product elements with their terrestrial counterparts because analyses are often conducted with quantities as small as 10^{-10} grams. Fortunately, the terrestrial relative isotopic abundances usually differ considerably from those of the fission products and, consequently, small amounts of contamination may, in nearly all cases, be assessed accurately. The mass spectrometric method has other advantages over the radiochemical method. Radionuclides that have half-lives too short to be easily measured with counting techniques always decay to long-lived or stable end-products which are measurable in a mass spectrometer. Not only has this increased the number of mass chains that have been studied, but it also has enabled the individual mass yields to be determined relative to their neighbours with a considerable degree of accuracy.

(a) Isotope Dilution

The absolute number of atoms of each isotope present can nearly always be determined by the isotope dilution technique. If a known weight of an element which has a certain isotopic constitution is added to an unknown quantity of the same element occurring in fission products but having a different isotopic constitution, then a determination of the ratios of the isotopes before and after the addition will give the concentrations of the fission product isotopes. When the isotope dilution method is to be employed, a special solution is made up to volume, containing weighed amounts of the various elements, each of which must have a different isotopic constitution from its fission product counterpart. A suitable fraction of this volume is then added to the solution containing the uranium and fission products, before any separations of the latter have been effected. In this way, the isotopic yields of the various elements occurring in fission may be normalized to each other.

(b) Integral Mass Spectrographic Determination

The Russians have developed a technique known as the "method of integral mass spectrographic determination" (Anikina, 1958) whereby the abundance of an isotope of one element relative to an isotope of another element can be found from the measurement of their relative ion currents in a mass spectrometer, and from the relative ionization efficiencies of these elements.

(c) The Isobaric Method

In this method the relative abundances of the isotopes of two different elements are related by means of a pair of isobars. This was first employed by Petruska et al. (1955) where the relative yield of Xe^{133} to the other xenon isotopes in fission was measured in young fission products and its daughter Cs^{133} was measured relative to Cs^{135} and Cs^{137} in old fission products. The xenon and cesium relative yields therefore were related to each other. The relative abundances of the isotopes of a given element can be determined with a precision of 1% or better and therefore this method can relate the isotope abundances of two elements with known precision. In the isotope dilution method, two mass spectrographic measurements are required for each element; once before and once after isotope dilution. Errors in these measurements may be accurately assessed, but four measurements are required to relate two elements rather than two as required by the isobaric technique. Furthermore, preferential loss or adsorption of either the fission product or the added isotope during the isotope dilution procedure may introduce other errors which are difficult to assess.

(d) Absolute Yield Determinations

The absolute number of fissions occurring in an irradiated sample may be determined in a number of ways. The depletion of the fissile nuclide relative to a non-fissile isotope of the same element, if one exists, can be determined with a mass spectrometer (Farrar, 1959). Alternatively, the depletion may be measured by separating and weighing the fissile material before and after irradiation. These methods are useful only after long irradiations where an appreciable amount of "burn-up" has occurred. Another method utilizes the fission counter (Katcoff and Rubinson, 1953) which records the ionization pulses produced by the fragments as they recoil from a thin layer of fissile material into a suitable counter gas. The usual method employed is the indirect measurement of the neutron flux with elements such as boron and cobalt, which have neutron capture cross sections known relative to the fission cross section for U^{235}. The flux may then be determined by measuring either the change in the B^{10}/B^{11} ratio or by counting the induced activity in the cobalt. The absolute cumulative yield of a particular mass chain (%) may then be obtained by multiplying the number of atoms produced in that chain by 100 and then dividing by the number of fissions.

E. Fine Structure

The accurate knowledge concerning the fission products and their individual yields is of considerable importance in the design and operation of nuclear reactors. This knowledge has, in addition, wide implications in theoretical physics because any quantitative theory of fission must explain their distribution. Therefore it is very important that the fission yield distribution be determined with as great precision as possible. Determination of the variation of yield with both the charge and mass are equally important; but the measurement of the former is hindered by the fact that essentially all the primary fragments have very short half-lives. With the advent of the mass spectrometer to fission product studies (Thode and Graham, 1947) numerous irregularities in the mass-yield curves for the thermal neutron fission of U^{235}, U^{233} and Pu^{239} were found.

This thesis establishes the extent of the fine structure in the heavy mass region for the fragments from U^{235} thermal neutron fission. Apart from the first mass spectrometric determination of the cumulative yields for masses 139, 141 and 153, it further relates the yields of cesium isotopes to barium isotopes, barium to cerium, cerium to neodymium, neodymium to samarium and samarium to europium through isobaric nuclides of masses 137, 140, 144, 147 and 151 respectively. This work, together with the previously reported relative xenon and cesium yields (Wanless and Thode, 1955; Petruska et al., 1955, 1955a) has made possible the determination of the relative yields of all mass numbers between 131 and 154. These relative yields along with extrapolated values amounting to about 4% have been normalized to total 100%, thus giving absolute cumulative fission yields.

As a result of these measurements, it has been possible not only to obtain accurate absolute yields of the heavy mass fragments occurring in the thermal neutron fission of U^{235} but also to obtain with even greater precision each yield relative to its adjacent yields. This latter feature has made the data particularly suitable for the study of fine structure in the mass-yield curve.

HISTORICAL INTRODUCTION

A. Discovery and Early Investigation of Fission

Nuclear fission was discovered by Hahn and Strassmann (1939). This discovery was the culmination of an intense investigation in a number of laboratories in the years following the discovery of the neutron (Chadwick, 1932). Fermi (1934) suggested that the bombardment of uranium with neutrons might lead to transuranic elements with atomic numbers higher than 92 or, alternatively, to elements of slightly lower atomic number, with the release of protons and alpha particles. These predictions were rapidly confirmed, although in some cases the products were not identified correctly. What were considered to be the new transuranic elements were studied with the intention of showing that they were not elements of lower atomic number than uranium. By various means, elements now known to be fission products were identified as transuranic "eka-elements". Other elements found in the product of the neutron irradiation of uranium, because of their similar chemical properties, were thought to be new isotopes of radium and actinium. These species are now known to be barium and rare earths; but much effort was expended in trying to attribute them to products of (n,p) and (n,α) reactions in uranium as Fermi had predicted.

Evidence of the production in fission of isotopes closely resembling barium and lanthanum in chemical properties, prompted Hahn and Strassmann in 1939 to undertake a series of very careful experiments. To the same solution, they added quantities of both barium and radium and observed that the unknown "radium" isotopes behaved identically to the isotopes of barium. Hahn and Strassmann (1939), after noting this and similar evidence for the presence of lanthanum, published their celebrated paper. It was then realized that the activities of many of the so-called transuranic elements corresponded to activities of isotopes of technetium, strontium, yttrium, xenon, krypton, cesium and rubidium.

Following Hahn and Strassmann's paper, Meitner and Frisch (1939) and Joliot (1939) recognized that the fission process would release a large amount of energy and was therefore possible. The release of about 200 MeV, principally in kinetic energy due to the mutual coulomb repulsion of the two fragments, was observed experimentally first by Frisch (1939), who recorded large ionization pulses from recoiling fission fragments in a gas chamber; and then by Joliot (1939), who measured the ranges of the fragments in air. Investigations of the size of these ionization pulses by Jentschke and Prankl (1939) and later by Booth et al. (1939) showed that the fragments had a continuous distribution of energies with two pronounced maxima at about 100 MeV and 60 MeV. This was the first evidence that the fission of uranium is predominently asymmetric. Hahn and Strassmann (1939) had noted the possibility that neutrons would be set free in the fission process, a prediction that was soon experimentally confirmed by Von Halban et al. (1939) and by Anderson et al. (1939). It was found that approximately 2.3 high energy neutrons were emitted per fission, and that most of them were emitted instantaneously.

On the basis of calculations of the binding energy released by neutron absorption in the different uranium isotopes, N. Bohr (1939) concluded that the comparatively rare isotope U^{235} was responsible for the low energy neutron fission of uranium. This was confirmed by Nier et al.

(1940) who performed experiments using uranium isotopes separated with a mass spectrometer. The knowledge that in the fission process an excess of neutrons is released and that a self-sustaining chain reaction could be produced, led to government classification of relevent publications. The work carried out by a large team of scientists in the U.S.A. during and after the war on the detailed analysis of the fission products of Th^{232}, U^{233}, U^{235}, U^{238} and Pu^{239} has been reported by Coryell and Sugarman (1951).

B. Early Studies of Fission Product Yields

Until 1945 the study of the mass distribution of the fission fragments from U^{235} fission was limited entirely to radiochemical methods. The concept of fission yield and their first experimental determinations were carried out by Anderson, Fermi and Grosse (1941) using radiochemical procedures. The ensuing detailed radiochemical investigations confirmed the preponderance of asymmetrical fission showing that the resulting mass-yield curve had two humps centering around mass numbers 96 and 138. Because the reproducibility of the results was generally no better than 10% and occasionally considerably worse, it was considered reasonable to draw a smooth curve of best-fit through the experimental data. Fission yield values that failed to lie near this smooth curve, when remeasured, were usually found to have been in error. Since 1946, the war-time data have been substantially improved. With great advances in experimental techniques and with time for careful study it has been possible to establish more detailed decay schemes and more precise yields for the fission product nuclides.

C. Mass Spectrometric Studies

Thode and Graham (1947), who were the first to apply a mass spectrometer to the study of fission products, showed that certain yields of krypton and xenon isotopes in the thermal neutron fission of U^{235} could not be reconciled with a smooth mass-yield curve. Later work by Macnamara, Collins and Thode (1950), showed that the yields of 5.27-day Xe^{133} and stable Xe^{134} were 20% and 30% higher than expected. Glendenin (1949) explained the irregularities or "fine structure" in the 133 and 134 mass chains by assuming that nuclei with 83 neutrons would have a high probability of "boiling off" a prompt neutron to form the more stable 82-neutron configuration. Wiles et al. (1953) realized that this mechanism would increase the yields of masses 133 and 134 at the expense of the 135 and 136 mass chains. Since the latter yields were not unexpectedly low, they postulated that in addition to the above mechanism high yields must result from an actual structural preference in the primary fission act for isotopes with 82 neutrons. Glendenin et al. (1951) reached a similar conclusion after observing a high yield of Mo^{100}, the complementary fragment to the 134 mass chain. Radiochemical yields determined by Pappas (1953) indicated that fine structure in the mass-yield curve was the rule rather than the exception. He then extended the Glendenin hypothesis of neutron "boil-off" to include nuclei with 1, 3, 5 and 7 neutrons in excess of the 82-neutron closed shell. On this basis, Pappas (1953) predicted submaxima of masses 134, 136, 138, 142 and 144.

From the currently published radiochemical yields, it is difficult to establish to what extent fine structure occurs in the mass-yield curve. The existence of numerous individual fission yield values suggests that many are indeed accurate. In this connexion, the 6.62% yield of Xe^{133} obtained by Katcoff and Rubinson (1953) and the 6.36% yield of Ba^{140} obtained by Santry and Yaffe (1960) are perhaps outstanding. The yields of Ba^{139}, Ba^{140}, Sr^{89}, Sr^{91} and Mo^{99} were also determined relative to each other (by radiochemical means) with considerable precision by Reed and Turkevich (1953).

The first major studies of the yields of the heavy fragments in U^{235} thermal neutron fission using the mass spectrometer with the isotope dilution technique were made by Petruska, Thode and Tomlinson (1955a) and by Steinberg and Glendenin (1956). The results of the latter seemed to confirm the existence of considerable fine structure in this mass region, whereas the results of Petruska et al. showed little fine structure other than that about mass 134. Recent work by Chu (1959) has essentially confirmed the relative yields obtained by Petruska et al. in the region between masses 140 and 154.

Recent surveys of the literature by Katcoff (1960) and Walker (1960) have summarized the current state of knowledge concerning fission yields for the thermal neutron fission of U^{235}. Their values are largely based on the absolute yields obtained by Petruska et al. (1955a) and by Steinberg and Glendenin (1956).

Accuracy in the values for the fission yields is still hard to obtain in spite of great advances in 4π counting and improved mass spectrometric techniques. Methods reported in this thesis have been designed to reduce the error in normalization of the mass spectrometric yields of the different fission product elements.

EXPERIMENTAL PROCEDURE

A. Mass Spectrometry

The measurements of isotope ratios reported in this thesis have been obtained using a 25-cm radius, ninety-degree sector, solid-source mass spectrometer. The ion detection was accomplished with a 12-stage Allen-type electron multiplier with copper-beryllium dynodes having a total gain of about 100,000. This instrument has been described by D. E. Irish (1956).

