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POSTIRRADIATION EXAMINATION OF THORIA-URANIA FUEL RODS

— Isotopic Analysis of the Fuel —

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POSTIRRADIATION EXAMINATION OF
THORIA-URANIA FUEL RODS

— Isotopic Analysis of the Fuel —

FINAL REPORT
Volume 2 of 3 Volumes

by

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1. INTRODUCTION

The objectives of this program were to determine the isotopic content of several heavy elements and fission products in irradiated thorium-uranium fuel and to perform metallurgical examinations on the fuel, stainless steel cladding, and Zircaloy fuel can. The specimens were taken from Core A of the Consolidated Edison Indian Point Reactor.

Thirteen fuel rods from six elements were selected for analysis. The rods were gamma-scanned and punctured for fission gas release. The results of the gamma scans were used to guide the selection of sampling positions for isotopic and metallurgical analysis.

The results of metallographic examinations on the thorium-uranium fuel, stainless steel cladding, and Zircaloy fuel can are reported in BAW-3809-6. The isotopic abundances of certain transactinium and fission product elements were determined and are reported herein. A complete list of the reports issued under this program is as follows:

BAW-3809-1, Progress Report No. 1, March - August 1967.

BAW-3809-2, Progress Report No. 2, September - November 1967.

BAW-3809-3, Progress Report No. 3, December 1967 - February 1968.

BAW-3809-4, Progress Report No. 4, March - May 1968.

BAW-3809-5, Progress Report No. 5, June - August 1968.

BAW-3809-6, Examination of Stainless Steel-Clad ThO_2 - UO_2 Fuel Rods and Zircaloy-2 Can After Operation for 442 Effective Full Power Days in the Indian Point Reactor.

BAW-3809-7, Postirradiation Examination of Thorium-Uranium Fuel Rods - Isotopic Analysis of the Fuel.

2. SUMMARY

Thirteen fuel rods from Core A of the Indian Point Reactor were analyzed for the isotopic content of selected actinide and fission product elements. Samples of the thoria-urania fuel were dissolved in 13 M nitric-0.05 M hydrofluoric acid, chemically separated and analyzed for the isotopic composition of thorium, uranium, neptunium, plutonium, americium-curium, neodymium, promethium, samarium and cesium. Neptunium was separated by a solvent extraction procedure in which Np(VI) was extracted into methyl isobutyl ketone-tetraheptylammonium chloride from a hydrochloric acid solution. A sequential separation procedure was developed for the separation of uranium, plutonium, neodymium, promethium, samarium and americium-curium. This procedure utilizes two extraction chromatography columns and two inorganic ion exchange columns. The separated samples were analyzed by mass, alpha and gamma spectrometry.

The rods showed a wide range of burnup and thus the isotopic compositions of the samples were varied. In one sample the ^{235}U was depleted to less than one-fourth of the initial amount, and in another sample the ^{235}U depletion was only about 15%. The samples that had experienced high burnups showed large amounts of ^{233}U produced from thorium. Plutonium-238 composed a major fraction of the plutonium in several fractions. This is attributed to the high ^{235}U enrichment.

The isotopic concentrations were calculated from reactor design and operating parameters. These results show fair agreement with the experimental values but consistently predict lower burnup than was obtained experimentally.

3. FUEL ROD SELECTION

The fuel rods examined under this program were taken from Core A of the Consolidated Edison Indian Point Reactor. The fuel consisted of thorium and highly enriched uranium (93% ^{235}U) oxides originally in pellet form.

3.1. Core Description

The Indian Point Reactor was described in detail during the design and construction phases,^{1,2} so only a synopsis is presented here. The reactor core contained 120 fuel elements, 137.50 inches long and 5.711 inches square. A cross section of the element is shown in Figure 3-1 and an overall view in Figure 3-2. The fuel element assembly consisted of a fuel bundle, a fuel can, an upper transition piece, a fuel element spring, and an upper nozzle and seal assembly.

The fuel was fabricated as high-density thoria-urania pellets of close dimensional tolerance, and was contained in modified type-304 stainless steel containing 200 to 250 ppm of boron. Each tube was closed at the ends with suitable end caps. The outside diameter of the fuel rod was 0.304 inch, and the cladding thickness was 0.0205 inch. Between the fuel pellets and the cladding was an initial gap (at room temperature), nominally 0.0015 inch thick, which was filled with helium gas. Each bundle (subassembly of tubes) contained 195 tubes which were spaced by ferrules (short, thin, tubular sections) in a 14×14 square array with one corner tube omitted. The ferrules were spaced axially 9 inches center-to-center in planes perpendicular to the longitudinal axis of the bundle. A row of smaller ferrules was spaced around the outside of the bundle at each plane of ferrules to support the outer row of tubes and to separate the fuel tubes from the can wall. The entire bundle was brazed as a unit.

The fuel element can—a Zircaloy-2 envelope surrounding the tube bundle—was fitted at each end with transition pieces. A spring in each

fuel element, locked in place by the top transition piece assembly, held down the fuel bundles.

Twenty-one movable control rods were mounted between the fuel element cans. Fixed control rods and flux depressor plates were also located between fuel elements. The core and the positions of fuel elements and control members are shown in Figure 3-3.

The core contained three zones with different fuel loadings. The arrangement of the three zones is shown in Figure 3-3, where Zone I is composed of elements 1 to 32, Zone II of elements 33 to 76, and Zone III of elements 77 to 120.

Internally, each fuel element comprised fuel rods of several different ^{235}U concentrations. The core contained fuel pellets of six different thorium-to-uranium ratios. Figure 3-4 shows the loading arrangement for each of the three different types of fuel elements. Table 3-1 shows the loading of the various fuel rods in the core. The uranium was enriched to 93% ^{235}U .

The average fuel pellet density was 9.25 g/cm^3 . The fuel pellet diameter was 0.260 inch. The pellets were stacked to an active fuel length of 98.5 inches at 68 F.

3.2. Core History and Rod Selection

An extensive gamma-scanning program was conducted at core midlife.³ One complete octant plus selected elements to show power symmetry were scanned. The elements for this program were taken from this same octant, which is shown in Figure 3-5. Elements adjacent to control rods that were not always out when the reactor was at power were not considered. Control rods 12, 13, and 17 were in this category, so elements 21, 22, 57, 58, 27, 28, and 65 were eliminated. From those remaining, elements 20, 61, 62, 66, 105, and 106 were chosen.

It was impractical to remove rods from the inner rows of the elements by the techniques used; and, when only one rod was to be taken, it was much simpler to remove a corner rod. Only type A and F rods were easily accessible, and F rods had no ^{235}U in the center portion. Thus, a type A rod was taken from each element. It was possible to obtain a rod of each type from a Zone III element by taking the two rods (A and B) adjacent to the index strip and five rods (F, C, D, D, E) from

the corner diagonally opposite. Since element 106 had a much lower burnup than element 105, the latter was chosen for obtaining one rod of each type. The fuel rod selection is summarized in Table 3-2.

The Zircaloy can was taken from element 61. This element was predicted to have the highest burnup of the six elements selected for this program; however, the analytical results did not verify this prediction.

The date of final operation for Core A was October 20, 1965; the core had operated for 442 effective full power days at 585 MWt. The core average burnup was estimated to be 14 GWd/tonne.

Table 3-1. Fuel Rod Loading

<u>Rod type</u>	<u>U/Th</u>	<u>U/(Th + U)</u>	<u>²³⁵U/(Th + U)</u>
A	0.032	0.031	0.029
B	0.047	0.045	0.041
C	0.063	0.059	0.055
D	0.088	0.081	0.075
E	0.11	0.10	0.093
F (Center 1/3 ThO ₂ only; outer two 1/3 type A)			

Table 3-2. Fuel Rod Selection

<u>Zone</u>	<u>Element No.</u>	<u>Rod type</u>
I	20	A
II	61	A
II	62	A
II	66	A
III	105	A, B, C, D, D, E, F
III	106	A, B

Figure 3-1. Fuel Element Cross Section

Fuel Rod Detail

Figure 3-2. Fuel Element

Figure 3-3. Core Hardware Position Designations

Figure 3-4. Fuel Element Loading Patterns

ZONE I

(ALL UNMARKED RODS ARE ENRICHMENT C)

ZONE II

(ALL UNMARKED RODS ARE ENRICHMENT D)

ZONE III

(ALL UNMARKED RODS ARE ENRICHMENT E)

LEGEND:

- A — LOWEST ENRICHMENT
- B — SECOND LOWEST ENRICHMENT
- C — THIRD " "
- D — FOURTH " "
- E — HIGHEST ENRICHMENT
- F — $1/3 \text{ THO}_2 + 2/3 \text{ ENRICHMENT A}$

Figure 3-5. Core Octant for Fuel Rod Selection

4. THEORETICAL ANALYSIS

Figure 3-3 is a diagram of the Consolidated Edison Core A showing numerical identifications for the fuel elements and other hardware. The fuel elements examined in this study are numbered 20, 61, 62, 66, 105, and 106. These elements were chosen so that they were representative of the different zones and were not adjacent to a moving control rod. Figure 4-1 gives the fuel element configuration and identifies the fuel pins that were selected for study. It can be seen that all the rods selected are "edge" rods, and that most of the rods are type A and share the same geometric location.

All fixed control rods were removed at midlife to compensate for fuel depletion. It was intended to insert Zircaloy-2 followers in place of the fixed rods; but in the process of handling, one of the followers was bent and could not be fitted into place. As a consequence, eight followers were left out so that the flux symmetry would not be disturbed. The omitted followers are numbered 1, 2, 7, 13, 12, 18, 23, and 24 (see Figure 3-3). Of the eight followers mentioned, only one was situated adjacent to an element being studied: follower number 18 by element number 62.

4.1. Calculational Methods Used for Asymmetry Test

Previous work on the Indian Point Reactor provided a starting point for this study. This work involved the calculation of induced power asymmetry within the reactor core at end-of-life conditions for comparison with the experimental study of the same phenomenon.⁴ Much of the data obtained even earlier, including critical experiment measurements,⁵ reactor operation, and calculated information,^{2,6} was also useful in the burnup calculations dealt with in this report. The calculational methods used for the asymmetry test are summarized in the following paragraphs. This study is reported in detail in BAW-1302.⁴

Analytic verification of the asymmetry experiment was attempted only at the reactor midplane. The effect of control rod motion through life on end-of-life conditions was calculated by the "window shade" method, in which control rods are simulated with a smeared poison. The actual rod position as a function of time, with the simulated rod position superimposed upon it, is shown in Figure 4-2. This information was obtained from weekly plant data tabulations.

The wide variation in power level from day to day during reactor operation made it impossible to obtain an exact representation in the calculation; instead, the operating history was represented by a series of power intervals, each at an average power level. This power interval representation is shown in Figure 4-3.

As a part of the window shade calculation, axial power shapes reflecting both the motion of the control rods and the axial depletion within the core were generated at many times in life. Therefore, the integral under the curve of the axial peaking factor for the central 20% of the core, plotted as a function of effective full power days (EFPD), represents the total EFPD experience by that portion of the core, assuming a radial peaking factor of one.

Radial peaking factors, which represent the ratio of the fuel element average power densities divided by the core average power density, were taken from measured and calculated radial power distributions. The radial peaking factors for the five basic control rod patterns used during operation are shown in Figure 4-4. The total number of EFPD for each fuel element is the summation, for all EFPD ranges, of the product of the element peaking factor and the portion of the axial peaking factor integral over the corresponding EFPD range. The EFPD in the calculative region are tabulated for each element in Table 4-1. Since eighth-core symmetry is assumed, only 17 elements are given; the locations of these elements are shown in Figure 4-4.

A point depletion calculation was run with the LIFE⁷ code for each of the six fuel rod types. The average burnup of all the rods within a single element was assumed to be the element's EFPD. This assumption could not be made in the individual rod burnup analysis, however, since the relative burnup between rods in an element had to be predicted.

Nevertheless, the element's EFPD predicted by the asymmetry calculations represented a very careful and accurate evaluation and thus served to provide a benchmark, at the level of the element, for other calculations showing both rod and element power distributions.

4.2. Calculational Methods for Pin Burnup Analysis

In order to determine the number of EFPD for an individual rod, it was necessary to apply power peaking factors to the element's EFPD. Two factors influence the power peaking of any individual rod: the power shape of the core (or global effect) and the power shape of the element. The global effect is due to the core geometry and the presence of three different enrichment zones and control rods. This effect corresponds to the core's gross radial power profile. Since the elements are loaded with rods of different enrichments, and since these rods are distributed in three different geometries, it is important to consider the elements' power shape. It is also particularly desirable to consider the softened spectrum experienced by "edge rods" because of their proximity to the water gap between elements. Although this effect was of no major consequence in the overall design, its importance was greatly emphasized in this study, since only edge rods were analyzed and not the large majority of interior rods which were correctly shown before in the asymmetry study.

4.2.1. Determination of Pin Peaking Factors

Two one-eighth core PDQs, which showed individual rods at average burnup conditions, were run as part of the study reported in BAW-1302. The geometric form used in these calculations maintains both element and rod identities in the calculation. One of these was an "A rods in" calculation and the other was a "C rods in" calculation. As noted previously in section 4.1, only two of the five control rod patterns are used in determining integrated element peaking factors. The remaining control rod patterns were calculated with less detailed PDQs. Close examination of the contribution of the remaining control rod patterns (numbers 2, 3, and 5), which are in effect for 40% of the core life span, indicated that their contribution was close to the combined integrated peaking factors for the elements of interest and therefore

would have a neutral effect. The then-controlling 60% of core life remaining was shared almost equally by the "A" and "C" control rod patterns. For this reason, a linear average of the "A" and "C" control rod PDQs was expected to adequately represent the true integrated value of element peaking. The results, shown in Table 4-2, indicate agreement to within 10%, and the one-eighth core PDQs were adopted as the calculational basis since their gross element power distribution was correct, and, in addition, they showed the pin power distribution within elements in the gross power distribution.

The pin power density-to-average core power density ratio was taken from the one-eighth core PDQs. Likewise, the average element power density-to-average core power density ratio was found. These power density ratios are given in Table 4-3 along with the ratio of pin power density to element power density.

