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Measurement Methods of Burnup and Heavy Element Isotopic Composition in Irradiated Power Reactor Fuels

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INTRODUCTION

The importance of the determination of burnup and heavy element isotopic composition can hardly be overestimated. These data are of prime importance in establishing the validity of the reactor constants and computer codes used. This in turn is an integral part of core design, reactor performance and core life. This, of course, is not unrelated to the economics of the problem. An error in the overprediction of the reactor core life could possibly result in severe economic loss to the reactor designer and manufacturer.

The term burnup has a rather broad meaning in that it includes both depletion of a fissionable species (U-235 depletion), and the energy produced by a fuel (number of fission events). The latter quantity, which is of interest here, can be expressed conveniently as atom percent fission which is defined as the number of fission events per 100 initial heavy element atoms. This is related to the energy released (i.e., MWD/T) by a constant. The methods of measurement of atom percent fission and heavy element isotopic composition are the subject of this paper.

MEASUREMENT METHODS

A. Heavy Element Isotopic Composition

The heavy element isotopic composition usually consists of the determination of the thorium and uranium isotopic content in thoria fuels, and that of uranium and plutonium in urania fuels. Thus far, the majority of the experience has been with urania fuels and so this system will be discussed, although other systems can be analyzed by the same technique. In the following discussion, the assumption is made that one starts with a dissolver solution which contains a representative portion of the fuel for analysis.

1. Mass Spectrometric Method

The basis of the method currently in use in this laboratory involves isotope dilution mass spectrometry. In essence this involves the addition of a known quantity of an isotope of the element preferably not found in the fuel, to a known volume of dissolver solution. Chemical separation and mass spectrometric analysis then yields the concentration per unit volume as well as the isotopic content. This technique is used for uranium and is also used for plutonium where the isotopic diluents (spikes) U-233 and Pu-242, respectively, are added to measured volumes of dissolver solution before any separations chemistry is performed. In the case of plutonium, the chemistry is preceded by a reduction-oxidation step to insure isotopic exchange.

The uranium and plutonium are isolated from the fission products by anion exchange techniques. The uranium is adsorbed onto Dowex-1 anion resin in high HCl concentration, washed free from plutonium and fission products with 5M HCl and eluted with dilute HCl. The uranium eluate is then evaporated to dryness, dissolved in 1 drop of dilute HCl and loaded onto a filament for mass spectrometric analysis. The quantity of uranium used is about 5×10^{-8} grams.

The plutonium is adsorbed onto Dowex-1 in 8M HNO_3. Washing with 3M HNO_3 removes nearly all fission products and uranium. The plutonium is eluted with 1M HCl for loading onto the mass spectrometer filament. The quantity of plutonium used is about 1×10^{-8} grams. Since most plutonium contains Pu-242, an unspiked plutonium sample must also be run.

Separation procedures other than ion exchange can be used as well. The main requirement is that they yield a product which is free of any extraneous mass. This is important especially for plutonium because the presence of extraneous mass results in poor and erratic emission characteristics in the mass spectrometric analysis.

Recently, the use of mixed spike has even eliminated the need for accurate pipetting. [1] The U-233 and Pu-242 spikes are mixed and calibrated by isotope dilution such that their ratio is accurately known. These are mixed with some dissolver solution and the separation ion exchange chemistry performed for the determination of the uranium and plutonium isotopic number density relative to U-238.

The accuracy of these methods depends upon the accuracy of the spiking isotope calibration and the mass spectrometric determination. The spikes (which are calibrated by isotope dilution) can be calibrated to an accuracy of 0.5 percent or better, depending on the number of sample filaments which are run and the number of scans per filament. The mass spectrometer filament-to-filament variation which is random, has been observed in this laboratory to be about ±0.5 percent. The precision which is obtained from the scan to scan variation is usually better than this for mass ratios within the range of 10 to 1. For calibration purposes, therefore, several filaments need to be run. A correction is made for mass discrimination bias (~ 0-2 percent) by comparison with known standards. The accuracy with which the isotopic content of a particular isotope can be determined is also dependent on the abundance of that isotope, where the error ranges from about ±0.5 percent for the higher abundance isotope to about 3 percent for isotopes having an abundance of 10^{-4}.