(a) Filament Assembly

All analyses were performed using the multiple-filament surface-ionization source described by Inghram and Chupka (1953) but with the beam centering plate omitted. In this source, provision is made for the simultaneous use of three filaments; two side filaments bearing the material to be analysed and a centre filament to ionize the atoms. This arrangement has the advantage over a single-filament assembly in that the emission rate of the sample is controlled by the relatively cool sample filament and the ionization by the hot ion filament. The filaments, which were made of tungsten ribbon 0.001" thick and 0.030" wide, were supported and insulated from the head assembly with standard 0.040" untinned kovar-glass feedthroughs. The nichrome source assembly was cleaned in aqua regia and a new set of filaments was made for each analysis. These filaments were heated in vacuo to approximately 2000°C for one hour to clean their surfaces. When two sample filaments were employed, about half the sample was placed on each. In this way, a complete isotopic analysis could be performed using only the first sample filament and then, at some later time, certain parts of the analysis could be repeated. This has the great advantage that measurements can be made, without removing the source from the mass spectrometer, at widely differing times between which the sample may undergo considerable isotopic change due to radioactive decay. Cross contamination precludes the possibility of putting different samples on the two filaments since, even when a sample filament is not purposefully heated, enough heat from the nearby ion filament is available for the volatilization of some elements.

(b) Sample Loading

A small drop of the concentrated solution to be analysed was added to one or both sample filaments, which are removable for this purpose from the source assembly. A current of between 1.4 and 2.0 amperes was passed through the filament at this time, heating it to about 150°C to hasten the evaporation. Because most of the samples analysed in this work were highly radioactive, this procedure was performed in an air-tight "dry-box" where care was taken to ensure that the droplets did not splutter as they evaporated.

(c) Ionic Species

By using suitable combinations of ion and sample filament temperatures, the spectrum of the isotopes of one fission product element could often be measured without interference from other elements. In this way, isotopes of an element such as barium could be obtained with no interference from nuclides with the same mass numbers such as isotopes of cesium. With the sample filament temperature ranging from 50°C to 1400°C and the ion filament between 1200°C and 2000°C, the order of appearance and the ionic form of the elements studied in this work was roughly Cs^+, Ba^+, CeO^+, Ce^+, Pr^+, LaO^+, Eu^+, Nd^+, Sm^+, NdO^+ and SmO^+.

(d) Mass Discrimination

Mass discrimination in this machine can be caused either by mass non-linearity in the electron multiplier, or by isotope fractionation at the ion source. Both these effects have been discussed by Irish (1956) who concluded from measurements made on neodymium that this electron multiplier caused no mass discrimination. Analyses of terrestrial barium isotopes, which were made in conjunction with the measurement of the half-life of Cs^{137} (Farrar et al., 1961), gave relative abundances in agreement with the values of Nier (1938) when obtained either with or without the electron multiplier as detector. In addition, since no change in the isotopic constitution of any element apart from that resulting from radioactive decay has been seen during the expenditure of any sample in this mass spectrometer, it has been concluded that the second effect is also negligible.

B. Chemical Reagents

Because considerable quantities of nitric acid were required for the dissolution of the various uranium compounds, and because the fission product elements analysed in this work were in submicrogram quantities, it was necessary to reduce impurities to an absolute minimum. The purification of nitric acid was accomplished by the triple distillation of concentrated CP reagent-grade acid in an all-quartz apparatus. The distillations were carried out at a very slow rate to eliminate bumping and mechanical carry-over. The hydrogen peroxide used was redistilled once in vacuo in an all-quartz apparatus. The hydrofluoric acid used was Baker and Adamson reagent grade, as provided by the manufacturer. Water was first distilled, then passed through a 100-cm dowex-50 resin exchange column to remove cationic impurities. This water was then redistilled twice in an all-quartz still. The reagents and water were stored in polyethylene bottles to minimize contamination.

C. Irradiation

(a) Sample Preparation

All the samples, excepting one, consisted of uranium enriched to 95% U^{235} in the chemical forms U_3O_8 or UO_2SO_4. These samples were sealed in quartz vials approximately 1 cm long and 0.2 cm wide which were then tightly wrapped in sheet aluminium to disperse the heat generated by the samples during irradiation. The vials were then sealed in aluminium cans and were irradiated in the McMaster swimming pool reactor. One sample, a disc of natural uranium metal, 3 cm in diameter and 0.4 cm thick, was irradiated in the Oak Ridge X-10 reactor sixteen years before analysis. Irradiation data concerning the samples is shown in Table I.

Samples F and H were prepared in the sulphate form in the hope that a rapid dissolution of the sample after irradiation could be effected. Heat generated during the irradiation, however, in both cases converted the sulphate to an insoluble compound necessitating a longer dissolution in nitric acid.

(b) Irradiation Times

The sizes of the samples and the lengths of irradiation were roughly governed by the following factors:

(i) The maximum size of the sample was limited to approximately 10 mg of U^{235} by consideration of the heat generated in the sample.

TABLE I

Irradiation Data

Sample	Mass	Form	Reactor	Neutron Flux $n/cm^2/sec$	t_1[a)]	t_2[b)]
A	10.4 mg	U_3O_8 93% U^{235}	McMaster	1.5×10^{13}	14.43 h	90 h
B	10.0 mg	U_3O_8 93% U^{235}	McMaster	1.5×10^{13}	14.43 h	90 h
C	9.0 mg	U_3O_8 93% U^{235}	McMaster	1.5×10^{13}	14.43 h	89 h
D	11.0 mg	U_3O_8 93% U^{235}	McMaster	1.5×10^{13}	14.43 h	103 d
E	30 g	Uranium Metal	Oak Ridge	6.0×10^{11}	18 d	5757 d
F	5.0 mg	UO_2SO_4 93% U^{235}	McMaster	1.5×10^{13}	1.417 h	4.6 h
G	5.0 mg	U_3O_8 93% U^{235}	McMaster	1.5×10^{13}	1.408 h	3.3 h
H	6.0 mg	UO_2SO_4 93% U^{235}	McMaster	1.5×10^{13}	1.433 h	3.4 h

a) length of irradiation

b) time from the end of irradiation until time of analysis by mass spectrometer

(ii) The irradiations had to be long enough to produce sufficient fission products for the mass spectrometric analysis. Isotopic analyses of some elements such as cesium and barium could be made with much smaller samples than others like samarium.

(iii) For the measurement of the yields of short-lived fission products, it was necessary to have as short irradiations as possible, in consideration of factor (ii).

For the study of 12.8-day Ba^{140} and 32.5-day Ce^{141}, a 14.43-hour irradiation produced a sufficient number of fissions. After "cooling" for about two days the activities of samples A, B and C were low enough to perform the chemical analyses. Improved chemical techniques and high sensitivity of the mass spectrometer for isotopes of barium made possible the analysis of 82.9-minute Ba^{139} in samples F, G and H only 3.3 hours after 85-minute irradiations.

D. Chemical Procedure

All polyethylene and glass pipettes, centrifuge tubes and teflon used in these experiments were thoroughly cleaned first which chromic oxide and sulphuric acid cleaning solution, then with aqua regia followed by repeated rinsings with deionized water. Because of the considerable radioactivity present in most of the samples, the entire chemical separations were carried out as quickly as possible within a lead-brick lined "dry-box".

(a) Treatment of Samples A and D

The remarkable properties of teflon that allow the adherence of very few substances, and the resistance to attack by most acids even with the application of a considerable amount of heat, make it very suitable for use in fission product studies. Capsules A and D were placed on a piece of teflon sheet and the outside surfaces were washed repeatedly in nitric acid. The capsules were then broken with a glass rod, and the contents were dissolved in about 2 ml of 6-molar redistilled nitric acid. The solution was evaporated to dryness under a heat lamp in order to remove the excess acid, leaving a yellow-brownish residue which was then dissolved quickly in about 3 ml of cold redistilled deionized water. This clear solution was then placed, with a pipette, into a centrifuge tube to which was added about 0.2 ml of redistilled hydrogen peroxide, which caused the formation of a dense white precipitate of $UO_4.2H_2O$. After centrifuging, the supernatant solution containing nearly all the fission products, was taken up with a new pipette and was deposited on the teflon sheet. This solution was then evaporated under the heat-lamp to one or two very small drops which were placed on the sample filaments of the mass spectrometer for isotopic analysis. This procedure took between five and six hours to perform, of which a total of perhaps fifteen minutes was spent in actual manipulations in the vicinity of the dry-box.

(b) Treatment of Samples B and C

The entire quartz capsules were dissolved in a mixture of equal parts nitric and hydrofluoric acid so that all the fission products, including those that had recoiled into the quartz, were obtained. To minimize the quantity of impurities imparted to the solution by the quartz, the acid was frequently replaced with fresh acid. This was continued until the acid penetrated the interior of the capsule after which the uranium, with its fission products, dissolved. Sufficient hydrofluoric acid was present to completely volatilize the silica as SiF_4. The procedure thereafter was identical to that used for samples A and D. Comparison of mass spectrometric data of samples B and C with those of A and D showed no apparent isotope discrimination as a result of failing to dissolve the quartz and any fragments tightly adhering to it. In the dissolution process for samples B and C, however, the fluoride in the samples caused the formation of BaF^+ ions in the mass spectrometer, which interfered considerably with the mass analysis of CeO^+ ions. Because cerium is analysed principally in this oxide form the latter method was discontinued.

(c) Treatment of Sample E

The whole piece of uranium was placed on a teflon sheet and a thin layer was removed by dissolution in redistilled nitric acid in order to clean the surface. This solution was then removed and the uranium was washed in deionized water. A larger sample was similarly dissolved and the uranium separated from the fission products in the manner described above.

(d) Treatment of Samples F, G and H

These samples were irradiated especially to measure the relative yields of 82.9-minute Ba^{139} and 12.8-day Ba^{140}. After irradiation, all three samples contained uranium in the oxide form and were treated as were samples A and D. By minimizing the quantities of acid and water used for the dissolutions, and generally hastening the procedure, these samples were placed in the mass spectrometer less than one hour after the breaking of the quartz. After removal from the reactor, seventy minutes were allowed to elapse before the start of the chemical separation of the uranium; in which time essentially all the 9.5-minute Cs^{139} had decayed to Ba^{139}. Had there been no such delay, there would have been the possibility that fractionation between the cesium and barium might have occurred.

E. Isotopic Analysis

With the high sensitivity from the electron multiplier, it was found that isotopes of cesium, barium, cerium, neodymium, samarium and europium, in quantities less than 10^{-10} grams were sufficient for isotopic analysis. In all the samples except D and E, the measured ratios changed during measurement due to radioactive decay. With a knowledge of the half-lives involved, each measured mass spectrogram was corrected to the time at which the given analysis started. The measured ratios, which are bracketed in the following tables, are presented only to show the magnitude of the decay corrections. The error shown with each measured isotopic ratio is the standard deviation from the mean of a large number of mass spectra.

Sample E, unlike the others, consisted of natural uranium containing only 0.720% U^{235}. It has been estimated from the flux data for the Oak Ridge X-10 reactor that approximately 3.0% of the fission products in this sample was produced by the fast fission of U^{238}. The few fission yield values known for U^{238}, principally radiochemical in nature, have been summarized by Katcoff (1960). The mass-yield curve for U^{238} fast fission fortunately does not differ appreciably from that of U^{235} except in regions of symmetric and very asymmetric fission and hence the corrections are significant in this work only for samarium and europium.

Equations used to convert measured ratios to cumulative fission yields are developed in the appendix.

RESULTS

A. Relative Cumulative Yields

(a) Barium

Measurements of the relative abundances of the barium isotopes occurring in the thermal neutron fission of U^{235} are summarized in Table II. Both Ba^{139} and Ba^{140} decayed appreciably during the analyses and, therefore, each measured mass spectrogram was corrected to the time at which the analysis started. Sample E, which was the oldest available sample, was used to obtain the ratio of Ba^{137} to Ba^{138}. Although the decay of Cs^{137} to Ba^{137} was incomplete, the total uncertainty in the ratio of the yields of the 137 to 138 mass chains was only 2%. This uncertainty was assessed from both the standard deviation of the measured barium ratios and the quoted standard deviation for the half-life of Cs^{137} (Farrar et al., 1961). Contamination of the samples with terrestrial barium made small contributions to the spectrograms at mass numbers 137 and 138; this, however, was easily subtracted since terrestrial barium also contains Ba^{135} and Ba^{136} which are not formed appreciably in fission. Thus, by measuring the abundances of Ba^{135} and Ba^{136} relative to the Ba^{137} and Ba^{138}, the contribution of the contamination was accurately obtained. A very small contribution to the Ba^{136} was produced by fission Ba^{136} that had decayed from the Cs^{136} which is formed in very low yield as a primary fission product. Allowance for this contribution was made with considerable precision because the independent yield and half-life of Cs^{136} have been recently determined (Farrar and Tomlinson; and Farrar, Bidinosti and Tomlinson - forthcoming publications).