Although a majority of the pins were shown, the one-eighth core PDQs did not show all pins in their proper location, nor did they properly show the effect of the softer spectrum experienced by edge pins. These exclusions were due to limitations in the capacity of the PDQ code. To correct the core PDQs, a detailed calculation was made for element 105 to determine the degree of error caused by the simplifications made in the core PDQs.

4.2.2. Detailed Study of Element 105

To determine the degree of error introduced by the simplified geometries used in the core PDQs, PDQ calculations of a single element were performed. Element 105 was chosen for these calculations since seven pins from this element were to be investigated. A PDQ calculation of element 105 as it appears at the top of Figure 4-5 was made first. A second calculation of element 105 correctly showed individual pins in their proper relation to followers and water gaps. The corrected geometry is shown at the bottom of Figure 4-5. Pins on the element's boundary were subjected to a softer spectrum because of their location between the "inside" pins and the water gap between elements. As a result, it was necessary to determine the correct spectrum to be used in the LIFE pin cell calculations so that the pin depletion could be performed correctly. A comparison of A-pin depletions using an "average"

core spectrum instead of the more thermal spectrum for the A pin indicated a 50% error in ^{233}U buildup.

The correct spectrum was obtained by iteration between LIFE⁷ and PDQ⁸ representations of each pin of interest in the element. The water number densities of the LIFE rod cell were modified, and the cross sections generated were used in PDQ; this process was repeated until the LIFE spectrum agreed with the spectrum produced by the subsequent PDQ. PDQ was the more meaningful spectrum indicator because it was performed in two dimensions over the entire element.

The correction factors listed in Table 4-3 can be written as ρ_A/P_A where ρ is the average rod power density (of the pin designated by the subscript) as calculated by the element PDQ, which has detailed geometry and corrected spectra, and P is the fuel rod power density (of the corresponding fuel rod location as designated by the subscript) as calculated by the simplified element PDQ, which is of the same form as that used in the core PDQ calculations. It should be noted here that the simplified geometry does not show any E-rod material in the location of the E rod being studied; this is also the case for the B-rod representation. The effect of multiplying the rod-to-element peaking numbers calculated by the core PDQs by ρ/P is that the former numbers are modified by an amount proportionate to the errors caused by the simplified geometry. The resultant rod-to-element peaking factors are given in Table 4-3 along with the associated rod EFPD.

The detailed study of element 105 was not repeated for other elements because of the magnitude of the task and the limited scope of this project. A method of approximating burnup in these elements relative to element 105 was derived. This move was prompted by the observation that pins of type A in different elements had nearly the same spectrum. A similar observation was made for the B rods. This spectral comparison is presented in Table 4-4. Since the rods of a given type had similar spectral environments, the only differences expected were in flux magnitude. The rod-to-average core peaking was taken as an indicator of relative flux level and was normalized for each rod to the corresponding rod type in element 105. This established the burnup relative to the A rod in element 105 for each rod, so that the rod burnup expressed as EFPD was taken to be the product of element 105

EFPD and the relative burnup, where relative burnup is defined as $P_{\text{rod}}/P_{105 \text{ rod}}$. These values are shown in Table 4-5.

4.2.3. Fuel Rod Depletions

After satisfactory agreement had been obtained between LIFE and PDQ spectra at time zero, a lifetime cell depletion was performed for each rod type. PDQ calculations were made at 300 days and 600 days so that the spectrum changes due to burnup could be observed and compared with the LIFE spectrum at those time steps. It was found that the LIFE spectrum did not vary greatly from that of PDQ over 600 days for all rods except the F rod which contained only thorium in the region of calculation.* The BOL and EOL spectra are compared in Tables 4-6 and 4-7. The final isotopic compositions of the well-behaved element 105 rods were then read from the LIFE depletion at time steps corresponding to the calculated rod EFPD. These final number densities appear in Table 4-8. Owing to the long period of time between the removal of these rods from the core and their disassembly for experimental analysis, the isotopes ^{233}Pa and ^{239}Np shown in the depletion results were decayed to ^{233}U and ^{239}Pu , respectively.

The F rod could not be depleted by the LIFE cell calculation because the true flux spectrum experienced by the F rod was due to the adjacent rods and not the F rod itself. In order to determine the correct burnup for this rod, the PDQ fluxes were normalized and used to generate one-group cross sections for the thorium chain isotopes. The F rod was depleted to 300 days using the LIFE depletion equations and the one-group cross sections that had been generated at time zero. At 300 days, the depleted number densities were used to run a LIFE (one time step) which was forced to the proper spectrum by inputting a modified hydrogen number density and then iterating with PDQ as previously described. When the flux spectra had converged, a new set of one-group cross sections was generated and the depletion was continued from 300 to 600 days. Since only pure ^{232}Th was present initially, the uranium chain was carried only through ^{236}U ; the higher uranium isotopes were negligible. The F-rod EOL number densities are listed in Table 4-8.

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* See Table 3-1 for description of F rod.

4.2.4. Determination of ^{238}Pu Concentration

The LIFE isotope chain stops at ^{236}U and continues with ^{238}U and ^{239}Pu . This approximation was due to the small amounts of the intervening isotopes generated and the lack of adequate cross-sectional data. It was of interest for this study to attempt to predict the ^{238}Pu buildup and take advantage of recent ENDF/B* cross-sectional data for ^{237}Np and ^{238}Pu . The position of ^{238}Pu in the decay chains for the ^{235}U and ^{238}U isotopes can be represented as follows:

The concentration of ^{238}Pu as a function of time can easily be found if certain simplifying assumptions are made: (1) ^{236}U goes directly to ^{237}Np with no losses or gains in the ^{237}U state; (2) ^{237}Np goes directly to ^{238}Pu with no losses in the ^{238}Np state; (3) the $(n, 2n)$ ^{239}Pu contribution to ^{238}Pu (much smaller than the (n, γ) reaction) can be ignored; and (4) the small amount of ^{234}U present initially and that accrued through buildup in the ^{232}Th chain do not make a significant contribution to the ^{235}U concentration and can be ignored. Given these assumptions, a simplified chain results:

* Evaluated Neutron Data File/B

These simplifications are quite justifiable in view of the half-lives of ^{237}U and ^{238}Np , which are very small relative to the times of interest (6.75 days and 2.1 days, respectively) versus times of hundreds of days, and the fact that ignoring the $(n, 2n)$ contribution from ^{239}Pu introduces less than 0.1% error in the ^{238}Pu concentration.

The Bateman equations are applicable here, starting with the ^{235}U isotope:

$$\frac{dN_{235}\text{U}}{dt} = N_{235}\text{U} a_{235}\text{U}.$$

$$\frac{dN_{236}\text{U}}{dt} = C_{235}\text{U} N_{235} - a_{236}\text{U} N_{236}\text{U}.$$

$$\frac{dN_n}{dt} = C_{n-1} N_{n-1} - a_n N_n$$

The time-dependent expression for the n th member of the decay chain can be expressed as

$$N_n(t) = K_1 e^{-a_1 t} + K_2 e^{-a_2 t} + \dots + K_n e^{-a_n t}$$

where

$$K_n = \frac{C_1 \cdot C_2 \cdot C_3 \dots C_{n-1}}{(a_1 - a_n)(a_2 - a_n) \dots (a_{n-1} - a_n)} N_1(o),$$

$$C_n = \text{production rate of the } n\text{th isotope} = \overline{\phi \sigma}_c,$$

$$a_n = \text{rate of loss of the } n\text{th isotope} = \overline{\phi \sigma}_a,$$

$$N_1(o) = \text{initial concentration of the parent isotope.}$$

This form of the solution assumes that all transitions are the result of a capture or fission event and that no decay occurs. This is quite valid since the assumption has already been made that the short-lived, radioactive isotopes may be ignored.

Since experimental data were available for the ^{238}Pu concentrations in the A and B rods, they were selected for the examination

of ^{23}Pu dependence on burnup. The A and B rods removed from the various elements covered exposures ranging from 200 to 994 EFPD. The four-group cross sections for ^{235}U and ^{236}U were flux weighted, using the LIFE 500-day A-rod and LIFE 500-day B-rod fluxes as effective averages for the full time span, and collapsed to one group. The 68-group cross sections for ^{237}Np and ^{238}Pu were obtained from a recent ENDF/B data file compilation, reduced to 68-group values, flux-weighted with the 68-group fluxes in the A-rod spectral calculation, and collapsed to one group. Table 4-9 gives the values of the effective flux and one-group cross sections. Also given are the concentrations of ^{235}U , ^{236}U , ^{237}Np , and ^{238}Pu as a function of effective full power days as calculated from the following equations:

$$N_{235\text{U}}(t) = N_{235\text{U}}(0) e^{-a_{235\text{U}}t},$$

$$N_{236\text{U}}(t) = N_{235\text{U}}(0)(e^{-a_{235\text{U}}t} - e^{-a_{236\text{U}}t})A,$$

$$N_{237\text{N}}(t) = N_{235\text{U}}(0)(Be^{-a_{235\text{U}}t} + Ce^{-a_{236\text{U}}t} + De^{-a_{237\text{N}}t}),$$

$$N_{238\text{P}}(t) = N_{235\text{U}}(0)(Ee^{-a_{235\text{U}}t} + Fe^{-a_{236\text{U}}t} + Ge^{-a_{237\text{N}}t} + He^{-a_{238\text{P}}t}),$$

where the constants A - H are as follows:

	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>	<u>E</u>	<u>F</u>	<u>G</u>	<u>H</u>
A-pins	-0.1972	0.0456	0.0308	-0.0764	0.0593	0.0138	-0.0563	-0.0168
B-pins	-0.2073	0.0473	0.0473	-0.0946	0.0492	0.0181	-0.0530	-0.0144

The results in terms of initial ^{235}U concentration of unity are illustrated in Figures 4-6 and 4-7. The one-group calculations of the ^{235}U and ^{236}U atomic densities agree with the LIFE depletion calculations to within 5% over the complete time span. Thus, the magnitude and time dependence of the ^{238}Pu concentration relative to the initial ^{235}U were judged to be on a relatively accurate basis for the A rods and the B rods. Therefore all ^{238}Pu atomic densities quoted in Table 4-8 were arrived at directly from the curves in Figures 4-6 and 4-7.

4.3. Theoretical Results

After having determined the EFPD for each rod being studied, the isotope number densities were taken from the LIFE rod depletions, F-rod depletions, and ^{238}Pu calculations. The EOL number densities for all rods are listed in Table 4-8. The percentage distribution of uranium isotopes is given in Table 4-10 to aid in direct comparison with experimental results. The plutonium isotopic distribution is presented in Table 4-11.

Group-one fluxes for all subject rods were normalized in the same manner as were the four-group fluxes in the F-rod study. From LIFE it was found that the fraction of group-one flux above one MeV was approximately 0.466 of the group-one flux, and this fraction of the normalized flux was used to calculate the nvt (>1 MeV) for each rod as listed in Table 4-12.

The capture-to-fission ratios (α) and fission-to-absorption ratios were calculated from LIFE data for each pin type studied in element 105. These ratios showed very little variation over 600 days and have, therefore, been compiled only at the midlife time of each rod. These parameters are presented in Table 4-13.

Table 4-1. Summary of Element Effective Full Power Days in Calculative Region

Element No.	Relative EFPD	Element No.	Relative EFPD
20	678	65	690
21	632	66	650
22	665	100	323
27	376	102	358
28	678	105	586
57	726	106	416
58	478	109	584
61	743	110	383
62	555		

Core average: 540 EFPD for core midplane section.

Table 4-2. Comparison of Element Peaking Factors, $P_{\text{element}}/\bar{P}_{\text{core}}$

Element	Individual PDQs (integrated)	1/8-core PDQs (average)
20	1.21	1.29
61	1.33	1.41
62	0.99	1.00
66	1.16	1.23
106	0.57	0.52
105	1.05	1.03

Table 4-3. Burnup Data for Element 105

Peaking	Control rod pattern	Fuel rod type				
		A	B	C	D	E
$P_{rod}/\bar{P}_{core}$	C	1.18	1.55	0.555	0.665	0.665 ^(a)
	A	1.36	1.80	0.635	0.763	0.763 ^(a)
	Average	1.27	1.68	0.595	0.714	0.714
$\bar{P}_{element}/P_{core}$	C	0.976	0.976	0.976	0.976	0.976
	A	1.086	1.086	1.086	1.086	1.086
$P_{rod}/\bar{P}_{element}$	C	1.21	1.51	0.569	0.681	0.649
	A	1.25	1.95	0.585	0.703	0.828
	Average	1.23	1.73	0.577	0.692	0.739
Correction factor		1.140	1.012	1.576	1.466	1.688
Corrected peaking		1.40	1.75	0.909	1.01	1.25
Rod EFPD ^(b)		795	994	516	574	708

(a) Value used since E is not shown in Core-PDQ
(see Figure 4-6).

(b) Rod EFPD = (Element EFPD) × (Corrected rod peaking)
= (568 EFPD) × (Corrected rod peaking).