It should be mentioned that of the methods available for the determination of uranium content per unit volume, the mass spectrometric and the fluorimetric methods are the most sensitive. The normal sample size is about 0.01 micrograms for the mass spectrometer and 1 microgram for the fluorimeter. This is important to minimize the radiation problem. The fluorimetric method, however, has an accuracy of only about ±20 percent for a single observation. The coulometric method is often employed and although it is less sensitive (sample size about 100 micrograms), it has an accuracy comparable to that of the mass spectrometric method.

2. Partial Mass Spectrometric Methods

The isotopic composition of uranium and plutonium can be determined accurately only by a mass spectrometer. However, the plutonium content relative to that of the uranium can be determined in other ways. Since high sensitivity is required for plutonium determination

(to minimize radiation hazard) most methods except alpha counting are excluded. In practice, to obtain a Pu/U ratio, the uranium content and the plutonium content per unit volume are measured. As stated previously, the U-233 spike technique is the best way to determine the uranium content for small amounts of uranium. For plutonium, the Pu-242 spike technique is also the best, but Pu-242 has only recently become somewhat available. Therefore, two other methods are commonly used. One involves mixing a known quantity of Pu-236 spike with a measured volume of dissolver solution. The plutonium is then separated from the fuel and mounted on a plate. From an alpha pulse-height analysis of the mounted sample, the peak ratio of Pu-236 (5.763 Mev) to Pu-239, 240 (5.15 Mev) is obtained. A combination of these data with the atom ratio of Pu-239 to Pu-240 obtained mass spectrometrically, and the half-lives of Pu-236, Pu-239, and Pu-240, gives the plutonium content per unit volume. This is combined with the uranium content per unit volume to yield the Pu-U ratios. Because of the constants involved and the combination of independent data, an error of about ±4 percent is assigned.

The other method involves aliquoting the dissolver solution, without chemistry, onto a plate for alpha counting. This plate is gross alpha counted in a counter of known geometry (usually about 51 percent) and then alpha pulse-height analyzed. From the alpha spectrum, the contribution of each alpha emitter to the total alpha count is determined and subtracted, such that the fraction, $\frac{\text{Pu-239 + Pu-240 alpha counts}}{\text{Total alpha counts}}$, is obtained. Other alpha emitters commonly found in irradiated UO_2 fuel include Pu-238, Am-241, and Cm-242.

The contribution from these alpha emitters becomes appreciable as the exposure increases. Very roughly, for exposures of 5000 MWD/T, their contribution is of the order 50 percent. By making use of the plutonium mass spectrometric data and the half-lives of Pu-239 and Pu-240, the plutonium content per unit volume is determined. The error involved here is comparable to that for the Pu-236 spike method but gradually becomes worse as the contribution from other alpha emitters increases.

3. Atom Percent Fission From Heavy Element Isotopic Data

The atom percent fission can be obtained by making use of the heavy element isotopic data. For low enrichment UO_2 power reactors, the main sources of fission are from U-235 and Pu-239 with a small contribution from U-238 and Pu-241. The atom percent fission from U-235, F_5, can be calculated by any of the three equations:

$$F_5 = N_8^\circ \left[\left(R_{5/8}^\circ - R_{5/8} \right) - \left(R_{6/8} - R_{6/8}^\circ \right) \right] \quad (1a)$$

$$F_5 = N_8^\circ \left[\frac{R_{5/8}^\circ - R_{5/8}}{1 + \alpha_5} \right] \quad (1b)$$

where $R_{5/8}^\circ$ and $R_{6/8}^\circ$ are the ratios of U-235 and U-236 atoms to U-238 atoms, respectively, in the sample before irradiation; $R_{5/8}$ and $R_{6/8}$ are the ratios after irradiation; and N_8° is the atom percent of U-238 before irradiation. The quantity, α_5 is the U-235 capture to fission ratio. These equations assume that the number of U-238 atoms remains constant throughout the exposure. This is normally a fairly good assumption up to high exposures (where the loss is less than 1 percent) but a correction can be made by adding in the plutonium formed in the irradiation. For low exposure samples (less than about 3000 MWD/T) the change in the isotopic composition is not enough to measure exposure accurately in this way. Some increased sensitivity can be obtained if the ratio of U-236/U-235, before, $R_{6/5}^\circ$, and after, $R_{6/5}$, is used

$$F_5 = N_5^\circ \left[\frac{R_{6/5} - R_{6/5}^\circ}{R_{6/5} + \alpha_5 \left(1 + R_{6/5} \right)} \right] \quad (1c)$$

where N_5° is the initial atom percent U-235.