TABLE II

Mass Spectrometric Data for Barium

Sample		Relative Abundances				
		Ba^{136}	Ba^{137}	Ba^{138}	Ba^{139}	Ba^{140}
A	Measured ratio[a]	0.0145	0.0206	1.000		(0.5995)
	S.D.	0.0005	0.0003			0.0032
	Corrected for contam.[b]	0.0003[c]		0.869_9		0.5995
	Relative yield[d]	0.001		1.062		1.000
A	Measured ratio[a]	0.0164	0.0237	1.000		(0.2584)
	S.D.	0.0003	0.0007			0.0020
	Corrected for contam.[b]	0.0006[c]		0.850_1		0.2584
	Relative yield[d]	0.001		1.061		1.000
C	Measured ratio[a]	0.0208	0.0298	1.000		(0.5804)
	S.D.	0.0006	0.0006			0.0058
	Corrected for contam.[b]	0.0002[c]		0.811_4		0.5804
	Relative yield[d]	0.001		1.074		1.000
E	Measured ratio	0.00940	0.270	1.000		
	S.D.	0.00003	0.002			
	Corrected for contam.[b]	0.001[c]	0.258	0.923		
	Relative yield[d]	0.001	0.987_6	1.069		
F	Measured ratio[e]				0.3080	1.000
	S.D.				0.0037	
	Relative yield[f]				1.034	1.000
G	Measured ratio[e]				0.3037	1.000
	S.D.				0.0040	
	Relative yield[f]				1.018	1.000
H	Measured ratio[e]				0.3058	1.000
	S.D.				0.0026	
	Relative yield[f]				1.031	1.000
Weighted relative yield			0.987_6	1.069	1.028	1.000

a) Ratio corrected for decay of Ba^{140} during measurement

b) Correction assuming natural barium isotope abundances obtained by Nier (1938)

c) Estimated amount of Ba^{136} that had decayed from Cs^{136} independently formed in fission

d) Assuming the half-lives of Cs^{137} and Ba^{140} are 30.4 years (Farrar et al. 1961) and 12.80 days (Engelkemeir et al. 1951) respectively

e) The ratio shown is the weighted average of measurements corrected for decay of Ba^{139} and Ba^{140} to a time 2 hours after removal from the reactor

f) Assuming that Ba^{139} has an independent yield 1.1% of the total chain yield (Ferguson 1959) and a half-life 82.9 minutes (Butler and Bowles 1958)

The ratio of Ba^{139} to Ba^{140} was measured for more than three half-lives of the Ba^{139}. A least squares analysis of the data gave a value of 83 minutes which is close to the value of 82.9 ± 0.1 minutes obtained by counting techniques (Butler and Bowles, 1958). It was considered, however, that the latter value which was obtained from measurements extending over ten half-lives was more accurate and hence was used for half-life corrections to the mass spectrometric data shown in Table II. No corrections for terrestrial contamination were, of course, necessary in these measurements since neither Ba^{139} nor Ba^{140} occur naturally. The measurement of the yield of Ba^{139} was unique in that it is by far the shortest-lived isotope to be measured in a solid-source mass spectrometer.

(b) Cerium

The mass spectrometric data and the relative yields of the various cerium isotopes produced in fission are shown in Table III. The Ce^{141} was measured relative to Ce^{142} and Ce^{144} in young fission products, whereas the Ce^{140} was obtained relative to Ce^{142} in older fission products in which essentially all the Ba^{140} had decayed to Ce^{140}. Where experimental values are bracketed the values shown are corrected for radioactive decay to the start of mass spectrometric analysis.

The presence of stable isotope contamination in fission product cerium would effect the ratios of Ce^{140} and Ce^{142} to Ce^{144}. The constant character of these ratios when obtained from several samples has led to the belief that terrestrial cerium was not a contributing factor in these measurements.

TABLE III

Mass Spectrometric Data for Cerium

Sample		Relative Abundances			
		Ce^{140}	Ce^{141}	Ce^{142}	Ce^{144}
A	Measured ratio[a)]		(0.841)	1.000	(0.904)
	Relative yield[b)]		0.988	1.000	0.920
	S.D.		0.009		0.008
B	Measured ratio[a)]		(0.810)	1.000	(0.880)
	Relative yield[b)]		0.983	1.000	0.910
	S.D.		0.019		0.005
C	Measured ratio[a)]			1.000	(0.908)
	Relative yield[b)]			1.000	0.921
	S.D.				0.017
D	Measured ratio[a)]	(1.073)		1.000	(0.712)
	Relative yield[b)]	1.076		1.000	0.914
	S.D.	0.015			0.018
	Weighted average	1.076	0.987	1.000	0.914
	S.D.	0.015	0.009		0.006

a) Due to radioactive decay, the mass spectrometric ratios changed over the duration of measurement (see text)

b) Assuming that the half-lives of Ba^{140}, Ce^{141} and Ce^{144} are 12.80 days (Engelkemeir et al., 1951), 32.5 days (Freedman and Engelkemeir, 1950) and 285 days (Merritt et al., 1957) respectively.

Complete separation of the rare earths in the mass spectrometer is not always possible by the adjustment of the filament temperatures. This is a disadvantage but, since each element has some isotope which has no corresponding isobar in the contaminant element, it is nevertheless possible to obtain the isotopic abundances. An example is the difficulty in separating praseodymium from cerium. In old fission products, this is of no consequence since praseodymium exists only at mass 141 at which the cerium has no isotope. In the young samples A, B and C, however, where the abundance of 32.5-day Ce^{141} was desired, both Ce^{141} and, to a lesser extent, Pr^{141} contributed to the mass spectrograms. The Pr^{141} contribution could be calculated from measurements of the peak at mass 143, which was principally Pr^{143}, and assuming a rough value for the relative yields of the masses 141 and 143. Subtractions of very small additional contributions to the 143 mass peak from Ce^{143} and Nd^{143} had to be made first; these being obtained by noting the relative magnitudes of the peaks at masses 142 (cerium only) and 145 (neodymium only). Corrections such as these were tedious since they had to be made to each of approximately fifty mass spectrograms. They were, however, always small and resulted in a knowledge of the relative abundances of Ce^{141}, Ce^{142} and Ce^{144} that were almost independent of small changes in the half-lives of Ce^{141} and Ce^{144} and that were even less dependent on small changes in the half-lives of Ce^{143} and Pr^{143}.

(c) Neodymium

Several measurements have essentially confirmed the isotopic ratios of Nd^{143}, Nd^{145}, Nd^{146}, Nd^{148} and Nd^{150} measured by Melaika et al. (1955). Measurements on sample E, which was sixteen years old, gave, however, a value for Nd^{144} which was considered more accurate than that previously obtained since no correction for growth from Ce^{144} was required. This new value is therefore shown in Table IV. The ratio of Nd^{147} to Nd^{146} was obtained from sample H where half-life corrections amounted to approximately 2%. Since Pm^{147} produced from the decay of Nd^{147} contributed to the mass spectrograms, a compensating correction almost cancelling the decay correction was required, the value of this being obtained by measurement of the Pm^{149} in a similar manner to that which was employed in the cerium analyses. Measurement of this ratio in sample G, but without corrections for Pm^{147} contamination, gave a yield ratio $0.732 \leqslant \frac{Nd^{147}}{Nd^{146}} \leqslant 0.750$, the lower and the upper limits being set by the conditions that promethium was being ionized equally efficiently to the neodymium, and was being not ionized at all, respectively. Because promethium has not been seen in this laboratory to ionize as efficiently as neodymium, the ratio should be closer to the upper limit, essentially confirming the observations on sample H.

(d) Samarium

The measured ratios of the samarium isotopes are shown in Table V. Since the absence of Sm^{148} precludes the possibility of natural samarium contamination in sample E, and since the beta decay of the mass 150 chain terminates at Nd^{150}, it may be assumed that the observed Sm^{150} was formed entirely as a result of neutron capture by Sm^{149}. The total relative yield of the 149 mass chain was therefore obtained by the direct addition of the abundances of Sm^{149} and Sm^{150}. The considerable age of sample E, in which the 2.52-year Pm^{147} had almost completely decayed to Sm^{147}, permitted the determination of the relative cumulative yield of Sm^{147} with essentially no dependence on the half-life of Pm^{147}.

TABLE IV

Mass Spectrometric Data for Neodymium

Sample		Relative Abundances				
		Nd^{143}	Nd^{144}	Nd^{145}	Nd^{146}	Nd^{147}
E	Measured ratio	1.000	0.929	0.668		
	Relative yield[a)]	1.000	0.928_5	0.665		
	S.D.		0.002	0.002		
H	Measured ratio				1.000	(0.725)[c)]
	Relative yield[b)]				1.000	0.749
	S.D.					0.015

a) Corrected for 3.0% contribution from U^{238} fast fission

b) Assuming the half-life of Nd^{147} is 11.06 days (Wright et al., 1957)

c) Due to radioactive decay of Nd^{147}, the mass spectrometric ratio changed over the duration of measurement

TABLE V

Mass Spectrometric Data for Samarium

Sample E	Relative Abundances					
	Sm^{147}	Sm^{149}	Sm^{150}	Sm^{151}	Sm^{152}	Sm^{154}
Measured ratios	2.088	1.000	0.0143	0.358	0.263	0.075
S.D.	0.008		0.0002	0.007	0.005	0.002
Ratios renormalized[a)]	2.059	1.000	0.000	0.353	0.259	0.074
Corrected for decay[b)]	2.085	1.000	0.000	0.395	0.259	0.074
Relative yield[c)]	2.118	1.000	0.000	0.391	0.255	0.071

a) The Sm^{150} in Sample E was formed entirely by the reaction $Sm^{149}(n\gamma)Sm^{150}$

b) Corrections assuming half-lives of Pm^{147} and Sm^{151} are 2.52 years (Melaika et al., 1955) and 98 years respectively (see text)

c) Corrected for 3.0% contribution from U^{238} fast fission

A critical study of the variation in the ratio of Sm^{151} to Sm^{149} over a period of several years suggests that the half-life of Sm^{151} should be about 98 years which is in good agreement with the 93 years reported earlier (Melaika et al., 1955). This method is, however, unsuitable for a proper half-life determination when applied for such a small fraction of the half-life. The relative yields of the samarium isotopes have been corrected for a small contribution from the fast fission of U^{238}.

(e) Europium

The measured isotopic abundances of the europium isotopes and the relative yields obtained from them are shown in Table VI.

TABLE VI

Mass Spectrometric Data for Europium

Sample E	Relative Abundances	
	Eu^{151}	Eu^{153}
Measured ratio	0.280	1.00
S.D.	0.005	
Corrected for hold-up[a)]	2.65	1.00
Relative yield[b)]	2.70	1.00

a) Corrected for 98-year half-life of Sm^{151}

b) Corrected for 3.0% contribution from U^{238} fast fission

A large correction to the measured ratio was necessary to obtain the yield at mass 151 because most of the fragments in this chain were held up at Sm^{151}. Because the measured ratio has a standard deviation of 1.8% and the half-life of Sm^{151}, estimated to be 98 years, is probably known only to within ±5 years, the total uncertainty in the relative yields is probably no better than 10%.

DISCUSSION

A. Relative Yields

Since the relative yields of the cesium and xenon isotopes had been recorded previously (Petruska et al., 1955; Wanless and Thode, 1955) with sufficient precision, no further measurements of these two elements were made in the present work.

No comparable work on the isotopic ratios of fission product barium has been conducted previously. From sample E, irradiated almost sixteen years before analysis, the Ba^{137} to Ba^{138} ratio was obtained and this made possible the normalization of the cesium and barium isotope abundances. Even in this old sample the Cs^{137} had decayed only by about 29% and therefore the error in the value obtained for the relative yields of Ba^{137} and Ba^{138} depends not only on the mass spectrometric ratio but also on the half-life of Cs^{137}. Should the value for the Cs^{137} half-life be different from 30.4 years it would be necessary to re-calculate the Ba^{137} to Ba^{138} ratio. It is believed, however, that the error in the ratio of the cumulative yields given in Table II could be no greater than 2%. There was no comparable difficulty in obtaining the Ba^{138} to Ba^{140} ratio since very young fission products were used and the correction needed to compensate for the decay of 12.8-day Ba^{140} was small. This ratio was of particular importance as it made possible the normalization of the barium isotopes to the relative yields of the cerium isotopes which were measured relative to Ce^{140}. The ratio of the yields of the 140 to 139 mass chains, obtained by measuring the abundance of Ba^{140} relative to 82.9-minute Ba^{139}, was found to be 0.973 ± .008; the error including a contribution from the Ba^{139} half-life given by Butler and Bowles (1958). If the half-life were as much as 84 minutes, which is greater than Butler's value by eleven times his quoted error, the ratio of Ba^{140}/Ba^{139} would be increased by 3.5%. It would be difficult, using the present techniques, to measure the Ba^{139} much sooner after its formation in fission and therefore reduce the half-life dependence of the calculated relative yield. Although it was possible to fix its yield in relation to the Ba^{140}, the value obtained did not determine the normalization of the barium isotopes to those of either the cesium or cerium and, consequently, the accuracy of the value is not as vital as that of the relative yields of the other barium isotopes. No equivalent mass spectrometric data have been published for the barium isotopes but there is good agreement with the data of Baerg and Bartholomew (1957) for the relative yields of Ba^{139} and Ba^{140} which were obtained using radiochemical techniques. Their data are presented along with the results of this work in Table VII.