Table 4-4. Neutron Spectra^(a)

<u>Rod</u>	<u>Control rod pattern</u>	<u>Group 1</u>	<u>Group 2</u>	<u>Group 3</u>	<u>Group 4</u>
20-A	C	0.4957	0.1573	0.1232	0.2239
	A	0.4979	0.1573	0.1227	0.2221
	Average	0.4968	0.1573	0.1230	0.2230
61-A	C	0.5132	0.1609	0.1226	0.2034
	A	0.5161	0.1609	0.1220	0.2009
	Average	0.5147	0.1609	0.1223	0.2022
62-A	C	0.5096	0.1602	0.1232	0.2070
	A	0.5130	0.1604	0.1227	0.2039
	Average	0.5113	0.1603	0.1230	0.2055
66-A	C	0.5155	0.1613	0.1227	0.2006
	A	0.5155	0.1608	0.1223	0.2013
	Average	0.5155	0.1611	0.1225	0.2010
105-A	C	0.5178	0.1602	0.1221	0.1989
	A	0.5196	0.1610	0.1217	0.1977
	Average	0.5187	0.1606	0.1219	0.1983
105-B	C	0.5791	0.1742	0.1207	0.1260
	A	0.5814	0.1742	0.1203	0.1241
	Average	0.5803	0.1742	0.1205	0.1251
106-A	C	0.4954	0.1585	0.1207	0.2254
	A	0.4977	0.1585	0.1203	0.2235
	Average	0.4966	0.1585	0.1205	0.2245
106-B	C	0.5448	0.1670	0.1158	0.1724
	A	0.5476	0.1668	0.1152	0.1704
	Average	0.5462	0.1669	0.1155	0.1714

(a) Group energy bounds:

<u>Group</u>	<u>Upper energy boundary</u>
1	10.0 MeV
2	9.12 keV
3	61.4 eV
4	0.414 eV

Table 4-5. Calculation of Rod Burnup

Peaking	Control rod pattern	20-A	61-A	62-A	66-A	106-A	106-B
$P_{\text{rod}}/P_{\text{core}}$	C	1.70	1.39	1.26	1.34	0.297	0.435
	A	1.26	1.61	1.50	1.50	0.337	0.501
	Average	1.48	1.50	1.38	1.42	0.317	0.468
P_{rod}/P_{105-A}	Average	1.17	1.18	1.09	1.12	0.250	
P_{rod}/P_{105-B}							0.264
Rod EFPD(a)		930	938	867	890	200	262

(a) For A pins:

$$\text{Rod EFPD} = (P_{\text{rod}}/P_{105-A}) \times (\text{Rod EFPD calculated for rod 105-A}).$$

For 106-B rod:

$$\text{Rod EFPD} = (P_{106-B}/P_{105-B}) \times (\text{Rod EFPD calculated for rod 105-B}).$$

Table 4-6. Calculated Neutron Spectra at Beginning of Core Life^(a)

<u>Rod</u>	<u>Case</u>	<u>Group 1</u>	<u>Group 2</u>	<u>Group 3</u>	<u>Group 4</u>
A	PDQ	0.5221	0.1598	0.1059	0.2122
	LIFE	0.5130	0.1506	0.1237	0.2077
B	PDQ	0.5526	0.1661	0.1011	0.1802
	LIFE	0.5468	0.1584	0.1265	0.1682
C	PDQ	0.5999	0.1803	0.1233	0.0966
	LIFE	0.6117	0.1778	0.1231	0.0874
D	PDQ	0.6145	0.1814	0.1204	0.0837
	LIFE	0.6153	0.1767	0.1236	0.0842
E	PDQ	0.6238	0.1820	0.1180	0.0762
	LIFE	0.6356	0.1761	0.1182	0.0700
F	PDQ	0.5851	0.1793	0.1257	0.1100
	LIFE	0.5934	0.1783	0.1154	0.1129

(a) Group energy bounds:

<u>Group</u>	<u>Upper energy boundary</u>
1	10.0 MeV
2	9.12 keV
3	61.4 eV
4	0.414 eV

Table 4-7. Calculated Neutron Spectra After 600 Effective Full Power Days^(a)

<u>Rod</u>	<u>Case</u>	<u>Group 1</u>	<u>Group 2</u>	<u>Group 3</u>	<u>Group 4</u>
A	PDQ	0.49656	0.15150	0.11682	0.23513
	LIFE	0.51156	0.14876	0.12015	0.21953
B	PDQ	0.52500	0.15725	0.11674	0.20100
	LIFE	0.53904	0.15645	0.12343	0.18108
C	PDQ	0.59889	0.18103	0.12225	0.09782
	LIFE	0.61088	0.17801	0.12065	0.09046
D	PDQ	0.61295	0.18221	0.11944	0.08541
	LIFE	0.61265	0.17672	0.12211	0.08852
E	PDQ	0.62168	0.18272	0.11718	0.07842
	LIFE	0.62448	0.17870	0.12057	0.07625
F	PDQ	0.58447	0.17992	0.12476	0.11084
	LIFE	0.61196	0.18301	0.11435	0.09069

(a) Group energy bounds:

<u>Group</u>	<u>Upper energy boundary</u>
1	10.0 MeV
2	9.12 keV
3	61.4 eV
4	0.414 eV

Table 4-8. Calculated Number Densities^(a) at End of Core Life
for Core Midplane Locations

Rod	²³² Th	²³³ U ^(b)	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U	²³⁸ Pu	²³⁹ Pu ^(b)	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu	¹⁴⁹ Sm
20-A	0.749 E-2	0.985 E-4	0.971 E-5	0.749 E-4	0.272 E-2	0.122 E-4	0.235 E-6	0.280 E-6	0.110 E-6	0.763 E-7	0.296 E-7	0.162 E-7
61-A	0.749 E-2	0.985 E-4	0.971 E-5	0.749 E-4	0.272 E-2	0.122 E-4	0.235 E-6	0.280 E-6	0.110 E-6	0.763 E-7	0.296 E-7	0.162 E-7
62-A	0.750 E-4	0.961 E-5	0.918 E-5	0.790 E-5	0.266 E-4	0.122 E-4	0.212 E-6	0.280 E-6	0.108 E-6	0.731 E-7	0.263 E-7	0.165 E-7
66-A	0.750 E-2	0.961 E-4	0.918 E-5	0.790 E-5	0.266 E-4	0.122 E-4	0.212 E-6	0.280 E-6	0.108 E-6	0.731 E-7	0.263 E-7	0.165 E-7
105-A	0.752 E-2	0.896 E-4	0.800 E-5	0.898 E-4	0.248 E-4	0.124 E-4	0.163 E-6	0.279 E-6	0.102 E-6	0.642 E-7	0.195 E-7	0.169 E-7
105-B	0.742 E-2	0.897 E-4	0.851 E-5	0.140 E-3	0.344 E-4	0.176 E-4	0.189 E-6	0.447 E-6	0.147 E-6	0.104 E-6	0.284 E-7	0.243 E-7
105-C	0.738 E-2	0.597 E-4	0.629 E-5	0.321 E-3	0.248 E-4	0.242 E-4	NC ^(c)	0.761 E-6	0.100 E-6	0.540 E-7	0.398 E-8	0.526 E-7
105-D	0.720 E-2	0.510 E-4	0.731 E-5	0.459 E-3	0.291 E-4	0.330 E-4	NC	0.904 E-6	0.111 E-6	0.527 E-7	0.327 E-8	0.721 E-7
105-E	0.704 E-2	0.538 E-4	0.882 E-5	0.587 E-3	0.371 E-4	0.411 E-4	NC	0.120 E-5	0.146 E-6	0.760 E-7	0.474 E-8	0.931 E-7
105-F	0.782 E-2	0.714 E-4	0.269 E-5	0.17 E-6	0.41 E-8	--	NC	NC	NC	NC	NC	NC
106-A	0.763 E-2	0.302 E-4	0.303 E-5	0.184 E-3	0.876 E-5	0.131 E-4	0.458 E-8	0.166 E-6	0.203 E-7	0.417 E-8	0.222 E-9	0.245 E-7
106-B	0.752 E-2	0.306 E-2	0.405 E-5	0.267 E-3	0.120 E-4	0.187 E-4	0.966 E-8	0.264 E-6	0.313 E-7	0.762 E-8	0.419 E-9	0.361 E-7

(a) Atomic densities in units of barn⁻¹-cm⁻¹.

(b) ²³³U and ²³⁹Pu corrected for decay of ²³³Pa and ²³⁹Np.

(c) NC = not calculated.

Table 4-9. Calculation of ^{238}Pu Atom Densities

A. Cross Sections — A Rod

Isotope	$\sigma_a(\text{b})$	$\sigma_c(\text{b})$	$\phi\sigma_a(\text{s}^{-1}) \times 10^{-8}$	$\phi\sigma_c(\text{s}^{-1}) \times 10^{-8}$
^{235}U	89.2	16.1	1.43	0.257
^{236}U	7.79	7.60	0.125	0.122
^{237}Np	56.4	55.9	0.904	0.896
^{238}Pu	132.0	107.0	2.12	1.72

B. Atomic Densities (No./barn-cm) — A Rod

Isotope	Time, EFPD					
	0	200	262	795	867	938
^{235}U	0.230 E-3	0.180 E-3	0.166 E-3	0.861 E-4	0.788 E-4	0.721 E-4
^{236}U	0.8 E-6	0.896 E-5	0.113 E-4	0.246 E-4	0.257 E-4	0.268 E-4
^{237}Np	0	0.927 E-7	0.153 E-6	0.985 E-6	0.112 E-5	0.125 E-5
^{238}Pu	0	0.458 E-8	0.966 E-8	0.163 E-6	0.192 E-6	0.235 E-6

C. Cross Sections — B Rod

Isotope	$\sigma_a(\text{b})$	$\sigma_c(\text{b})$	$\phi\sigma_a(\text{s}^{-1}) \times 10^{-8}$	$\phi\sigma_c(\text{s}^{-1}) \times 10^{-8}$
^{235}U	74.0	13.7	1.02	0.190
^{236}U	7.76	7.56	0.107	0.105
^{237}Np	40.9	40.2	0.565	0.555
^{238}Pu	113.0	90.6	1.56	1.25

D. Atomic Densities (No./barn-cm) — B Rod

Isotope	Time, EFPD					
	0	200	262	795	867	938
^{235}U	0.329 E-3	0.275 E-3	0.261 E-3	0.153 E-3	0.153 E-3	0.143 E-3
^{236}U	1.1 E-6	0.98 E-5	0.124 E-4	0.295 E-4	0.312 E-4	0.327 E-4
^{237}Np	0	0.88 E-7	0.147 E-6	0.105 E-5	0.121 E-5	0.137 E-5
^{238}Pu	0	0.26 E-8	0.56 E-8	0.113 E-6	0.140 E-6	0.169 E-6

Table 4-10. Calculated Uranium Composition (Atom Percent) for Core Midplane Locations

Rod	Composition				
	233	234	235	236	238
20-A	44.3	4.4	33.7	12.2	5.5
61-A	44.3	4.4	33.7	12.2	5.5
62-A	43.1	4.1	35.4	11.9	5.5
66-A	43.1	4.1	35.4	11.9	5.5
105-A	39.9	3.6	40.0	11.1	5.5
105-B	30.9	2.9	48.2	11.9	6.1
105-C	13.7	1.4	73.6	5.7	5.6
105-D	8.8	1.3	79.2	5.0	5.7
105-E	7.5	1.2	80.3	5.2	5.8
105-F	96.2	3.6	0.2	--	--
106-A	12.6	1.3	77.0	3.7	5.5
106-B	9.2	1.2	80.3	3.6	5.6

Table 4-11. Calculated Plutonium Composition (Atom Percent) for Core Midplane Locations

Rod	Composition				
	238	239	240	241	242
20-A	32.2	38.3	15.1	10.4	4.0
61-A	32.2	38.3	15.1	10.4	4.0
62-A	30.3	40.0	15.4	10.5	3.8
66-A	30.3	40.0	15.4	10.5	3.8
105-A	26.0	44.5	16.2	10.2	3.1
105-B	20.6	48.8	16.1	11.3	3.1
106-A	2.3	85.0	10.4	2.1	0.1
106-B	3.1	84.4	10.0	2.4	0.1

Table 4-12. Calculated Cladding Fluence

Rod	Normalized group 1 flux	Normalized group 1 flux above 1 MeV	Element EFPD, s	nvt above 1 MeV
20-A	10.2 E 13	0.456 E 14	0.568 E 8	0.259 E 22
61-A	9.35 E 13	0.417 E 14	0.624 E 8	0.260 E 22
62-A	8.69 E 13	0.388 E 14	0.466 E 8	0.181 E 22
66-A	9.34 E 13	0.417 E 14	0.545 E 8	0.227 E 22
105-A	8.05 E 13	0.359 E 14	0.491 E 8	0.176 E 22
105-B	8.28 E 13	0.369 E 14	0.491 E 8	0.181 E 22
105-C	8.83 E 13	0.394 E 14	0.491 E 8	0.193 E 22
105-D	8.69 E 13	0.388 E 14	0.491 E 8	0.190 E 22
105-E	8.53 E 13	0.381 E 14	0.491 E 8	0.187 E 22
105-F	8.39 E 13	0.374 E 14	0.491 E 8	0.184 E 22
106-A	9.15 E 13	0.408 E 14	0.265 E 8	0.108 E 22

Table 4-13. Midlife Values of Capture/Fission and Fission/Absorption

	Isotope	Fuel Rod Type					
		A	B	C	D	E	F
$\alpha = \frac{\text{Capture}}{\text{Fission}}$	²³³ U	0.115	0.120	0.134	0.135	0.138	0.127
	²³⁵ U	0.221	0.230	0.266	0.269	0.277	0.251
$\frac{\text{Fission}}{\text{Absorption}}$	²³² Th	0.0132	0.0151	0.0188	0.0199	0.0211	0.0140
	²³⁴ U	0.0139	0.0157	0.0203	0.0209	0.0222	0.0167
	²³⁶ U	0.0226	0.0237	0.0242	0.0261	0.0274	0.0197

Figure 4-1. Location of Selected Fuel Rods

Zone I - Element 20
(All Unmarked Rods are Enrichment C)

Zone II - Element 61
(All Unmarked Rods are Enrichment D)

Zone II - Element 62
(All Unmarked Rods are Enrichment D)

Zone II - Element 66
(All Unmarked Rods are Enrichment D)

LEGEND:
A - LOWEST ENRICHMENT
B - SECOND LOWEST ENRICHMENT
C - THIRD " "
D - FOURTH " "
E - HIGHEST ENRICHMENT
F - $1/3\text{THO}_2 + 2/3\text{ ENRICHMENT A}$

Zone III - Element 105
(All Unmarked Rods are Enrichment E)

Zone III - Element 106
(All Unmarked Rods are Enrichment E)

Figure 4-2. Control Rod Position History, Actual and Simulated

Figure 4-3. Simplified Reactor Operating History

Figure 4-4. Fuel Element Peaking Factors for Five Basic Control Rod Patterns

0.80 20 1.47 1.44 1.50 1.18	0.67 21 1.39 1.27 1.43 1.19	0.94 22 1.56 1.13 1.26 1.22	1.62 57 1.59 0.88 0.86 1.32	0.96 58 0.77 0.71 0.59 1.12	0.59 100 0.45 0.65 0.51 0.69
	0.91 27 1.44 0.84 1.40 1.18	1.19 28 1.27 0.83 1.36 1.21	1.44 61 1.30 1.28 1.27 1.28	1.06 62 0.76 1.13 0.95 1.04	0.74 102 0.53 0.73 0.57 0.62
		1.12 65 1.06 1.00 1.52 1.30	1.12 66 0.89 1.38 1.32 1.14	1.22 105 0.95 1.21 0.98 0.88	0.72 106 0.52 0.65 0.48 0.44
			1.09 109 0.84 1.32 1.12 0.92	0.77 110 0.64 0.85 0.66 0.55	

Fuel Element No.

x.xx zz
x.xx
x.xx
x.xx
x.xx

1. Group A + fixed shims.
2. Fixed shims only.
3. Groups B and C.
4. Group C.
5. No rods.

Figure 4-5. PDQ Element Geometries

Artificial element geometry
used to represent zone 3
elements in core PDQ.