The atom percent fission of Pu-239, F_9, is normally computed from the equation:

$$F_9 = \frac{N_8^\circ}{\alpha_9} \left[R_{0/8} + R_{1/8} + R_{2/8} \left(\frac{1 + \alpha_1}{\alpha_1} \right) \right] \quad (2)$$

where $R_{0/8}$, $R_{1/8}$, and $R_{2/8}$ are the respective atom ratios of Pu-240, Pu-241, and Pu-242 to U-238 and α_9 and α_1 are the capture to fission ratios of Pu-239 and Pu-241, respectively. A decay correction for Pu-241 is usually included.

The Pu-241 atom percent fission F_1, is computed from the equation:

$$F_1 = \frac{N_8^\circ}{\alpha_1} \left[R_{2/8} \right] \quad (3)$$

Finally, the atom percent fission for U-238, F_8, cannot be calculated from the measured isotopic data, since there is no way to measure the change in U-238. This quantity is usually estimated by machine calculation and is in the range of 2-6 percent of the total fissions.

4. Accuracy of Atom Percent Fission Determination in Heavy Element Analysis

It is seen that this method of measuring atom percent fission is not applicable to low exposure fuel. At intermediate exposures (~ 0.5 atom percent fission) it is most accurate, because the change in the U-235 content is appreciable and the majority of the fission comes from this source. As the exposure increases, however, the proportion of fission due to Pu-239 and Pu-241 increases which also introduces a larger error due to uncertainties in the values used for α_9 and α_1 in equations (2) and (3). In order to illustrate this more clearly, a hypothetical UO_2 fuel of 1.9 percent enrichment was used. An IBM-650 program was used to calculate the isotopic composition of a fuel element in a water-moderated reactor after various exposure times. The program included self-shielding and spectrum shift effects, but did not treat spatial variation. In Table I, the atom percent fission for each species at 0.5, 1.0 and 1.5 percent fission is tabulated. The errors include realistic uncertainties in the experimental data and uncertainties in the constants used to obtain the various atom percent fission contributions.

The errors are more clearly shown in Table II.

TABLE I

ATOM PERCENT FISSION FOR IRRADIATED FUEL

Fission Source	0.5 Atom % Fission	1.0 Atom % Fission	1.5 Atom % Fission
F_5	0.42 ± 0.03	0.71 ± 0.02	0.95 ± 0.02
F_9	0.06 ± 0.02	0.22 ± 0.05	0.45 ± 0.11
F_8	0.03 ± 0.01	0.06 ± 0.02	0.09 ± 0.03
F_1	0.00 ± 0.00	0.02 ± 0.01	0.05 ± 0.03
F_T	0.51 ± 0.03	1.01 ± 0.07	1.54 ± 0.12

TABLE II

DISTRIBUTION OF ERRORS (%)

Fission Source	0.5 Atom % Fission (%)	1.0 Atom % Fission (%)	1.5 Atom % Fission (%)
F_5	± 4.4	± 2.0	± 1.2
F_9	± 4.0	± 6.0	± 7.0
F_8	± 2.0	± 2.0	± 2.0
F_1	± 0.3	± 0.9	± 1.6
F_T	± 6.3	± 6.7	± 7.6

It is seen that a determination of exposure from heavy element data requires a significant change in the isotopic content which also implies the need for accurate pre-irradiation data which is assumed in the above example. The reliance on α_9 and α_1 introduces increasing uncertainties at higher exposures. The big advantage of this method is that, in principle, the contribution to total fission from each fissioning species can be determined.

B. Radioactive Fission Product Method

The more widely used method for the determination of exposure is the use of a fission product. By use of this method, the atom percent fission is given by:

$$\text{Atom \% Fission} = \frac{N_{FP} \times 100}{N^{\circ}_{HE} \times Y_{eff}} \qquad (4)$$

where N_{FP} = number of fission product atoms of a selected nuclide

N°_{HE} = number of initial heavy element atoms

Y_{eff} = effective fission yield of the selected fission product nuclide

In principle, this quantity is not difficult to obtain in that the only measurements necessary are the determination of the heavy element and fission product concentration per unit volume of dissolver solution. These data combined with a fission yield give the atom percent fission.

1. Properties of Selected Fission Products and Their Determination

The determination of the heavy element content per unit volume has been previously discussed. The selection of a fission product and the accuracy of its determination are the more serious problems.