TABLE VII

Relative Yields of Barium Isotopes

	Relative Yields			
	Ba^{137}	Ba^{138}	Ba^{139}	Ba^{140}
This Work	0.987_6	1.069	1.028	1.000
Baerg and Bartholomew[a]			1.039	1.000

a) Relative yields using counting techniques and assuming 84.0 minute half-life for Ba^{139} (Baerg and Bartholomew, 1957)

A variety of fission product samples has been used to obtain the cerium isotope ratios. An old sample, in which the Ba^{140} had completely decayed through its daughter La^{140} to stable Ce^{140}, has been used to obtain the Ce^{140} to Ce^{142} ratio. Young fission products have been used to obtain the yields of 32.5-day Ce^{141} and 285-day Ce^{144} in relation to stable Ce^{142}. In this way it has been possible to obtain the ratios of the fission yields in the mass chains 140, 141, 142 and 144 whose values are practically independent of the half-lives of Ce^{141} and Ce^{144} as well as the half-lives of the precursors to Ce^{140}. The ratios obtained in this work are compared with previous data from this and other laboratories in Table VIII.

The ratios obtained for the isotopes Ce^{140}, Ce^{142} and Ce^{144} are in good agreement with those of Petruska et al. (1955a), Chu (1959) and Anikina et al. (1958) but not with those of Steinberg and Glendenin (1956). No comparable method has been used to determine the relative yield of mass 141 but Anikina, using the method of integral mass-spectrographic determination, obtained a value from Pr^{141} which is in excellent agreement with this work. This method, which relies on the integration of the ion currents of the isotopes of the different elements being normalized, would be very hard to employ with the mass spectrometers used in this laboratory because, for instance, both praseodymium and cerium appear in the mass spectra as Pr^+ and Ce^+ and simultaneously PrO^+ and CeO^+. In view of this, and because the magnitude of the ion currents in these mass spectrometers can be made to vary over a factor of 10^6 by simple filament temperature adjustment, it is difficult to see, from the description given by Anikina et al. (1958), how this method

TABLE VIII

Relative Yields of Cerium Isotopes

	Relative Yields			
	Ce^{140}	Ce^{141}	Ce^{142}	Ce^{144}
This Work	1.076	0.987	1.000	0.914
Petruska et al. (1955a)	1.058		1.000	0.893
Chu (1959)	1.05		1.000	0.90
Anikina et al. (1958)	1.086	0.966[a]	1.000	0.966[a]
Steinberg and Glendenin (1956)	1.119		1.000	1.061[b]

a) Obtained by the "method of integral mass spectrographic determination"

b) Using 282 day half-life for Ce^{144}

can obtain normalization of elements with the accuracy of the method used in this work. The present studies of neodymium isotopes have concentrated on both very old fission products where essentially all the Ce^{144} had decayed to Nd^{144} and very young fission products where Nd^{147} was measured relative to Nd^{146} and Nd^{148}. From these measurements it was possible to relate the yields of cerium isotopes to those of the neodymium at the 144 mass chain with no dependence on the value of the Ce^{144} half-life chosen. The Nd^{147} yield was likewise used to relate the neodymium and samarium yields through its isobar Sm^{147}.

The results quoted in this work are identical to those of Melaika et al. (1955) except at mass 144, where the new value is considered to be more accurate, and at mass 147 which has not been measured before as neodymium.

The relative yields of the neodymium isotopes have been compared with some literature values in Table IX. It is clear that the relative yields quoted by Steinberg and Glendenin (1956), Anikina et al. (1958) and Inghram et al. (1950) differ considerably from those reported in this work and in that of Melaika et al. (1955) and Chu (1959). There are several possible reasons for the difference between these results and those of Inghram et al. The fission products analysed by Inghram et al. were obtained from a fuel rod where a considerable fraction of the neutrons have energies of several MeV, whereas in this work, the neutron energies were essentially thermal. It is well known that for high energy neutron induced fission, the mass-yield curve becomes broader. This effect would tend to raise the yield of Nd^{150} relative to that of Nd^{144}. Other possibilities that would have a similar effect are the contributions

TABLE IX

Relative Yields of Neodymium Isotopes

	Relative Yields						
	Nd^{143}	Nd^{144}	Nd^{145}	Nd^{146}	Nd^{147}	Nd^{148}	Nd^{150}
This work	1.000	0.928_5	0.665	0.505	0.378_2	0.281	0.110
S.D.		0.002	0.002	0.003	0.005	0.002	0.002
Melaika et al. (1955)	1.000	0.930	0.665	0.505		0.281	0.110
Steinberg and Glendenin (1956)	1.000	0.966	0.649	0.523		0.287	0.115
Chu (1959)	1.00	0.90	0.66	0.50		0.282	0.110
Anikina et al. (1958)	1.000	0.966	0.690	0.552		0.293	0.121
Inghram et al. (1950)	1.000	0.860	0.670	0.520		0.304	0.122

to the fission products from the fast neutron fission of U^{238} and also the thermal neutron fission of the Pu^{239} that is formed as a result of neutron capture by U^{238}. There is no obvious reason why the data of both Anikina and Steinberg differ from the others; but it has been noted (Farrar and Tomlinson, 1962) that if their data were corrected assuming 8% and 4.5% terrestrial neodymium contamination respectively, the results from the different laboratories would become more consistent.

The relative yields of the samarium isotopes, which are compared with some literature values in Table X are in better agreement with the results of Melaika et al. (1955) and Chu (1959) than with those of Inghram et al. (1950).

TABLE X

Relative Yields of Samarium Isotopes

	Relative Yields					
	Sm^{147}	Sm^{149}	Sm^{150}	Sm^{151}	Sm^{152}	Sm^{154}
This Work	2.118	1.000	0.000	0.391	0.255	0.071
Melaika et al. (1955)	2.10	1.000	0.000	0.398	0.251	0.068
Chu (1959)	2.04	1.000	0.000	0.38	0.24	0.067
Inghram et al. (1950)	1.95	1.000	0.000	0.405	0.254	0.083

The reasons put forward to account for the differences in the neodymium ratios as measured by Inghram et al. (1950) and as obtained in this work, apply also to the differences observed for the samarium isotope abundances.

Since in the present work the correction to the Sm^{147} for the fraction of the mass chain remaining at 2.52-year Pm^{147} is small, the error in this relative yield should be small also and, therefore, the yields of the samarium isotopes can be normalized to those of neodymium almost independently of small changes in the values of the half-lives of Nd^{147} and Pm^{147}. Although the yield of the 153 chain may probably be obtained as accurately by taking a value intermediate to the Sm^{152} and Sm^{154} yields, it has been possible to obtain a measured value from the ratio of Eu^{151} to Eu^{153}. Taking 98 years for the Sm^{151} half-life, the yield of the 153 chain relative to the 151 chain has been obtained but is probably only accurate to 10%. No equivalent work on the yield of Eu^{151} has been published although Chu (1959) obtained the ratio of Eu^{155} to Eu^{153} in relatively young fission products, with a mass spectrometer. In compiling the yields of all the fission products in Table XII the mass 155 yield was obtained from the yield of Eu^{153} found in this work and the relative yield of Eu^{155} as found by Chu (1959).

The relative yields shown in Tables II to VI are summarized in Table XI. Included in this table are the xenon and cesium isotope abundances. The data for the latter isotope ratios have been changed by about 0.2% to become consistent with a 30.4-year half-life for Cs^{137} (Farrar and Tomlinson, 1961).

B. Absolute Yields

There is no single method which is obviously preferable for obtaining absolute fission yields from the relative yields summarized in Table XI. In Table XII, five different approaches have been given which

TABLE XI

Relative Yields of Heavy Mass Fragments

Mass Number	Relative Yields		
	Xenon[a]		
131	1.000		
132	1.496	Cesium[b]	
133	2.260	1.074	
134	2.750		
135		1.046	
136	2.207		Barium
137		1.000	0.987_6
138			1.069
139	Cerium		1.028
140	1.076		1.000
141	0.987		
142	1.000	Neodymium	
143		1.000	
144	0.914	0.928_5	
145		0.665	
146		0.505	Samarium
147		0.378_2	2.118
148		0.281	
149			1.000
150	Europium	0.110	
151	2.70		0.391
152			0.255
153	1.00		
154			0.071

a) Relative yields of xenon isotopes obtained by Wanless and Thode (1955)

b) Relative yields of cesium isotopes obtained by Petruska et al. (1955) corrected using 30.4-year Cs^{137} half-life obtained by Farrar et al. (1961)

TABLE XII

Results of Five Methods of Normalization to Obtain Absolute Yields

Mass Chain	Yield %				
1	2a)	3b)	4c)	5d)	6e)
117 - 130f)	3.984	3.984	3.984	3.984	3.984
131	2.98	2.92	2.92	2.91	2.93
132	4.45	4.36	4.36	4.35	4.38
133	6.72	6.59	6.59	6.57	6.62
134	8.18	8.02	8.02	8.00	8.06
135	6.55	6.42	6.42	6.40	6.45
136	6.57	6.44	6.44	6.42	6.47
137	6.26	6.14	6.14	6.12	6.17
138	6.78	6.64	6.64	6.63	6.68
139	6.52	6.39	6.39	6.37	6.42
140	6.34	6.21	6.21 (6.34)	6.20 (6.32)	6.25
141	5.82	5.70	5.82	5.80	5.73
142	5.89	5.77	5.89	5.88	5.80
143	5.80	5.68	5.80	5.79	5.71
144	5.39	5.28	5.39	5.37	5.30
145	3.86	3.78	3.86	3.85	3.80
146	2.93	2.87	2.93	2.92	2.89
147	2.19	2.15	2.19	2.19	2.16
148	1.63	1.60	1.63	1.63	1.61
149	1.04	1.02	1.04	1.03	1.02
150	0.638	0.625	0.638	0.637	0.628
151	0.405	0.397	0.405	0.404	0.399
152	0.264	0.259	0.264	0.263	0.260
153	0.150	0.147	0.150	0.150	0.148
154	0.0735	0.0721	0.0735	0.0734	0.0724
155g)	0.0296	0.0290	0.0296	0.0295	0.0291
156 - 162f)	0.025	0.025	0.025	0.025	0.025
Summation	101.47	99.52	100.25	100.00	100.00

a) Relative yields of Table XI normalized to Nd^{143} = 5.80% obtained by Petruska et al. (1955a)

b) Relative yields of Table XI normalized to Cs^{133} = 6.59% obtained by Petruska et al. (1955)

c) Relative yields of mass numbers 131 - 140 normalized to Cs^{133} = 6.59% and mass number 141 - 154 normalized to Nd^{143} = 5.80%

d) The relative yields of xenon, cesium and barium isotopes related to the relative yields of cerium, neodymium, samarium and europium by isotope dilution (1955a) and normalized to total 100%

e) Relative yields of Table XI normalized to total 100%

f) Values suggested by Walker (1960)

g) Obtained using Eu^{155}/Eu^{153} = 0.197 obtained by Chu (1959) differ from each other by only a few percent at individual fission yields. Using the absolute yield of 5.80% obtained by Petruska et al. (1955a) for the mass 143 chain, the relative yields of Table XI have been normalized and tabulated as absolute yields in column 2 of Table XII. The yields normalized in this way total 101.47% when some 4% of extrapolated yields in the region of 117 to 130 and 155 to 162 are included. In a similar way the yields may be normalized to the absolute yield of 6.59% (Petruska et al., 1955) for the mass 133 chain as shown in column 3, resulting in a yield total of 99.52%. A further normalization uses the value of 6.59% for Cs^{133} to normalize the yields of all chains of mass up to and including 140 and the value of 5.80% for Nd^{143} to normalize the relative yields of 140 and higher masses. This is shown in column 4 to Table XII and leads to a total of 100.25% fission yield. It also leads to two values for the 140 mass chain differing by some 2%. This could be either a measure of the error in the relative yields of Cs^{133} and Nd^{143} as measured by Petruska using the isotope dilution technique, or an accumulation of errors in the present work which results from relating the isotopic ratios of one element to those of another. The method used to obtain column 4 of Table XII is dependent not only on Petruska's values for the isotope dilution of cesium and neodymium but also on his estimate for the number of fissions. If, instead, the isotope dilution is used to obtain only the Nd^{143} yield relative to that of Cs^{133}, the isobaric technique may be used to relate xenon and barium isotopes to Cs^{133}; and cerium, samarium and europium to Nd^{143}. The resultant relative yields may be normalized to 100% as shown in column 5 of Table XII.