Letters indicate pin types.

Detailed element geometry
showing exact location of
all pins.

This geometry used in cor-
rection study of element 105.

Primed letters indicate pins
that were shown with modified
water densities.

Figure 4-6. ^{235}U Neutron Capture Chain for A Rod

Figure 4-7. ^{235}U Neutron Capture Chain for B Rod

5. ISOTOPIC ANALYSIS OF THORIA-URANIA FUEL

5.1. Dissolution Studies

Complete dissolution of the selected thoria and thoria-urania fuel specimens was essential to the success of this investigation. Because refractory oxides of this type frequently resist the usual acid dissolution techniques, it was first necessary to determine whether satisfactory dissolution could be obtained.

In two recent studies, the effects of many variables on the rate of thoria dissolution were surveyed.^{9,10} The results of both indicate that 13 M nitric-0.05 M hydrofluoric acid approaches the optimum dissolution rate. However, these reports and others on the same subject disagree on how the dissolution rate is affected by (1) the burnup level of irradiated thoria fuel, and (2) the presence of other cations that form strong fluoride complexes. Since there was not enough data to allow the behavior of unirradiated thoria to be extrapolated to that of irradiated thoria at varying burnup levels, a two-part approach to the dissolution problem was taken. First, the adequacy of the selected solvent was demonstrated with unirradiated thoria and thoria-urania fuels. Next, a more drastic alternate technique was devised to dissolve any irradiated fuel specimens that might prove to be much less reactive.

5.1.1. Dissolution of Unirradiated Thoria-Urania Fuel

The dissolution of unirradiated thoria-3% urania pellets was evaluated with 13 M HNO₃-0.05 M HF under reflux conditions. Complete dissolution of 1.5-gram specimens was obtained in 6 to 8 hours using 50 ml of acid. Pure thoria pellet specimens required more time but were usually completely dissolved within 12 hours. A small amount of flocculent white residue was observed after these dissolutions. Because this precipitate seemed to increase rather than diminish when boric acid was added (to complex the residual fluoride), it was tentatively

identified as silica etched from the glass flask. The residue from one thorium-uranium solution was filtered, weighed, and counted. The 25 mg of residue contained less than $10^{-3}\%$ of the total solution activity; this result tends to verify that it was silica rather than undissolved fuel. (In later work, where dissolution times were about 1 hour, little or no residue was observed.)

The second dissolution technique involves a variation of the sealed-tube technique of Metz and Waterbury.¹¹ Refractory oxides are dissolved at an increased temperature made possible by a completely sealed system. Our modification of this technique replaces the one-piece sealed tube with a two-piece Pyrex dissolving vessel sealed with a Teflon O-ring. The glass parts are commercially available O-ring joints that have been sealed to the appropriate size; an outer stainless steel vessel contains water to provide a compensating external pressure. The water vapor pressure, registered on a pressure gage, may be converted to the internal temperature. In practice, a pressure of 50 to 60 psi, corresponding to about 150 C, is maintained.

When the high-temperature dissolver was evaluated with unirradiated thorium and thorium-uranium fuels, it was found that it required two or three times as long as the reflux system for complete dissolution. The poor performance was attributed to the lack of mixing in this non-boiling system. The addition of a magnetic stirring bar, which was operated during heating by an external magnetic stirrer, remedied the problem, and complete dissolution was then obtained within 1 hour.

Following the dissolution of irradiated fuel specimens reported in section 5.1.2, studies were initiated using a lower fluoride concentration. Reduction of the fluoride concentration to 0.025 M did not appreciably alter the dissolution times, but it did prevent any formation of silica.

5.1.2. Dissolution of Irradiated Thorium-Uranium Fuel

The results of the studies described in section 5.1.1 indicated that successful dissolution might be obtained in a reasonable time by using 13 M HNO_3 -0.05 M HF under reflux. The first sample, weighing approximately 3 grams, was still in the cladding; it was completely dissolved in 8 to 12 hours. The exact time of dissolution could not be

determined because of the deep coloration that resulted from dissolution of the cladding. Subsequent samples, each weighing approximately 0.8 gram, were declad. Considerable fragmentation of the fuel occurred in the decladding operation. All of the declad samples were dissolved in 13 M HNO₃-0.05 M HF, and in all cases complete dissolution was obtained in less than 1 hour.

5.2. Separation Procedures

5.2.1. Uranium, Plutonium, Americium-Curium, Neodymium, Promethium and Samarium

A four-column procedure for uranium, plutonium, americium-curium, neodymium, promethium, and samarium, which employs two extraction columns and two inorganic exchange columns in the sequential separation of these elements, has been devised. The first extraction column, di-(2-ethylhexyl)phosphoric acid (D2EHP) in diisopropylbenzene on a Chromosorb support, retains thorium, uranium, plutonium, and zirconium from 1N nitric acid. Subsequently, uranium and plutonium are sequentially eluted with 6N hydrochloric acid and 6N hydrochloric acid containing iodide. The nitric acid effluent from the first column flows directly onto a column of lead dioxide-lead iodate, which removes unwanted cerium and ruthenium. The second-column effluent passes through a column of potassium cobaltoferrocyanide (KCFC), which quantitatively retains cesium. The third-column effluent is buffered to ensure complete retention of cerium-free lanthanides and trivalent actinides on the final D2EHP extraction column. The americium-curium fraction is eluted with 1 M glycolic acid-0.016 M diethylenetriaminepentaacetic acid (DTPA), after which neodymium, promethium and samarium are eluted successively with increasing concentrations of dilute nitric acid. The separation scheme is outlined in Figure 5-1.

The procedure described is based on the versatility of D2EHP as an extractant and some recent advances in inorganic ion exchangers. The high distribution values of trivalent and tetravalent cations into D2EHP are well known.^{12, 13} (See Figure 5-2.) The initial column in the procedure depends on the acid-dependent selectivity of D2EHP to retain thorium, plutonium(IV), and zirconium, the most strongly held elements, from 1N nitric acid. Hexavalent uranium also shows a high

distribution from even strong nitric acid although the uranyl ion behaves more like a typical divalent cation from most other acids.¹⁴ Nemodruk and Gluhova¹⁵ have postulated that uranyl extraction is governed not by solvation, but rather by ion exchange of a quadruply charged UO^{+4} ion. Although the mechanism has not been investigated as a part of this project, neutral uranyl nitrate extracts well into many oxygenated solvents and may be partly responsible for the observed behavior. Regardless of the mechanism, uranium is quantitatively retained from 1N nitric acid on the first column and subsequently removed with 6N hydrochloric acid. A second pass through this column with 6N hydrochloric acid containing iodide reduces tetravalent plutonium to the nonretained trivalent state, which then elutes. Thorium and zirconium, exhibiting only tetravalence in solution, remain on the column and are discarded.

Some cross contamination between the uranium and plutonium has been noted. Generally this is of no consequence, but in this work both ^{238}Pu and ^{238}U were determined mass spectrometrically. The possible isobaric interference was eliminated by performing an additional anion exchange separation. The plutonium fraction in 8 M HNO_3 was loaded onto a column of Dowex 1-X2 (200 - 400 mesh nitrate form). Any uranium contamination was removed with 3 M HNO_3 , after which plutonium was eluted with 0.6 M HNO_3 .

The second column serves to remove unwanted cerium and ruthenium activity. Besides contributing a large fraction of the total activity in nuclear fuels of reprocessing age, cerium is a potential mass interference in the analysis of neodymium because both have isotopes of mass 142. Among materials having useful ion exchange properties, iodate and periodate salts are particularly interesting because they have sufficient potential to oxidize many cations. An example is the reported use of silver paraperiodate to retain selected elements from nitric acid solutions of fission products.¹⁶ Silver paraperiodate was not satisfactory for this application for two reasons: ruthenium retention is low from 1N nitric acid, and the salt is sufficiently soluble at this acidity to contribute residue which interferes at a later point in the procedure. Ruthenium retention was improved with either silver iodate or lead iodate; however, neither retained much cerium. The addition of lead dioxide to either of these provided the potential necessary to oxidize cerium to the tightly

held tetravalent state. The other elements normally expected to form insoluble iodates previously were retained on the first column. The choice of lead dioxide-lead iodate over lead dioxide-silver iodate was based on higher ruthenium retention.

Potassium cobaltoferrocyanide (KCFC), as proposed by Prout¹⁷ and Boni,¹⁸ retains cesium on the third column. The frequently encountered problem of small amounts of elements other than cesium being retained does not arise in the present application since these elements have been removed on previous columns. Thus, the KCFC column is specific and quantitative for cesium removal, and the separated ¹³⁷Cs may be counted without additional purification.

The final column is a longer version of the initial extraction column. The lanthanides and trivalent actinides, while not retained from 1 N nitric acid, may be completely extracted into D2EHP from 1 M glycolic acid at a pH of 2.5 to 3.5.¹⁹ Therefore, the effluent of the third column is adjusted to this pH range with an ammonium glycolate buffer* to ensure complete retention of the lanthanides and trivalent lanthanides on the final column. The buffering ability of the ammonium glycolate solution is shown in Table 5-1.

Di(2-ethylhexyl)phosphoric acid also has application for separating the lanthanide and actinide groups. Weaver and Kappelmann¹⁹ have reported a solvent extraction process for recovering large amounts of the transplutonium elements, in which large group separations from the lanthanides were obtained. A small amount of diethylenetriamine-pentaacetic acid (DTPA) was reported to preferentially complex the actinides in the aqueous phase while the lanthanides extract into the organophosphate. In the proposed separation, the aqueous phase is 1 M glycolic acid-0.016 M DTPA. The pronounced effect of this small amount of DTPA is shown in Figure 5-3. After the final extraction column is washed free of trivalent actinides with the glycolic acid-DTPA, the lanthanides can be eluted with dilute nitric acid. Neodymium is eluted first with 1:100 HNO₃ and then promethium with 1:80 HNO₃ followed by samarium with 1:50 HNO₃.

- - - - -
* 5 M ammonium glycolate adjusted to pH 5 with ammonium hydroxide.

The trivalent actinides, while free of most other elements, are separated in a solution that contains large amounts of organic salts. If this is undesirable, the DPTA complex may be weakened by acidifying the solution to pH 1.5 and passing it through a cation exchange column. The trivalent actinides will now be retained, the objectionable organic may be removed with water washing, and then the actinides can be eluted in 5N nitric acid.

The distribution of activity from a typical fuel sample is shown in Figures 5-4 through 5-8.

5.2.2. Neptunium

Neptunium-237 is produced during the irradiation of uranium-containing fuels by the fast-neutron-induced (n, 2n) reaction on ^{238}U , and by the (n, γ) reaction on ^{236}U followed by beta decay of the 6.75-day ^{237}U product. Because the ^{236}U concentration initially may be lower than the ^{236}U produced by (n, γ) on ^{235}U , production by two successive (n, γ) reactions on ^{235}U may be the major production mode for ^{237}Np in highly enriched fuels. All of these processes are of sufficiently low cross section to make ^{237}Np a minor constituent of the irradiated fuel. The high concentration of major fuel constituents relative to neptunium imposes stringent decontamination requirements on the neptunium separation procedure.

5.2.2.1. Ion Exchange

Thorium is known to be strongly retained on cation exchange resin from dilute nitric acid.²⁰ Zolotov and Nishanov²¹ report the separation of neptunium from uranium and plutonium with 1N nitric acid on KU-2 cationite. These authors claim complete reduction of Np(VI) to Np(V) by the resin. Pentavalent neptunium is less strongly retained than hexavalent or tetravalent actinides and is therefore the first eluted from a mixture.

Since KU-2 resin is not commercially available, a study of neptunium-uranium separation was undertaken using Dowex 50-X4 with 1N nitric acid as the eluant. The variables were resin mesh size and column height. The results are shown in Figure 5-9. Uranium-235 and ^{239}Np were used as tracers, and the gamma activity in 1-ml

fractions was measured to determine the elution curves. Gamma spectra indicated whether the activity was due to ^{235}U or ^{239}Np . Neptunium-239 had been prepared by irradiating depleted uranium in the BAWTR and separating ^{239}Np by the same cation exchange procedure being tested. Gamma spectra taken after the separation showed no impurities.

A 15-cm column of Dowex 50-X4 (100 - 200 mesh) gave adequate separation in a reasonable time and was selected to test the reproducibility and completeness of neptunium recovery. Five columns of this type were loaded with replicate samples of ^{239}Np in a simulated thorium-uranium dissolver fuel solution. Three-ml fractions of the eluate were collected. The ^{239}Np content of each fraction, determined by gamma counting, was compared to an aliquot identical to that loaded onto each column. The results are shown in Table 5-2.

The low recovery of ^{239}Np , ranging from 94 to 98%, was thought to be due to the presence of some tetravalent neptunium, which would be retained strongly by the resin. To eliminate this possibility, excess potassium permanganate was added to the nitric acid solution of neptunium tracer. A portion of this hexavalent neptunium was then made 0.1 M with sodium nitrite and heated to form pentavalent neptunium.²¹ The recovery of neptunium in six replicate cation exchange separations with permanganate and permanganate-nitrite treatment is shown in Table 5-3.

Recovery of neptunium seemed somewhat more consistent with the combined permanganate-nitrite pretreatment; however, it was still considerably less than quantitative recovery. Nelson and Michelson²² have studied the cation exchange behavior of neptunium in hydrobromic acid systems and found that neptunium is reduced to (IV) on the resin, even in the presence of bromine. Neptunium(IV) is undoubtedly stabilized by the high affinity of the resin for tetravalent cations. It is likely that a portion of the neptunium could be reduced to (IV) even in 1N nitric acid, resulting in the observed low recovery.