Among the radioisotopes Cs-137 is the most ideally suited and widely used fission product for measurement. Its half-life is long, 29.2 years [2] compared with an exposure time of, for example, 5 years. It has a low capture cross section of less than 2 barns, a simple and relatively well known decay scheme, a low formation cross section from mass 136 chain members, and a high fission yield.

The isotope Sr-90 is also used occasionally since its half-life is comparable with that of Cs-137. However, its constants, half-life and fission yields, are less well known than those of Cs-137.

For shorter exposures, fission product isotopes such as Ce-144 and Zr-95 with half-lives of 285 and 65 days, respectively, are used. There are fission product isotopes available having shorter half-lives (i.e., Ba-140, 12.8 days) but these are normally of no use in the case of power reactors which have fuel exposure of the order of years. The former four isotopes then, are the ones commonly used and, in fact, nearly exhaust the supply of available radioisotopes. The pertinent properties are summarized in Table III. [3]

The properties in Table III are used in the radiochemical determination of each of these nuclides. All the radiochemical procedures involve the addition of a known quantity of the same natural element (carrier) as the radioactive nuclide in milligram amounts to a measured volume of the dissolver solution. For Cs-137, the procedure involves precipitation of the cesium as the chloroplatinate or perchlorate after removal of other fission products by scavenging. The precipitate is weighed to correct for chemical losses and the 662 kev gamma ray is counted relative to that of a standard whose Cs-137 content is known. After decay corrections, these data yield the number of Cs-137 atoms per unit volume of solution.

The procedures for Sr-90 involve repeated iron hydroxide scavenges and precipitation of the strontium fraction as the oxalate. In order to avoid Sr-89 interference, the yttrium is separated from the aged and dissolved oxalate precipitate by hydroxide precipitation and is finally β^- counted as the oxalate.

TABLE III

PERTINENT NUCLEAR CONSTANTS OF SOME FISSION PRODUCT NUCLIDES

Fission Product Nuclide	Half-life	Decay Chain	Predominant Radiation (Used for Measurement)
Cs-137	29.2 y	I-137 (24.4 s) ↓ Xe-137 (3.9 m) ↓ Cs-137 (29.2 y) ↓ Ba-137 (stable)	0.662 Mev γ ray from Cs-137
Sr-90	~ 28 y	Kr-90 (32 s) ↓ Rb-90 (2.9 m) ↓ Sr-90 (~ 28 y) ↓ Y-90 (64.2 h) ↓ Zr-90 (stable)	2.26 Mev β^- from Y-90
Ce-144	285 d	Ce-144 (285d) ↓ Pr-144 (17.4 m) ↓ Nd-144 (5×10^{15} y)	2.98 Mev β^- from Pr-144
Zr-95	65 d	Y-95 (11 m) ↓ Zr-95 (65 d) ↓ Nb-95 (35 d) ↓ Mo-95 (stable)	0.723 and 0.760 Mev γ rays from Zr-95 and 0.768 Mev γ ray from Nb-95

The rare earth fraction of fission products, in which essentially all the activity is due to Ce-144, is separated from other activities by hydroxide precipitation and finally precipitated as the oxalate for β^- counting. The sample is counted through an aluminum absorber such that only the high energy Pr-144 β^- particles are counted. The count rate is converted to dpm by a counter geometry factor to yield the number of atoms of Ce-144 per unit volume.

The Zr-95 can be gamma counted from a direct mount of a measured volume of fuel solution. It can also be separated by barium fluorozirconate and hydroxide precipitations, and gamma counted finally as the hydroxide or mandelate.

2. Accuracy of the Fission Product Method

a. In Fission Product Determination

In the case of the fission product determination of atom percent fission there is no practical lower limit to measurement of an exposure and actual measurements have been made using 12.8d Ba-140 to measure an exposure of less than 0.1 MWD/T. As stated previously, the upper limit and length of exposure is a function of the individual fission products selected. The relatively short half-lives of Zr-95 and Ce-144 preclude their use as long exposure indicators. The fission product Cs-137 is useful up to an exposure where the Cs-134 contribution from Cs-133 neutron capture becomes appreciable in the gamma spectrum. It is estimated that the contribution of the Cs-134 amounts to about 50 percent of that due to Cs-137 when approximately 60 percent of the initial U-235 has fissioned. Thus, for an initial enrichment of 2 percent, this would be about 10^4 MWD/T. The isotope Sr-90 does not have this limitation but is subject to others (discussed in section c).