These yields are essentially the yields of column 4 normalized to 100%. The final method which is shown in column 6 takes the relative yields from Table XI and forces them to total 100%. It is not possible to make a positive choice between the yields given in the five columns of Table XII, perhaps because they are so similar. A change in the accepted half-life of Cs^{137} would alter all the values given by the method of column 5, whereas in the other methods it would only alter groups of yields. On the other hand, it was considered unlikely that the absolute yield of either Cs^{133} or Nd^{143} was known to better than 2%, or, in fact, that even their relative yield was known with this accuracy from the work of Petruska et al. (1955, 1955a).

The normalization in column 6 of Table XII has been arbitrarily selected as the most favoured for several reasons. Firstly, the absolute values are not dependent on other work where uncertainty of values is arbitrarily assigned. Secondly, this method gives an accurate yield for each mass chain relative to that of its neighbours without dual values at mass 140 as was found with two of the above methods. This is therefore the most useful mass-yield data for consideration of fine structure and it has been presented in Figure 1. It is unlikely that any yield in the present summation with over 1% value is in error by as much as 2% from its true value. Also, each yield relative to its neighbours is given with a certainty that the true value lies within 1% of the value given, with the possible exception of masses 137 relative to 138, and 139 and 153 relative to their neighbouring mass chains. In Table XIII the present data are compared with previous experimental work and with the compilations of various reviewers. Except for the normaliza-

FIGURE I

TABLE XIII

Absolute Yields of the Heavy Fragments from U^{235} Thermal Neutron Fission

Mass Chain	Element	Absolute Fission Yield				
		Walker (1960)	Katcoff (1960)	Steinberg and Glendenin (1956)	Petruska et al. (1955a)	This Work
117	Cd	0.010	0.011	0.010		
118		0.010				
119		0.011				
120		0.011				
121	Sn	0.012	0.015	0.014		
122		0.013				
123		0.014	0.013	0.014		
124		0.017				
125	Sb	0.036	0.021	0.023		
126	Sn	0.10		0.1		
127	Sb	0.25		0.25		
128		0.50				
129	I	1.00	0.8	1.0		
130		2.0				
131	Xe	2.9	2.93	2.9	2.92	2.93
132	Xe	4.3	4.38	4.3	4.37	4.38
133	Xe (Cs)	6.5	6.62	6.5	6.59	6.62
134	Xe	8.0	8.06	7.5	8.03	8.06
135	Cs	6.4	6.41	6.3	6.41	6.45
136	Xe	6.4	6.46	6.2	6.44	6.47
137	Cs (Ba)	6.0	6.15	5.9	6.15	6.17
138	Ba	5.8	5.74	5.7		6.68
139	Ba	6.4	6.55	6.2		6.42
140	Ba (Ce)	6.4	6.44	6.4	6.33	6.25
141	Ce	5.8	6.0	5.7		5.73
142	Ce	5.9	6.01	5.9	6.03	5.80
143	Nd	5.9	6.03	6.2	5.80	5.71
144	Ce (Nd)	5.6	5.62	6.0	5.39	5.30
145	Nd	4.0	3.98	4.0	3.86	3.80
146	Nd	3.1	3.07	3.2	2.93	2.89
147	Nd (Sm)	2.6	2.36	2.6	2.38	2.16
148	Nd	1.7	1.71	1.8	1.63	1.61
149	Sm	1.3	1.13	1.3	1.13	1.02
150	Nd	0.70	0.67	0.71	0.64	0.628
151	Sm	0.45	0.44		0.45	0.399
152	Sm	0.28	0.281		0.285	0.260
153	Eu	0.14	0.169	0.14		0.148
154	Sm	0.08	0.077		0.077	0.0724
155	Eu	0.03	0.033	0.031		
156	Eu	0.015	0.014	0.013		
157	Eu	0.007	7.8×10^{-3}	7.4×10^{-3}		
158	Eu		2×10^{-3}	2×10^{-3}		
159	Gd		1.1×10^{-3}	1.1×10^{-3}		
160						
161	Tb		7.6×10^{-5}	7.8×10^{-5}		

tion of the samarium isotopes the present work is in excellent agreement with the work of Petruska et al. (1955a) but has given additional yields at masses 138, 139, 141 and 153. The samarium isotopes have been normalized to the neodymium isotopes in this work through their isobars at mass 147. Although 9% disagreement exists between the relative yields of Nd^{143} and Sm^{149} as obtained by the isotope dilutions of Petruska and by the isobaric technique of this work, the latter method has fewer possible sources of error; the relative yield of these isotopes being known to better than 2.5%. It is interesting to note that using the neodymium-samarium normalization of this work the fission yield curve between the masses 146 and 152 becomes considerably smoother than the curve obtained using Petruska's values. Comparison of the present work in Table XIII with the work of Steinberg and Glendenin which is presented in Figure 2 is considerably less favourable. It is apparent that the present data do not indicate the extensive fine structure suggested by the work of Steinberg and Glendenin. The main difference occurs in the normalization of the cerium isotopes with those of neodymium and barium which Steinberg and Glendenin accomplished by isotope dilution. Significantly different values for their yield of Nd^{144} relative to the other neodymium isotopes and Ce^{144} relative to the other cerium isotopes give rise to fine structure not found in the present work. Finally, these authors have chosen radiochemical values for the yields at masses 139 and 141 which, although not very different from the values obtained in the present work, introduce further fine structure into their "recommended total chain yield" curve. The value obtained for Nd^{144} in the present work is not likely to be in error since no half-life

Heavy Mass-Yield Distribution Obtained
by Steinberg and Glendenin (1956)

FIGURE 2

correction to the measured isotope abundance was required and the ratio of Ce^{140} and Ce^{142} to Ce^{144} not only confirms the earlier work of Petruska et al. (1955a) but also the work of Chu (1959) and Anikina et al. (1958). Comparisons of the present data with reviews by Walker (1960) and Katcoff (1960) are more favourable than with the data of Steinberg and Glendenin (1956). This may be expected, however, since the reviews are largely based on an average of the values of Petruska et al. and those of Steinberg and Glendenin.

The yield of Ba^{140} was long considered to be one of the best known fission yield values and normalization of radiochemical yields were frequently made to 6.32% obtained by Yaffe et al. (1954). In the present work the yield of the 140 chain comes to 6.25% which is consistent with the more recent value of (6.36 ± 0.12)% obtained by Santry and Yaffe (1960). Excellent agreement exists between the present work and that of Katcoff and Rubinson (1953) for the yield of the 133 mass chain; both reporting an identical value of 6.62%. Although such agreement must be considered somewhat fortuitous, the careful measurement of the absolute yield of Xe^{133} obtained by Katcoff and Rubinson using a fission counter to determine the total number of fissions has long been considered to be one of the most accurately determined absolute fission yields. Examination of Figure 1 shows that apart from the well-established "fine structure" in the region of mass 134 the remainder of the fission yield curve is fairly smooth. The values for the yields in the heavy mass region are plotted in Figure 3 along with the hypothetical fission yield curve deduced by Pappas (1953). The latter curve was obtained by assuming a smooth mass-yield curve for the primary fission fragments and

Heavy Mass-Yield Distribution Predicted by Pappas (1953)

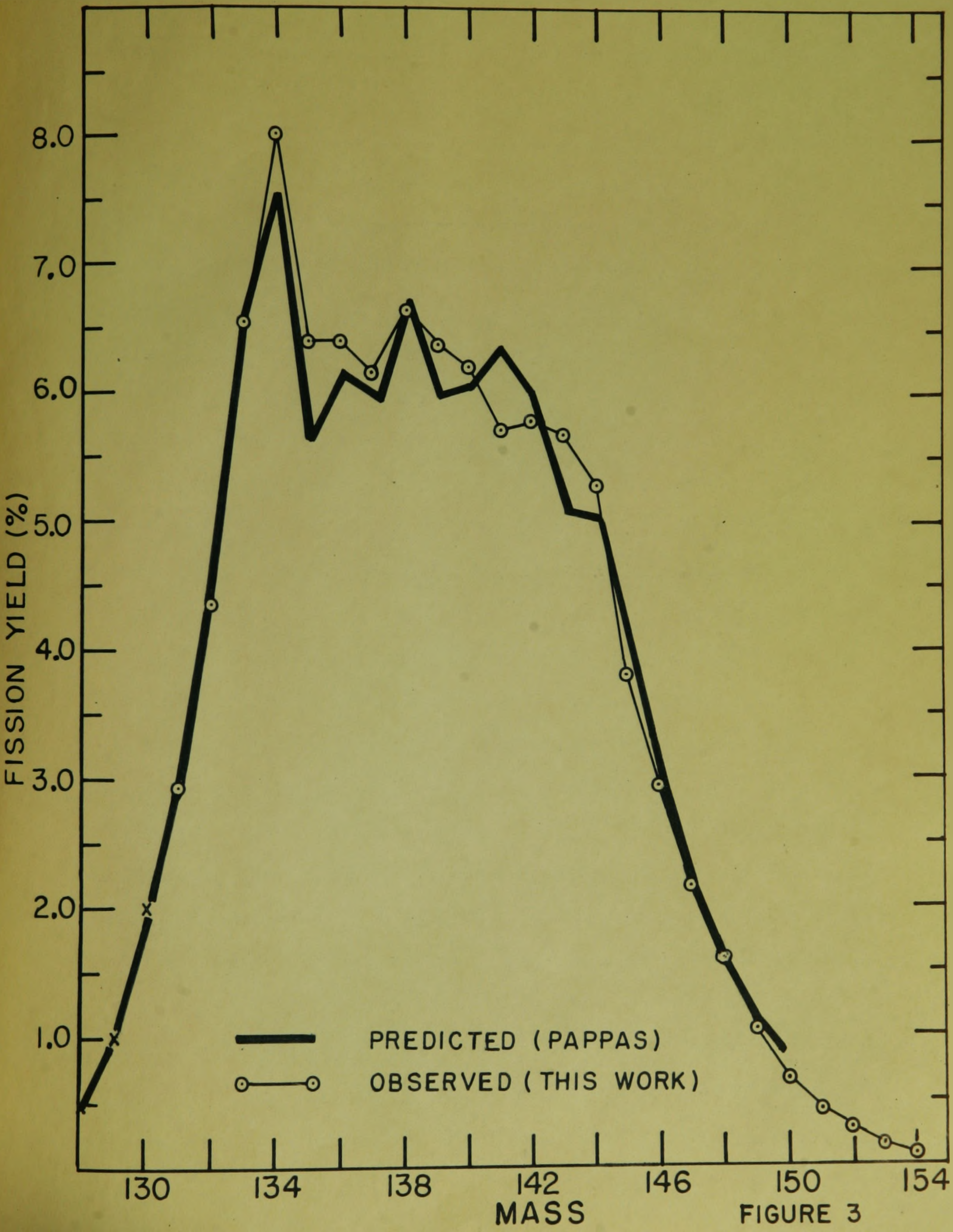


FIGURE 3

then superposing on it the effects of fragments with 83, 85, 87 and 89 neutrons emitting a neutron to form the more energetically favourable 82, 84, 86 and 88 neutron configurations. From Figure 3 it is readily apparent that the yields do not show any of the "fine structure" predicted by Pappas (1953), and in fact, where Pappas predicts a maximum at mass 141, a minimum is observed.

C. Interpretation

In order to discuss the fine structure in the mass-yield curve, it is important to review four possible processes that can influence the observed cumulative fission yields.

(i) Structural preference in the fission act for isotopes of a given mass and charge.

(ii) Variation in the neutron emission from the primary fragments.

(iii) Delayed neutron emission from some fission products that have undergone one or more beta decay processes.

(iv) Neutron capture by fission products.

Since beta decay does not change the mass number of the members of a beta decay chain, any explanation of the origin of the cumulative yields need not be concerned with the charge distribution of the complementary fragments. Any preference in the primary fission act must result, for binary fission, in equal yields for the complementary primary fragments. If both these de-excite by the emission of the same number of neutrons, then the observed fission yields must be identical. The other three processes, because they result in simple chain branching, affect one fragment but not the complementary fragment, and hence introduce differences between the light and heavy mass-yield distributions. Neutron absorption increases the yield of the mass chain one mass unit higher than that of the neutron absorber, whereas prompt and delayed neutron emission increase the yield of the mass chain one unit lower than the emitter. Thus it can be seen that the latter two processes increase the yield of a particular mass chain but always at the expense of an adjacent chain. In this work, the effects of neutron capture have been minimized by the use of appropriately low neutron fluxes and short irradiation times. Since the data have been corrected for this process where necessary, and since it is of no fundamental importance to the actual fission process, neutron capture will not be discussed further.