It is worth emphasizing that cation exchange is satisfactory for those applications where radiochemically pure neptunium is more important than quantitative recovery. However, for this work another approach was desirable.

5.2.2.2. Solvent Extraction

Of the more versatile solvent extraction systems for the actinides are those of Maeck,^{23,24,25} which involve tetraalkylammonium complexes of these elements. The procedure for neptunium²⁴ is based on extraction of the tetrapropylammonium nitrate neptunium(VI) ion association complex into methyl isobutyl ketone (MIBK) from acid-deficient aluminum nitrate salting solution. Tetravalent neptunium is then back-extracted into a ferrous chloride-hydroxylamine reducing solution and reextracted into thenoyltrifluoroacetone (TTA)-xylene. Uranium, which accompanies neptunium in the first extraction, neither back-extracts into the aqueous strip solution nor reextracts into TTA and is thus separated. However, this procedure cannot tolerate large amounts of thorium, which extracts well even from unsalted nitric acid.²⁶

In a comprehensive study of 57 metal-tetraalkylammonium complexes from various acids, Maeck²⁶ reported little or no extraction of thorium from hydrochloric acid. Although little neptunium(VI) extracted from this medium as the tetrapropylammonium or tetrabutylammonium complex, nearly 75% extraction was reported with the tetrahexylammonium salt. It seemed likely that further lengthening of the alkyl chain would enhance neptunium extraction without significantly increasing the extraction of thorium from a hydrochloric acid system. Solutions of tetraheptylammonium chloride (Eastman Organic Chemicals) were prepared in MIBK and nitropropane for evaluation. A relatively strong oxidant is required to ensure that all of the neptunium is in the hexavalent state as required for extraction. Some of the common oxidants, such as permanganate and argentic oxide, are not useful in a hydrochloric acid system. Three oxidants that seemed compatible with chloride were evaluated. With cerium(IV), added as ceric ammonium nitrate, neptunium extraction did not exceed 90%. When the sample was fumed with perchloric acid, before adding the hydrochloric acid, neptunium extraction remained below 12%. Chlorate ion, added as potassium chlorate, proved to be a satisfactory oxidant. Extraction behavior of neptunium with chlorate oxidation, determined by measuring the ²³⁹Np gamma activity in each phase, is shown in Table 5-4.

Neptunium extraction was found to exceed 99.5% from all of these systems at 5N or greater hydrochloric acid concentration. Higher than 6N HCl was not studied with MIBK because of high mutual solubility of the two phases. Although neptunium is more extractable in nitropropane, it is also more difficult to back-extract. The recovery of neptunium in a 10-minute contact of the nitropropane phase with the reducing-strip solution was only about 95%. With MIBK as the diluent, back-extraction exceeded 99%. Therefore, MIBK was selected as the diluent.

Decontamination from extraneous activity was evaluated by processing mixed fission products in a simulated thorium-uranium dissolver fuel solution.* Most of the ruthenium and about 10% of the zirconium were extracted from 5N hydrochloric acid. Repeated contacts with 5N hydrochloric acid removed very little of this activity; however, essentially all of the zirconium activity back-extracted into the reducing-strip solution.

Scrubbing the organic phase with acid-deficient aluminum chloride,** analogous to the aluminum nitrate used in nitrate systems,²⁴ successfully removed the zirconium activity, leaving only ruthenium as shown by gamma spectral analysis. A portion of the ruthenium does back-extract into the reducing-strip solution*** with neptunium (Figure 5-10), but is of little consequence since ruthenium volatilizes during the preparation of neptunium counting sources.

Because of the improved decontamination from fission products with an aluminum chloride scrub, it seemed reasonable that extracting initially from aluminum chloride would result in even

- - - - -
* A simulated thorium-uranium dissolver solution was prepared by adding thorium, uranium, HF, and H_3BO_3 to a uranium-fission product solution. The resultant solution was 1 M HNO_3 , 0.02 M HF, 0.09 M H_3BO_3 and contained 10 mg/ml of thorium and 1 mg/ml of uranium in addition to mixed fission products. An aliquot of this solution was evaporated to dryness and taken up to the original volume in 5N HCl.

** 2.8 M $AlCl_3$, 2.0 M NH_4OH .

*** 0.25 M $FeCl_2$ - 0.5 M $NH_2OH \cdot HCl$ - 1.0 M HCl.

better decontamination. This indeed was the case. However, extraction of neptunium was always low in aluminum chloride, presumably because of incomplete oxidation to Np(VI). The initial extraction from 5 N hydrochloric acid was therefore retained.

Thorium decontamination was evaluated by adding ^{234}Th tracer to the simulated thoria-urania dissolver fuel solution. The ^{234}Th was obtained by scavenging with iron carrier from a uranium carbonate solution. The iron and small amounts of uranium that were occluded were then retained on Dowex 1-X8 from 9 N hydrochloric acid, while carrier-free ^{234}Th was eluted.

Thorium decontamination was found to exceed 99.8% per contact from either 5 N hydrochloric acid or acid-deficient aluminum chloride salting solution. Since the proposed separation procedure combines one contact with each of these media, the overall thorium decontamination factor is $(0.998/0.002)^2$ or 2×10^5 .

The overall recovery of neptunium in this procedure is shown in Table 5-5. A very consistent recovery of 98.9% is seen, and the largest loss is encountered in incomplete back-extraction from the organic phase. The loss has been shown to be small enough and consistent enough to permit a bias correction to be applied with confidence.

The final step in the neptunium separation is the extraction of neptunium(IV) from the reducing strip solution into 0.5 M TTA in xylene. This organic phase, as previously reported,²⁴ was used directly to prepare the neptunium alpha-counting plate for radio-metric assay.

5.2.3. Cesium

Cesium was separated satisfactorily by the sequential separation described in section 5.2.1; however, the level of ^{137}Cs activity was greater than desirable. Separate aliquots of the dissolver solutions were processed by the ASTM perchlorate method.²⁷

5.3. Measurement Techniques

Most of the analyses were made by isotope dilution mass spectrometry or by the measurement of alpha or gamma radioactivity. The high specific activity of spent nuclear fuels and the accompanying hazards to

personnel make it desirable to work with the smallest sample aliquot consistent with the required accuracy and the capabilities of the measurement technique. The preferred technique for high-accuracy measurement of individual stable and long-lived isotopes is isotope dilution mass spectrometry, in which microgram samples are adequate for many elements. However, when using an enriched isotope of the same element, the concentration of the enriched isotope should approximate that of the sample, and it must be accurately known. Because chemical methods of analysis are not sufficiently reliable at these very low concentrations, it is necessary to prepare standardized solutions of the natural element at the milligram per gram level from high-purity material. The standard solutions are diluted accurately by weight to the microgram per gram level. The enriched isotope solutions, at the microgram per gram level, are then calibrated against the diluted natural element solutions by mass spectrometry. Spike calibrations are described below in the discussion of measurement techniques for individual elements.

Alpha radioactivity was measured using a silicon surface-barrier detector, capable of better than 30 keV resolution (FWHM), and a 400-channel pulse height analyzer. A single standard is satisfactory for alpha counting since detection efficiency is not a function of the alpha-particle energy. Two standard sources were used for this program: ^{238}Pu and ^{239}Pu . The activity of each of these sources was known to $\pm 1.5\%$. The gamma activity measurements and standards are described in the sections below.

5.3.1. Thorium

Thorium is determined by ethylenediaminetetraacetic acid (EDTA) titration. Fluoride is first removed by heating to dense fumes of HClO_4 . The solution is cooled, diluted with water, and adjusted to pH 2.5 with an ammonium hydroxide-ammonium chloroacetate buffer. An excess of 0.01 M EDTA is added. A 0.01 M thorium nitrate solution is used to back-titrate to a methylthymol blue endpoint.

A commercial analytical reagent solution of 0.01 M EDTA was used. The standardization was verified by titrating with a standard zinc solution which had been prepared from 99.999% zinc metal on a

weight basis. The standard thorium solution was prepared from ignited ThO_2 and was cross-calibrated with the standard EDTA solution.

5.3.2. Uranium

Uranium analyses were made by both isotope dilution mass spectrometry and alpha spectrometry. The amounts of ^{233}U , ^{234}U , ^{235}U , ^{236}U , and ^{238}U were determined mass spectrometrically. Depleted ^{238}U was used as the isotopic spike. A standard solution of the natural element was prepared from NBS 950a U_3O_8 after repeated ignition at 900 C to ensure stoichiometry. This material was used to calibrate a solution of 98% ^{233}U , which in turn was used to calibrate the 99.96% depleted ^{238}U to be used in this work. The ratio of ^{232}U to the sum of ^{233}U and ^{234}U was determined by alpha spectrometry.

5.3.3. Neptunium

The only isotope of neptunium present in detectable amounts was ^{237}Np . This isotope was determined by alpha spectrometry.

5.3.4. Plutonium

Isotope dilution mass spectrometry was the measurement technique used to determine ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu . The spike solution was prepared from a sample of 99.88% ^{242}Pu and calibrated against a standard solution prepared from NBS 949a plutonium metal standard. A small amount of ^{236}Pu was present in the fuel, and the ratio of this isotope to ^{238}Pu was measured by alpha spectrometry.

5.3.5. Americium-Curium

The combined americium-curium fraction was analyzed by alpha spectrometry.

5.3.6. Cesium

The separated cesium fraction was analyzed by gamma spectrometry using a sodium iodide (TI) detector. The interference due to ^{134}Cs was removed by subtracting a pure ^{134}Cs spectrum from the unknown spectrum until the 0.8 MeV ^{134}Cs peak was removed. The ^{137}Cs photopeak counts in the resolved spectrum were compared with those in spectra from standard ^{137}Cs sources to obtain the ^{137}Cs activity in the

sample. The standard sources were prepared from Nuclear Chicago Standard B-142.

After the separated cesium samples had been analyzed, aliquots of the dissolver solutions were analyzed, without a separation, using a lithium-drifted germanium detector. The much better resolution of this detector obviates the need for a separation. Essentially identical results were obtained by the two methods.

5.3.7. Neodymium

In this program only ^{143}Nd and ^{148}Nd were to be determined. These isotopes were measured by isotope dilution mass spectrometry using a ^{150}Nd spike solution.

A standard solution was prepared from 99.8% natural element oxide after repeated ignition at 900 C to insure stoichiometry. This solution was additionally standardized by ethylenediaminetetraacetic acid (EDTA) titrimetry. The diluted natural element solution was used to calibrate the 96.0% enriched ^{150}Nd used in this work.

The amount of ^{142}Nd also was measured to ascertain the contribution of nonfission product neodymium contribution. Although the separation procedure gave adequate removal of other lanthanides, the possible interference of cerium and samarium was checked by measuring the mass 140 and 154 peaks. No problem with cerium and samarium was found, nor was there any significant amount of nonfission product neodymium in the samples.

5.3.8. Promethium

Promethium-147 was determined by measuring the bremsstrahlung radiation accompanying its beta decay using a sodium iodide well crystal. The absence of other gamma activity was verified by gamma spectrometry. The activity of the unknown samples was compared with a standard source prepared from a standardized solution of ^{147}Pm obtained from The Radiochemical Center, United Kingdom Atomic Energy Authority.

5.3.9. Samarium

The samarium fraction was analyzed by isotope dilution mass spectrometry. The isotopes determined were ^{148}Sm , ^{149}Sm , and

^{150}Sm . The mass peaks at 142 and 143 were also measured to ensure that no neodymium was present. A spike solution was prepared from 94.5% ^{144}Sm and calibrated against a standard solution prepared from a sample of the 99.99% natural element oxide after repeated ignition at 900 C to ensure stoichiometry.

5.4. Results

5.4.1. Sample Location

The locations of samples taken for isotopic analysis are shown in Figures 5-11, 5-12 and 5-13. These figures are representations of gamma scans of the fuel rods and indicate the relative activity at the various sample locations. These figures do not include many of the details of the gamma scans, such as activity from ferrules and spacers, voids, and cracked and misplaced pellets. Detailed gamma scans are shown in BAW-3809-6.²⁸

5.4.2. Isotopic Concentrations

Experimental values of isotopic concentrations are listed in Tables 5-6 through 5-10. The values are expressed as postirradiation atoms per 10^5 preirradiation atoms of uranium plus thorium. Uranium and plutonium isotopic compositions (atom percent) are also listed in Tables 5-11, 5-12 and 5-13.

In general the low enrichment rods, type A, showed the most drastic change in uranium composition from the preirradiation values, since these rods were exposed to higher neutron fluxes. The different uranium contents were used to flatten the power distribution.

The uranium results show a large depletion in ^{235}U accompanied by an increase in ^{233}U , ^{234}U and ^{236}U . The transmutation of ^{238}U was small; a change of about 10% of the initial concentration was the greatest found. More than 75% of the initial ^{235}U was burned out of rod 20-A, resulting in a 236/235 ratio greater than 0.5. In several rods the most abundant isotope of uranium was ^{233}U , which accounted for more than 35% of the fissions in rods 20-A and 66-A.

The ratio of ^{238}Pu to ^{239}Pu is different than that usually found in power reactor fuel. This is due to the high enrichment of ^{235}U in this core. The source of ^{238}Pu is ^{235}U via ^{236}U and ^{237}Np , and the

source of ^{239}Pu is ^{238}U . The ^{235}U content in this fuel is typical of power reactor fuel, and the amount of ^{238}Pu is not greatly different from that found in fuel from other power reactors. The ^{238}U content is less than 1% of this fuel, whereas a ^{238}U content greater than 95% is typical of power reactor fuel. Thus, the amount of ^{239}Pu and higher plutonium isotopes would be expected to be only a small fraction of that usually found. The determination of ^{236}Pu was complicated by the low concentration resulting in a very low counting rate. The uncertainty at the 95% confidence level is estimated to be 20%.

The values for ^{237}Np also include an appreciable uncertainty. The amount of ^{237}Np in the fuel was not extremely small, but because of the long half-life, the disintegration rate was low. The amount of ^{237}Np is consistent with the ^{238}Pu found.

Approximately three years elapsed between reactor shutdown and analysis. This resulted in appreciable decay of ^{241}Pu to ^{241}Am . This correction amounted to about 90% of the ^{241}Am found at the time of analysis. Thus, the amount produced during reactor operation is the difference between two large numbers. A similar problem was encountered with the ^{242}Cm results. Approximately seven half-lives had passed since reactor shutdown, and any error in the half-life of ^{242}Cm contributes a five-fold greater error in the decay factor for ^{242}Cm .