The isotope Cs-137 can be determined with the greater accuracy than other radioisotopes because more work has been done with this than other long-lived fission products. This is due in a large part to the accuracy with which the Cs-137 standard is known which is better than 1 percent.

Although the Zr-95 is also gamma counted in comparison with the Cs-137 standard, a correction must be made for the difference in the counting crystal photopeak efficiencies which increases the total error. In addition, because of its relatively short half-life, the exposure history must be taken into account fairly accurately to make the proper decay correction.

The Ce-144 and Sr-90 are both β^- counted. In general, the effective geometry for this type of counting is difficult to determine accurately, because normally a standard with the same β^- energy distribution as that of the sample is not available, and the geometry is somewhat dependent on the β^- energy which increases the uncertainty. This uncertainty can, of course, be decreased with calibration by means of 4π counting or beta-gamma coincidence counting. This, however, does not account for the variation of the absorption of β^- particles with the thickness of the precipitate

(chemical yield). In addition to the above disadvantages, the Sr-90 determination involves two chemical yield determinations which increases its uncertainty.

b. Migration of Fission Products Relative to Fuel

The measurement of fission product to uranium ratio assumes that the fission product distribution remains unaltered throughout the exposure of the fuel, i.e., there is no movement of the fission product atom relative to the heavy element atoms. It is known that this is a poor assumption for very volatile species, such as rare gases which migrate out of the fuel. It has also been found recently that other species which are volatile at the operating fuel temperatures move under the temperature gradient toward the cooler fuel regions. [4, 5, 6] Their rate of movement is, of course, a function of the operating fuel temperature. This is true of Cs-137 (the boiling point of cesium metal is 670°C) which has been found to undergo extensive radial migration to the cooler peripheral location on the fuel rods. Non-volatile species having volatile precursors of appreciable half-life are also subject to migration.

The gross movement of nonvolatile species, such as cerium or zirconium, has not been observed to any appreciable extent in fuels that have not undergone melting. [5] However, consideration should be given to the possibility of migration in sampling. For fuel rods where radial migration is possible, the samples for analysis consist of transverse cuts about 1/2 inch long.

c. Fission Yields

It was seen that fission yields must be used to convert the fission product-heavy element atom ratio to atom percent fission. Since fission yields are dependent on the fissioning species, the contribution to total fission from each fissioning species must be known in order to obtain an effective fission yield for each sample. Table IV shows the thermal neutron fission yields of the fission products discussed above for U-235 and Pu-239 and the fast fission yield for U-238 (fission neutron spectrum). [7]

These data indicate that the fission products least sensitive to the fission species are Cs-137 and Zr-95. The difference is on the order of 10 percent for U-235 and Pu-239. The sensitivity of the Sr-90 fission yield with fissioning species limits its usefulness unless accurate additional data are available. The sensitivity of Ce-144 with the fissioning species is intermediate between that of the Zr-95 and Sr-90.

TABLE IV

FISSION YIELD

Fission Product	Fission Yields (%) for		
	U-235	Pu-239	U-238
Cs-137	6.15	6.63	6.2
Sr-90	5.77	2.25	3.2
Ce-144	5.62	3.93	4.5
Zr-95	6.2	5.8	5.7

An estimate of the contribution to the total error from the heavy element and fission product determinations as well as the fission yield is tabulated in Table V. These errors are felt to be fairly realistic within the following assumptions and limitations:

(a) The period of exposure is within two half-lives of the shorter lived species when these are used for the burnup indicators;

(b) There is some reasonably accurate way to estimate an effective fission yield;

(c) The error in the constant to convert atom percent fission to MWD/T is not included;

(d) The error in the absolute fission yield for each species is small compared to that of the effective fission yield.

d. Illustrative Data

The data shown in Table VI were obtained from low enrichment UO_2 fuel irradiated in two boiling water reactors, Dresden and VBWR. These data illustrate the agreement typically obtained among the various fission product indicators discussed above, except Sr-90, and the mass spectrometric method. In those cases where the mass spectrometric method could be used, the contribution from each fissioning species

TABLE V

ESTIMATED ERROR ASSOCIATED WITH EACH FISSION PRODUCT (%)