The effects of delayed neutrons are more difficult to assess. In 1956 Keepin and Wimett (1956) reviewed the subject of delayed neutrons. They gave complete tabulations of all determinations of the delayed neutron periods and abundances for U^{235} and for several other fissionable nuclides. More recently Keepin and Wimett (1957) have obtained delayed neutron data for the fast fission of Th^{232}, U^{233}, U^{235}, U^{238}, Pu^{239} and Pu^{240} and for the thermal neutron fission of U^{233}, U^{235} and Pu^{239}. These authors programmed a least-squares analysis of their delayed neutron data on an IBM704 computer and found that in all cases the data could be described by six neutron periods, although slight differences in the values of these were found for each fissioning isotope. The differences in the relative and absolute abundances for different fissioning nuclei are reasonable in view of the shifts in the mass and charge distribution of their fission products. However, radiochemical studies have shown that the six periods which constantly recur in studies of gross delayed neutron radioactivity must be attributed to more than six beta-active nuclides, meaning that some of the periods are complex, representing two or more nuclides with similar half-lives. Perlow and Stehney (1959) have shown from experiments where the iodine and the bromine fission products are separated quickly after a very short irradiation, that most of the delayed neutron precursors are isotopes of these two elements. The actual neutron emitters are isotopes of xenon and krypton respectively, but when neutron emission occurs, it does so promptly after the nuclide is formed, so the emission follows the half-life of the precursor isobar. From the experiments of Perlow and Stehney (1959), it appears that the relative yields of neutrons from the precursors I^{137}, I^{138} and I^{139} are 1, 0.47 and 0.38 respectively, and from the precursors Br^{87}, Br^{88}, Br^{89} and Br^{90}, the relative yields are 0.37, 1.0, 1.9 and 1.5 respectively. It must be pointed out that the identification and yields of all the precursors except perhaps Br^{87} and I^{137} are still somewhat uncertain. The yield for the delayed neutrons with the 2.30-second period found by Keepin et al. (1957) is considerably higher than can be explained by the yields from the delayed neutron precursors Br^{90} and I^{139} obtained by Perlow and Stehney (1959). Keepin has predicted that in addition to Br^{90} and I^{139}, the nuclides Br^{91} and Cs^{144} might be precursors contributing to this period.

It is apparent that the mass-yield curve may be corrected only roughly for these effects, the result being that the yields of the 137 and 139 mass chains are raised approximately 0.1% and 0.16% fission yield whereas the yield of mass 138 drops very slightly and the yield of mass 136 drops about 0.2%. The effect of these changes on the mass-yield curve for the heavy fragments is that the irregularities between masses 135 and 139 are largely removed. Instead there is a broad peak extending from masses 136 to 141 and a sharp peak between masses 132 and 136. The remaining fine structure cannot be attributed to the effects of presently known delayed neutron emitters.

There have been several suggestions as to why there is fine structure in the mass-yield curve around mass 134. Glendenin (1949) has postulated that the primary fission fragments have a smooth mass-yield distribution but that fragments reaching the 83-neutron configuration tend to "boil off" a further neutron to reach a more stable 82-neutron shell structure. Pappas (1953) has extended this postulate to include nuclides with 85, 87 and 89 neutrons boiling-off one neutron to reach the more stable 84, 86 and 88 neutron configurations. Wiles et al. (1953) postulated that high yields near mass 134 must result from an actual structural preference for isotopes with 82 neutrons, in the primary fission act. After noting that in the thermal neutron fission of U^{235}, U^{233} and Pu^{239}, the number of neutrons emitted for a near-symmetrical split is different than for a very asymmetrical split, Fickel (1959) and Bidinosti (1959) implied that fine structure in the mass 134 region might be the result of the transition in the number of neutrons emitted by fragments between these mass regions.

The postulates used by Glendenin (1949) and Pappas (1953) to account for the fine structure in the heavy mass region, assume a mechanism whereby neutron emission from a specific nuclide is greater than for neighbouring nuclides. By this mechanism the forming of a maximum such as that at mass 134 must be accompanied by a region of low yield in the mass region directly above the peak. The results of this work and that of Petruska et al. (1955a) indicate that the mass region above 134 is not abnormally low.

In connexion with the postulate of Wiles et al. (1953) it must be noted that any irregularity in the primary fragment mass-yield distribution must appear symmetrically in the complementary half of the distribution. The observation by Glendenin et al. (1951) of a high yield for Mo^{100}, the complementary fragment to mass 134 for the usual two-neutron emission, led these authors to a similar conclusion. One would expect on these grounds that the mass yield distributions for Pu^{239} and U^{233} should also have high yields at mass 134.

Present determinations in this laboratory of the thermal neutron fission yields of both U^{233} by Bidinosti et al. (1961) and Pu^{239} by Fickel and Tomlinson (1959, 1959a) have revealed that, in both cases, a pronounced maximum occurs at mass 134. It is of great interest to note that in both the fission of U^{233} and Pu^{239} there are maxima at mass 100 but in neither case is this the complementary fragment to mass 134, and, furthermore, the complementary fragment to mass 134 in each case is not significantly different from other yields in its respective region. From this it is evident that the peak at mass 134 is not formed primarily as a result of a preference in the primary fission act and must therefore result from some effects occurring after fission. The postulate of Wiles et al. (1953) might be criticized on theoretical grounds. It is well known that the level density of a magic nucleus is much the same as any other nucleus in the nearby mass region, the main difference being that the ground state is depressed. It is hard to believe therefore that the fission act could show a preference for the 82-neutron configuration because the fragments when formed are highly excited with perhaps several nucleons in excited levels; thus completely destroying any shell effects.

There are, however, ways of measuring the yields of the primary fragments directly. Using back-to-back ionization chambers in which the fissionable material was mounted on a thin film serving as a common cathode, and employing counting techniques, Brunton and Hanna (1950) and Brunton and Thompson (1950) were able to calculate the most probable mass ratio of two fragments from the ratio of the energy of their pulses. Stein (1957) using time-of-flight techniques to determine the velocities of the fragment pairs, was able to provide data concerning the absolute fragment energies and primary mass distributions for the thermal neutron fission of U^{233}, U^{235} and Pu^{239}. The statistical fluctuation in the data resulting from the 3050 fragment pairs studied in U^{235} fission, however, make the data unsuitable for studying fine structure. Stein showed, nevertheless, that the data could be fairly well represented by a smooth curve. Recently Gibson et al. (1961) and Milton and Fraser (1961) have used solid-state detectors and an improved time-of-flight apparatus, respectively, to study the prompt fission yields for the thermal neutron fission of U^{235}. Both groups, unlike Stein (1957), report fine structure in the mass-yield curve but they disagree on its extent. Present methods for measuring the yields of the primary fragments have not yet produced sufficient mass resolution to study the primary mass-yield distribution with the precision that is obtained in measurements of the cumulative mass-yield distribution.

A sensitive test for the accuracy of a complete set of cumulative fission yields is the evaluation of $\bar{\nu}$ the average number of prompt and delayed neutrons emitted per fission. The value of $\bar{\nu}$ may be obtained (Fickel, 1959) from the measured fission yields from the relation

$$\bar{\nu} = 236 - 2 \frac{\Sigma \text{ mass x yield}}{\Sigma \text{ yield}}$$

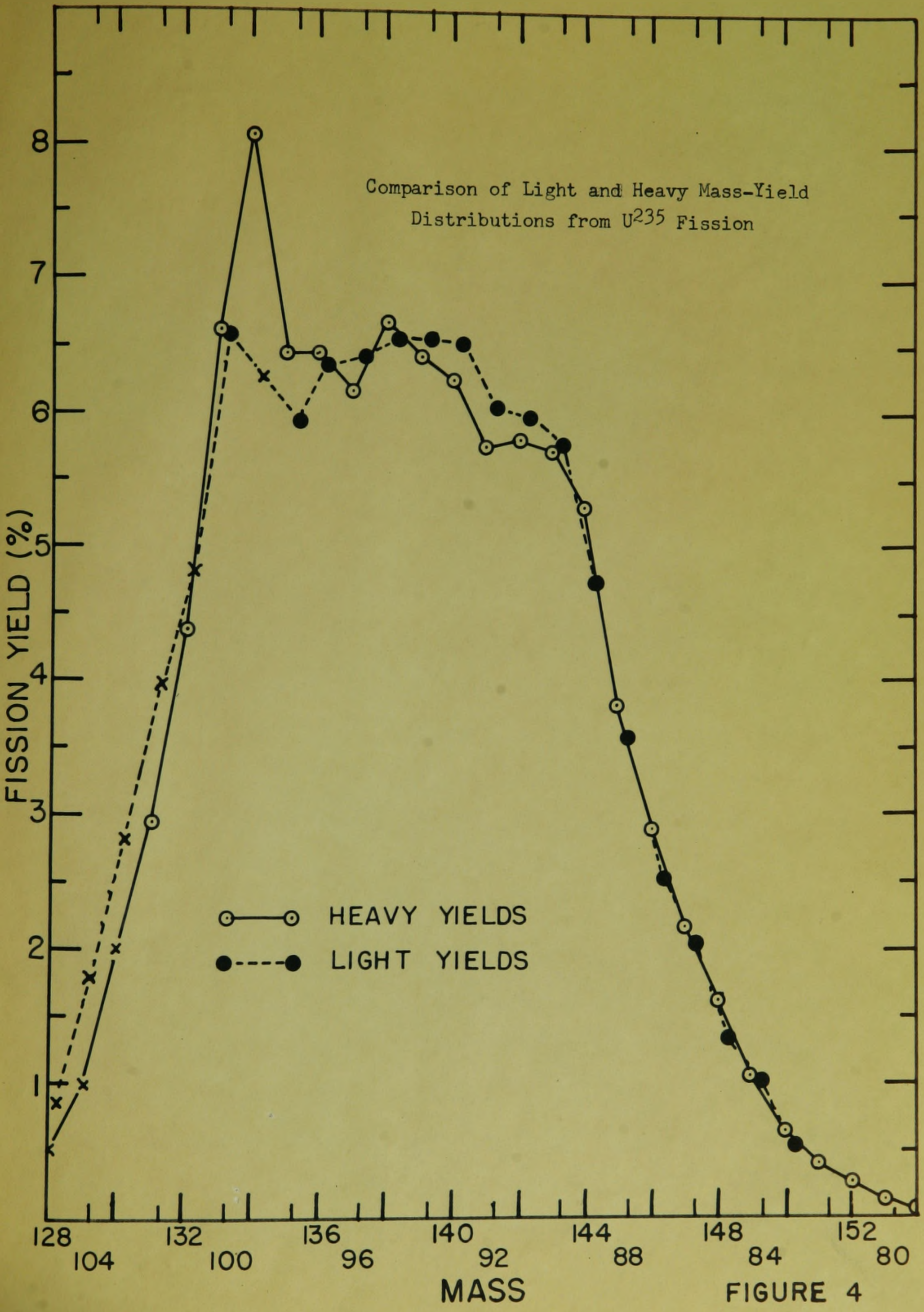
Using the results of this work for the heavy mass yields and using recent data (Farrar et al., 1961a) for the light mass yields, a value of $\bar{\nu}$ = 2.65 has been obtained. The value of the summation term is nearly equal to 236 and therefore small variations in this term will result in large percentage changes in $\bar{\nu}$. Because of this sensitivity, the value is considered to be in excellent agreement with the accepted value of $\bar{\nu}$ = 2.43 (Hughes et al., 1960).

It is instructive at this point to fold the mass-yield curve for the light fragments (Farrar et al., 1961a) over that for the heavy fragments. Since the sum of the mass numbers of correspodning fission fragments, together with the number of emitted neutrons must total 236 (U^{235} + n) and since the average number of neutrons that are emitted is non integral, there is no <u>a priori</u> way to decide which masses should be made to correspond. There are, however, several interesting ways in which to do this. The cumulative yield of each light mass chain could be plotted over that of a heavy mass chain in such a manner that the sums of their masses total 234 corresponding to 2 neutron emission, or alternatively, they could be plotted to total 233 for three neutron emission. Although the emission of 2.43 neutrons in any single event is impossible, there is some justification for summing the yields to total 233.57.

Because the light mass-yield curve in the region between masses 90 and 80 shows close similarity to the form of the heavy mass-yield curve between masses 144 and 154, the curves have been arbitrarily folded in Figure 4 for correspondence in these regions. The implications of the fact that this fold corresponds to an average emission of 2.75 neutrons will be discussed later. A similarity, although not so pronounced, exists between the light mass-yield curve between masses 105 and 101 and the heavy curve between masses 128 and 132. A fold that would make the curves correspond in these regions would be equivalent to the emission of an average of 2.2 neutrons.

In Figure 4 it can be seen that where the folded yield curves correspond, the yield of a given light mass chain does not exactly equal the yield of a single heavy counterpart, but instead lies between the yields of the complementary fragments for 2 and 3 neutron emission. This is to be expected in view of the following considerations on prompt neutron emission. The average number of neutrons emitted in the thermal neutron fission of U^{235} is 2.43 (Hughes et al., 1961). In addition, it has been found by Diven et al. (1956) that although in most cases either 2 or 3 neutrons are given off with approximately equal probability, events with 1 and also 4 neutron emission have been observed with probabilities of approximately 0.16 and 0.13 respectively. Events when either 0 neutrons or more than 4 neutrons occur have been observed but the probability is considerably lower.