The fission product concentrations are given in Table 5-10, but the results are perhaps more illustrative when presented as atoms per 100 fissions (Table 5-14) and as the ratio of this last value (atoms per 100 fissions) to the fission yield (Table 5-15).

The data shown in Tables 5-14 and 5-15 indicate that only ^{137}Cs and ^{148}Nd show concentrations that are consistent with the fission yields in all samples. This constancy is one of the reasons that these two nuclides are commonly used as fission monitors. The variations for the other nuclides are due to production or loss by neutron capture. The large capture cross sections for ^{143}Nd , ^{147}Pm , $^{148}\text{Pm}^{\text{m}}$, ^{149}Sm , and ^{150}Sm can be used to explain these results, at least qualitatively. The neutron flux is not well known, either in intensity or spectral distribution, but relative values can be obtained from the calculations in section 4 (see Tables 4-4 and 4-12) and from the ratio of the ^{235}U fissions to the ^{235}U content. The relative flux values from these two sources (Table

5-16) give an indication of the relative importance of the capture processes in each sample.

In Table 5-15 the fission product concentrations are listed as the ratio of atoms per 100 fissions to the percentage fission yield. If production or depletion by neutron capture processes is not important, this ratio will be approximately unity. The ratios for ^{137}Cs and ^{148}Nd are approximately unity for all samples. The burnout of ^{143}Nd is evidenced by the low ratios, and the trend is consistent with the relative neutron flux.

The relationship of ^{147}Pm and the samarium isotopes is more complex. Promethium-147 and ^{149}Sm are fission products, but ^{148}Sm and ^{150}Sm are not. Promethium-147 is a relatively long-lived fission product and has an appreciable capture cross section to produce $^{148}\text{Pm}^{\text{m}}$ and $^{148}\text{Pm}^{\text{g}}$. The half-life of $^{148}\text{Pm}^{\text{m}}$ is fairly long (41 d), and the capture cross section is quite large; thus, there will be competition between the processes of isomeric transition to $^{148}\text{Pm}^{\text{g}}$ and neutron capture to form ^{149}Pm . Although $^{148}\text{Pm}^{\text{g}}$ has a large cross section, the half-life is only 5.4 days, and beta decay to ^{148}Sm is the principal removal process. Similarly, the predominant process for ^{149}Pm is beta decay to ^{149}Sm . Samarium-149 also is a fission product. This isotope has a very large cross section (4×10^4 b) and is largely transmuted to ^{150}Sm . Thus the ^{147}Pm formed as a result of fission may appear as ^{147}Pm , ^{148}Sm , ^{149}Sm , and ^{150}Sm . Fission product ^{149}Sm will appear as ^{149}Sm , ^{150}Sm , and possibly ^{151}Sm .

Our results show that for rods exposed to a relatively high flux, some of the ^{147}Pm is transmuted to ^{150}Sm . In rods exposed to a lower flux, the isomeric transition of $^{148}\text{Pm}^{\text{m}}$ prevails over neutron capture, and the sum of ^{147}Pm and ^{148}Sm is equivalent to the fission yield of ^{147}Pm .

5.4.3. Burnup

In this fuel, both ^{233}U and ^{235}U contributed a significant number of fissions. A small contribution came from ^{232}Th and ^{239}Pu . The burnup was evaluated in three ways: change in the actinide isotopic composition, production of fission product ^{148}Nd , and production of fission product ^{137}Cs .

The number of ^{235}U fissions can be calculated from the uranium isotopic composition in several ways.²⁹ When the preirradiation isotopic composition of the uranium is well known, the change in the number of ^{235}U and ^{236}U atoms can be used to indicate the number of fissions. Alternately the change in either the ^{235}U or ^{236}U content and the capture-to-fission ratio for ^{235}U can be used to calculate the fissions. Frequently the isotopic content of each rod is not well known, and often neither is the value of the capture-to-fission ratio. In this study, the number of fissions of ^{235}U were calculated using the change from the preirradiation content and from the change in ^{235}U and ^{236}U and the values for capture-to-fission given in Table 4-13. Agreement between the two methods was good.

The number of ^{233}U fissions was calculated as the ratio of ^{234}U produced during irradiation to the capture-to-fission ratio of ^{233}U . Since the change in ^{234}U content was at most four times the initial value, both post- and preirradiation ^{234}U content must be accurately known. The values of capture-to-fission ratio are given in Table 4-13.

Fissions of ^{232}Th constituted about 1% of the total and were calculated from the depletion of ^{232}Th (the sum of ^{233}U , ^{234}U , and fissions of ^{233}U) and the capture-to-fission ratio of ^{232}Th . The ^{239}Pu fissions made a very small contribution and were calculated using an estimated value for capture-to-fission ratio.

The fission product most commonly used as a fission monitor is ^{148}Nd . For ^{235}U and ^{239}Pu fissions the fission yields are very similar, but the yields are significantly different from ^{235}U and ^{233}U fissions. It was necessary to calculate the fissions of ^{235}U and ^{233}U by the heavy element method to obtain a weighted fission yield. The agreement in the literature values for ^{148}Nd fission yields is good,³⁰ and the values used are 1.68 ± 0.03 for ^{235}U and 1.31 ± 0.04 from ^{233}U .

Neodymium is not usually found as an impurity in uranium; however, lanthanide impurities are found in thorium. Significant impurity levels of ^{148}Nd would complicate the burnup determination. The amount of ^{142}Nd , which is not a fission product, was measured to investigate this problem. There was no significant amount of nonfission product neodymium in the samples.

Another fission product frequently used as a burnup monitor is ^{137}Cs ; however, ^{137}Cs has been shown to redistribute in the fuel. It was felt that this would not be a significant problem in this study since the maximum heat rating of the fuel was only about 11 kW/ft. The fission yield of ^{137}Cs also is appreciably different for ^{235}U and ^{233}U , although the difference is considerably less than for ^{148}Nd . The ^{137}Cs fission yield from ^{235}U is quite well established as $6.22 \pm 0.07\%$, but the yield from ^{233}U is less certain. Values range from 6.02 to 7.33,^{30,31} and the mean value is 6.68 ± 0.21 .

The burnup values by the three methods are listed in Tables 5-17 and 5-18. There is no apparent bias between the three sets of values, and the agreement between the methods for all of the rods is good.

Burnups range from 6 to 30 GWd/T for the midplane positions of rods containing uranium and 2 GWd/T for the ThO_2 portion of the F rod. The burnup values for the other positions correlate fairly well with the gamma scan data. Values in Tables 5-17 and 5-18 can be converted to GWd/T by multiplying by 0.0098.

Table 5-1. Final pH of Various Ammonium Glycolate Buffer — 1N Nitric Acid Mixtures^(a)

<u>1N HNO₃: buffer volume ratio^(a)</u>	<u>Final pH</u>
1.0	4.2
1.5	3.9
2.0	3.7
2.5	3.5
3.0	3.3
3.5	3.1
4.0	2.8
4.5	2.4

(a) 5 M ammonium glycolate adjusted to pH 5 with ammonium hydroxide.

Table 5-2. Distribution of ²³⁹Np in Cation Exchange Column Effluent (%) in Five Replicate Separations

<u>No. ml</u>						<u>Total ml</u>
<u>1 - 3</u>	<u>4 - 6</u>	<u>7 - 9</u>	<u>10 - 12</u>	<u>13 - 15</u>	<u>15 - 18</u>	
0.14	92.4	5.2	0.06	0.02	0.01	97.8
0.03	90.1	3.5	0.01	-0.01	-0.03	93.6
0.22	86.0	10.2	0.07	-0.05	0.02	96.5
0.06	88.4	5.1	0.01	-0.05	-0.05	93.5
0.06	87.7	7.6	-0.03	0.02	-0.04	95.4

Table 5-3. Reproducibility of Recovery of Preheated ²³⁹Np From Cation Exchange Separation (%)

<u>Pretreated with KMnO₄</u>	<u>Pretreated with KMnO₄, then NaNO₂</u>
97.7	96.6
96.6	95.2
96.3	95.9
95.2	95.5
92.2	96.1
97.2	95.5

Table 5-4. Extraction (%) of $^{239}\text{Np(VI)}$ From Hydrochloric Acid into Various Tetraheptylammonium Chloride-Diluent Mixtures

Tetraheptylammonium chloride concentration	Diluent			
	MIBK		Nitropropane	
	0.25 <u>M</u>	0.5 <u>M</u>	0.25 <u>M</u>	0.5 <u>M</u>
4 <u>N</u> HCl	76	89	98.9	99.6
5 <u>N</u> HCl	99.6	99.8	--	--
6 <u>N</u> HCl	99.9	99.8	99.8	99.9
8 <u>N</u> HCl	--	--	99.9	99.7

Table 5-5. Distribution (%) of ^{239}Np in Solvent Extraction Procedure^(a)

Not extracted from 5 <u>N</u> HCl-KClO ₃	0.33 ± 0.05
Removed in AlCl ₃ scrub	0.12 ± 0.04
Remaining in organic phase after FeCl ₂ -NH ₂ OH strip	0.72 ± 0.03
Recovery in final strip phase	98.88 ± 0.05

(a) Based on six replicates.

Table 5-6. Experimental Values for Uranium Isotopic Concentrations^(a) at Core Midplane Positions

<u>Rod</u>	<u>²³²U</u>	<u>²³³U</u>	<u>²³⁴U</u>	<u>²³⁵U</u>	<u>²³⁶U</u>	<u>²³⁸U</u>
20-A	2.4	1430	167	646	381	154
61-A	1.7	1140	130	896	331	155
62-A	2.2	1170	132	777	349	154
66-A	2.2	1260	151	724	367	154
105-A	1.8	1160	126	877	338	155
105-B	1.8	1160	130	1400	485	222
105-C	0.6	574	66	3680	367	306
105-D	0.7	592	94	5700	406	424
105-D ₂	0.5	590	95	5760	392	424
105-E	0.7	563	105	7040	448	526
105-F	0.6	720	33	2	--	--
106-A	0.2	337	36	2220	128	166
106-B	0.3	437	52	3290	185	236

(a) Expressed as atoms per 10⁵ preirradiation atoms of thorium plus uranium.

Table 5-7. Experimental Values for Uranium Isotopic Concentrations^(a) at Low Flux Positions^(b)

<u>Rod</u>	<u>²³²U</u>	<u>²³³U</u>	<u>²³⁴U</u>	<u>²³⁵U</u>	<u>²³⁶U</u>	<u>²³⁸U</u>
20-A	0.78	864	63	1395	259	160
61-A	0.53	705	55	1680	213	162
62-A	1.2	1040	97	1100	315	158
66-A	1.2	972	87	1170	295	158
105-A	1.3	1030	93	1120	305	158
105-B	1.2	915	91	1830	390	225
105-C	0.40	476	61	4160	248	311
105-D ₁	0.41	467	88	6250	310	430
105-F	0.64	691	52	1990	192	164
106-A	0.17	326	35	2340	103	166
106-B	0.14	312	47	3480	133	237

(a) Expressed as atoms per 10⁵ preirradiation atoms of thorium plus uranium.

(b) Sample locations shown in Figures 5-11, 5-12, and 5-13.

Table 5-8. Experimental Values for Plutonium Isotopic Concentrations^(a) at Core Midplane Positions

<u>Rod</u>	<u>²³⁶Pu</u>	<u>²³⁸Pu</u>	<u>²³⁹Pu</u>	<u>²⁴⁰Pu</u>	<u>²⁴¹Pu</u>	<u>²⁴²Pu</u>
20-A	2×10^{-4}	4.9	4.1	1.4	1.1	0.52
61-A	1×10^{-4}	3.4	3.8	1.2	0.78	0.36
62-A	1×10^{-4}	4.0	3.9	1.4	1.1	0.42
66-A	2×10^{-4}	4.0	3.7	1.2	1.0	0.42
105-A	7×10^{-5}	3.0	3.7	1.2	0.90	0.32
105-B	5×10^{-5}	4.6	6.4	2.0	1.6	0.52
105-C	1×10^{-4}	0.83	9.1	1.3	0.69	0.05
105-D ₁	1×10^{-5}	0.90	12.7	1.5	0.83	0.05
105-E	1×10^{-5}	1.1	17.0	1.9	1.0	0.06

(a) Expressed as atoms per 10^5 preirradiation atoms of thorium plus uranium.

Table 5-9. Experimental Values for Isotopic Concentrations^(a) of ²³⁷Np, ²⁴¹Am, ²⁴²Cm and ²⁴⁴Cm at Core Midplane Positions

<u>Rod</u>	<u>²³⁷Np</u>	<u>²⁴¹Am</u>	<u>²⁴²Cm</u>	<u>²⁴⁴Cm</u>
61-A	12	0.014	0.007	0.005
105-A	15	0.030	0.009	0.005
105-B	18	0.017	0.01	0.006
105-C	--	0.001	0.0007	0.00007
105-D ₁	--	0.003	0.002	0.0002
105-E	--	0.006	0.002	0.0002

(a) Expressed as atoms per 10^5 preirradiation atoms of thorium plus uranium.

Table 5-10. Experimental Values for Fission Product Concentrations^(a) at Core Midplane Positions

<u>Rod</u>	<u>¹³⁷Cs</u>	<u>¹⁴³Nd</u>	<u>¹⁴⁸Nd</u>	<u>¹⁴⁷Pm</u>	<u>¹⁴⁸Sm</u>	<u>¹⁴⁹Sm</u>	<u>¹⁵⁰Sm</u>
20-A	193	120	46.4	--	--	--	--
61-A	158	102	38.0	33.2	7.8	0.27	26.2
62-A	167	110	40.2	--	--	--	--
66-A	180	116	43.2	--	--	--	--
105-A	157	105	38.3	33.3	11.8	0.29	26.1
105-B	190	137	48.2	41.3	13.7	0.38	31.7
105-C	84	76	22.0	23.2	3.9	0.45	13.3
105-D ₁	98	87	25.8	28.5	4.9	0.57	16.1
105-D ₂	--	93	25.5	--	--	--	--
105-E	108	96	28.8	31.3	5.7	0.80	17.4
105-F	14.8	11.1	2.82	--	--	--	--
106-A	38.3	33	9.8	--	--	--	--
106-B	51.4	38	13.2	--	--	--	--

(a) Expressed as atoms per 10⁵ preirradiation atoms of thorium plus uranium.