Fission Product	Estimated Errors			
	Heavy Element	Fission Product	Fission Yield	Total
Cs-137	± 1	± 3	± 4	± 5
Sr-90	± 1	± 6	± 8	± 10
Ce-144	± 1	± 5	± 6	± 8
Zr-95	± 1	± 5	± 5	± 7

TABLE VI

COMPARISON OF EXPOSURES (MWD/T) OBTAINED BY DIFFERENT METHODS

Heavy Element MWD/T	Initial Enrichment (%)	Cs-137		Ce-144		Zr-95	
		MWD/T	F.Y. (%)	MWD/T	F.Y. (%)	MWD/T	F.Y. (%)
---	2.8	104	6.16	100	5.6	95	6.2
---	2.8	399	6.16	420	5.6	372	6.2
---	2.8	700	6.16	693	5.6	620	6.2
1091	1.5	1080	6.16	1100	5.6	---	---
1617	1.5	1495	6.17	1573	5.5	---	---
2209	1.5	2180	6.20	2215	5.4	---	---
3197	1.5	3130	6.23	3250	5.3	---	---

to the total fission was used to obtain an effective fission yield for the fission product burnup. These effective fission yields (F. Y.) are also tabulated. It is seen that the variation especially in the Cs-137 fission yield is relatively small (1 percent) while that of Ce-144 is already 6 percent for exposures up to only 3200 MWD/T.

The data are in reasonably good agreement within the error previously discussed.

DISCUSSION AND CONCLUSION

It was seen in the previous section that by mass spectrometric methods, the heavy element isotopic composition could be determined to an accuracy of ± 1 percent, which is the present day reasonable limit of accuracy. However, a calculation of atom percent fission from these data is limited in accuracy (to about 6 percent) by uncertainties in the pre-irradiation data and capture to fission ratios. Radioactive fission product burnups are also limited in accuracy by the errors associated with the determination of the fission product and the fission yield. Although a 6 percent error is not large for some purposes, greater accuracy than this is needed to determine reactor constants and reactor performance.

An obvious solution to the problem is to measure the fission product more accurately and use more accurate fission yields. The latter can be achieved by mass spectrometric analysis of a selected fission product, which suggests the use of a stable nuclide. In this case, many of the errors associated with the radioactive species such as uncertainties in the decay scheme, counter geometry, and chemical yield, as well as irradiation history are eliminated.

Although the number of possible stable fission product indicators is much greater than the number of radioactive indicators (limited to four as previously discussed), the ideal indicator must satisfy certain requirements which greatly reduce the number available. Some of the requirements are: (a) the nuclide should not undergo migration relative to the heavy element atoms; (b) the element of the chosen nuclide must have at least one shielded nuclide, preferably two, one of which is used for correction of natural contamination, and the other of which is used for the spiking isotope; (c) a low destruction cross section and a low formation cross section from adjacent mass chains is required; (d) the element of the nuclide should have good emission characteristics for high sensitivity in the mass spectrometric analysis; and (e) the fission yield must be well known and constant for all fissioning species and independent of neutron energy (from thermal to fission neutrons).

The last requirement of fission yields restricts the choice of fission product nuclides to two regions near mass 99 and mass 148 for U-235 and Pu-239 fission. This is illustrated in Figure 1 where the estimated fission yields of the more important fissioning species are plotted relative to the thermal neutron fission yield of U-235 as a function of the fission product mass number. [8]

The Vallecitos Atomic Laboratory, in conjunction with the Phillips Petroleum Company (NRTS, Idaho Falls) under the sponsorship of the AEC, has been engaged in research in the development of methods for the accurate determination of burnup. Since fission product neodymium satisfies most of the requirements listed above and spans the 148 mass region where the ratio of fission yields for U-235 and Pu-239 are close to 1.00, [Fig. 1] appreciable work has been done in this laboratory on neodymium. A carrier-free chemical separation procedure involving ion exchange and reverse phase chromatographic column techniques [9, 10] has been devised for the neodymium separations [11]. The isotope Nd-150 is used as the spike (analagous to U-233 for uranium) and the concentration determination can be made to ± 1 percent mass spectrometrically.

Neodymium, in particular Nd-148, appears very promising for the U-235-Pu-239 system with respect to the small fission yield variation [Fig. 1]. However, Nd-148 may not be the "ideal" nuclide for all systems.