As mentioned above, the curves have been folded so that they correspond to an average emission of 2.75 neutrons. It may be assumed that for a pair of fragments in these mass regions, either 2 or 3 neutrons



Comparison of Light and Heavy Mass-Yield Distributions from U^{235} Fission

FIGURE 4

are emitted, the latter occurrence being three times more probable than the former. This argument may be extended to include the additional effects of 0, 1, 4 and 5 neutron emission, but if the proportions are chosen to give an average of 2.75 neutrons, and if the contribution from the latter modes is not too great, the result does not differ greatly from that obtained in the simple case.

It would be advantageous now to consider a specific example. Every event in which the formation of mass 146 occurs, mass 87 occurs if 3 neutrons are emitted, and mass 88 occurs if 2 are emitted. Because fragments with adjacent masses to 146 each have two corresponding masses in the light mass region, the yield of mass 146 is not equal to the yield of mass 87 or mass 88 but lies somewhere between these yields. Furthermore, if a smooth curve is drawn through the yields in the neighbourhood of masses 87 and 88 and the curve is folded for the emission of 2.75 neutrons, then it can be shown that the yield at mass 146 will lie on this curve. It is apparent from Figure 4 that such is the case. Slight fine structure in the light mass-yield curve in the region between masses 83 and 88 has been attributed to delayed neutron emission (Petruska et al., 1955a). It can be seen that if this fine structure were eliminated, the yields in this mass region would also lie between the yields of their various complementary fragments. All these considerations would apply if the curves were matched on the left-hand side of Figure 4, where it can be seen that the light fragment yields would lie between their complementary heavy yields, at least in the mass regions 128 to 132 and 105 to 102. Since such a fold corresponds to an average of 2.2 neutrons per fission, it may be concluded from these data that this average number of neutrons is emitted whenever one of the fragments is in the mass region 128 to 132.

To summarize, the average neutron emission for a fragment ending in the 128-132 mass region and its complementary fragment is 2.2 neutrons, and for a fragment ending in the 144 to 154 mass region, and its complementary fragment, the average is 2.75 neutrons. What might happen in between these two mass regions will be the object of the ensuing discussion.

The above arguments allow one to conclude the total number of neutrons emitted by the light and heavy fragment in a given mass region, but do not give any indication as to the proportion that come from the light or heavy fragments. For example, there is no way of deciding from cumulative yield data alone whether the 2.75 neutrons, that are given off by fragments of mass 80 to 90 and masses 144 to 154, are given off entirely by the light or entirely by the heavy fragment, or whether the proportions are changing as a function of mass ratio. To be consistent with the observed data, however, the total number of neutrons at each mass split must remain constant in these or any extended mass regions where the fission yield curves match.

It was pointed out by Fickel (1959) and Bidinosti (1959) that any change in the total neutron emission characteristics as a function of mass must mean that the neutron emission is changing either in the light mass region, in the heavy mass region or in both, and that fine structure can be expected in the fission-yield curve where the change occurs. For example, if primary fragments of mass A and higher masses emit an average of two neutrons, and fragments of mass A - 1 and lower masses emit one, then the yield of the mass chain A - 2 will be approximately twice as great as the yields in nearby chains. In a similar way, if there is a transition from a low neutron yield to a higher neutron yield as the mass decreases, a minimum will be observed.

It may be stated that where the total number of neutrons emitted per fission is changing as a function of mass, it is possible to obtain from the cumulative yield data alone, assuming a smooth primary distribution, whether the increase or decrease in the total neutron emission as a function of mass is to be associated with the light or heavy fragments. This statement may be clarified with a further example. Let us assume that from masses 136 to 132 and the corresponding masses 98 to 102, there is a reduction in the total neutron yield. Three possibilities could exist.

(i) All the reduction in the total is to be found in the heavy mass region. That is, the reduction in neutron emission in passing from masses 136 to 132 exceeds the increase in the neutron yield in passing from masses 98 to 102. This would result in the yields between masses 136 and 132 being higher than those between 98 and 102. The yields in the latter region would be either on, or slightly above, a hypothetical smooth mass-yield curve depending on whether the neutron yield were constant or increasing slightly with mass respectively.

(ii) All the reduction in the total neutron yield is associated with a relative decrease between masses 98 and 102 which is greater than the relative increase between 136 and 132. This would lead to a depression in the measured cumulative yields in the 98 to 102 mass region relative to the smooth curve, and no change or a slight decrease in the measured yields in the region between masses 136 and 132.

(iii) The neutron yield of both the light and heavy fragments decreases in the same proportion in the two mass regions. This would result in the enhancement of the cumulative yields in the 136 to 132 mass region and the depression of the yields in the 98 to 102 mass region.

Now it is known from the cumulative yield data that the total neutron yield drops from 2.75 to 2.2 neutrons between masses 144 and 132 and their light mass counterparts. What form the total neutron yield takes in this mass region cannot be easily deduced,but changes over a limited mass range might result in fine structure of one of the three types mentioned above. A superficial examination of Figure 4, where it is seen that there is a large peak at mass 134 and a smaller peak at mass 100, indicates quite clearly that condition (i) applies in the mass regions used in the example. Hence it can be deduced from the cumulative yield data alone that the neutron yield from the heavy fragments between masses 136 and 132 decreases faster than does the total neutron yield in this region, and that the neutron yield from the light fragments between masses 102 and 98 also decreases with decreasing mass, although not as sharply as in the heavy mass region.

It is now evident that there are three observable quantities involved in the explanation of the cumulative mass yields. These are

(i) The primary fragment mass-yield distribution before prompt neutron emission,

(ii) The distribution of prompt neutrons emitted from the individual primary fragments,

(iii) The cumulative mass-yield distribution corrected for the effect of delayed neutrons.

It is also evident that if any two of the above three distributions are known, then the third may be obtained, except in the case of (ii) being obtained from (i) and (iii) where it is only possible to get the mean of the distribution.

Primary fragment mass-yield data are available from the work of Stein (1957), Milton and Fraser (1961) and Gibson et al. (1961). However, great accuracy is needed in such data if the cumulative yield data is to be subtracted from it in order to get the average neutron yield as a function of mass, because this operation involves the subtraction of two comparably large numbers.

There have been several measurements of the neutron multiplicity as a function of the mass ratio. Fraser and Milton (1954) measured the number of neutrons emitted in the fission of U^{233}, that were coincident with fission pulses in a double ionization chamber. By measuring the relative energies of the two fragments in each pulse the neutron yield as a function of mass number was deduced. These workers concluded that neutron emission from the light fragment predominates at low mass ratios and that emission from the heavy fragment predominates at high mass ratios. They also found that the total number of neutrons emitted shows much less dependence on the mass ratio. Whetstone (1959) studied the variation in prompt neutron emission from Cf^{252} as a function of the mass number of the fragment from which the neutrons were emitted. For this, the source was placed at the end of a large cadmium-loaded liquid scintillator tank for neutron detection, and the time-of-flight method was used to determine the fission mode. Because the neutrons are emitted principally in the direction of the fragment, in the laboratory frame of reference, the detected neutrons could be attributed almost entirely to one of the fragments. The resulting neutron distribution was similar to that found by Fraser and Milton (1954) for the thermal neutron fission of U^{233}. Recently Apalin et al. (1960), using a liquid organic scintillator have measured the number of neutrons emitted by individual fission fragments of U^{235}. The results of their work show a close similarity to the data found for U^{233} and Cf^{252}. They found that the number of neutrons emitted from the light fragment slowly varies from an average of 0.6 neutrons at mass 82 to about 2.5 at mass 109, and from the heavy fragment, it varies from 0.4 at mass 130 to about 2.7 at mass 158. The precision of the data at the ends of these mass ranges is however poor, because of the low probability of these fission modes. The mass dispersion in the data of Apalin et al. (1960), however, do not make it directly applicable to the cumulative yields of this work in order to obtain the primary yields. It therefore appears that the currently available primary yield data are not accurate enough to give useful neutrons yield distributions, and that the neutron distributions that are currently available are not sufficiently accurate to be used with the cumulative yields to give useful primary yield information.

It is of interest, nevertheless, to assume a smooth primary fission yield curve and to see what neutron distribution would result when taken with the cumulative yields obtained in the present work. This choice has been made, in spite of recent evidence (Gibson et al., 1961; Milton and Fraser, 1961) which indicates that some fine structure in the primary curve may exist, because it is not yet clear to what extent this may occur. For this analysis, it was assumed that the primary fission fragments, before the occurrence of prompt neutron emission, form a smooth double-humped mass distribution normalized, and symmetrical about mass 118. Any number of smooth distributions can be made, but the particular one used was one obtained from the experiments of Stein (1957) by drawing a smooth normalized curve of best-fit through his data. From this curve an analysis was made to find out what changes in the neutron distribution from the different fragments would be necessary to result in the exact cumulative mass-yield curve found in this work. The smooth primary mass-yield distribution, the final cumulative yields corrected for the effect of delayed neutrons, and the resulting neutron yield as a function of the heavy primary fragment mass number are shown in Figure 5. In Figure 6 are shown the results of a similar operation performed with the light fragment cumulative yields obtained by Farrar et al. (1961a), using the same primary distribution. In both Figures 5 and 6, the neutron distributions obtained from these analyses are compared with the experimentally-determined neutron distributions of Apalin et al. (1960).

It is indeed surprising that the neutron distributions resulting from these analyses show such simple variation with mass number. Large maxima and minima in the cumulative mass-yield curve can result from a small systematic change in the neutron multiplicities over four or five mass numbers. Furthermore, if the data of Apalin et al. (1960) were corrected for mass dispersion, the slope of the measured neutron distribution would be increased and the distribution would be contracted horizontally, thus making the curves almost exactly coinciding with those obtained empirically in this work.