Table 5-11. Atom Percent Composition of Uranium Isotopes
at Core Midplane Positions

<u>Rod</u>	<u>^{233}U</u>	<u>^{234}U</u>	<u>^{235}U</u>	<u>^{236}U</u>	<u>^{238}U</u>
20-A	49.8	6.2	24.2	14.1	5.8
61-A	43.1	4.9	33.7	12.5	5.8
62-A	45.5	5.1	30.0	13.5	5.9
66-A	47.4	5.7	27.3	13.8	5.8
105-A	43.7	4.8	33.0	12.7	5.8
105-B	34.2	3.8	41.2	14.2	6.6
105-C	11.6	1.3	74.7	6.2	6.2
105-D ₁	8.2	1.3	79.0	5.6	5.9
105-D ₂	8.1	1.3	79.4	5.4	5.8
105-E	6.5	1.2	81.1	5.1	6.1
105-F	95.4	4.4	0.2	--	--
106-A	11.6	1.2	76.9	4.4	5.8
106-B	11.4	1.2	78.3	4.4	5.7

Table 5-12. Atom Percent Composition of Uranium Isotopes
at Low Flux Positions (a)

<u>Rod</u>	<u>^{233}U</u>	<u>^{234}U</u>	<u>^{235}U</u>	<u>^{236}U</u>	<u>^{238}U</u>
20-A	31.6	2.3	50.9	9.4	5.8
61-A	25.0	2.0	59.6	7.6	5.8
62-A	38.4	3.5	40.5	11.6	6.0
66-A	36.3	3.2	43.6	11.0	5.9
105-A	37.9	3.5	41.5	11.3	5.8
105-B	26.5	2.7	53.0	11.3	6.4
105-C	9.0	1.2	79.1	4.7	5.9
105-D ₁	6.1	1.2	81.9	4.1	5.7
105-F	22.3	1.7	64.5	6.2	5.3
106-A	11.0	1.2	78.4	3.5	5.9
106-B	7.4	1.1	82.7	3.2	5.6

(a) Sample locations shown in Figures 5-11, 5-12,
and 5-13.

Table 5-13. Atom Percent Composition of Plutonium Isotopes at Core Midplane Positions

<u>Rod</u>	<u>²³⁸Pu</u>	<u>²³⁹Pu</u>	<u>²⁴⁰Pu</u>	<u>²⁴¹Pu</u>	<u>²⁴²Pu</u>
20-A	40.5	34.0	11.7	9.5	4.3
61-A	34.9	39.3	12.6	9.5	3.7
62-A	36.8	36.5	12.7	10.2	3.8
66-A	38.5	35.6	12.0	9.8	4.1
105-A	33.0	40.4	13.1	10.1	3.4
105-B	30.4	42.7	13.2	10.3	3.4
105-C	6.9	76.0	10.6	6.0	0.5
105-D ₁	5.7	79.2	9.4	5.4	0.3
105-E	5.1	80.6	9.0	5.0	0.3

Table 5-14. Values of Fission Product Atoms per 100 Fissions at Core Midplane Positions

<u>Rod</u>	<u>¹³⁷Cs</u>	<u>¹⁴³Nd</u>	<u>¹⁴⁸Nd</u>	<u>¹⁴⁷Pm</u>	<u>¹⁴⁸Sm</u>	<u>¹⁴⁹Sm</u>	<u>¹⁵⁰Sm</u>
20-A	6.42	4.0	1.53	--	--	--	--
61-A	6.40	4.1	1.54	1.34	0.31	0.01	1.05
62-A	6.40	4.2	1.54	--	--	--	--
66-A	6.40	4.1	1.54	--	--	--	--
105-A	6.35	4.2	1.54	1.34	0.47	0.01	1.05
105-B	6.34	4.6	1.61	1.38	0.46	0.01	1.06
105-C	6.26	5.7	1.64	1.74	0.29	0.03	1.00
105-D ₁	6.33	5.6	1.66	1.83	0.31	0.04	1.04
105-E	6.25	5.6	1.67	1.81	0.29	0.05	1.01
105-F	6.7	5.0	1.3	--	--	--	--
106-A	6.48	5.6	1.66	--	--	--	--
106-B	6.42	4.7	1.65	--	--	--	--

Table 5-15. Ratios of Fission Product Atoms per 100
Fissions to the Fission Yield at Core
Midplane Positions

<u>Rod</u>	<u>^{137}Cs</u>	<u>^{143}Nd</u>	<u>^{148}Nd</u>	<u>^{147}Pm</u>	<u>$^{147}\text{Pm} + ^{148}\text{Sm}$</u>	<u>^{149}Sm</u>	<u>$^{149}\text{Sm} + ^{150}\text{Sm}$</u>
20-A	1.00	0.68	0.99	--	--	--	--
61-A	1.00	0.70	0.99	0.65	0.80	0.01	1.13
62-A	1.00	0.71	0.99	--	--	--	--
66-A	1.00	0.70	1.00	--	--	--	--
105-A	0.99	0.71	0.99	0.65	0.87	0.01	1.13
105-B	1.00	0.78	0.01	0.66	0.88	0.01	1.10
105-C	1.00	0.97	0.98	0.81	0.95	0.03	1.01
105-D ₁	1.01	0.95	1.00	0.86	1.0	0.04	1.03
105-E	1.00	0.95	0.99	0.84	0.98	0.05	1.03
105-F	1.0	0.8	1.0	--	--	--	--
106-A	1.03	0.95	0.99	--	--	--	--
106-B	1.03	0.80	0.99	--	--	--	--

Table 5-16. Relative Effective Thermal Flux

<u>Rod</u>	<u>Relative flux $\times 10^{-13}$</u>	
	<u>Calculated</u>	<u>Experimental</u>
20-A	0.45	0.47
61-A	0.41	0.36
62-A	0.39	0.38
66-A	0.41	0.41
105-A	0.36	0.36
105-B	0.30	0.31
105-C	0.14	0.11
105-D	0.12	0.09
105-E	0.10	0.08

Table 5-17. Burnup^(a) at Core Midplane Positions

<u>Rod</u>	<u>Heavy element</u>	<u>¹⁴⁸Nd</u>	<u>¹³⁷Cs</u>	<u>Average</u>
20-A	3050	3010	2990	3020
61-A	2490	2460	2460	2470
62-A	2620	2600	2610	2610
66-A	2830	2810	2800	2810
105-A	2480	2480	2470	2480
105-B	2980	3030	3000	3000
105-C	1340	1320	1350	1340
105-D ₁	1530	1550	1570	1550
105-D ₂	1490	1540	--	1520
105-E	1730	1720	1740	1730
105-F	230	220	220	220
106-A	590	580	610	590
106-B	790	790	820	800

(a) Burnup expressed as fissions per 10^5 pre-irradiation atoms of thorium plus uranium.

Table 5-18. Burnup^(a) at Low Flux Positions^(b)

<u>Rod</u>	<u>Heavy element</u>	<u>¹⁴⁸Nd</u>	<u>¹³⁷Cs</u>
20-A	1510	--	--
61-A	1210	--	1250
62-A	2060	--	--
66-A	1900	--	--
105-A	2000	--	2060
105-B	2230	--	--
105-C	1040	--	--
105-D ₁	1080	--	--
105-F	950	960	980
106-A	470	--	--
106-B	560	--	--

(a) Burnup expressed as fissions per 10⁵ preirradiation atoms of thorium plus uranium.

(b) Sample locations shown in Figures 5-11, 5-12, and 5-13.

Figure 5-1. Outline of Sequential Separation Procedure Adapted for Thoria-Urania Fuel

Figure 5-2. Extraction of Various Metal Ions into Di-(2-ethylhexyl)phosphoric Acid (D2EHP) ^{12, 13}

Figure 5-3. Separation of Lanthanides From Am-Cm¹⁹

Figure 5-4. Gamma Spectrum of Typical Dissolved Fuel Sample

Figure 5-5. Gamma Spectrum of Fission Products Retained on Initial D2EHP Column

Figure 5-6. Gamma Spectrum of Fission Products Retained on $\text{PbO}_2\text{-Pb}(\text{IO}_3)_2$ Column

Figure 5-7. Gamma Spectrum of Fission Products Retained on KCFC Column

Figure 5-8. Gamma Spectrum of Fission Products Retained on Final D2EHP Column

Figure 5-9. Cation Exchange Separation of Neptunium and Uranium (Dowex 50-X4)

Figure 5-10. Distribution of Fission Product Activity in Solvent Extraction Procedure for Neptunium

Figure 5-11. Idealized Gamma Scans Showing Sample Location for Isotopic Analysis — Rods 20-A, 61-A, 62-A, 66-A

Figure 5-12. Idealized Gamma Scans Showing Sample Locations for Isotopic Analysis — Rods 105-A, 105-B, 105-C, 105-D₁

Figure 5-13. Idealized Gamma Scans Showing Sample Locations for Isotopic Analysis — Rods 105-D₂, 105-E, 105-F, 106-A, 106-B

6. COMPARISON OF EXPERIMENTAL AND CALCULATED VALUES

All rods examined in this program were taken from the outer row of the bundles; as discussed in section 4, there are severe problems in assessing the effect of the water gap on the neutron spectrum. Type A rods were taken from the corners and thus are more subject to the moderating effect of the water gap.

Values for the isotopic composition were calculated for the core midplane positions and are given in Table 4-9 in units of $\text{barn}^{-1} \text{cm}^{-1}$ or atoms $\times 10^{-24}/\text{cc}$. The calculated and experimental values are shown in Tables 6-1 and 6-2 in units of atoms per 10^5 preirradiation atoms of uranium plus thorium. It is perhaps easier to see the overall comparison in Tables 6-3 and 6-4, where the results are shown as the ratio of experimental to calculated values. In general, the agreement is fair.

The worst agreement for uranium isotopes is for ^{235}U . In every case, the calculated value for ^{235}U is greater than the experimental value indicating that the fuel was exposed to a greater fluence than had been calculated or that the neutron spectrum, and thus the cross sections, was different from those used in the calculations. Also, the results for ^{234}U are not in good agreement. Here the calculated values are generally less than the experimental values. This trend is consistent with that for ^{235}U ; a greater change in fuel composition was found experimentally than had been calculated. The ^{233}U results agree fairly well, probably because of greater production accompanied by greater burnout.

When comparing the calculated and experimental results for uranium, there are apparently two groups: rods 20-A, 61-A, 62-A, 66-A, 105-A, and 105-B in one group, and 105-C, 1050, 1052, 106-A, and 106-B in the other. The first group of rods experienced higher fluxes than those in the second group.

The most serious disagreement between calculated and experimental values is for ^{238}Pu , where the experimental results are about 50% greater than the calculated results. Since ^{238}Pu results from three successive captures and the ^{237}Np cross section was not well established, the disagreement is not too surprising. The other plutonium nuclides show fairly good agreement, but the experimental values are generally higher than the calculated values.

Almost all of the experimental results indicate that the change in fuel composition was greater than was calculated, and thus the neutron flux levels were probably greater than calculated.

Table 6-1. Comparison of Calculated and Experimental Values of Uranium Isotopic Concentrations^(a) at Core Midplane Locations

<u>Rod</u>		<u>²³³U</u>	<u>²³⁴U</u>	<u>²³⁵U</u>	<u>²³⁶U</u>	<u>²³⁸U</u>
20-A	Experimental	1430	167	646	381	154
	Calculated	1250	123	947	344	153
61-A	Experimental	1140	130	896	331	155
	Calculated	1250	123	947	344	153
62-A	Experimental	1170	132	777	349	154
	Calculated	1220	116	999	336	155
66-A	Experimental	1260	151	724	367	154
	Calculated	1220	116	999	336	155
105-A	Experimental	1160	126	877	338	155
	Calculated	1130	101	1130	314	156
105-B	Experimental	1160	130	1400	485	222
	Calculated	1130	108	1770	435	223
105-C	Experimental	574	66	3680	367	306
	Calculated	755	79	4060	314	306
105-D ₁	Experimental	592	94	5700	417	424
	Calculated	645	92	5800	369	418
105-E	Experimental	563	105	7040	448	526
	Calculated	680	111	7240	469	520
105-F	Experimental	720	33	2	--	--
	Calculated	903	34	2	--	--
106-A	Experimental	337	36	2220	128	166
	Calculated	381	38	2330	111	166
106-B	Experimental	437	52	3290	185	236
	Calculated	381	51	3370	152	236

(a) Expressed as atoms per 10⁵ preirradiation atoms of thorium plus uranium.

Table 6-2. Comparison of Calculated and Experimental Values for Plutonium and ^{149}Sm Isotopic Composition^(a) at Core Midplane Locations

<u>Rod</u>		<u>^{238}Pu</u>	<u>^{239}Pu</u>	<u>^{240}Pu</u>	<u>^{241}Pu</u>	<u>^{242}Pu</u>	<u>^{149}Sm</u>
20-A	Experimental	4.9	4.1	1.4	1.1	0.52	--
	Calculated	3.0	3.5	1.4	1.0	0.37	--
61-A	Experimental	3.4	3.8	1.2	0.78	0.36	0.27
	Calculated	3.0	3.5	1.4	1.0	0.37	0.22
62-A	Experimental	4.0	3.9	1.4	1.1	0.42	--
	Calculated	2.7	3.5	1.4	1.0	0.33	--
66-A	Experimental	4.0	3.7	1.2	1.0	0.42	--
	Calculated	2.7	3.5	1.4	1.0	0.33	--
105-A	Experimental	3.0	3.7	1.2	0.90	0.32	0.29
	Calculated	2.1	3.6	1.3	0.81	0.25	0.22
105-B	Experimental	4.6	6.4	2.0	1.6	0.52	0.38
	Calculated	2.4	5.6	1.9	1.3	0.36	0.34
105-C	Experimental	--	9.1	1.3	0.69	0.05	0.45
	Calculated	--	9.4	1.2	0.68	0.05	0.65
105-D ₁	Experimental	--	12.7	1.5	0.83	0.05	0.57
	Calculated	--	11.6	1.4	0.68	0.04	0.93
105-E	Experimental	--	17.0	1.9	1.0	0.06	0.80
	Calculated	--	15.2	1.8	1.0	0.06	1.17

(a) Expressed as atoms per 10^5 preirradiation atoms of uranium plus thorium.