The problem of fission yield uncertainty is also being investigated. The thermal fission yields from several fissionable nuclides are being determined in order to improve the accuracy in the regions of possible interest. The variation of fission yield with neutron spectrum will be studied by irradiation in different neutron spectra.

It is anticipated that this work will result in methods for the stable fission product determination of atom percent fission comparable in accuracy to those of the heavy element determinations. This then will provide a much more accurate evaluation of fuel performance and reactor physics parameters.

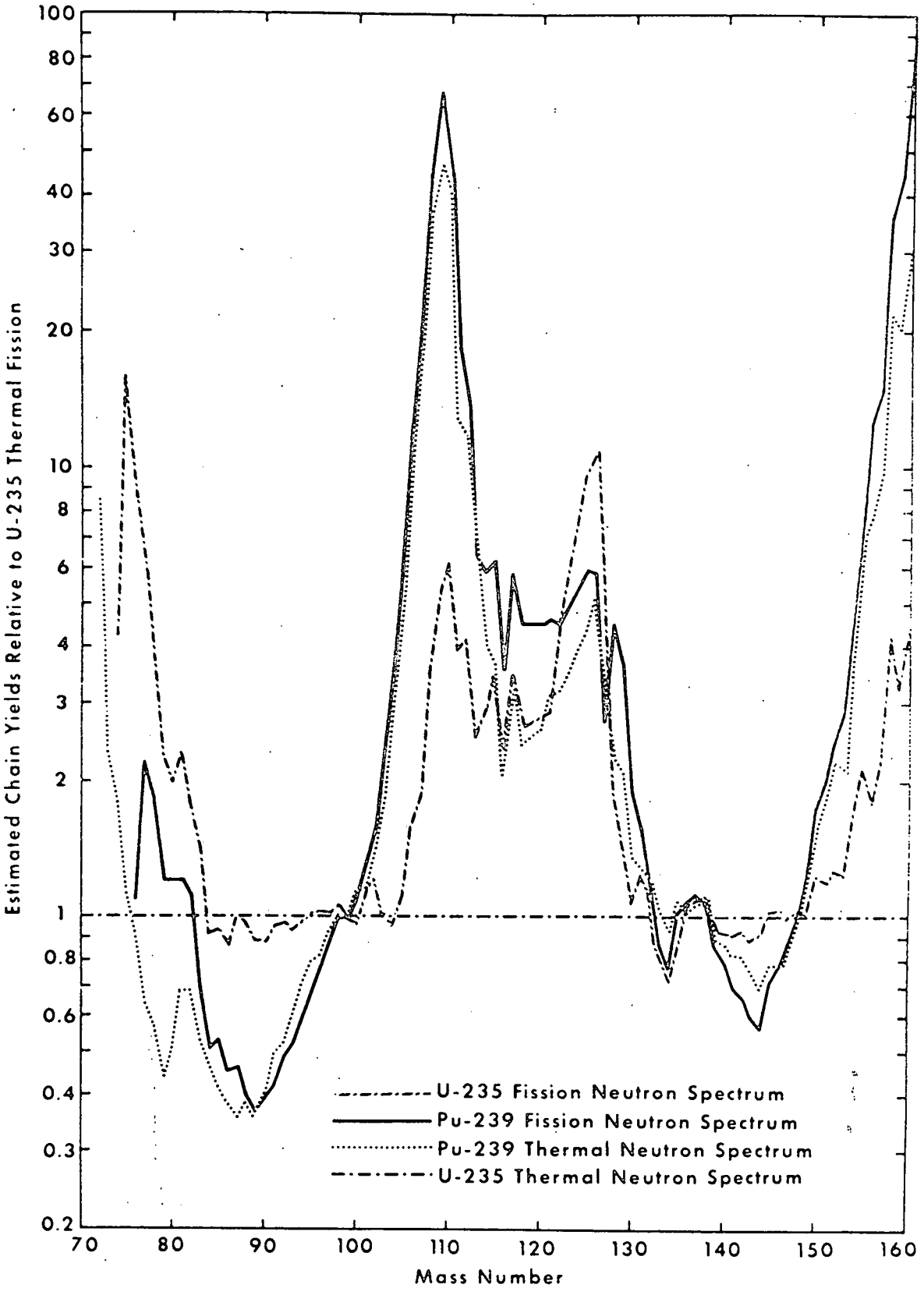

FIGURE 1 RELATIVE FISSION YIELDS FOR U–235 AND Pu–239.

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