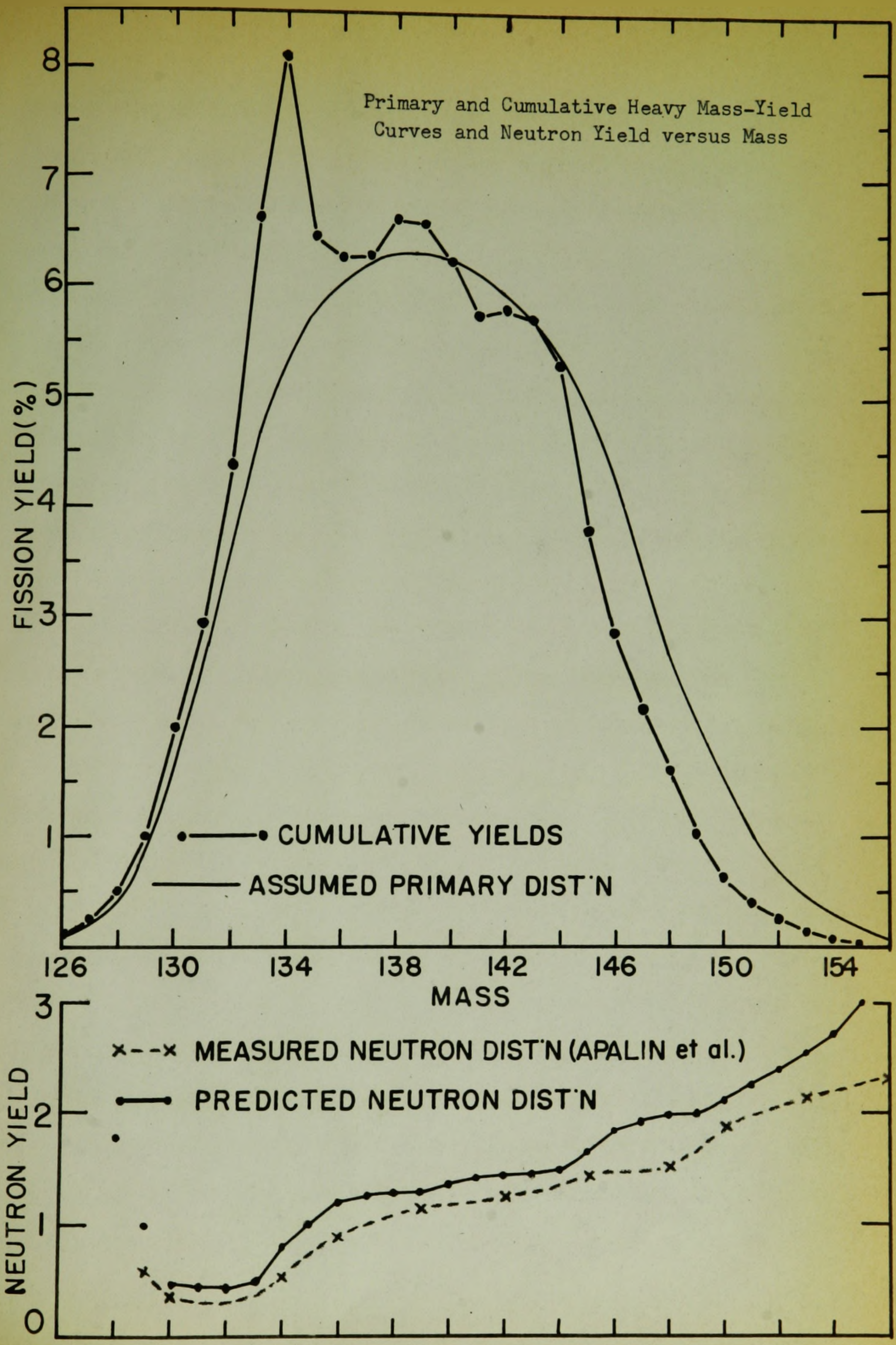
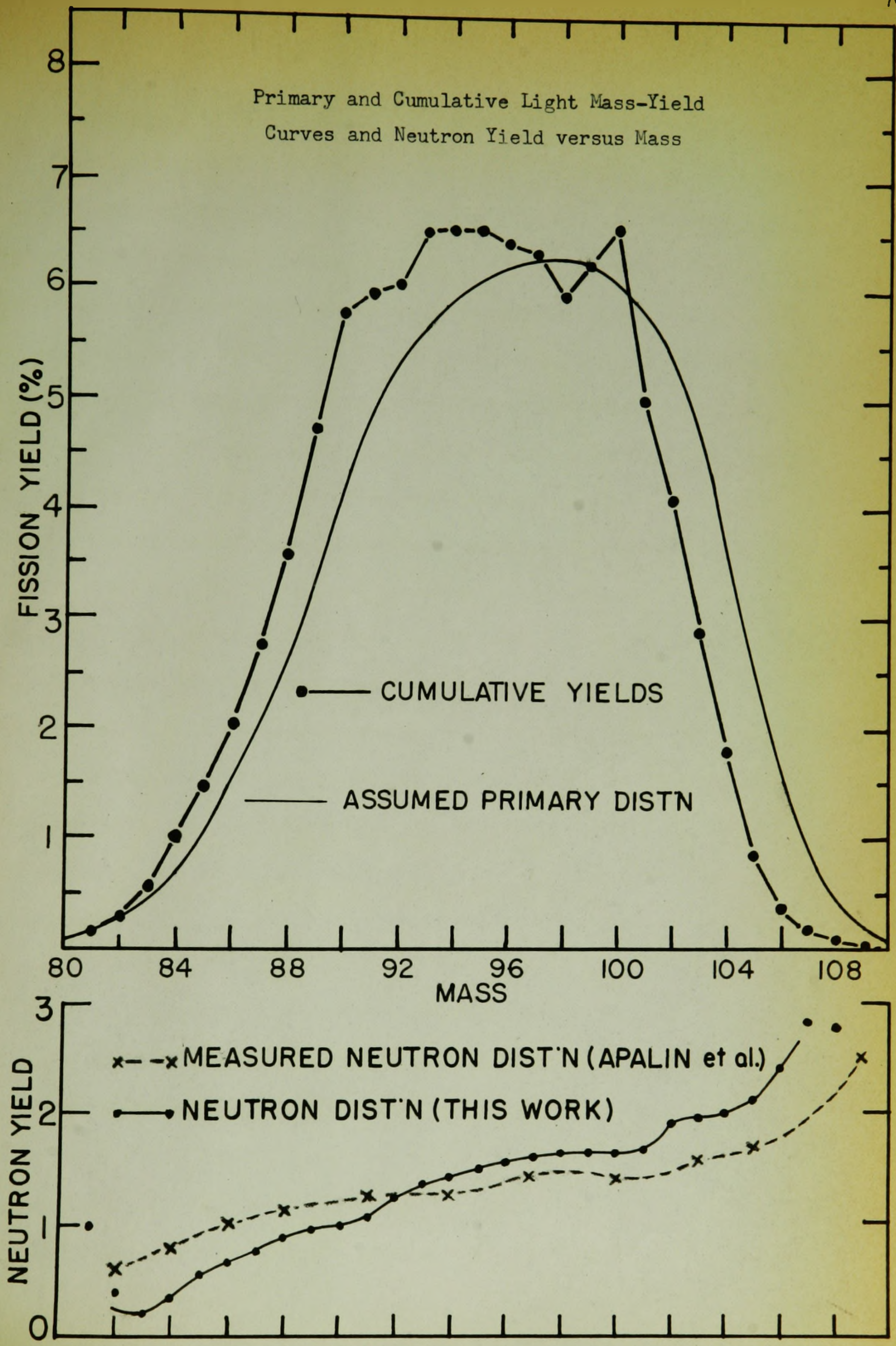

Primary and Cumulative Heavy Mass-Yield Curves and Neutron Yield versus Mass
FISSION YIELD (%)
8
7
6
5
4
3
2
1
CUMULATIVE YIELDS
ASSUMED PRIMARY DIST'N
126
130
134
138
142
146
150
154
MASS
NEUTRON YIELD
3
2
1
0
×--× MEASURED NEUTRON DIST'N (APALIN et al.)
PREDICTED NEUTRON DIST'N

FIGURE 5



Primary and Cumulative Light Mass-Yield Curves and Neutron Yield versus Mass

FIGURE 6

Brubaker (1960) has recently calculated the neutron emission from U^{235} fission fragments. The calculations were based on the fragment velocity data of Stein (1957), the primary fragment masses given by Cameron (1957) and evaporation theory. The results of these calculations are inconsistent with both the experimental data of Apalin et al. (1960) and the results of this work. They do, however, substantiate the method he employed, the data used being probably insufficiently accurate.

It must be pointed out that the resulting neutron distributions shown in Figures 5 and 6 are by no means unique. Any number of neutron distributions that are sharply varying functions of mass can be formed, but those distributions that are chosen so that there is a smooth variation with mass do not differ from one another in general shape. Had another primary fragment mass-yield curve been chosen, the resulting neutron distribution would, of course, be different. Fine structure on the top of the distribution would not affect the trends as much as widening the primary distribution. Any choice of primary mass-yield distribution must of course be normalized and must, for the case of U^{235}, be consistent with a total of 2.2 neutron emission for near-symmetric mass split, and 2.75 neutron emission for the more asymmetric splits.

Thus, assuming a smooth primary fragment mass-yield curve, the fine structure in the cumulative mass-yield curves can be easily explained on the basis of a slowly varying neutron yield with mass. Since the neutron distributions are so similar to those measured by Apalin et al. (1960), it is attractive to think that it is possible that all the fine structure in the cumulative mass-yield curve occurs as a result of the neutron variation with mass number. It is clear that there is no necessity for considering preference in the fission act for nuclides with closed shell configurations, or fine structure in the primary mass-yield distribution in order to account for fine structure in the cumulative mass-yield curve.

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APPENDIX

A. Equations for Converting Measured Ratios to Cumulative Fission Yields

(a) Simple Decay Sequence

A single set of equations suffice to describe the quantities of the members of a decay chain at various times after an irradiation. A typical beta-decay chain may be represented by the following sequence:

$$\begin{tikzcd} U^{235} \arrow[d, Rightarrow, "1 - f"] & \text{Fission} \arrow[d, "f"] & \\ A \arrow[r, "\lambda'"] & B \arrow[r, "\lambda"] & C_{stable} \end{tikzcd}$$

Where the primary yield of nuclide B is fraction f of the total chain yield, and the primary yield of nuclide A includes the yields of isobars that rapidly decay to A. λ' and λ are the decay constants of nuclides A and B respectively.

If the quantity of U^{235} is represented by U at any time t during the irradiation then

$$\frac{dU}{dt} = - \hat{\sigma}_a \emptyset U \quad \text{whence} \quad U = U_o e^{-\hat{\sigma}_a \emptyset t}$$

where U_o = the quantity of uranium at t = 0

$\hat{\sigma}_a$ = the effective thermal neutron capture cross section for U^{235}

$\emptyset$ = the 2200 metre/second neutron flux

For short irradiations such as those used in this work

$$\hat{\sigma}_a \emptyset t \ll 1 \quad \text{and hence} \quad U \approx U_o$$

The differential equations representing A, B and C at time t during the irradiation are as follows:

$$\frac{dA}{dt} = (1 - f)Uy\hat{\sigma}_F\varnothing - A\lambda'$$

$$\frac{dB}{dt} = A\lambda' + fUy\hat{\sigma}_F\varnothing - B\lambda$$

$$\frac{dC}{dt} = B\lambda$$

where $\hat{\sigma}_F$ = the effective thermal neutron fission cross section for U^{235}

y = the probability that a fission event will result in a fragment in this mass chain

hence $\hat{\sigma}_F y$ = the effective cross section for the formation of a fragment in this mass chain

Solution of the differential equations has been undertaken in two steps. The quantities of nuclides A, B and C at any time t during, or at the end of, a constant irradiation are:

$$\left.\begin{aligned}
A_t &= \frac{yF}{\lambda'}(1 - e^{-\lambda' t})(1 - f) \\
B_t &= \frac{yF}{\lambda}(1 - e^{-\lambda t}) - \frac{yF}{\lambda - \lambda'}(e^{-\lambda' t} - e^{-\lambda t})(1 - f) \\
C_t &= yFt + \frac{yF}{\lambda}(e^{-\lambda t} - 1) + \frac{yF\lambda}{\lambda - \lambda'}\left[\frac{(e^{-\lambda' t} - 1)}{\lambda'} - \frac{(e^{-\lambda t} - 1)}{\lambda}\right](1 - f)
\end{aligned}\right\}\text{(a)}$$

where $F = U_o\hat{\sigma}_F\varnothing$

At any time T after the end of an irradiation of duration t the quantities of A, B and C are:

$$\left.\begin{aligned}
A_T &= A_t e^{-\lambda' T} \\
B_T &= \frac{\lambda' At}{\lambda - \lambda'}(e^{-\lambda' T} - e^{-\lambda T}) + B_t e^{-\lambda T} \\
C_T &= \frac{At}{\lambda - \lambda'}(\lambda' e^{-\lambda T} - \lambda e^{-\lambda' T}) + B_t(1 - e^{-\lambda T}) + A_t + C_t
\end{aligned}\right\}\text{(b)}$$

where A_t, B_t and C_t are given by equations (a).

From equations (a) and (b) and a knowledge of f, λ', λ, t and T the quantities A_T/yF, B_T/yF and C_T/yF may be calculated. For a typical adjacent decay chain containing nuclides B*, C* and D* where B* and C* are the same elements as B and C respectively, but different isotopes, the quantities B^*_T/y^*F and C^*_T/y^*F can be determined for the same periods t and T.

To illustrate the procedure for obtaining y/y^*, the ratio of the cumulative yields of the two decay chains, let it be supposed that a measurement at time T of the ratio C_T/C^*_T produced a value J; and that calculations showed that $C_T = kyF$ and $C^*_T = my^*F$ where J, k and f are pure numbers

then $$y/y^* = mJ/k$$

(b) Sequence Containing a Nuclide with High Cross Section

Should nuclide B have a high thermal neutron cross section σ, the equations (a) and (b) may be used with slight modification.

The decay chain may be represented by the following sequence:

$$\begin{tikzcd} \text{Fission} \arrow[r] & A \arrow[r, "\lambda'"] & B \arrow[r, "\Delta"] \arrow[dr, "\sigma\phi"] & E_{\text{stable}} \\ & & & D_{\text{stable}} \end{tikzcd}$$

where the primary yield of nuclide B is fraction f of the total chain yield. Δ = the decay constant of nuclide B.

At any time t during, or at the end of a constant irradiation, it is immediately apparent that $E_t + D_t = C'_t$

where C'_t is the C_t found in equations (a) but with λ replaced by $(\Delta + \sigma\phi)$. The equations for the quantities of nuclides D or E at time t may be most easily obtained from C'_t, noting that:

$$D_t = \frac{C'_t \, \sigma\phi}{\sigma\phi + \Delta} \qquad E_t = \frac{C'_t \, \Delta}{\sigma\phi + \Delta} \qquad \ldots\ldots\ldots\ldots \quad (c)$$

At any time T after the end of an irradiation of duration t the quantities of nuclides D and E are:

$$D_T = D_t \quad \text{and} \quad E_T = C'_T - D_t$$

where D_t is given by equations (c)and

$$C'_T = \frac{A_t}{\Delta - \lambda'} (\lambda' e^{-\Delta T} - \Delta e^{-\lambda' T}) + B'_t (1 - e^{-\Delta T}) + A_t + C'_t$$

where B'_t and C'_t are given by equations (a) but with λ replaced by $(\Delta + \sigma\phi)$.