Table 6-3. Ratios of Experimental to Calculated Values of Uranium Isotopic Composition at Core Midplane Locations

<u>Rod</u>	<u>^{233}U</u>	<u>^{234}U</u>	<u>^{235}U</u>	<u>^{236}U</u>	<u>^{238}U</u>
20-A	1.15	1.36	0.68	1.11	1.01
61-A	0.92	1.06	0.95	0.96	1.01
62-A	0.96	1.14	0.78	1.04	0.99
66-A	1.04	1.30	0.72	1.09	0.99
105-A	1.02	1.25	0.77	1.08	0.99
105-B	1.03	1.20	0.79	1.11	1.00
105-C	0.76	0.84	0.90	1.17	1.00
105-D ₁	0.92	1.02	0.99	1.13	1.01
105-E	0.83	0.94	0.97	0.96	1.01
105-F	0.76	0.97	--	--	--
106-A	0.88	0.95	0.95	1.15	1.00
106-B	1.13	1.02	0.98	1.22	1.00

Table 6-4. Ratios of Experimental to Calculated Values of Plutonium and ^{149}Sm Isotopic Composition at Core Midplane Location

<u>Rod</u>	<u>^{238}Pu</u>	<u>^{239}Pu</u>	<u>^{240}Pu</u>	<u>^{241}Pu</u>	<u>^{242}Pu</u>	<u>^{149}Sm</u>
20-A	1.6	1.2	1.0	1.2	1.4	--
61-A	1.1	1.1	0.88	0.82	1.0	1.2
62-A	1.5	1.1	1.0	1.1	1.3	--
66-A	1.5	1.0	0.91	1.0	1.3	--
105-A	1.5	1.0	0.94	1.1	1.3	1.3
105-B	1.9	1.2	1.1	1.2	1.4	1.1
105-C	--	1.0	1.0	1.0	1	0.7
105-D ₁	--	1.1	1.1	1.2	1	0.6
105-E	--	1.1	1.0	1.0	1	0.7

APPENDIX
Sequential Separation Procedures

1. Scope and Principle

A four-column procedure for uranium, plutonium, americium-curium, cesium, neodymium, promethium, and samarium from irradiated nuclear fuel is described. Two extraction columns and two inorganic ion exchange columns are used in the sequential separation of these elements. The first extraction column, di-(2-ethylhexyl)phosphoric acid (D2EHP) in p-diisopropylbenzene on a Chromosorb support, retains thorium, uranium, plutonium, and zirconium from 1N nitric acid. The retained uranium and plutonium are sequentially eluted with 6N hydrochloric acid and 6N hydrochloric acid containing iodide as a later step in the procedure. The nitric acid effluent from the first column flows directly onto the second column of lead iodate-lead dioxide, which removes unwanted cerium and ruthenium. The effluent from the second column then passes through a third column of potassium cobaltoferrocyanide (KCFC), which quantitatively removes cerium. The effluent from the third column is then buffered to a higher pH to ensure the complete retention of cerium-free lanthanides and trivalent actinides on the final D2EHP extraction column. The americium-curium fraction is eluted from the final column with 1 M glycolic acid-0.016 M diethylenetriamine-pentaacetate, after which the lanthanides are individually eluted with dilute nitric acid.

2. Apparatus

All glassware is first cleaned with hot 8 M nitric acid to remove surface and acid-leachable impurities. The glassware is then rinsed with distilled water, oven-dried, and stored in a clean closed container.

- 2.1. Glass, 6 mm ID × 100 mm (six-inch Pasteur pipets serve nicely for this purpose after the tips are shortened.)
- 2.2. Glass, 6 mm ID × 250 mm. These are lengths of 6 mm-ID Pyrex tubing with a heat-drawn tapered tip.
- 2.3. Glass wool or quartz wool.
- 2.4. Test tubes, assorted sizes.
- 2.5. Conical tubes, 5-ml size.

- 2.6. Pipets, assorted Kirk-type glass micropipets and plastic disposable transfer pipets.

3. Reagents

- 3.1. Sodium nitrate solution, 0.1 M.
- 3.2. Ferric nitrate solution, 10^{-5} M in 2N nitric acid.
- 3.3. Boric acid, saturated solution.
- 3.4. Nitric acid; concentrated 8N, 5N, 1N and weight dilutions with water of 1:160, 1:125, 1:100, 1:80, and 1:50.
- 3.5. Hydrochloric acid, 6N and 6N-1% ammonium iodide (freshly prepared).
- 3.6. Chromosorb inert support material, acid and alcohol washed, siliconized (Analabs, Inc., North Haven, Connecticut).
- 3.7. Di-(2-ethylhexyl)phosphoric acid, (D2EHP).
- 3.8. Lead iodate, 60 to 80 mesh.
- 3.9. Lead dioxide, 60 to 80 mesh.
- 3.10. Potassium cobaltoferrocyanide, (KCFC) 50 to 100 mesh (Bio-Rad Laboratories, Richmond, California).
- 3.11. Para-diisopropylbenzene, (p-DIPB), (City Chemical Corporation, New York).
- 3.12. Tygon tubing, 1/4 inch ID by 1/16 inch wall thickness.
- 3.13. Ion exchange resin, Dowex 50-X4, 200 - 400 mesh (H⁺ form).
- 3.14. Buffer solution, 5 M glycolic acid adjusted to pH 5.0 with ammonium hydroxide.
- 3.15. Actinide eluant solution, 1 M glycolic acid-0.016 M pentasodium diethylenetriaminepentaacetate (Na₅DTPA).

4. Preparation of Column Materials

4.1. Extraction Columns

4.1.1. Purification of D2EHP

Heat the D2EHP with an equal volume of 8 M nitric acid under reflux for 2 hours to hydrolyze any pyrophosphates that may be present. Cool and transfer to a separatory funnel, discarding the aqueous phase. Dilute the D2EHP with a threefold volume of diethyl ether. Scrub the organic phase with an equal volume of 1 M sodium hydroxide for 2 minutes. Drain and discard the aqueous phase. Filter the organic phase into a clean separatory funnel to remove the hydrolyzed iron usually present as an impurity. Scrub the organic phase four more times with an equal volume of 1 M sodium carbonate, discarding the aqueous phase each time. Wash the organic phase once with 1N nitric acid, once with 0.1N nitric acid, and once with water. Drain the organic phase, as waterfree as possible, into a clean beaker. Partially cover the beaker and allow the ether to evaporate in a well-ventilated hood. Complete evaporation, normally requiring 2 to 3 days, is verified when constant weight is obtained for successive weighings at 4-hour intervals. Note: D2EHP must be freshly purified before each batch of column material is prepared.

4.1.2. Preparation of D2EHP Column Material

Take care not to transfer any of the water droplets that usually remain with the D2EHP after the ether evaporation is complete. Add 7.5g of p-DIPB to 5g of D2EHP and disperse both in sufficient chromatographic-grade methanol to completely immerse the 100g of Chromosorb that is added. Allow the methanol to evaporate in a well-ventilated hood until the material is visually dry. Final evaporation is conveniently accomplished by drawing air through the material in a Buchner funnel. Note: The prepared material has a shelf life of up to 4 months if it is completely dry, heat-sealed in plastic, and stored in a dark container.

4.2. Preparation of Lead Iodate-Lead Dioxide Column Material

Lead iodate and lead dioxide are sieved separately to obtain the 60 to 80 mesh fractions. These are blended on a $2\text{PbIO}_3:1\text{PbO}_2$ basis and mixed thoroughly.

4.3. Preparation of KCFC Column Material

The KCFC column material is air-dried of excess moisture and otherwise used as received.

5. Column Preparation

5.1. First Column (D2EHP)

Place a small plug of glass wool in the restriction of a 6 mm $\times$ 100 mm column. Prepare a water slurry of the D2EHP-p-DIPB-Chromosorb material. Pipet the slurry into the water-filled column until a settled bed height of 6 cm is attained. Wash the column with 5 ml of 8 M HNO_3 followed by 2 ml of 1N HNO_3 .

5.2. Second Column ($\text{PbIO}_3\text{-PbO}_2$)

Place a small plug of glass wool in the restriction of a 6 mm $\times$ 100 mm column. Prepare a water slurry of 2 $\text{PbIO}_3:1\text{PbO}_2$. Mix well and decant the fine particles several times. Pipet the slurry into the water-filled column until a settled bed height of 3 cm is attained. Wash the column with 3 ml of 1N nitric acid.

5.3. Third Column (KCFC)

Place a small plug of glass wool in the restriction of a 6 mm $\times$ 100 mm column. Prepare a water slurry of the air-dried KCFC. Pipet the slurry into the water-filled column until a settled bed height of 3 cm is attained. Wash the column with 5 ml of 1N nitric acid.

Note: The KCFC tends to decompose slightly during prolonged storage in acid and therefore should not be washed with acid until just before use.

5.4. Final Column (D2EHP)

Place a small plug of glass wool in the restriction of a 6 mm $\times$ 250 mm column. Prepare a water slurry of the D2EHP material. Pipet the slurry into the water-filled column until a settled bed height

of 15 cm is attained. The material should be settled by tapping on the column, if necessary, until 1 ml of water requires $2 \text{ min} \pm 5 \text{ seconds}$ to drain completely. Wash the column with 10 ml of 8N nitric acid followed by 5 ml of water.

5.5. Assembly of Columns

The first, second, and third columns are coupled with about 2-cm lengths of Tygon tubing as shown in Figure A-1, and are thereafter handled as a unit. A small portion of liquid must be left on columns 2 and 3 before coupling to ensure a smooth flow of liquid. Wash the 3-column unit with 5 ml of 1N nitric acid just before using.

6. Sample Handling

This procedure is designed for samples of irradiated nuclear fuel; because of the high levels of alpha, beta, and gamma activity associated with these samples, they must be handled only in well-ventilated hoods or glove boxes, by personnel wearing appropriate protective clothing.

7. Sample Pretreatment

Add the appropriate enriched isotopes required for the isotope dilution mass spectrometric analysis to a sample containing not more than 3 mg of total actinides. Adjust the nitric acid concentration of the sample to 2N, and add 0.1 ml of NaNO_2 and 0.1 ml of $10^{-5} \text{ M Fe(NO}_3)_3$. Mix thoroughly and heat under a heat lamp for 30 minutes to convert all plutonium to the tetravalent state. Cool and adjust the nitric acid concentration between 0.5 and 1N with water. If fluoride is present, add a 100-fold mole excess of boric acid to complex the fluoride.

8. Separation Procedure

- 8.1. Load the pretreated sample on the 3-column unit. Allow the liquid to drain to the top of the bed. Wash with five 1-ml portions of 1N nitric acid, using them first to rinse the pretreatment tube and thus ensure complete sample transfer. Collect all of the effluent from this step for step 8.2. Separate the columns, saving No. 1 for step 8.10 and No. 3 for cesium analysis.

- 8.2. Add 2 ml of the glycolic acid buffer solution to the 1N nitric acid effluent from the 3-column unit. Mix thoroughly and transfer this solution to the final (D2EHP) column. Rinse the container with 8 ml of 1:160 nitric acid to ensure a complete sample transfer. Discard all effluents from this step.
- 8.3. Elute the trivalent actinides with four 4-ml portions of the glycolic acid Na_5DPTA actinide eluting solution. Collect each portion separately and save for step 8.11.
- 8.4. Wash the final column with 8 ml of water, discarding the effluent.
- 8.5. Elute the final column with four 4-ml portions of 1:125 nitric acid. Collect each portion separately.
- 8.6. Elute the final column with four 4-ml portions of 1:100 nitric acid. Collect each portion separately.
- 8.7. Elute the final column with four 4-ml portions of 1:80 nitric acid. Collect each portion separately.
- 8.8. Elute the final column with four 4-ml portions of 1:50 nitric acid. Collect each portion separately.
- 8.9. Neodymium, promethium, and samarium will usually be found in the 1:100, 1:80, and 1:50 nitric acid, respectively. The exact position of each element in the various effluent portions may be verified by gamma spectrometry to locate activities of Pr, Pm, and Eu. Label the tubes containing the desired elements. Gently evaporate the solutions under a heat lamp until all of the portions for each element may be combined in a single tube. The elements to be mass analyzed are evaporated to dryness in a 5-ml conical tube and flamed lightly over a bunsen burner.
- 8.10. Elute uranium from the first column with 6 ml of 6N hydrochloric acid. Elute plutonium next with 6 ml of 6N hydrochloric acid—1% ammonium iodide. Evaporate to near dryness. Add 8 M nitric acid and repeat twice, transferring to a 5 ml conical tube for the final evaporation. Note: If ^{238}Pu

is present in sufficient quantity for mass spectrometric analysis, it may be necessary to further purify the plutonium fraction by anion exchange to remove ^{238}U .

- 8.11. If analysis of the trivalent actinides is planned, they can be separated readily from the organic salts present in the eluting solution by cation exchange. Add 0.1 ml of concentrated nitric acid to each 10 ml of actinide effluent from step 8.3. This reduces the pH from 2.53 to 1.5, from which the trivalent actinides are retained on a 6 mm $\times$ 35 mm column of Dowex 50-X4 (200 - 400 mesh) resin. Wash the column with 6 ml of water, discarding the effluent. Elute the trivalent actinides into a suitable vessel with 5 ml of 5N nitric acid.

Figure A-1. Three Columns Coupled for Separation

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C. Brassfield
J. McGurty
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17. E.I. du Pont de Nemours and Company (2)
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Attn: S. Rideout (2)
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18. Westinghouse Electric Corporation (3)
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Attn: T. Stern (2)
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19. Westinghouse Electric Corporation (5)
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20. Idaho Nuclear Corporation (2)
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21. Babcock & Wilcox (90)

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Lewis, RH
Library, NPGD (2)
Littrell, LW (2)
Lynch, ED
Marsh, SF (2)
McDaniel, GT
Moncrief, EC
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Rosenberger, GG
Schmitt, DH
Travis, CC/TRG
Vorres, KS/Alliance
Williams, RL
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